Sources and Effects of Errors in Vector Field Electron Tomography

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Abstract

Vector field electron tomography is a relatively new technique for quantitative three-dimensional imaging, combining phase retrieval with vector tomography to reconstruct electromagnetic fields, potentials, and sources, from transmission electron micrographs. Vector field electron tomography reconstructs electromagnetic vector fields (i.e., the vector potential, magnetic induction field, and current density) associated with magnetic nanomaterials, such as magnetic recording media, spintronics devices, grain boundaries in hard magnets, and magnetic particles for biomedical applications.

Although there is a range of techniques for characterising magnetic nanomaterials, such as Kerr microscopy, magnetic force microscopy, and Lorentz microscopy, these techniques only provide projections or surface components of the vector field. Vector field electron tomography takes projections of the potential obtained using electron holography, acquired over two or more tilt series, and reconstructs complete vector fields associated with the particle.

In the present work, we consider the reconstruction of the vector potential of magnetite nanoparticles, a material of significant interest in the study of nanomagnetism. This work addresses errors, both in the recorded micrographs and those incurred during the reconstruction process, and examines the effect that these errors have on the accuracy of the reconstructed vector field. We use simulated micrographs in order to have complete control over the nature of the errors, and to be able to precisely quantify the accuracy of the reconstruction.

In this thesis, we use a phase retrieval algorithm based on the transport-of-intensity equation, and reconstruct the vector potential of the simulated magnetite specimen using two tilt series and a filtered backprojection algorithm. We use three different root-mean-square error metrics to determine the accuracy of the reconstruction in terms of the total vector difference, the difference in magnitude, and the difference in direction. We then compare these results with analytical predictions.

We derive expressions to predict the error in a vector field electron tomography
reconstruction as functions of image noise and initial specimen orientation, and test the applicability of these expressions under a range of conditions. There is strong, quantitative agreement between our analytical and numerical results regarding the effects of image noise. Our work on orientation-dependent errors provides a semi-quantitative analysis of the total root-mean-square error as a function of magnetic moment orientation. We also present a method for reducing orientation-dependent errors by averaging reconstructions from multiple pairs of tilt series. We find that the reconstruction of the magnetic vector potential of uniformly magnetised specimens can be significantly improved by distributing the acquired images over additional tilt series.
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Declaration of Originality

This thesis contains no material which has been accepted for the award of any other degree or diploma at any university or equivalent institution and, to the best of my knowledge and belief, this thesis contains no material previously published or written by another person, except where due reference is made in the text of the thesis.

Signed,

Zachary David Cleary Kemp

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Chapter 1

Introduction

1.1 Motivation and research goals

Magnetic nanomaterials are a critically important feature of twenty-first century technology and a crucial aspect of a vast range of topical areas of research, including spintronics [1, 2]; biomedical nanotechnology [3–6]; magnetic information storage [7, 8]; the study of shape memory alloys [9], magnetic vortices [10], and nanoscale grain boundaries in hard magnets [11]; and the development of nanomotors [12].

In order to fully understand these materials, it is essential to have techniques to accurately characterise their magnetic properties at the nanoscale. There are many experimental techniques that have been developed for this purpose [13], such as Kerr microscopy [14], magnetic exchange force microscopy [15], and spin polarised scanning tunneling microscopy [16], each of which can be employed to examine components of the magnetisation on the surface of the sample. Lorentz transmission electron microscopy (LTEM) [17] has significant advantages over these techniques in that it can be used to probe the internal magnetic structure of a sample [18]. Using LTEM, the phase of the electron wavefunction at the exit surface of the magnetic specimen can be measured from out-of-focus micrographs or electron holograms, but this provides only two-dimensional projections of the potentials.

Tomography reconstructs an object function from its projections. This relies on the principle of the projection-slice theorem [19] which, in three dimensions, states that the two-dimensional (2D) Fourier transform of a projection of a function is equal to a slice through the origin of the three-dimensional (3D) Fourier transform of the object [20, 21]. There are a large number of techniques that can utilise this principle to reconstruct the object function.

The simplest method for tomographic reconstruction is direct Fourier inversion, in which the Fourier transform of the object is constructed from the transformed
projections. This method results in errors due to interpolation of the projection data (which is in polar coordinates) onto Cartesian coordinates [22]. Because of the lower density of samples further away from the origin, in the polar representation, the interpolation results in errors that are greater at higher spatial frequencies. This results in degradation of the high spatial frequency components of the reconstruction [23].

Backprojection algorithms, as the name implies, reproject the acquired images back into the reconstruction domain [24]. A simple, unfiltered backprojection causes erroneous non-zero values to occur outside the object function [25]. Because the projections are not negative anywhere (in conventional absorption contrast tomography), additional projections do not remove these artefacts. Filtered backprojection (FBP) corrects this problem by high pass filtering the projections prior to backprojection [24] using a cropped ramp filter, known as the Ram-Lak filter [26,27]. However, reconstructions that utilise FBP can result in streaking artefacts [28]. These artefacts can be reduced by combining the Ram-Lak filter with a low pass filter [26], at the expense of contrast, or by including additional tilt angles in the reconstruction [29].

Iterative techniques [30, 31] can produce improved reconstructions, particularly when the angular sampling is sparse [32], but this comes at a computational cost [33]. This class of techniques includes simultaneous iterative reconstruction techniques, commonly used in seismic tomography [34], and so-called algebraic reconstruction techniques [35, 36]. They begin with an estimate for the object function, typically obtained using an FBP algorithm. At each iteration, projections of this estimate are compared with the experimental projections, and the estimate is corrected based on the differences between these two projections.

Conventional scalar computed tomography (CT) has been employed for decades as a means to probe the internal structure of 3D objects [37,38]. X-ray computed tomography and positron emission tomography are routinely used as medical diagnostic tools, but scalar tomography has also proved extremely useful in a diverse range of other fields. Examples include the imaging of microscopic biological samples using synchrotron radiation [39], structural characterisation of semiconductor devices using scanning transmission electron microscopy [40], 3D imaging of binary stars using Doppler measurements [41], delineation of magma bodies using seismic tomography [42], 3D imaging of macroscopic mechanical parts using neutron tomography [43], and a wide range of non-destructive techniques using X-ray tomography [44–46].

Initially developed for the 3D imaging of biological specimens [47,48] using the transmission electron microscope (TEM), electron tomography has now been adopted for use by materials scientists [49,50]. Examples of its use in this domain are the
imaging of the electrostatic potential of p-n junctions [51], integrated circuits [52], and carbon nanotubes [53].

One well-known limitation [54–58] of electron tomography is the restricted tilt range typically accessible in experiments, which results in a missing wedge of information in the Fourier transform of the reconstruction. Recently, full ±90° rotation has been achieved in scalar electron tomography [56, 59]. However, the projected thickness of a thin specimen can increase with increasing tilt angle, and this causes a decrease in the transmission of electrons, which adversely affects the tomographic reconstruction [59, 60]. Thus, acquiring this full range of images places strong constraints on the specimen geometry. Additionally, the sample holder and supporting grid obscure the electron beam at high tilt angles, requiring that modifications are made for acquisition of the full tilt range [59].

The theory of scalar tomography has been extended to the reconstruction of vector fields [61]. Vector field tomography (VFT) reconstructs vector fields from projection data. The majority of the work on VFT relates to the use of acoustic time of flight or Doppler measurements to reconstruct fluid flows. Such applications include the imaging of blood flow [62], flue gas velocity in coal fired power stations [63], and water velocity in lakes [64]. Recently, VFT has been shown to be effective in the reconstruction of electric fields arising from brain activity, using electroencephalographic measurements [65].

Electron holography [66] reconstructs the complete electron wavefunction at the image plane. Early electron holography utilised interference between the object wave and a coaxial reference wave [67–69]. The object wave interacts with the specimen, and the reference wave continues unimpeded. Interference between the two waves, at the detector, is used to infer the phase of the object wave that results from interactions with the specimen potential. A more recent method, known as off-axis electron holography [70], utilises an electron biprism to split the electron beam into the two coherent waves [71]. The reference wave in off-axis holography is tilted with respect to the optic axis, and overlaps the object wave at a slightly defocussed image plane to produce the hologram [72].

Phase retrieval is the process of reconstructing the phase of the exit surface electron wavefunction. Through-focal methods for phase retrieval typically require the acquisition of at least two images at different planes of defocus [73], although a single image can be used if the specimen is sufficiently homogeneous [74, 75]. Iterative methods, such as the Gerchberg-Saxton algorithm [76], are effective at reconstructing the phase, but can be computationally demanding. Recent through-focal phase retrieval algorithms make use of the transport-of-intensity equation (TOI), which relates longitudinal derivatives of the intensity of the electron wavefunction to trans-
verse derivatives of its phase \[77,78\]. In the present work, we use a method based on the TIE due to its simplicity and computational efficiency. One noteworthy drawback of this method is that the specimen must remain unchanged over the course of acquiring a through-focal series \[79\]. However, an unchanging specimen is also a requirement for the acquisition of tomographic tilt series, so this is not an additional constraint in our work.

Electron holographic tomography (EHT) \[51,80\] combines off-axis electron holography with electron tomography to reconstruct the electrostatic potential of a specimen from its projections. EHT has been utilised for the reconstruction of the electrostatic potential of semiconductor nanowires \[58\] and silicon p–n junctions \[81\]. Automation of the EHT process has been used to obtain measurements of the mean inner potential of a latex sphere and a needle-shaped germanium specimen \[82\].

Vector field electron tomography (VFET) utilises at least one additional tilt series to enable a complete, 3D magnetic characterisation of an electromagnetic specimen using the TEM \[83–86\]. VFET can be used to reconstruct the vector potential, magnetic induction, and current density of a magnetic nanoparticle at all points inside, and around, a sample \[83,87\]. VFET requires information of the phase shift induced in the electron beam as it traverses an electromagnetic sample. The exit phase of the beam is measured over two or more tilt series and is used to tomographically reconstruct the vector fields in three dimensions. This requires the phase to be reconstructed from intensity measurements, which can be performed using any of the holographic methods described in the previous paragraphs.

The theoretical foundations of VFET are well established, and the process has been employed to experimentally reconstruct electromagnetic vector fields associated with magnetic nanomaterials using both propagation based phase retrieval \[84, 88\] and off-axis EHT \[89, 90\]. However, there are numerous artefacts in these reconstructions and, while there are many known sources of error, including image noise, image misalignment \[91\], spatial sampling, and the missing wedge, the propagation of these errors through the reconstruction process is not well understood.

This thesis addresses the errors in magnetic vector potentials reconstructed using VFET. Specifically, our work considers errors in the reconstruction of the magnetic vector potential of nanoparticles using propagation based phase retrieval and an FBP reconstruction algorithm. Due to the complexity of VFET, there is a great number of errors that can be introduced at various stages of the image acquisition and reconstruction processes. A major aim of this work is to benchmark the accuracy of the FBP reconstruction algorithm for VFET under a range of conditions, and to enable accurate reconstructions of the vector potential of magnetic nanomaterials using the TEM. We use simulations to provide quantitative data on the effect that
errors have on VFET reconstructions, and compare our results with analytical models. Additionally, we present a reconstruction method that can reduce errors by utilising additional pairs of tilt series. A detailed outline of the thesis is given in Sec. 1.2.

1.2 Thesis outline

Chapter 2 details the theoretical foundations of VFET. We begin with an explanatory overview of the formation of images in LTEM, highlighting the way in which electromagnetic information is encoded in the phase of the electron beam, due to the Aharonov-Bohm effect, and how this information is transferred to the recorded electron micrographs. We then explore the methods by which this information can be recovered from these micrographs, with special attention paid to propagation based phase retrieval using the TIE, which is used for the remainder of this work. Then, we address what is arguably the core aspect of the entire VFET process: the tomographic reconstruction itself. For this work, we consider the reconstruction of the magnetic vector potential using an FBP algorithm. We also address regularisation of singularities in the reconstruction algorithms. The primary focus of this work is on the accuracy of VFET reconstructions. Concluding Ch. 2, we define error metrics that will be used throughout this work to quantify the differences between exact and reconstructed vector potentials.

Some significant sources of error in VFET are discussed in Ch. 3. Specifically, we address: shot noise, which is an unavoidable variation in image intensity that arises from the quantum mechanical nature of electrons; the truncation error that occurs due to the use of a finite difference approximation when calculating the through-focal derivative of the intensity from out-of-focus micrographs; diffraction artefacts, which are deviations from the mean-field projection approximation caused by scattering of the electrons by the crystal lattice of the specimen; the effects of spatial resolution, including that of the voxel grid on which the reconstruction is performed, as well as the spatial sampling of the tilt series in terms of both the number of pixels per image, and the number of images per tilt series.

The processes involved in the simulation of micrographs for VFET are described in Ch. 4. We discuss the simulation of specimens of uniform, and arbitrary, magnetisation. The calculation of defocussed micrographs, using both the projection approximation and the more sophisticated multislice method, are explained. A model for the inclusion of realistic image (shot) noise is also described in this chapter.

Chapter 5 reports on an in-depth study of the propagation of shot noise through the phase retrieval and tomographic reconstruction processes. Shot noise is present
in all transmission electron micrographs, but can be reduced by increasing acquisition time and, consequently, dose to the specimen. Our analysis enables highly accurate predictions of noise-induced root-mean-square (RMS) errors in a vector potential reconstructed from two tilt series using FBP, allowing for a more informed choice of acquisition time.

We observe that initial specimen orientation has a significant effect on reconstruction errors. Chapter 6 discusses the causes of this phenomenon, and the analysis therein provides an estimate of the total RMS error as a function of magnetisation orientation.

In Ch. 7 we explore the reduction of orientation-dependent errors using additional tilt series. The experimental acquisition of three mutually orthogonal tilt series poses significant challenges [85]. We circumvent this technical limitation by utilising orthogonal pairs of tilt series, and averaging the reconstructions resulting from each respective pair. This method can significantly reduce systematic errors in the reconstruction.

Chapter 8 discusses avenues for future research, and some experimental recommendations, and other concluding remarks, are provided in Ch. 9.
Chapter 2

Theory of vector field electron tomography

2.1 Introduction

In the TEM, electromagnetic information about the sample is encoded in the phase of the electron wavefunction at the exit surface via the Aharonov–Bohm effect [92, 93]. This phase is a linear combination of two components which, under the projection approximation, can be interpreted as projections of the electrostatic potential and the magnetic vector potential, respectively. Here we consider projections to be straight line integrals of the respective potentials in the electron propagation direction. This definition is consistent with the projection approximation of electron holography [94], ensuring applicability of the projection-slice theorem, which underpins the reconstruction algorithm.

The exit surface phase is retrieved from micrographs at multiple angles over two tilt series, and VFET allows us to reconstruct the vector potential from the magnetic phase, which is extracted from the total phase using additional measurements. In our work, this is achieved using a sample-flipping technique. A schematic of the experimental setup is shown in Fig. 2.1. In the remainder of this chapter, we discuss the theory of the processes used to perform the reconstruction, from phase retrieval to vector tomography, in more detail.
Figure 2.1: Schematic of the experimental setup. The exit phase of the electron wave is modified by the potentials of the sample, and is measured by recording out-of-focus micrographs and applying a phase retrieval algorithm. The focal plane image (indicated by a dashed border) is approximated from the underfocus (left) and overfocus (right) micrographs. The exit phase is measured for multiple angles about two orthogonal axes ($\alpha$ and $\theta$), enabling reconstruction of the vector potential associated with the sample.

2.2 Image formation in Lorentz transmission electron microscopy

The electron beam in the TEM originates from an electron source of one of three types. A thermionic source utilises the thermal emission of electrons from a heated element. For a field emission source, a large potential is applied between the source and an anode, exploiting the increased magnitude of the electric field at sharp edges, by using a source that comes to a fine point. This large electric field causes electrons to be ejected toward the anode. The third type of source, the Schottky source, is a hybrid of the other two types, consisting of a heated cathode placed in a strong electric field. Whichever type is used, the electron source is housed in an assembly called the electron gun, which produces a collimated beam of electrons. The field emission gun produces a highly coherent beam, making it the source of choice for electron holography and LTEM.

The beam passes through a series of lenses and apertures, as well as the specimen itself. Although exact configurations vary between microscopes, the basic design begins with a condenser lens, which focuses the electron beam onto the sample, followed by the sample itself. After passing through the specimen, the beam is focussed by the objective lens, and then passes through the back focal plane, where
an objective aperture can be placed to select beams of different scattering angles. Further along, the beam forms an image (the intermediate image), where an aperture can be inserted for use in selected area diffraction. An intermediate lens focuses another intermediate image (or a diffraction pattern, depending on the mode of operation), and a projector lens focuses it onto the phosphor screen or charge-coupled device (CCD). A diagram showing the main components, as well as the diffraction and imaging modes, is shown in Fig. 2.2. A detailed description of the TEM and its various modes of operation can be found in Ref. [95].

**Figure 2.2:** The diffraction (left) and imaging (right) modes of the TEM. (After Williams and Carter [95].)

**LTEm** is the imaging of magnetic materials in the TEM, and can be used to produce either Foucault images or Fresnel images. Foucault images are formed by observing the diffraction spots caused by the electron beam scattering from each magnetic domain of the specimen in a different direction, and selecting individual diffraction spots using the objective aperture [96]. This results in the domains corresponding to the selected diffraction spots appearing brighter than the other domains at the image plane [97]. Foucault images are useful for estimating domain size, but cannot be used for quantitative measurements of the magnetic properties of the specimen. Fresnel images are produced by defocusing the objective lens, resulting in phase contrast in parts of the image corresponding to domain boundaries in the specimen [96, 98].
Another form of Lorentz imaging, known as differential phase contrast (DPC), makes use of a scanning TEM and a segmented detector [99–101], or utilises a series of Foucault images taken in a conventional TEM, equipped with an electron biprism [102, 103] or electron trapezoidal prism [104]. Phase gradients in the electron wavefunction cause a signal difference across the detector, and this difference is a direct measure of the angle of deflection of the electron beam [105]. DPC has been used to reconstruct all three components of the magnetic induction field of a magnetic force microscopy tip in the specimen plane [106,107].

Under the projection approximation, the phase contrast that appears in Fresnel images arises from the phase shift acquired by the electron beam, as it traverses the specimen, due to the Aharonov-Bohm effect. We can utilise the contrast in the Fresnel images to determine the phase of the electron wavefunction at the image plane.

The contribution to the phase shift due to the electrostatic potential \( V(r_\perp, z) \) is given by [93]

\[
\varphi_e(r_\perp) = \frac{\pi}{E\lambda} \int V(r_\perp, z) dz, \tag{2.1}
\]

where \( z \) is the distance along the optical axis, \( r_\perp \) is the plane normal to the optical axis, \( E \) is the electron accelerating potential, and \( \lambda \) is the electron wavelength. The contribution to the phase shift due to the magnetic vector potential \( A(r_\perp, z) \) is given by [77]

\[
\varphi_m(r_\perp) = -\frac{e}{\hbar} \int A(r_\perp, z) \cdot dz, \tag{2.2}
\]

where \( e \) is the magnitude of the electron’s charge, and \( \hbar \) is the reduced Planck constant. The magnetic Aharonov-Bohm effect is illustrated in Fig. 2.3.

The phase shifts given in Eqs. (2.1) and (2.2) result in out-of-focus contrast upon defocus. Properties of the specimen can be inferred from this phase contrast, and
Phase retrieval using the transport-of-intensity equation

In order to obtain projections of the vector potential for its reconstruction, the phase must be inferred from recorded micrographs. This can be achieved using any of a number of phase reconstruction methods, including linear [73] and iterative [76,109] propagation based methods, as well as off-axis [110] holographic techniques. An in-line method based on the TIE [73,77] is used in this work to compute the phase from simulated out-of-focus micrographs. In addition to being computationally efficient, this method is linear under the phase object approximation. This simplifies the analysis of the propagation of errors from the micrographs to the phase, relative to iterative methods, such as the Gerchberg-Saxton algorithm [76]. The TIE relates transverse phase gradients to the longitudinal derivative of the intensity [77,78,111]:

\[-k \frac{\partial I_0}{\partial z} = \nabla_\perp \cdot (I_0 \nabla_\perp \varphi_0),\]  

(2.3)

where \( k \) is the wavenumber, \( I_0 \) and \( \varphi_0 \) are the intensity and phase at the image plane \((z = 0)\), respectively, \( \nabla_\perp \) is the two-dimensional gradient operator, and

\[\frac{\partial I_0}{\partial z} \equiv \frac{\partial I(r_\perp,z)}{\partial z} \bigg|_{z=0}.\]  

(2.4)

To obtain the phase, given measurements of both \( I_0 \) and \( \frac{\partial I_0}{\partial z} \), Eq. (2.3) can be solved using a Fourier transform method [73,112]:

\[\varphi(r_\perp) = \frac{k}{4\pi^2} \mathcal{F}^{-1} \left\{ \frac{k}{|k|^2} \cdot \mathcal{F} \left[ \frac{1}{I_0} \left( \mathcal{F} \left( \frac{\partial I_0}{\partial z} \right) \right) \right] \right\},\]  

(2.5)

where \( k \) is the spatial frequency vector, and \( \mathcal{F} \) and \( \mathcal{F}^{-1} \) denote the two-dimensional Fourier transform and its inverse, respectively. Under the phase object approximation, when the object is very weakly attenuating, the in-focus intensity \( I_0 \) can be replaced with the incident intensity \( I_{\text{in}} \), hence [113]

\[\varphi(r_\perp) = \frac{k}{4\pi^2 I_{\text{in}}} \mathcal{F}^{-1} \left\{ \frac{\mathcal{F} \left( \frac{\partial I_0}{\partial z} \right)}{|k|^2} \right\}.\]  

(2.6)
This approximation is sufficient for the purposes of noise analysis [114]. For the numerical implementation of Eq. (2.5), we utilise the approximation
\[ I_0 \approx \frac{I(r, \Delta f) + I(r, -\Delta f)}{2\Delta f}, \]
which reduces the number of images required from three to two. The longitudinal derivative of the intensity can be approximated from out-of-focus micrographs. For this work, due to its simplicity and computational efficiency, a two image central difference method is employed, i.e.,
\[ \frac{\partial I_0}{\partial z} = \frac{I(r, \Delta f) - I(r, -\Delta f)}{2\Delta f}. \]

Figure 2.4: Obtaining the magnetic phase using the time-reversal method. From left to right: Orientation of the particle, through-focal series, retrieved phases, and magnetic phase extracted from the two retrieved phases.

As discussed in Sec. 2.2, the phase obtained using the TIE contains information about both the magnetic and electrostatic properties of the sample. For the purposes of reconstructing the vector potential, it is only the magnetic component that is of interest. There are multiple methods to obtain this. The dependence of the electrostatic phase shift on the electron wavelength can be exploited to obtain the magnetic phase by varying the accelerating potential [115]. Another method assumes that the electrostatic phase shift is proportional to the sample thickness, and uses the in-focus image to calculate the electrostatic contribution and remove it from the total retrieved phase [98]. The separation of the magnetic and electrostatic phases
can also be achieved by flipping the sample and recording additional micrographs with the electron beam now traveling in the opposite direction relative to the sample. This allows the magnetic component of the phase to be extracted by exploiting the different time-reversal symmetries of the electrostatic and magnetic Aharonov–Bohm shifts [116]. For phase retrieval using the TE, this separation can be applied to the micrographs themselves [117], or to the retrieved phases [118]. We use the latter method, for which

\[ \varphi_m = \frac{\varphi_f - \varphi_r}{2}, \]  

(2.9)

where \( \varphi_f \) and \( \varphi_r \) are the phase shifts imparted by the sample before and after flipping, respectively. Experimentally, \( \varphi_r \) is obtained by rotating the sample by 180° about an axis orthogonal to \( z \), and reversing the resulting phase maps about the axis of rotation. A diagram highlighting how the magnetic phase is obtained is shown in Fig. 2.4.

### 2.4 Vector tomography

Tomography is the reconstruction of an object function from its projections. Scalar tomography typically takes a series of projections about a single tilt axis and reconstructs a scalar function from these projections. Vector tomography requires the acquisition of additional tilt series to obtain enough information about the specimen to reconstruct a vector field in three dimensions.

Vector fields can be reconstructed from their projections using a variety of techniques [119], including direct Fourier inversion [85], simultaneous iterative reconstruction techniques [120], and algebraic reconstruction techniques [121]. For this work, the vector potential \( \mathbf{A}(x, y, z) \) is reconstructed using an FBP algorithm, which uses the projection-slice theorem in cylindrical coordinates, in combination with the Coulomb gauge condition. This algorithm is given by [85]

\[
\mathbf{A}(x, y, z) = \int_0^\pi \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \tilde{T}^\theta |k_r^\theta|^2 |k_r^\theta|^2 \begin{bmatrix} k_x^2 + k_y^2 \\ k_y \\ -k_x \\ k_x k_z \\ k_y \\ k_x k_z \\ k_y \\ k_y \end{bmatrix} e^{2\pi i(k_x^\theta(x \cos \theta + y \sin \theta) + k_z z)} dk_r^\theta dk_z d\theta \\
+ \int_0^\pi \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \tilde{T}^\alpha |k_r^\alpha|^2 |k_r^\alpha|^2 \begin{bmatrix} k_x \\ k_y \\ k_z \\ -k_x -k_z \\ k_x k_y \\ k_y \\ k_x k_y \\ k_y \end{bmatrix} e^{2\pi i(k_x^\alpha(y \cos \alpha + z \sin \alpha) + k_x x)} dk_r^\alpha dk_x d\alpha,
\]  

(2.10)
where $\theta$ and $\alpha$ are positive angles of rotation about the $z$ and $x$ axis, respectively, $T^\theta$ and $T^\alpha$ are the tilt series associated with these directions, whilst tildes represent Fourier transformed quantities, and $k_r^\theta = k_y / \sin \theta$ and $k_r^\alpha = k_y / \cos \alpha$ are radial spatial frequencies. $T^\theta$ and $T^\alpha$ are given by [85]

$$T^\theta(x, z) = -\frac{\hbar}{e} \varphi_m(x, z),$$

and

$$T^\alpha(y, x) = -\frac{\hbar}{e} \varphi_m(y, x).$$

The geometry for the tilt series acquisition is shown in Fig. 2.5.

Figure 2.5: Acquisition of tilt series $T^\theta$ and $T^\alpha$. The curved arrows indicate the direction that the object $O$ is rotated in each tilt series, and the double arrows indicate the propagation direction of the electron beam. (After Yu et al. [85].)

In practice, a discrete form of Eq. (2.10) is used for the reconstruction. For the present work, we perform the reconstructions on a cubic voxel grid, and it is convenient to make a change of variables which enables the algorithm to be expressed as three summations. Given this, the algorithm we use can be expressed as:

$$A_{\text{rec}, m,n,p}^{\text{rec}} = \frac{\pi}{n_t - 1} \sum_{\mu=0}^{n_t - 1} \frac{1}{M^2} \sum_{l,j=-M/2}^{M/2} \left( \tilde{T}_l^{\theta} \eta_{l,j,\mu} e^{2\pi i(ln' + jp)/M} + \tilde{T}_l^{\alpha} \eta_{l,j,\mu} e^{2\pi i(ln'' + jm)/M} \right),$$

(2.13)

where $A_{\text{rec}}^{\text{rec}}$ is the reconstructed vector potential; $M$ is the number of pixels along each direction of the input images; $n_t$ is the number of magnetic phase maps in each tilt series; $m$, $n$, and $p$ are the voxel indices in real space; $n' = m \cos \theta + n \sin \theta$; and
\( n'' = n \cos \alpha + p \sin \alpha \). The coefficients \( \eta^\theta_{l,j,\mu} \) and \( \eta^\alpha_{l,j,\mu} \) are given by:

\[
\eta^\theta_{l,j,\mu} = \frac{l^2}{aM(l^2 + j^2)} \begin{bmatrix}
  \frac{l^2 \sin^2 \theta + j^2}{l \sin \theta} \\
  -l \cos \theta \\
  -j \cot \theta
\end{bmatrix},
\]

(2.14)

and

\[
\eta^\alpha_{l,j,\mu} = \frac{l^2}{aM(l^2 + j^2)} \begin{bmatrix}
  j \tan \alpha \\
  l \sin \alpha \\
  \frac{l^2 \cos^2 \alpha + j^2}{l \cos \alpha}
\end{bmatrix}.
\]

(2.15)

Here, \( a \) is the width of the input micrographs, and \( \theta \) and \( \alpha \) are both given by \( \pi \mu/(n_t - 1) \). Bilinear interpolation is used to transform from radial to Cartesian coordinates. The inverse discrete Fourier transforms implicit in Eq. (2.13) can be computed using a fast Fourier transform (FFT) algorithm (see, for example, Ref. [122]).

### 2.5 Regularisation of singularities

The phase retrieval and FBP algorithms used here contain Fourier-space singularities, resulting in numerical instability and amplification of noise. In order to overcome these problems, solutions can be forced to be finite at the singularities by modifying the algorithms using regularisation. In previous work [123], Tikhonov regularisation [124] has been employed to deal with the singularity in Eq. (2.5) at \( |k| = 0 \), and we follow this approach. This is achieved by applying the following transformation to Eq. (2.6):

\[
|k|^2 \rightarrow \frac{|k|^4 + \delta_{\text{TIE}}^4}{|k|^2},
\]

(2.16)

where \( \delta_{\text{TIE}} \) is a regularisation parameter having dimensions of a reciprocal length. We obtain:

\[
\varphi(r_\perp, 0) = \frac{k}{4\pi^2 I_m} \mathcal{F}^{-1} \left\{ \frac{|k|^2 \varphi A \left( \frac{\partial \varphi}{\partial z} \right)}{|k|^4 + \delta_{\text{TIE}}^4} \right\}.
\]

(2.17)

This makes the solution finite at \( |k| = 0 \), stabilising the numerical computation, and also suppressing amplification of noise near \( |k| = 0 \). For \( |k|/\delta_{\text{TIE}} \gg 1 \), Eq. (2.17) reduces to Eq. (2.6), but at (and near) \( |k| = 0 \), the regularised \( \text{TIE} \) deviates from the exact solution. Because the regularisation does not arise naturally from the theory, it induces errors in the reconstruction. Finding an appropriate choice for a regularisation parameter is a compromise between choosing a large enough value to
adequately suppress errors, while keeping it small enough to remain consistent with the theory and avoiding unnecessarily introducing artefacts from the regularisation itself [125]. Tikhonov regularisation is employed wherever singularities occur in the algorithms. For Eq. (2.5), the transformation used is

\[ |k|^2 \rightarrow \frac{|k|^4 + \delta_{\text{TIE}}^4 / 2}{|k|^2}. \tag{2.18} \]

We have introduced the factor of two for consistency in the scale of the regularisation parameters. That is, we require that the results obtained using Eq. (2.5) match those of Eq. (2.6) when \( I_0 = I^{\text{in}} \). The use of Eq. (2.18) for the former and Eq. (2.16) for the latter ensures this, provided that \( \delta_{\text{TIE}} \) is small.

It is also possible to separate the electrostatic and magnetic components of \( \partial I_0 / \partial z \), prior to using Eq. (2.17) to retrieve the phase [117]. This allows for the more appropriate selection of regularisation parameters for recovery of the electrostatic and magnetic phases. In the present work, we are not concerned with the accurate reconstruction of the electrostatic phase, so we utilise only a single regularisation parameter.

Equation (2.10) contains singular surfaces at \( k_y = 0 \) for \( A_x \) and \( A_z \). In terms of the tilt angles, these singular surfaces are at \( \theta = 0 \) and \( \alpha = \pi/2 \). Due to the practical limitations of recording tilt series over the entire range of \( \pi \) radians, it is natural to remove the singular surface at \( \theta = 0 \) by simply omitting this image from the reconstruction process. For the singular surface at \( \alpha = \pi/2 \), we again use Tikhonov regularisation:

\[ \frac{1}{\cos \alpha} \rightarrow \frac{\cos \alpha}{\cos^2 \alpha + \delta_{\text{FBP}}^2}, \tag{2.19} \]

where \( \delta_{\text{FBP}} \) is the regularisation parameter.

The division by \( I_0 \) in Eq. (2.5) can also cause numerical instability in the presence of noise. We address this by regularising with

\[ I_0 \rightarrow \frac{I_0^2 + \delta_{\text{INT}}^2}{I_0}, \tag{2.20} \]

where \( \delta_{\text{INT}} \) is the regularisation parameter. Because noise in \( I_0 \) does not impact greatly on the errors in the reconstruction, the choice of \( \delta_{\text{INT}} \) does not affect the noise-induced errors examined in this work. We arbitrarily set \( \delta_{\text{INT}} = 0.08I^{\text{in}} \), which is small enough to avoid introducing significant systematic errors in the retrieved phases.
2.6 Error metrics used for this work

Quantifying errors in reconstructed vector fields differs from quantifying those of scalar reconstructions in that the total error involves errors due to differences in the magnitude of the input and output vector at each voxel, and differences in the orientation of the vectors.

In determining the vector potential of a specimen, the accuracy of a reconstruction method can be quantified in different ways depending on what properties of the specimen are of most interest. What makes a reconstruction accurate depends on the application. For example, for some applications, the magnitude of the vector potential may be of little interest, and accuracy is only required for the direction of the reconstructed vectors. For other applications, it may be important to know the accuracy of the magnitude of the vectors. Other quantities that may be of interest include the curl and divergence of the vector field, and even though the magnetic induction, and the magnetic vector potential in the Coulomb gauge, are divergence free, the reconstructed field may not be. The metrics can also be chosen to highlight the accuracy of a reconstructed field in terms of these quantities. Because the optimum value of experimental parameters (e.g., initial specimen orientation, defocus, or image acquisition time) can depend on which metric is used, the choice of metric can directly influence which sample preparation techniques and experimental procedures are used.

2.6.1 Root-mean-square error metrics

Here, we restrict our attention to three different metrics; namely, the total RMS error, the RMS error in vector magnitude, and the RMS error in vector direction. The total normalised RMS error is defined by

\[
\langle \text{tot} E \rangle = \sqrt{\frac{\sum_{i,j,k} |A_{i,j,k}^{\text{rec}} - A_{i,j,k}|^2}{\sum_{i,j,k} |A_{i,j,k}|^2}}. \tag{2.21}
\]

This can be written as

\[
\langle \text{tot} E \rangle = \sqrt{\frac{\sum_{i,j,k} |E_{i,j,k}|^2}{\sum_{i,j,k} |A_{i,j,k}|^2}}, \tag{2.22}
\]

where \(E\) is the error in the reconstructed vector field, and

\[
A_{i,j,k}^{\text{rec}} = A + E. \tag{2.23}
\]


Our directional RMS error is defined as the RMS value of the angle between the exact and reconstructed vectors at each voxel and is given by

\[
\langle \text{dir} E \rangle = \sqrt{\frac{1}{M^3 \pi^2} \sum_{i,j,k} \text{dir} E_{i,j,k}^2},
\]

(2.24)

where

\[
\text{dir} E_{i,j,k} = \cos^{-1} \left( \frac{A_{i,j,k} \cdot A_{i,j,k}^{\text{rec}}}{|A_{i,j,k}| |A_{i,j,k}^{\text{rec}}|} \right)
\]

(2.25)

is the angle between the exact and reconstructed vectors at the \((i, j, k)\) voxel. The factor of \(\pi^{-2}\) in Eq. (2.24) is included so that \(\langle \text{dir} E \rangle\) is expressed as a fractional error rather than an angle.

The normalised RMS error in the magnitude of the vector potential is given by

\[
\langle \text{mag} E \rangle = \sqrt{\sum_{i,j,k} \text{mag} E_{i,j,k}^2 \sum_{i,j,k} |A_{i,j,k}|^2},
\]

(2.26)

where

\[
\text{mag} E_{i,j,k} = |A_{i,j,k}^{\text{rec}}| - |A_{i,j,k}|.
\]

(2.27)

2.6.2 Quantifying systematic errors in the vector potential

Equation (2.26) quantifies the error in the magnitude of the vector potential, but it does not differentiate between systematic and random errors. To measure systematic errors in the vector magnitude, we define a mean scaling factor:

\[
F_s = \frac{\sum_{i,j,k} |A_{i,j,k}^{\text{rec}}|}{\sum_{i,j,k} |A_{i,j,k}|}.
\]

(2.28)

Equation (2.28) provides a measurement of the mean scaling of the vector potential, which causes the magnetisation of the specimen to appear weaker or stronger in the reconstructed vector potential than in the exact potential.
Chapter 3

Sources of error

3.1 Introduction

In this chapter, several sources of error in VFET are briefly introduced. This is not a comprehensive list of every possible source of error, but several of the most significant sources of error are addressed here.

We start with a discussion of shot noise in Sec. 3.2. Section 3.3 provides a brief introduction to the truncation errors involved in our reconstructions. In Sec. 3.4, a significant diffraction artefact—the delocalisation of dark field images—is discussed. Errors arising from image misalignment are discussed in Sec. 3.5. In Sec. 3.6, we briefly discuss image, and reconstruction, resolution. Section 3.7 covers errors that may arise due to the way the micrographs are simulated, and in Sec. 3.8, errors caused by attenuation of the electron beam are discussed.

3.2 Shot noise

The recorded micrographs are subject to shot noise due to the finite number of electrons recorded at the detector [126]. The noise is dependent on beam current and acquisition time, and noise in the images propagates through the reconstruction process and affects the accuracy of the reconstructed vector fields. Singularities in the phase retrieval and tomographic reconstruction algorithms can amplify the noise, and the methods of regularisation, as well as the parameters used, can suppress this amplification.

Work by Yu et al. [85] has used simulated data to examine the noise stability of both direct and FBP methods of reconstruction, for two and three tilt series. It was found that the reconstruction errors (quantified using an RMS error metric) were smaller when three tilt series were used in the reconstruction, rather than two,
because the three tilt series method circumvents singular surfaces that arise in two components of the vector potential reconstruction. This is achieved by reconstructing each component from the two tilt series that do not contain singular surfaces in the reconstruction algorithm for that particular component. Because both the two and three tilt series methods actually use only two tilt series to reconstruct each component of the vector potential, the reduction in error for the three tilt series method is not due to improved statistics achieved by including more information in the reconstruction.

When the simulated micrographs contain no added noise, the direct method is superior to the FBP method, but when noise is added, the FBP is more accurate than the direct method, highlighting the fact that the choice of reconstruction method can be motivated by knowledge of the sources of error in the input micrographs, and the way in which these errors propagate through different reconstruction algorithms.

In this work, we utilise an incident noise level defined by

$$\sigma_{\text{in}} = \frac{\sqrt{N}}{N},$$

which is the fractional noise level in a micrograph recorded in the absence of a sample. Here \(N\) is the mean number of detected electrons (again, in the absence of a sample). This results in slightly underestimated noise levels when the sample is highly attenuating, but more closely matches an experimental scenario where the beam current, acquisition time, and detector behaviour is typically the same for every image. The simulation of shot noise is discussed in Sec. 4.6, and a thorough analysis of its effects in VFET is presented in Ch. 5.

### 3.3 Truncation error

Because the CCD detector in the electron microscope is sensitive only to the intensity and not the phase of the wavefield, the phase must be determined indirectly. In this thesis, a propagation based method (the TIE) is used to recover phase maps from out-of-focus images. As mentioned earlier, the TIE (Eq. (2.3)) relates transverse derivatives of the phase to the longitudinal derivative of the intensity [113]. The out-of-focus images are used to calculate the derivative of the intensity using the approximation

$$\frac{\partial I(r_\perp,0)}{\partial z} \approx \frac{I(r_\perp,\Delta f) - I(r_\perp,-\Delta f)}{2\Delta f}$$

$$= \frac{\partial I(r_\perp,0)}{\partial z} + \frac{(\Delta f)^2}{3!} \frac{\partial^3 I}{\partial z^3} + O((\Delta f)^4),$$

(3.2) (3.3)
where $\Delta f$ is the defocus, which is then used to calculate the phase. Because the calculated derivative relies on a finite difference approximation, this procedure induces errors, which then propagate to the reconstructed vector potential. Similarly, a truncation error is incurred if the in-focus image is approximated from the defocussed images, rather than recorded directly. These errors are dependent upon the number of images used, as well as their respective defoci. For two images, used in this thesis, the in-focus image is approximated by

\[
I(r_\perp, 0) \approx I(r_\perp, \Delta f) + I(r_\perp, -\Delta f) + \frac{(\Delta f)^2}{2!} \cdot \frac{\partial^2 I}{\partial z^2} + O((\Delta f)^4).
\]

(3.4)

Note that a truncation error of $O((\Delta f)^2)$ is incurred due to each of these approximations.

In practice, approximating in-focus micrographs using Eq.(3.4) does not increase the computational overhead significantly compared to using the approximation $I(r_\perp, 0) \approx I^{in}$, and requires no additional acquisition of images. For these reasons, Eq. (3.4) will be used for the reconstruction of attenuating samples from two defocussed images per tilt angle.

### 3.4 Diffraction artefacts

VFET reconstruction algorithms require that the retrieved phase maps be projections of the potential. Deviations from the projection approximation can occur due to dynamical scattering from both the electrostatic atomic potentials and the magnetic vector potential, as well as Bragg diffraction. Diffraction artefacts in the micrographs can be significant, and can cause errors in otherwise reliable tomographic reconstructions [127]. These artefacts can be particularly strong near zone axes of crystals, and because they are very sensitive to changes in crystal orientation, these artefacts will combine when the phase maps are subtracted via Eq. (2.9) to extract the magnetic components. In VFET, where two tilt series—each comprising multiple images—are acquired, it can be difficult to avoid problematic specimen orientations. A method for the simulation of micrographs that exhibit these artefacts is explained in Sec. 4.4, and their contribution to reconstruction errors, as well as potential methods of amelioration, are discussed in Sec. 8.3.
3.5 Image alignment

Experimental images must be aligned in order to perform the phase retrieval and tomographic reconstruction [91, 128]. Translational alignment of images in a tilt series can be achieved using a cross-correlation technique [129]. In this method, each successive image is translated based on the location of the peak in the cross-correlation of the image with the preceding image [130]. This process results in a translational error that increases with increasing angle [129]. An angular cross-correlation method can be used for the rotational alignment of images taken at the same tilt angle, but this is not easily applied to the rotational alignment of subsequent images in a tilt series. Rotational alignment for tomography can also be achieved by examining features of the specimen [131].

The effect of image misalignment is not explored in this thesis, and image registration is not implemented in our simulations. Because the simulated images naturally display perfect alignment, there is no error incurred due to misalignment in our simulations.

3.6 Spatial resolution

The spatial resolution of the micrographs and phase maps affects the accuracy and resolution of the resulting reconstruction. The CCD will typically record images with a relatively high degree of spatial sampling (for example, the Gatan UltraScan 1000 P CCD camera of the FEI Titan 80-300 FEG-TEM at the Monash Centre for Electron Microscopy has a resolution of 2048 × 2048 pixels [132]), so the primary concern in the choice of resolution for the reconstruction is the processor speed, CPU time and random access memory available. The backprojected phases must be interpolated at voxel grid points, and an error is incurred that is dependent on the method of interpolation. An FFT algorithm is used for the reconstruction and, because performance of the transform is typically highest for a radix 2 FFT [133], an increase in resolution usually involves doubling the resolution in each direction. This results in an eightfold increase in memory usage.

The number of phase maps in each tilt series (the angular resolution) has an effect on the accuracy of the reconstruction, but higher angular resolution entails longer acquisition time, increased sample degradation, longer computation time, and increased computer memory requirements. In addition to this, as discussed in Ch. 1, the limited range of angles available in a tilt series (up to about ±70° [134]) due to the constraints of the tilt stage used, results in a missing wedge of information in the acquired data. In scalar tomography this causes an elongation, in the reconstruction,
of features that are aligned perpendicularly to the axis of rotation [54,135]. In VFET, the missing wedge causes errors in the reconstructed vector potential [84], although the nature of the resulting artefacts has not been thoroughly investigated. In the present work, the effect of the missing wedge is not explored. In our simulations, we vary the number of images in each tilt series, as well as the number of tilt series themselves, to explore the effect of variations in angular sampling.

3.7 Errors arising from imperfectly simulated micrographs

There are errors that arise in the simulations that are due to the method of micrograph simulation rather than to the reconstruction process. In other words, these are errors that will not occur when using experimental micrographs. Examples include wraparound errors caused by the use of the FFT in computing defocussed images from \( \varphi(r_\perp, 0) \), and numerical errors resulting from the method of integration used to compute \( \varphi(r_\perp, 0) \) from the vector potential. These errors should be identified and reduced as much as possible in order to provide accurate error estimates. Some of these errors are addressed further in Ch. 4.

3.8 Attenuation

Attenuation can be a source of error in VFET for a variety of reasons. The reduced signal strength due to attenuation decreases the signal-to-noise ratio. For a large, highly attenuating specimen, the effect can be significant if the object covers a high percentage of the field of view.

We include attenuation in our specimen simulations, but we do not perform a detailed analysis of the effects of attenuation on reconstruction errors. However, some preliminary work on attenuation, as well as potential future work on the subject, is discussed in Sec. 8.5.
Chapter 4

Simulation of transmission electron micrographs for vector field electron tomography

4.1 Introduction

Simulations are a core aspect of the work presented in this thesis. Computational work allows us to investigate properties of the VFET reconstruction process not readily accessible to analytical examination, and in cases where an analysis is performed, these computational methods provide results that we can compare it with.

The advantage of utilising simulations instead of experiments, in the context of error analyses, is that in simulations we can know, \textit{a priori}, the exact value of the vector potential at every point in space. This enables precise calculation of errors (such as those described in Sec. 2.6), which provides a means to quantify the effect of changing acquisition and reconstruction methods, as well as specimen properties. Future work in VFET can make use of the findings of numerical studies, to enable more accurate experimental reconstructions of electromagnetic vector fields. In addition, qualitative features of reconstruction errors observed using simulations can provide researchers with a more complete understanding of the artefacts they observe in experimental reconstructions.

In the remainder of this chapter, the methods used for simulating transmission electron micrographs are described.
4.2 Specimen simulation

The simulated uniformly magnetised samples used in this work exist as binary 3D shape function arrays, with the voxels containing the sample being set to 1, and voxels of empty space set to 0. These arrays can easily be constructed for simple geometric objects, such as a cube or sphere, using an analytical representation [136]. For more complex geometries, the samples are modelled using the 3D modelling software Blender 2.67 [137], and a Python implementation of a ray-casting algorithm is employed to convert the Blender object into a binary mask file. This method enables the construction of arbitrary sample shapes. To illustrate the flexibility of this approach, a micrograph simulated using a complicated sample shape (a human head) is shown in Fig. 4.1.

![Image of simulated micrograph](imageurl)

Figure 4.1: Simulation of a uniformly magnetised object using a human head as the sample shape. (Top row: exit phases. Bottom row: defocussed micrographs.)

The magnetic vector potential is constructed from the shape function $D(r)$, which corresponds to the binary array described above, using a Fourier transform method given by [138]
where $M_0$ is the (uniform) magnetisation of the particle, $\hat{m}$ is a unit vector in the direction of the magnetisation, $\mu_0$ is the vacuum permeability, and the tilde indicates a Fourier transformed quantity. We can also construct the vector potential of an arbitrary magnetisation $M$ via 

$$A(\mathbf{r}) = \mathcal{F}^{-1} \left\{ -\frac{i\mu_0 M_0}{|k|^2} \widetilde{D}(k)(\hat{m} \times k) \right\},$$

(4.1)

In our simulations, we achieve this by placing arrow objects\(^1\) in our Blender scene, and we utilise a Python script to construct a vector field from these arrows inside the specimen. This is done using a nearest neighbour method, which gives us the flexibility to simulate large magnetic domains using a small number of arrows, or to add more detail to the magnetisation distribution using a larger number of arrows.

### 4.3 The projection approximation

The simulations presented in this thesis follow the work of Yu et al. [85], which involved the modelling of a pure phase object with no electrostatic component to the phase shift induced by the sample. In order to investigate the effects of electrostatic phase shifts, and the required modification to the acquisition and reconstruction process (see Eq. (2.9) and the preceding explanation), we add a constant scalar potential to the region bounded by the simulated sample. This quantity corresponds to the mean inner potential, which describes the volume average of the scalar potential of a specimen [139].

The electrostatic and magnetic components of the phase at the exit surface of the simulated specimen are calculated using Eqs. (2.1) and (2.2), respectively. We use the composite Simpson’s rule for the numerical integration [140]. Treating the electron beam as a monochromatic plane wave, the electron wavefunction at the image plane is given by

$$\Psi(\mathbf{r}_\perp) = e^{-i\varphi(\mathbf{r}_\perp)},$$

(4.3)

where $\varphi(\mathbf{r}_\perp) = \varphi_m(\mathbf{r}_\perp) + \varphi_e(\mathbf{r}_\perp)$ is the total exit phase. Absorption is then modelled by adding an imaginary part to the mean inner potential.

\(^{1}\)Each object placed in a Blender scene is associated with a rotation property that describes the global orientation of the object, and can be accessed via Python scripting.
In order to include the effects of microscope defocus in our simulations, we utilise a transfer function formalism. We obtain the electron wavefunction at a defocus of $\Delta f$ using a multiplication in Fourier space:

$$\Psi(r_\perp, \Delta f) = \mathcal{F}^{-1} \left\{ \tilde{\Psi}(k, \Delta f = 0) H(k) \right\}, \quad (4.4)$$

where $H(k)$ is the transfer function. In the absence of other aberrations, such as spherical aberration, which we ignore here, the transfer function is given by [141,142]

$$H(k) = e^{i(\pi \Delta f \lambda |k|^2)}. \quad (4.5)$$

The image intensity is then calculated from the electron wavefunction:

$$I(r_\perp) = |\Psi(r_\perp)|^2. \quad (4.6)$$

The projection approximation is valid when the electron wavelength is small compared to the characteristic length scale of the specimen [85]. However, here we do not take into account the variation of the electrostatic potential inside the specimen; this omission results in an absence of artefacts that arise from the electron beam interacting with the individual atomic potentials. We address this limitation in the following section.

### 4.4 Multislice simulations

In Sec. 4.3, the use of the projection approximation to calculate the exit phase was discussed. This method approximates the sample as being infinitely thin, ignoring dynamical scattering of the electron beam as it traverses the sample. In order to provide more realistic simulations, a multislice approach was incorporated into the vfet code. The multislice method solves the problem of dynamical scattering by dividing the sample into slices. Each slice alters the phase of the incident wavefunction, which is then propagated to the next slice and the process is repeated for each successive slice [143]. In addition to this, the use of a mean inner potential in the phase object approximation simulations is replaced, in the multislice simulations, with a realistic electrostatic potential that varies within the specimen due to the individual atomic potentials. This results in more realistic electron scattering, which leads to diffraction artefacts in the micrographs that better match experimental results. The multislice code used for this project\(^2\) computes a complete electrostatic

---

\(^2\)Source code by Earl J. Kirkland, used for the multislice image simulation, is available at [http://people.ccmr.cornell.edu/~kirkland/cdownloads.html](http://people.ccmr.cornell.edu/~kirkland/cdownloads.html) (accessed February 6, 2016). For the theory behind the code, and detailed instructions on usage, see Ref. [144].
potential when atom locations and atomic numbers are provided for every atom in
the sample [144], producing realistic diffraction artefacts in the simulated micro-
graphs. For accurate simulations, it is important to ensure the integrated intensity
across each image remains close to unity. According to Kirkland [144], an integrated
intensity of > 0.9 is sufficient for this purpose. In our simulations, we ensure that
this value does not drop below 0.99. Third order spherical aberration of 0.5 mm is
included, which is consistent with experiments in aberration corrected LTEM [145],
but we do not model partial coherence in our simulations.

Figure 4.2: Unit cell of magnetite structure used for multislice simulations in this
work. The colours represent Fe$^{2+}$ (green), Fe$^{3+}$ (red), and O$^{2-}$ (white).

We model magnetite [146] in all our simulations. The crystal structure used in
the multislice simulations for this work is shown in Fig. 4.2. We simulate a vector
potential of resolution $M$ (the reconstruction resolution), utilising the methods dis-
cussed in Sec. 4.2. For each 0.5 nm slice of the atomic potential, calculated from the
crystal structure, we introduce a magnetic phase shift. This is achieved by creating
a 2D array representing a slice of the vector potential, which is selected from the
3D vector potential using a nearest neighbour method. The slice is upscaled to the
image resolution $M_{ms}$ using a Gaussian filter with a small standard deviation (in our
simulations, we use 0.05 pixels). The filter ensures that there are no unrealistically
steep gradients in the slice, which would cause erroneous fresnel diffraction effects.
Combining the magnetic and electrostatic phase shifts in this way ensures that the
dynamical scattering modelled by the multislice method, typically used to simulate non-magnetic specimens, is also applied to the magnetic scattering.

In the multislice simulations used in our work, a carbon support film is modelled in addition to the magnetite specimen. The support film is assumed to be completely amorphous, so we construct it out of carbon atoms placed randomly within the mask, and with an average density of $2.1 \times 10^3$ kg m$^{-3}$ [147]. A drawback of the way this has been implemented is that the simulated carbon film is only as wide as the voxel grid. Because of this, as the specimen is rotated, the carbon film no longer fills the entire field of view. This could be overcome by storing the carbon film in a separate, higher resolution array, and rotating the film prior to cropping it. This would only cause problems for very high angles, which could be omitted due to being inside the missing wedge. An annotated out-of-focus micrograph, simulated using these methods, is shown in Fig. 4.3.

![Simulated out-of-focus micrograph of an octahedral nanoparticle. This image highlights some of the main features of the simulations, such as the delocalisation of dark field images, the realistic sample shape, and the shot noise. Magnetic contrast is also present in this image but, at this vector potential strength, it is not visible without further image processing.](image)

Figure 4.3: Simulated out-of-focus micrograph of an octahedral nanoparticle. This image highlights some of the main features of the simulations, such as the delocalisation of dark field images, the realistic sample shape, and the shot noise. Magnetic contrast is also present in this image but, at this vector potential strength, it is not visible without further image processing.
4.5 Amelioration of aliasing artefacts

Aliasing effects can result in high frequency artefacts in the micrographs. This can cause inaccuracies in our simulations. To avoid this, we filter the simulated micrographs using a Gaussian kernel with a standard deviation of 0.01 pixels.

Figure 4.4 illustrates this by comparing an analytically derived error surface (as a function of image noise $\sigma_{in}$ and defocus $\Delta f$) with the results of simulations using both the aliased, and anti-aliased, micrographs. These results show that Gaussian blurring is effective at ameliorating the aliasing artefacts that are present for small defoci. For the theory underpinning the analytical error surfaces, see the noise analysis presented in Ch. 5.

This aliasing is discussed by Kirkland [144] (p. 118), and bandwidth limiting similar to the Gaussian filtering used here is included in his multislice code (see Sec. 4.4).

4.6 Shot noise model

Early work on the effects of noise in VFET used a white Gaussian [83] or Poisson [85] noise model. These models treat the noise level as a constant, with no variation
across the image. In this work, a more realistic shot noise model is used. This model takes into account the change in noise level with changing intensity and provides a more accurate distribution of noise over the simulated images, particularly for large defocus and/or when there is significant attenuation.

The scale used for the intensity is arbitrary, and is proportional to the mean number of detected electrons per pixel. Thus, we can define a constant of proportionality $\kappa$ such that

$$I^\text{in} = \kappa N,$$  \hspace{1cm} (4.7)

where $N$ is the mean number of detected electrons per pixel in the absence of a sample. The noise-free intensity $I_{i,j}^{\text{ideal}}$ at a given pixel is related to the mean number of detected electrons $N_{i,j}$ at that pixel by

$$I_{i,j}^{\text{ideal}} = \kappa N_{i,j}.$$ \hspace{1cm} (4.8)

The actual intensity, taking shot noise into account, is then

$$I_{i,j} = \kappa \Pi(N_{i,j}),$$ \hspace{1cm} (4.9)

where $\Pi(N_{i,j})$ produces a random integer selected from a Poisson distribution with mean $N_{i,j}$. This leads to the relation

$$\frac{I_{i,j}}{I_{i,j}^{\text{ideal}}} = \frac{\Pi(N_{i,j})}{N_{i,j}}.$$ \hspace{1cm} (4.10)

From Eqs. (4.7) and (4.8), the mean number of detected electrons is:

$$N_{i,j} = \frac{I_{i,j}^{\text{ideal}}}{I^\text{in}} N.$$ \hspace{1cm} (4.11)

Substituting this into Eq. (4.10), the intensity is then given by

$$I_{i,j} = \Pi\left(\frac{I_{i,j}^{\text{ideal}}}{I^\text{in}} N\right) \frac{I^\text{in}}{N}.$$ \hspace{1cm} (4.12)

Making use of Eq. (3.1), the intensity in the noisy image is then

$$I_{i,j} = \Pi\left(\frac{I_{i,j}^{\text{ideal}}}{\sigma^2_{\text{in}} I^\text{in}}\right) \sigma^2_{\text{in}} I^\text{in}.$$ \hspace{1cm} (4.13)
4.7 Apodisation

To improve sampling in Fourier space [85] and to remove wraparound artefacts in the simulated micrographs, an apodisation function is used; i.e.,

\[ w_{i,j} = \begin{cases} 
1 & (i - \frac{M}{2})^2 + (j - \frac{M}{2})^2 < (r_w M)^2 \\
0 & \text{elsewhere}
\end{cases}, \]  

(4.14)

where \( r_w \) is the radius of the apodisation function as a fraction of the image size. A windowed micrograph is then obtained by multiplying the image pointwise by \( w \):

\[ I_{i,j} \rightarrow w_{i,j} I_{i,j}. \]  

(4.15)

In this thesis we use a value of \( r_w = 0.3 \).

4.8 Simulation parameters used in this work

There are several parameters required for the simulation of transmission electron micrographs that can be adjusted to emulate various microscope operating conditions. Many of these parameters remain constant throughout most of the work presented in this thesis. Except where stated otherwise, our simulations use the parameters shown in Table 4.1. Quantities that are varied often, such as defocus and micrograph width, are not shown in this table.

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Table 4.1: Parameters used in simulations unless stated otherwise.
Chapter 5

Effect of image noise on vector field electron tomography reconstruction errors

5.1 Introduction

In this chapter, one significant source of error in VFET is addressed; namely, shot noise in the recorded electron micrographs. The effect of image noise on the reconstructed vector field has been briefly investigated by Yu et al. [85]. Their work tested the stability of various VFET algorithms when white Poisson noise was added to simulated micrographs, noting qualitative features of the noise-induced artefacts in the reconstructed vector fields. Owing to both the large amount of data required and technical considerations, such as alignment of the tilt series, VFET can be a time consuming and computationally demanding process; consequently, it is important to have a thorough understanding of how variables such as spatial resolution and image acquisition time affect accuracy. This is a key motivation for the present work.

The effects of image noise on the use of the TIE in phase retrieval have been explored by Paganin et al. [114]. They found that the optimum defocus is proportional to the cube root of the RMS value of the noise level. As the image noise increases, the defocus must also be increased to compensate for the lower signal-to-noise ratio. In VFET, errors in the retrieved phase contribute to errors in the reconstructed magnetic vector potential. This causes the reconstruction errors to have a similar functional dependence on noise and defocus, to those of the retrieved phase: at low noise levels, the reconstruction errors are minimised with small defoci, and at higher noise levels, larger defoci must be used to reduce the noise-induced errors in the phase and, consequently, in the reconstructed magnetic vector potential. We
explore this effect in detail in Sec. 5.2.

A significant source of error in VFET reconstructions is the amplification of noise near Fourier-space singularities in both the phase retrieval and vector tomography algorithms. In the FBP algorithm, singular surfaces arise due to the Fourier inversion used to recover the vector potential. In theory, these singular surfaces can be avoided by utilising a total of three mutually orthogonal tilt series. However, acquisition of the third tilt series is improbable using contemporary electron microscopy instrumentation [85]. As mentioned earlier in this thesis, the effects of noise can be ameliorated by regularisation of these singularities. Humphrey et al. [117] have described a scheme that separates the electrostatic and magnetic components of the longitudinal derivatives of the micrographs, allowing different regularisation parameters to be applied separately to each. Here, we are not concerned with accurate recovery of the electrostatic component of the phase, and utilise a single regularisation parameter in the TIE.

The results obtained by Yu et al. [85] provide a means to identify certain noise induced artefacts, which they found can occur in the form of streaking in the reconstructed vector potential, which results from the presence of the previously mentioned singularities in the reconstruction algorithms. However, their work did not provide a quantitative analysis of the propagation of noise-induced errors through the reconstruction. Indeed, prior to the work (by the author of this thesis) on which this chapter is based [148], a quantitative analysis of the propagation of noise through these algorithms had not been presented in the literature. In this chapter, the limitations of previous work are addressed, by providing a thorough, quantitative and analytical study of the propagation of image noise through a VFET algorithm.

Here, we derive expressions to calculate expected VFET reconstruction errors as a function of image noise. We use the three different RMS error metrics defined in Sec. 2.6 to quantify the total reconstruction error, and the errors in the direction and magnitude of the reconstructed vectors, separately. We compare the errors predicted using these analytical models with those measured from VFET reconstructions performed using simulated micrographs.

We close this introduction with a brief overview of the remainder of the chapter. In Sec. 5.2, we derive analytical expressions for VFET reconstruction errors as a function of image noise. In Sec. 5.3, we present errors calculated from tomographically reconstructed vector fields using simulated micrographs, and compare these with our analytical results. Section 5.4 explores the use of our analysis for comparing the noise-stability of VFET reconstructions using different sets of parameters. We make some concluding remarks in Sec. 5.5.
5.2 Propagation of shot noise through a vector field electron tomography algorithm

In the following sections, we derive expressions for the total and directional root-mean-square errors, as well as the root-mean-square error in magnitude (defined in Sec. 2.6).

5.2.1 Effect of shot noise on total root-mean-square errors

Ignoring other sources of error, we can express the reconstructed vector field at a given voxel as

\[ A_{\text{rec}}_{i,j,k} = A_{i,j,k} + A_{\text{noise}}_{i,j,k}, \]  

(5.1)

where \( A_{\text{noise}}_{i,j,k} \) is the noise-induced component of the vector field at \((i, j, k)\). Substituting Eq. (5.1) into Eq. (2.21), \( \langle \text{tot} E_{\text{noise}} \rangle \) can be obtained by calculating

\[ \langle \text{tot} E_{\text{noise}} \rangle = \sqrt{\sum_{i,j,k} \frac{|A_{\text{noise}}_{i,j,k}|^2}{\sum_{i,j,k} |A_{i,j,k}|^2}}, \]  

(5.2)

where \( \sigma_{\text{out}} \) is the standard deviation of the noise-induced reconstruction error and \( M \) is the number of voxels along each direction of the reconstruction. Equation (5.2) is valid when the reconstruction resolution \( M \) is large. In deriving an analytical noise model for \( \sigma_{\text{out}} \), we utilise the noise variance analysis of McDowell et al. [149]. In the case of phase retrieval using the TIE, the input signal is the longitudinal derivative, \( \partial_z I \), of the intensity across the image plane. The error in \( \partial_z I \) due to noise, when two defocused micrographs are used to approximate the derivative, is given by [114]

\[ \sigma'_{\text{in}} = \frac{\sigma_{\text{in}}}{\sqrt{2\Delta f}}. \]  

(5.3)

To determine \( \sigma_{\text{out}} \), we express the noise in the Fourier transform of the through-focal derivatives as a random complex number \( c_{l,j} \) with variance

\[ \langle c_{l,j}c_{l,j}^* \rangle = I_0^2 \sigma_{\text{in}}^2, \]  

(5.4)

where \( \langle \cdots \rangle \) denotes the expectation value. Note that, by virtue of being the Fourier transform of a real function (the noise in the through-focal derivatives), \( c_{l,j} \) has the
Hermitian property
\[ c_{l,j} = c_{-l,-j}, \] (5.5)

but this does not affect our derivation. In practice, a white noise model will not be sufficient to accurately predict errors from all sources of noise that may be present. For this reason we introduce the filter functions, \( H^\theta \) and \( H^\alpha \), to account for the spectrum of the image noise in each tilt series; if this spectrum is independent of defocus, these filters applied to the approximated through-focal derivatives are identical to those applied to the images used to calculate them. These functions can also incorporate any additional filtering that is performed in an effort to reduce the effects of noise on the reconstruction. Considering only errors due to noise, the Fourier transformed through-focal derivatives, constructed from experimental micrographs, are then given by

\[ \partial_z \tilde{I}_{l,j,\mu}^\theta = \partial_z \tilde{I}_{l,j,\mu}^{\text{ideal},\theta} + \frac{\pi}{e} H_{l,j,\mu}^\theta c_{l,j,\mu}^\theta, \] (5.6)
and
\[ \partial_z \tilde{I}_{l,j,\mu}^\alpha = \partial_z \tilde{I}_{l,j,\mu}^{\text{ideal},\alpha} + \frac{\pi}{e} H_{l,j,\mu}^\alpha c_{l,j,\mu}^\alpha, \] (5.7)

where \( \partial_z \tilde{I}_{l,j,\mu}^{\text{ideal},\theta} \) and \( \partial_z \tilde{I}_{l,j,\mu}^{\text{ideal},\alpha} \) are the noise-free components of \( \partial_z \tilde{I}^\theta \) and \( \partial_z \tilde{I}^\alpha \), respectively.

Noting the linearity of the reconstruction algorithm (Eq. (2.13)), and considering Eqs. (5.1), (5.6), and (5.7), we can immediately write down an expression for the noise-induced component of the reconstructed vector field:

\[ A_{\text{noise}}^{m,n,p} = \frac{\pi}{(n_t - 1)e} \sum_{\mu=0}^{n_t-1} \frac{1}{M^2} \sum_{l,j=-M/2}^{M/2-1} \eta_{l,j,\mu} c_{l,j}, \] (5.8)

where

\[ \eta_{l,j,\mu} = \left( H_{l,j,\mu}^\theta c_{l,j} e^{2\pi i (n' + j p)/M} + H_{l,j,\mu}^\alpha c_{l,j} e^{2\pi i (n'' + j m)/M} \right). \] (5.9)

Here, \( n' = m \cos \theta + n \sin \theta \), and \( n'' = n \cos \alpha + p \sin \alpha \). The variance \( \sigma_{\text{out}}^2 \) in each voxel of the reconstruction is the second moment of \( A_{\text{noise}} \):

\[ \sigma_{\text{out}}^2 = \frac{\pi}{(n_t - 1)e^2 q} \left[ \frac{1}{M^2} \sum_{\mu=0}^{n_t-1} \left( \sum_{l,j=-M/2}^{M/2-1} \eta_{l,j,\mu} c_{l,j} \right)^2 \right], \] (5.10)
where \( q \) is the number of phase maps, each corresponding to a single measurement of the total exit phase, used to obtain a single magnetic phase map (typically \( q = 2 \)), and

\[
\zeta_{l,j} = \frac{a^2 M^2 k}{I_0 4\pi^2 (l^2 + j^2)}
\]  

(5.11)

is the spatial frequency response of the phase retrieval algorithm under the phase object approximation. The noise is uncorrelated between phase maps both within, and between, tilt series, which leads to the result

\[
\sigma_{\text{out}}^2 = \frac{I_0^2 \sigma^2_{\text{in}} 2\pi^2 \hbar^2}{q M^4 (n_t - 1)^2 e^{2}} \sum_{\mu=0}^{n_t-1} \sum_{l,j=-M/2}^{M/2} H_{l,j}^2 \zeta_{l,j}^2 \left( |\eta_{l,j,\mu}^\theta|^2 + |\eta_{l,j,\mu}^\alpha|^2 \right),
\]

(5.12)

where we have made use of Eq. (5.4).

In practice, the spatial frequency spectrum of the noise may be a function of tilt series and angle, but for simplicity, we assume that it is a function only of spatial frequencies, and have removed these dependencies in \( H \). We treat the Poisson noise in our simulations as being approximately uniform, and set \( H = 1 \) in our analytical calculations.

The summations in Eq. (5.12) can be computed very quickly\(^1\), and require no knowledge of the properties of the sample. Any regularisation scheme can also be incorporated, which enables the effects of regularisation on errors due to noise to be calculated.

5.2.2 Effect of shot noise on directional root-mean-square errors

In this section we derive approximations for the contribution to \( \langle \text{dir} \rangle \) due to noise. Substituting Eq. (5.1) into Eq. (2.24), the directional error is given by

\[
\langle \text{dir} \rangle_{\text{noise}} = \sqrt{\frac{1}{M^3 2\pi} \sum_{i,j,k} \left[ \cos^{-1} \left( \frac{|A_{i,j,k}| + \hat{A}_{i,j,k} \cdot A_{i,j,k}^{\text{noise}}}{|A_{i,j,k} + A_{i,j,k}^{\text{noise}}|} \right) \right]^2},
\]

(5.13)

\(^1\)The analytical error surfaces shown later in this chapter take approximately ten seconds to compute, each, on a single core of an Intel\textsuperscript{®} i5-4670K processor at stock speed (3.4 GHz). On the same hardware, the numerical results, which will be discussed in Sec. 5.3, take several days.
where the circumflex on $A$ is used to denote a unit vector. The magnitude of the reconstructed vector can be determined using the cosine rule, and is given by

$$|A_{i,j,k} + A_{i,j,k}^{\text{noise}}| = |A_{i,j,k}| \sqrt{1 + \frac{|A_{i,j,k}^{\text{noise}}|^2}{|A_{i,j,k}|^2} + \frac{2 \cdot \hat{A}_{i,j,k} \cdot A_{i,j,k}^{\text{noise}}}{|A_{i,j,k}|}}.$$  

Substituting this into Eq. (5.13), and using the binomial approximation, gives

$$\langle \text{dir} \ E_{\text{noise}} \rangle = \sqrt{\frac{1}{M^3 \pi^2} \sum_{i,j,k} \{\cos^{-1} \varpi\}^2},$$  

where

$$\varpi = \left(1 + \frac{\hat{A}_{i,j,k} \cdot A_{i,j,k}^{\text{noise}}}{|A_{i,j,k}|} \right) \left(1 - \frac{|A_{i,j,k}^{\text{noise}}|^2}{2|A_{i,j,k}|^2} - \frac{\hat{A}_{i,j,k} \cdot A_{i,j,k}^{\text{noise}}}{|A_{i,j,k}|} \right).$$  

We now introduce the notation:

$$R_{i,j,k} \equiv \frac{A_{i,j,k} \cdot A_{i,j,k}^{\text{noise}}}{|A_{i,j,k}| |A_{i,j,k}^{\text{noise}}|}.$$  

Here $R_{i,j,k}$ is a random variable on the interval $[-1, 1]$ which, if the orientation of $A_{i,j,k}^{\text{noise}}$ is uniformly random, has a probability density given by

$$p(R_{i,j,k}) = \frac{1}{\pi} \cdot \left| \frac{d}{dx} \cos^{-1} x \right|_{x=R_{i,j,k}} = \begin{cases} \frac{1}{\pi \sqrt{1 - R^2_{i,j,k}}} & -1 < R_{i,j,k} < 1 \\ 0 & \text{elsewhere} \end{cases}.$$  

Discarding terms of $\mathcal{O}(|A_{i,j,k}^{\text{noise}}|^3)$ in Eq. (5.15) gives

$$\langle \text{dir} \ E_{\text{noise}} \rangle = \sqrt{\frac{1}{M^3 \pi^2} \sum_{i,j,k} \left\{\cos^{-1} \left[ -\frac{R^2_{i,j,k} + 1}{\frac{|A_{i,j,k}^{\text{noise}}|^2}{|A_{i,j,k}|^2}} \right] \right\}^2},$$  

and the approximation

$$\cos^{-1}(1 - x) \approx \sqrt{2x},$$  

valid for small $x$ (i.e. low noise levels), simplifies Eq. (5.19) to

$$\langle \text{dir} \ E_{\text{noise}} \rangle = \sqrt{\frac{1}{M^3 \pi^2} \sum_{i,j,k} \left| A_{i,j,k}^{\text{noise}} \right|^2 \left( \frac{\sqrt{2R^2_{i,j,k} + 1}}{|A_{i,j,k}|} \right)^2}.$$  

The rms value of $|A_{i,j,k}^{\text{noise}}|$ is $\sigma_{\text{out}}$, while that of $\sqrt{2R_{i,j,k}^2 + 1}$ is $\sqrt{2}$, and because these are assumed to be independent, they can be evaluated separately to give

$$\langle \text{dir} E_{\text{noise}} \rangle = \sqrt{\frac{2}{M^3\pi^2}} \sum_{i,j,k} \frac{\sigma_{\text{out}}^2}{|A_{i,j,k}|^2}$$

$$\approx \sqrt{\frac{2}{\pi}} \langle \text{tot} E_{\text{noise}} \rangle,$$  \tag{5.22}

where we have made use of the approximation

$$\frac{1}{M^3} \sum_{i,j,k} \frac{\sigma_{\text{out}}^n}{|A_{i,j,k}|^n} \approx \langle \text{tot} E_{\text{noise}} \rangle^n,$$  \tag{5.23}

which is valid when $|A_{i,j,k}|$ is approximately constant for all $i, j, k$. This requirement implies that Eq. (5.22) may be inaccurate for geometries where the magnitude of the vector potential varies significantly over the domain of reconstruction. For example, a pair of uniformly magnetised particles with antiparallel moments has large $|A_{i,j,k}|$ near each particle, but decays quickly outside the specimen, and directional errors calculated in this way may be significantly less accurate than for a single dipole.

Because we have dropped higher order terms in the derivation of $\langle \text{dir} E_{\text{noise}} \rangle$, the accuracy of Eq. (5.22) in approximating noise-induced errors is expected to be poor for large $\sigma_{\text{in}}/\Delta f$. For accurate results in this region, the $O(|A_{i,j,k}^{\text{noise}}|^3)$ terms in Eq. (5.15) can be retained, to give the improved approximation

$$\langle \text{dir} E_{\text{noise}} \rangle = \frac{\langle \text{tot} E_{\text{noise}} \rangle}{\pi} \sqrt{2 + \langle \text{tot} E_{\text{noise}} \rangle / 2},$$  \tag{5.24}

where we have made use of Eq. (5.23).

### 5.2.3 Effect of shot noise on root-mean-square errors in magnitude

Substituting Eq. (5.14) into Eq. (2.26), we get

$$\langle \text{mag} E_{\text{noise}} \rangle = \sqrt{\sum_{i,j,k} |A_{i,j,k}|^2 \left[ (1 + |A_{i,j,k}^{\text{noise}}|^2 / |A_{i,j,k}|^2 + 2R_{i,j,k} |A_{i,j,k}^{\text{noise}}| / |A_{i,j,k}|)^{1/2} - 1 \right]^2} / \sum_{i,j,k} |A_{i,j,k}|^2.$$  \tag{5.25}
Using the binomial approximation again, and discarding terms of $O(∥A_{i,j,k}^{\text{noise}}∥^2)$, gives

$$\langle \text{mag} E_{\text{noise}} \rangle = \sqrt{\sum_{i,j,k} R_{i,j,k}^2 A_{i,j,k}^{\text{noise}} |A_{i,j,k}|^2}.$$

$$= \sigma_{\text{out}} \sqrt{\sum_{i,j,k} R_{i,j,k}^2 \sum_{i,j,k} |A_{i,j,k}|^2}.$$

$$= \langle \text{tot} E_{\text{noise}} \rangle \sqrt{2}.$$

(5.26)

5.3 Comparison of reconstruction errors – simulations and analytical results

Here, we are concerned with the contribution to the RMS errors in the reconstructed vector potential due only to image noise. For the purposes of comparing analytical and numerical results, the noise-induced component $\langle \text{tot} E_{\text{noise}} \rangle$ of the normalised total RMS error $\langle \text{tot} E \rangle$ (defined in Sec. 2.6) can be extracted from simulations using

$$\langle \text{tot} E \rangle_{\text{noise}} = \sqrt{\langle \text{tot} E \rangle^2 - \langle \text{tot} E_0 \rangle^2},$$

(5.27)

where $\langle \text{tot} E_0 \rangle$ is the value of $\langle \text{tot} E \rangle$ when $\sigma_{\text{in}} = 0$. The noise-induced components of the directional and magnitude errors, $\langle \text{dir} E_{\text{noise}} \rangle$ and $\langle \text{mag} E_{\text{noise}} \rangle$, respectively, can be obtained in the same manner; i.e.,

$$\langle \text{dir} E_{\text{noise}} \rangle = \sqrt{\langle \text{dir} E \rangle^2 - \langle \text{dir} E_0 \rangle^2},$$

(5.28)

and

$$\langle \text{mag} E_{\text{noise}} \rangle = \sqrt{\langle \text{mag} E \rangle^2 - \langle \text{mag} E_0 \rangle^2}.$$  

(5.29)

We now present analytical results for reconstruction errors, based on the analysis presented in Sec. 5.2, and compare these with the errors in reconstructions performed on simulated TEM data, extracted using Eqs. (5.27)-(5.29). There are a multitude of parameters that affect reconstruction errors, some of which are outlined in Ch. 3. Here, we restrict our attention to a small number of parameters. In this section, we address the variation of two parameters of significance; namely, defocus and image noise. Later, in Sec. 5.4, we investigate the effect of changing sampling and regularisation parameters.

For our simulations, the sample shape was measured from experimental scalar tomograms of an octahedral magnetite nanoparticle. The nanoparticles were produced by K. Spiers [150], with the tomography performed by T. Petersen [151]. For further details, see Ref. [148].
constructed by tracing the surface of the particle in three dimensions using 3D modelling software (Blender 2.67). The shape of the simulated sample, along with the experimental tomograms from which it was constructed, is shown in Fig. 5.1.

![Figure 5.1](image.png)

Figure 5.1: Shape of sample mask used for our simulations (gray) and surface point cloud obtained using scalar tomography [148,151] applied separately to two orthogonal tilt series (pink and green, respectively). Points outside the particle result from errors caused by the carbon support. Four different orientations are shown.

A mean inner potential of 17 V [152], mass magnetisation of 80 emu/g [153], and density of 5.18 g/cm³ [154], were chosen to simulate a magnetite sample. An imaginary potential of 0.8i V, inferred from the experimental micrographs of the sample being simulated, was added to the mean inner potential to simulate attenuation. These parameters were used to calculate total phase shifts using the projection approximation, and defocused images were then obtained by means of a transfer function [155]. Shot noise of the form given by Eq. (4.13) was then added to the simulated micrographs.

Figure 5.2 shows a through-focal derivative computed from simulated micrographs, along with one computed from experimental micrographs of the actual sample.

Underfocus and overfocus micrographs were simulated for every θ and α, with M = 64 and nt = 31. After the addition of noise, the micrographs were used to retrieve the phases that comprise Tθ and Tα. The vector potential A(r) was
 CHAPTER 5. EFFECT OF IMAGE NOISE

Figure 5.2: Example of through-focal derivatives computed from micrographs with \( \Delta f = -10 \mu m \) and \( \sigma_{in} = 1\% \) using (a) simulated micrographs, and (b) experimental micrographs. 

reconstructed using Eq. (2.13), and then used to calculate \( \langle \text{tot}_E \rangle_{\text{noise}}, \langle \text{dir}_E \rangle_{\text{noise}}, \) and \( \langle \text{mag}_E \rangle_{\text{noise}} \). These simulations were performed for a range of \( \sigma_{in} \) and \( \Delta f \). Each simulation was performed ten times for each \( \sigma_{in} \) and \( \Delta f \), with the results averaged, to suppress fluctuations about the mean RMS error for large \( \sigma_{in} \). The \( x \)-component of the exact vector potential is shown alongside that of the reconstructed vector potentials, for various combinations of \( \Delta f \) and \( \sigma_{in} \), in Fig. 5.3.

The predictive power of our analysis is quantified using a normalised residual metric defined by

\[
\chi^\text{tot} = \left( \langle \text{tot}_E \rangle_{\text{noise}} - \langle \text{tot}_E \rangle \right) / \langle \text{tot}_E \rangle_{\text{noise}},
\]

(5.30)

where the macron denotes the analytically derived estimate. For the accuracy of \( \langle \text{dir}_E \rangle_{\text{noise}} \) and \( \langle \text{mag}_E \rangle_{\text{noise}} \), the metrics \( \chi^\text{dir} \) and \( \chi^\text{mag} \) are similarly defined.

Figure 5.4 shows \( \langle \text{tot}_E \rangle_{\text{noise}} \) and \( \langle \text{tot}_E \rangle_{\text{noise}} \), respectively, as contour plots over \( \sigma_{in} \) and \( \Delta f \). It is evident that increasing the image noise will result in increased reconstruction errors, as the errors due to noise in the micrographs propagate through the reconstruction algorithm and translate to noisy reconstruction data. The reduction in error as the defocus increases is less intuitive. This occurs because phase contrast in the micrographs increases with increasing defocus, provided that the defocus is not too large in magnitude. When \( \sigma_{in} \) remains constant, the ratio of the noise level to the phase contrast signal drops as the defocus is increased, which results in a reduced noise-induced error in the phase [114]. It is this error that then propagates to the reconstructed vector field.
5.3. SIMULATIONS AND ANALYTICAL RESULTS

![Image of vector potential slices with varying parameters]

<table>
<thead>
<tr>
<th>$\Delta f$ $(\text{µm})$</th>
<th>$\sigma_{in} = 0$</th>
<th>$\sigma_{in} = 2.5%$</th>
<th>$\sigma_{in} = 5%$</th>
</tr>
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<tr>
<td>5</td>
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<td><img src="image2.png" alt="Image slice" /></td>
<td><img src="image3.png" alt="Image slice" /></td>
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<td>25</td>
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<td>50</td>
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<td><img src="image8.png" alt="Image slice" /></td>
<td><img src="image9.png" alt="Image slice" /></td>
</tr>
</tbody>
</table>

Figure 5.3: A one voxel ($\sim 5\text{ nm}$) thick slice of the $x$-component of the exact vector potential at $z = 0$ (top), and the same slice taken from reconstructions using various values of $\Delta f$ and $\sigma_{in}$. 
Figure 5.4: Contour plots of $\langle \text{tot} E_{\text{noise}} \rangle$ as a function of $\sigma_{\text{in}}$ and $\Delta f$ for (a) reconstructions using simulated data, in which the phase was obtained using Eq. (2.5), and (b) analytical results obtained using Eqs. (5.2) and (5.12). The numbers given on the contour lines are total RMS errors expressed as a percentage.

The contour plots in Fig. 5.4 are presented to provide an overview of how the total reconstruction error depends on image noise and defocus, in both the simulated and analytical results. For the results that follow, we use 3D surface plots to represent the error data, because they provide a clearer comparison between simulated and analytical results, and serve to better highlight the regions in which these results deviate from each other.

Figure 5.5 shows surface plots comparing the analytically derived estimates $\langle \text{tot} E_{\text{noise}} \rangle$, obtained using Eq. (5.12) in conjunction with Eq. (5.2), with $\langle \text{tot} E_{\text{noise}} \rangle$ obtained from the reconstruction of simulated micrographs by applying Eqs. (2.21) and (5.27). These represent the same data presented in Fig. 5.4, with the addition of simulated results where the reconstruction relied on the phase object approximation.

Because $I_{\text{ideal}}^{i,j}$ varies across the image, the noise level is not, in practice, constant. However, for the analytical calculations presented here, $H_{i,j}$ is set to unity for simplicity, approximating the noise as uniformly distributed.

The strong agreement between analytical results and simulations shows that Eq. (5.12) can be used to accurately predict RMS errors from the signal-to-noise ratio of transmission electron micrographs, and can be used to determine minimum experimental acquisition times required to keep the reconstruction errors due to noise below a desired limit. It can also be used to quickly compute the noise-induced errors for a variety of experimental parameters. These results also show that, under the conditions used in these simulations, shot noise is adequately modelled as white noise.
5.3. SIMULATIONS AND ANALYTICAL RESULTS

Figure 5.5: (a) Analytically derived values $\langle \bar{E}_{\text{noise}} \rangle$ (red) obtained using Eqs. (5.2) and (5.12), along with the results of simulations $\langle E_{\text{noise}} \rangle$ using Eq. (2.5) for the phase retrieval (blue) and Eq. (2.6) (green). (b) Normalised residual $\chi_{\text{tot}}$ plotted against $\sigma_{\text{in}}/\Delta f$ for each method (blue and green dots, respectively).

Figures 5.6(a) and 5.6(c) show a comparison of $\langle E_{\text{noise}} \rangle$ and $\langle \bar{E}_{\text{noise}} \rangle$, calculated using Eq. (5.22). The analytical results agree well with the simulations for small $\sigma_{\text{in}}/\Delta f$ but, as expected, deviate for larger $\sigma_{\text{in}}/\Delta f$. Figures 5.6(b) and 5.6(d) show that the analytical estimates can be significantly improved by employing Eq. (5.24).

The results for $\langle E_{\text{noise}} \rangle$ are shown in Fig. 5.8, again highlighting that there is good agreement between the analytical results and those obtained by reconstructing the vector potential from simulated micrographs.

The total errors $\langle E_{\text{tot}} \rangle$, $\langle E_{\text{dir}} \rangle$, and $\langle E_{\text{mag}} \rangle$ are shown as surface plots in Fig. 5.7. These can be compared with the extracted noise induced errors shown in Figs. 5.5, 5.6, and 5.8, to provide insight into the significance of the noise induced component of the total error, in relation to the combined contribution of all other sources of error, at various noise levels and defoci.
Figure 5.6: Analytically derived values $\langle \text{dir} E_{\text{noise}} \rangle$ (red), along with the results of simulations $\langle \text{dir} E_{\text{noise}} \rangle$ using Eq. (2.5) for the phase retrieval (blue), and Eq. (2.6) (green). (a) Using the approximation given by Eq. (5.22), and (b) using the higher order approximation given by Eq. (5.24). (c) and (d) are the normalised residuals $\chi_{\text{dir}}$ plotted against $\sigma_{\text{in}}/\Delta f$ for each approximation, respectively, showing both the results obtained using Eqs. (2.5) (blue) and (2.6) (green).
Figure 5.7: Surface plots showing the reconstruction errors (a) $\langle \text{tot} E \rangle$, (b) $\langle \text{dir} E \rangle$, and (c) $\langle \text{mag} E \rangle$. Each subfigure shows both the results obtained using Eq. (2.5) for the phase retrieval (blue) and Eq. (2.6) (green).
Figure 5.8: (a) Analytically derived values $\langle \text{mag} E_{\text{noise}} \rangle$ obtained using Eq. (5.26) (red), along with the results of simulations $\langle \text{mag} E_{\text{noise}} \rangle$ using Eq. (2.5) for the phase retrieval (blue), and Eq. (2.6) (green). (b) Normalised residuals $\chi_{\text{mag}}$ plotted against $\sigma_{\text{in}}/\Delta f$ for each method (blue and green dots, respectively).

5.4 Comparison of noise-induced errors using different reconstruction parameters

Normalisation of calculated RMS errors for a given reconstruction method requires knowledge of the RMS value of the vector potential. This limits the usefulness of this technique in determining the accuracy of a given method for a particular sample. However, in comparing the accuracy of two algorithms or regularisation schemes used to reconstruct the same sample, the relative errors are not dependent on the RMS value of the vector field being reconstructed, so these comparisons can be made without any a priori knowledge of the sample. From Eq. (5.2), we have

$$\frac{\langle \text{tot} E \rangle}{\langle \text{tot} E \rangle'} = \frac{\sigma_{\text{out}}}{\sigma'_{\text{out}}},$$

where the primed symbols indicate a different reconstruction parameter set to the unprimed symbols. In this section, we explore the use of Eq. (5.31) for comparing the propagation of noise through the FBP algorithm using different reconstruction parameters.

We determine the relative errors of different reconstruction methods applied to simulated micrographs, and compare these results to the analytical predictions. The sample chosen for these simulations is a uniformly magnetised magnetite specimen.
5.4. COMPARISON OF ERRORS USING DIFFERENT PARAMETERS

Figure 5.9: Simulated micrograph of the sample used for comparison of reconstruction methods (left) and an experimental image of a magnetite nanoparticle, observed using an FEI Tecnai T20, equipped with an LaB$_6$ source, operating at 200 kV (right). The edge contrast on the simulated micrograph is a result of the negative defocus. The experimental image was recorded near Gaussian focus.

(as in Sec. 5.3) oriented in the $z$—direction. An irregular shape (shown in Fig. 5.9) was chosen for these simulations. In the remainder of this section, we present results showing the effect on the accuracy of reconstructions when altering regularisation parameters and sampling, respectively, and compare these results with those predicted.

5.4.1 Effect of changing regularisation parameters

To compare results for reconstructions using different regularisation parameters, RMS errors were calculated for two different reconstructions; the first (which we will refer to as “method 1”) using $\delta_{\text{TIE}} = 0.1\text{px}^{-1}$ and $\delta_{\text{FBP}} = 0.0001$, and the second (which we will refer to as “method 2”) using $\delta_{\text{TIE}} = 1\text{px}^{-1}$ and $\delta_{\text{FBP}} = 0.01$. Here, the units, $\text{px}^{-1}$, are inverse pixels. Both reconstructions were repeated for shot noise levels in the simulated micrographs in the range $0\% - 5\%$. The noise-induced errors were extracted from the total RMS errors using the method discussed in Sec. 5.2. The errors for method 1 were then plotted against the those for method 2, and a least squares linear regression [156] used to find the slope. The exact and reconstructed magnetic phases for both methods are shown in Fig. 5.10, with the orientation chosen to provide the maximum magnetic phase contrast. The comparison of the errors is shown in Fig. 5.11.

The experimental and reconstruction parameters used in the simulations were
CHAPTER 5. EFFECT OF IMAGE NOISE

Figure 5.10: Comparison of exact and retrieved phase using methods 1 and 2, respectively. The grayscale range of each subfigure is normalised separately, so no colour bar is provided. Note that the micrographs are shown with the electrostatic component removed to provide a direct comparison of the magnetic signal strength and the noise.
then employed in Eq. (5.12) to obtain the theoretical expected value for the slope in Fig. 5.11. The results show that the theoretical calculation can provide accurate predictions of the relative contributions of noise to the total RMS errors for different regularisation parameters. However, because this does not take into account sources of error other than noise, the errors incurred due to increasing $\delta_{\text{TIE}}$ and $\delta_{\text{FBP}}$, discussed in Sec. 2.5, cannot be accounted for here.

![Graph showing normalised RMS errors using method 1 ($\delta_{\text{TIE}} = 0.1$ and $\delta_{\text{FBP}} = 0.0001$), and method 2 ($\delta_{\text{TIE}} = 1$ and $\delta_{\text{FBP}} = 0.01$). The theoretical model produces a slope that agrees well with the simulations. Here, we assume that the line passes through the origin because the noise-induced component of the error is zero, for any reconstruction method, when the micrographs are noise-free.](image)

Figure 5.11: Plot of normalised RMS errors using method 1 ($\delta_{\text{TIE}} = 0.1$ and $\delta_{\text{FBP}} = 0.0001$), and method 2 ($\delta_{\text{TIE}} = 1$ and $\delta_{\text{FBP}} = 0.01$). The theoretical model produces a slope that agrees well with the simulations. Here, we assume that the line passes through the origin because the noise-induced component of the error is zero, for any reconstruction method, when the micrographs are noise-free.

### 5.4.2 Effect of changing spatial and angular sampling

Also of interest in Eq. (5.31) are the effects of changing the number of tilt angles $n_t$ in each tilt series [157] and the pixel-width $M$ of the reconstruction grid. Because these variables occur in the limits of the summations as well as the prefactor, it is difficult to intuit the effect that changing them has on the noise-induced error. The simulations performed in Sec. 5.4.1 were repeated, this time keeping the regularisation parameters the same as method 1, but using a different combination of $n_t$ and $M$. We refer to this as method 3.

Projections of the exact and reconstructed vector potentials using methods 1 and 3 are shown in Fig. 5.12. The results of these simulations, along with those predicted by Eq. (5.31), are shown in Fig. 5.13. The theoretical model for the propagation of
noise through the reconstruction algorithm accurately predicts the noise-induced component of the total RMS error.

Figure 5.12: Projections of exact and reconstructed vector potentials using methods 1 and 3, with $\sigma_m = 1\%$ and $\sigma_m = 5\%$.

The images of the vector potential in Fig. 5.12 show that the reduction in angular and spatial sampling in method 3 has little effect on the appearance of the vector potential, other than the obvious pixelation due to the poorer spatial sampling. However, Fig. 5.13 shows that the total RMS error in the poorly sampled reconstruction is approximately twice that of the well sampled reconstruction.
5.5. IMPLICATIONS

5.5 Implications

Shot noise is an unavoidable source of error in any imaging process. In VFET, shot noise in the micrographs results in errors in both the magnitude and direction of the reconstructed vector potential. An understanding of the relationship between image noise and the accuracy of VFET reconstructions provides various benefits. For a particular application, there may be a maximum tolerable error in a tomographic reconstruction. Understanding the propagation of noise through the reconstruction algorithm, combined with an understanding of other sources of error, will provide a maximum allowable noise level in the micrographs. This in turn will enable determination of a minimum acquisition time per image, allowing for minimal sample degradation and microscope time.

The results presented in Secs. 5.3 and 5.4 show that the propagation of noise in VFET can be accurately quantified for a given reconstruction algorithm, including any regularisation scheme, and a known noise level and spatial frequency spectrum. For our simulations, we have used a shot noise model in which the variance of the noise at each pixel in each image used in the reconstruction is dependent on the intensity of the electron beam. For this reason, the spatial dependence of the noise level is also a function of tilt angle, so the power spectrum of the noise is, in practice, a complicated function of spatial frequencies and tilt angle. However, we have shown shot noise to be adequately modelled as white noise for the purposes of analysing...
propagation of errors in the reconstruction. There are other factors that can be
taken into account in determining a value for the noise filters used for analytical
calculations of VFET reconstruction errors, in cases where setting it to unity is not
sufficient. One possible example is the blurring of shot noise in the detector, which
is commonly modelled using a modulation transfer function [158] or noise transfer
function [159], which could be incorporated into these filter functions. A filtered
noise model may be useful for determining the contribution of other stochastic error
sources, such as thermal detector noise, and the power spectral density of these noise
sources would typically be independent of tilt series and tilt angle, allowing a simple
two-dimensional filter to be used in the noise analysis.

With modification, the noise analysis presented here can be used for many other
calculations of noise-induced errors in reconstructions where linear, Fourier trans-
form methods have been employed. This includes, but is not limited to, those arising
from other VFET algorithms, as well as scalar tomography and phase retrieval algo-

rithms.

We have derived expressions for the total, directional, and magnitude RMS noise-
induced errors in vector fields reconstructed from transmission electron micrographs
using an FBP algorithm. The results of simulations presented here show that noise-
induced errors can be quickly computed, to accurately estimate their contribution
to the total RMS errors in a VFET reconstruction, and that these analytical estimates
agree well with numerical simulations under a wide range of conditions.
Chapter 6

Effect of specimen orientation on the accuracy of vector field electron tomography

6.1 Introduction

The defining feature of a vector field is that, in addition to its magnitude, it has a direction associated with each point in space. The magnitude of the projected magnetic vector potential, and hence the magnetic phase contrast, changes as the sample is rotated through at least one of the tilt series, regardless of the geometry of the specimen. By way of example, Fig. 6.1 shows the magnetic phase (computed using the projection approximation) being held approximately constant throughout the $\theta$-tilt series, by initially orienting the magnetisation in the $z$-direction. The consequent large variation in magnetic phase contrast, as the particle is rotated through the $\alpha$-tilt series, is illustrated in Fig. 6.2.

For a reconstruction algorithm employing two tilt series, the initial orientation of a uniformly magnetised particle can be chosen to prevent orientations of zero magnetic contrast occurring in either of the tilt series; however, this necessarily prevents the acquisition of micrographs at angles of maximal magnetic contrast if the tilt series are orthogonal. Therefore, the choice of magnetisation orientation is a compromise between these two competing error sources. Although the effect is more complicated for a specimen with a non-uniform magnetisation, for many specimens the multipole expansion of the magnetisation will contain a dominant dipole moment. In such cases, the specimen can be considered to possess a well-defined direction given by this dipole moment. The principles we discuss here regarding magnetic dipoles and, more generally, uniformly magnetised particles, are relevant
In this chapter, we aim to quantify the effect that specimen orientation has on the resulting errors for a reconstruction algorithm utilising two orthogonal tilt series. In Sec. 6.2 we demonstrate how the concept of contrast can be used to quantify the error contribution from a single phase map due to specimen orientation. In Sec. 6.3 we use these results to derive equations that explain the changes in reconstruction error as a function of initial specimen orientation, and in Sec. 6.4 we present results of simulations which we compare with the analytical results. Finally, in Sec. 6.6, we discuss some of the implications of this work for electron microscopists, who employ VFET techniques for the reconstruction of the vector potential of magnetic nanoparticles.
6.2. ORIENTATION-DEPENDENT ERRORS

Figure 6.2: Magnetic phase (gray) induced in the electron beam (yellow) as the particle, with magnetisation (red) initially oriented in the $z$-direction, is rotated through the $\alpha$-tilt series (shown here starting at $\alpha = \pi/2$). This produces a large variation in the magnetic phase.

6.2 Orientation-dependent errors

In VFET, magnetic phase contrast in the micrographs is the source of the information that is used to reconstruct the vector field. Because the projection of the magnetic vector potential changes with specimen orientation, the magnetic phase contrast varies as the specimen is rotated through each tilt series. The phase maps used in the filtered backprojection reconstruction algorithm (see Eq. (2.10)) are not uniformly distributed over the sphere. Because of this, the average magnetic phase contrast can vary with initial orientation of the specimen.

If the errors in the phase maps are small, maximising the average contrast in the micrographs minimises the error in the reconstruction. However, when the errors are significant (e.g., for large defocus), the increased contrast results in an increase in the reconstruction error.

In the remainder of this section, we describe the choice of coordinate system used for the error analysis, and discuss these competing sources of orientation-dependent errors.
6.2.1 Coordinate system used in the error analysis

In our simulations, the two tilt series are acquired as the specimen is rotated around the $x$- and $z$-axes, respectively. Consequently, it is natural to use spherical coordinates with the pole aligned with the $-y$ direction for describing the magnetisation direction. In this work, we define our coordinate system such that the polar angle is given by

$$\gamma = \arccos\left(\frac{-y}{\sqrt{x^2 + y^2 + z^2}}\right),$$

and the azimuthal angle is given by

$$\gamma' = \arctan\left(\frac{z}{x}\right).$$

The geometry of the tilt series acquisition, as well as the corresponding coordinate system given by Eqs. (6.1) and (6.2), is shown in Fig. 6.3. For consistency with previous work [85,148], the reconstruction algorithm we use involves a different electron propagation direction for each of the tilt series, but for visual clarity, we show only one direction in our diagrams. The two methods are equivalent, amounting only to a different start and end point in the $\alpha$-tilt series. Note, also, that the chosen coordinate system for the tilt series acquisition is arbitrary, and our analysis

\[\gamma = \arccos\left(\frac{-y}{\sqrt{x^2 + y^2 + z^2}}\right),\]

\[\gamma' = \arctan\left(\frac{z}{x}\right).\]

Figure 6.3: (a) Geometry used for the acquisition of tilt series. In the $\alpha$ series, the object $O$ is rotated about the $x$-axis and, in the $\theta$ series, it is rotated about the $z$-axis. (b) Spherical polar coordinate system used to describe magnetisation directions. Here $\mathbf{m}(\gamma, \gamma')$ is the magnetisation vector of the particle, which is assumed to be uniformly magnetised, $\gamma$ is the polar angle measured from the negative $y$-axis, and $\gamma'$ is the azimuthal angle, measured from the $x$-axis to the projection of $\mathbf{m}$ onto the $x-z$ plane.
is valid for any reconstruction using two orthogonal tilt series, provided that $\gamma$ and $\gamma'$ are defined relative to the orientation of the tilt series in the given coordinate system.

### 6.2.2 Image contrast as a source of error

In vector tomography, for each vector in the field to be reconstructed, there are orientations for which this vector contributes no contrast in the projection. For a dipole, under the projection approximation, the image contrast vanishes if the moment direction is parallel to the optic axis of the TEM, and contributes the maximum possible contrast if it is orthogonal. In the remainder of this section, we derive an expression for the magnetic phase contrast in terms of moment orientation for a dipole, which we use in Sec. 6.3 to obtain an expression for the rms reconstruction error in terms of the magnetisation direction.

The phase object approximation is valid when the electron wavelength is significantly shorter than the characteristic length scale of the sample [85]. Under this approximation, $I_0$ is constant at $z = 0$, and Eq. (2.3) becomes [160]:

$$-k \frac{\partial I_0}{\partial z} = I_0 \nabla^2 \phi_0.$$  \hspace{1cm} (6.3)

Utilising a forward difference approximation for $\partial I/\partial z$, the tie under the phase object approximation (Eq. (6.3)) can be written

$$\frac{I^+ - I_0}{\Delta f} = - \frac{1}{k} \nabla^2 \phi_m.$$  \hspace{1cm} (6.4)

This gives us an expression for the overfocus intensity:

$$I^+ = -\frac{\Delta f}{k} \nabla^2 \phi_m + I_0.$$  \hspace{1cm} (6.5)

For our purposes, we use the rms definition of contrast [161,162]. In the context of VFET, we are concerned with the rms contrast in the magnetic component of the defocused micrographs, which is given by

$$K = \sqrt{\frac{1}{I_0^2 (M^2 - 1)} \sum_{i,j} (I^+_{i,j} - I_0)^2}.$$  \hspace{1cm} (6.6)

$$= \sqrt{\frac{1}{I_0^2 (M^2 - 1)} \sum_{i,j} \left[ \frac{\Delta f}{k} \nabla^2 \phi_m (r_{\perp}) \right]^2},$$  \hspace{1cm} (6.7)

where $M$ is the width of the micrograph in pixels. Here, we have made use of the fact that, under the phase object approximation, $I_0$ is a constant for $z = 0$, and is equal
CHAPTER 6. EFFECT OF SPECIMEN ORIENTATION

to the mean value of $I^+$. This latter property is a consequence of the conservation of intensity [78, 112]. The phase shift induced in the electron wavefunction by a magnetic dipole is given by

$$\varphi_m = -\frac{e}{\hbar} \int A \cdot d\mathbf{z},$$

$$= -\frac{\mu_0 e}{2\pi\hbar} \frac{(\mathbf{m} \times \mathbf{r}) \cdot \mathbf{z}}{x^2 + y^2},$$

where $\mu_0$ is the permeability of free space, and $\mathbf{m}$ is the magnetic dipole moment.

Equation (6.9) describes the phase shift of a spatially infinitesimal (i.e., point-like) magnetisation density, and the Laplacian of $\varphi_m$ vanishes. To model the magnetic phase contrast arising from a small, but finite, uniformly magnetised particle, we add a regularisation parameter $\tau$ to the analytic form of the phase, giving

$$\nabla^2(\varphi_m) = -\frac{\mu_0 e}{2\pi\hbar} \nabla^2 \left( \frac{\mathbf{z} \cdot \mathbf{m}}{x^2 + y^2 + \tau^2} \right),$$

$$= \frac{4\mu_0 e}{\pi\hbar} \left( \frac{\mathbf{z} \cdot \mathbf{m}}{(x^2 + y^2 + \tau^2)^3} \right),$$

where $\mathbf{r} \equiv (x, y, z)$ is the position vector. In terms of the magnetisation, utilising Eqs. (6.7) and (6.11), the contrast is given by

$$K = \frac{4\mu_0 e \Delta f |\mathbf{m}|}{\pi\hbar k I_0 \sqrt{M^2 - 1}} \sqrt{\sum_{i,j} \left( \frac{\mathbf{z} \cdot \mathbf{m}}{x^2 + y^2 + \tau^2} \right)^2},$$

$$= \frac{4\mu_0 e \tau^2 \Delta f |\mathbf{m}| |\mathbf{z} \times \hat{\mathbf{m}}|}{\pi\hbar k I_0 \sqrt{M^2 - 1}} \sqrt{\sum_{i,j} \left( \frac{\cos \xi}{(x^2 + y^2 + \tau^2)^3} \right)^2},$$

where $\mathbf{r}_\perp$ is the vector projection of $\mathbf{r}$ onto the $x$-$y$ plane, and $\xi$ is the angle between $\mathbf{z} \times \hat{\mathbf{m}}$ and $\mathbf{r}_\perp$. Because the contrast is low far away from the dipole, we can approximate Eq. (6.13) by constraining the summation to a circular region of radius $M/2$ pixels. Under this approximation, $\cos^2 \xi$ takes on all values in the range $[0, 1]$, irrespective of the direction of $\mathbf{m}$, and we can write

$$K \propto |\mathbf{z} \times \hat{\mathbf{m}}|.$$  

We use a normalised error metric to quantify the accuracy of the reconstructed vector potential, and expect this to be a function of the fractional RMS contrast $K/K_{\text{max}}$, where $K_{\text{max}}$ is the maximum value $K$ can take as we vary $\hat{\mathbf{m}}$, i.e., the
6.3. DERIVATION OF ERROR ESTIMATES

value of $K$ with $|\hat{z} \times \hat{m}| = 1$. Using Eq. (6.14), we can then express the fractional mean square contrast as

$$\frac{K^2}{K_{\text{max}}^2} = |\hat{z} \times \hat{m}|^2$$  \hspace{1cm} (6.15)

$$= 1 - (\hat{m} \cdot \hat{z})^2.$$  \hspace{1cm} (6.16)

6.2.3 Truncation error due to the finite difference approximation

In order to calculate the longitudinal derivative of the intensity for use in the TIE, a finite difference approximation must be employed. This results in a truncation error in the derivative, which contributes errors to the retrieved phases and, ultimately, to the reconstructed vector fields. To first order, the truncation error arising from the use of Eq. (2.8) is [114]:

$$\text{trunc} E \approx \frac{(\Delta f)^2}{6} \frac{\partial^3 I_0}{\partial z^3}.$$  \hspace{1cm} (6.17)

This implies that the mean square error in the phase map due to this contribution will be predominantly a function of $(\Delta f)^4$.

6.3 Derivation of error estimates

In this section, we derive analytical estimates $\langle E_{\text{tot}} \rangle$ for the total reconstruction error $\langle E \rangle$ (see Eq. (2.21)) as a function of specimen orientation via a Taylor series expansion of $\langle E \rangle$.

Under the assumption that the rms error contribution $\text{rms} E_{\varphi}$ of a single phase map is a function of contrast, we make use of Eqs. (6.16) and (6.17), and expand $\text{rms} E_{\varphi}^2$ as a Taylor series about $1 - (\hat{m} \cdot \hat{z})^2 = 0$ and $(\Delta f)^4 = 0$:

$$\text{rms} E_{\varphi}^2 ((\hat{m} \cdot \hat{z})^2, (\Delta f)^4) = \text{rms} E_{\varphi}^2 (1, 0)$$  \hspace{1cm} (6.18)

$$+ \frac{1}{2} \left[ \frac{\partial^2 (\text{rms} E_{\varphi}^2)}{\partial p^2} ((\hat{m} \cdot \hat{z})^2 - 1)^2 \right.$$

$$+ 2 \frac{\partial^2 (\text{rms} E_{\varphi}^2)}{\partial p \partial q} ((\hat{m} \cdot \hat{z})^2 - 1) (\Delta f)^4$$

$$\left. + \frac{\partial^2 (\text{rms} E_{\varphi}^2)}{\partial q^2} (\Delta f)^8 \right],$$  \hspace{1cm} (6.19)
where $p \equiv (\hat{m} \cdot \hat{z})^2$ and $q \equiv (\Delta f)^4$. Here, we have set the partial derivatives of first order to zero, because we expect extrema in the error function at $(\hat{m} \cdot \hat{z})^2 = 1$ and $(\Delta f)^4 = 0$.\(^1\) We replace the partial derivatives in Eq. (6.19) with the constants

\[ A = \frac{\partial^2}{\partial p^2} (\text{rms} \bar{E}_\varphi^2), \]
\[ B = \frac{\partial^2}{\partial p \partial q} (\text{rms} \bar{E}_\varphi^2), \]
\[ C = \frac{1}{2} \frac{\partial^2}{\partial q^2} (\text{rms} \bar{E}_\varphi^2). \]

The mean square error in the phase is then given by

\[
\text{rms} \bar{E}_\varphi^2 ((\hat{m} \cdot \hat{z})^2, (\Delta f)^4) = \min E^2
+ (B(\Delta f)^4 - A) (\hat{m} \cdot \hat{z})^2
- B(\Delta f)^4 + \frac{1}{2} A(\hat{m} \cdot \hat{z})^4
+ C(\Delta f)^8,
\]

where we have combined the constant terms via $\min E^2 = \text{rms} \bar{E}_\varphi^2(1, 0) + \frac{1}{2} A$. In general, the error contribution of the respective tilt series can be different. We use the subscripts $\alpha$ and $\theta$ to denote the tilt series to which each error and undetermined constant belongs. Equation (6.23) then evaluates to

\[
\text{rms} \bar{E}_\varphi^2(\alpha, \gamma, \gamma')
= (m_y \sin(\alpha) + m_z \cos(\alpha))^2 (B_\alpha (\Delta f)^4 - A_\alpha) - B_\alpha (\Delta f)^4
+ \frac{A_\alpha}{2} (m_y \sin(\alpha) + m_z \cos(\alpha))^4 + C_\alpha (\Delta f)^8 + \min E^2_\alpha
\]

and

\[
\text{rms} \bar{E}_\varphi^2(\theta, \gamma, \gamma')
= (m_x \sin(\theta) + m_y \cos(\theta))^2 (B_\theta (\Delta f)^4 - A_\theta) - B_\theta (\Delta f)^4
+ \frac{A_\theta}{2} (m_x \sin(\theta) + m_y \cos(\theta))^4 + C_\theta (\Delta f)^8 + \min E^2_\theta
\]

\(^1\)As we noted in Sec. 6.2.2, the magnetic phase contrast vanishes, under the projection approximation, when the dipole moment is parallel to the electron propagation direction. In addition, the truncation error due to defocus, and hence the total defocus dependent error in the absence of noise, vanishes for small defocus.
for the respective tilt series. Here we have made use of the fact that the direction
of the magnetic moment, in terms of our spherical polar coordinates, $\gamma$ and $\gamma'$, is
given by

$$\hat{m} = \sin \gamma \cos \gamma' \hat{x} + \cos \gamma \hat{y} + \sin \gamma \sin \gamma' \hat{z}.$$  \hspace{1cm} (6.26)

Integrating Eqs. (6.24) and (6.25) over the respective tilt series gives the mean square
errors

$$\langle \hat{E}^2 \rangle_\alpha = \frac{\pi}{2} (m_y^2 + m_z^2) (B_\alpha (\Delta f)^4 - A_\alpha) - \pi B_\alpha (\Delta f)^4$$
$$+ \frac{3\pi A_\alpha}{16} (m_y^2 + m_z^2)^2 + \pi C_\alpha (\Delta f)^8 + \pi^{min} E_\alpha^2$$  \hspace{1cm} (6.27)

and

$$\langle \hat{E}^2 \rangle_\theta = \frac{\pi}{2} (m_y^2 + m_z^2) (B_\theta (\Delta f)^4 - A_\theta) - \pi B_\theta (\Delta f)^4$$
$$+ \frac{3\pi A_\theta}{16} (m_y^2 + m_z^2)^2 + \pi C_\theta (\Delta f)^8 + \pi^{min} E_\theta^2.$$  \hspace{1cm} (6.28)

Because the orientation-dependent in each tilt series are not independent, the total
reconstruction error due to this alignment is obtained by summing the square-roots
of Eqs. (6.27) and (6.28):

$$\langle \hat{E} \rangle = \langle \hat{E} \rangle_\alpha + \langle \hat{E} \rangle_\theta$$  \hspace{1cm} (6.29)

In the simulations we present in this thesis, the error contributions from the two
tilt series are similar. Because of this, we drop the $\alpha$ and $\theta$ subscripts to reduce the
number of unknown constants to a total of four; i.e., $A$, $B$, $C$, and $^{min} E$.

6.4 Numerical simulations

The error estimate derived in Sec. 6.3 assumes the object to be dipole-like (i.e., small
and uniformly magnetised). To test our analytical predictions for a realistic spec-
imen, we compute micrographs from a simulated, uniformly magnetised magnetite
specimen. The micrographs are produced using the projection approximation, ac-
cording to the processes detailed in Ch. 4, and we reconstruct the magnetic vector
potential of the simulated object using the filtered backprojection algorithm given
by Eq. (2.10). Slices of the vector potential—exact, and reconstructed from samples
with $\gamma = 0$ and $\gamma = \pi/2$, respectively—of a 100 nm magnetite sphere, are shown in
Fig. 6.4. The reconstruction error is then calculated using the RMS metric defined in
Eq. (2.21). For a given defocus, we vary the magnetisation direction over 11 polar
Figure 6.4: A one voxel (3.125 nm) thick slice through the origin of each component of \( \mathbf{A} \) for a 100 nm diameter magnetite sphere reconstructed from simulated micrographs with a defocus of 100 \( \mu \)m. Top row: The exact simulated vector potential. Middle row: Reconstruction with \( \mathbf{m}(\gamma, \gamma') \) chosen to maximise \( \langle \text{tot} E \rangle \). Bottom row: Reconstruction with \( \mathbf{m}(\gamma, \gamma') \) chosen to minimise \( \langle \text{tot} E \rangle \).

angles and 21 azimuthal angles, a total of 191 orientations of the magnetisation, and perform the entire simulation and reconstruction process once for each orientation. This gives us the error as a function of specimen orientation. We compare these simulations with our analytical results based on Eqs. (6.27)–(6.29). The results for a single defocus (using the 100 nm sphere) are shown as grayscale plots in Fig. 6.5.

We compare these simulations with our analytical results. These analytical re-
Figure 6.5: Grayscale plots of error as a function of specimen orientation for the $\Delta f = 25 \mu m$ case, using simulations (left) and analytical results (right). The errors are shown as a function of $\gamma$ and $\gamma'$ at the top. The spheres show the same results mapped back onto spherical coordinates, with the axes indicating the orientation of these spheres in Cartesian coordinates.
Table 6.1: Choice of constants used to match the analytical results to the results of simulations using the 100 nm sphere. The results, themselves, can be seen in Figs. 6.5 and 6.6.

<table>
<thead>
<tr>
<th>$A$</th>
<th>$B$</th>
<th>$C$</th>
<th>$\text{min} E$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$5.00 \times 10^{-2}$</td>
<td>$-450 \text{mm}^{-4}$</td>
<td>$-1.4 \times 10^6 \text{mm}^{-8}$</td>
<td>$1.33 \times 10^{-4}$</td>
</tr>
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</table>

Our purpose here is to verify the functional form of Eq. (6.29), so we set these to values that best match the results of simulations. Nevertheless, our analysis, combined with the numerical results we report here, can provide a broad understanding of the nature of orientation-dependent errors, and how they are affected by various experimental parameters, such as specimen diameter and microscope defocus.

There are various numerical errors that affect the simulations for some parameter combinations. These includes sampling errors when $dM/a$ is small, where $d$ is the length scale of the object, and wraparound artefacts when $d\Delta f/a$ is large. In other words, $a$ must be made large enough, relative to $d$, that the Gibbs phenomenon does not result in wraparound artefacts, and $M$ must consequently be increased to improve sampling of the particle. These errors are difficult to quantify, and the sampling would typically be high in an experimental set up. The wraparound effects are a numerical artefact, and do not occur in experimental micrographs. For these reasons we avoid these errors in our simulations, via the appropriate selection of these parameters, and focus on contrast dependent errors and truncation errors in the finite difference approximation (Eq. (2.8)) used for the Tie.

In the remainder of this this section, we report on results of simulations using three different simulated specimens: a 100 nm sphere, a 40 nm sphere, and a 40 nm cube, respectively.

For our first set of results, we use a spherical sample with a diameter of $d = 100 \text{nm}$, centred at the origin. This shape is chosen to decouple the magnetisation orientation from the specimen geometry. In addition to the representation already provided in Fig. 6.5, our results, both simulated and analytical, for four different defoci, are presented as two-dimensional scatter plots in Fig. 6.6; the values chosen for the constants $A$, $B$, and $C$, are shown in Table 6.1.

We then repeat these simulations with a 40 nm sphere. The constants $A$, $B$, $C$, and $\text{min} E$, chosen to achieve an approximate match between simulations and the analysis, are shown in Table 6.2. Figure 6.7 shows slices of the exact and reconstructed vector potentials, for two different orientations, and the corresponding scatter plots, showing the total RMS error as a function of specimen orientation, are
Figure 6.6: Scatter plots showing the total RMS errors in the reconstructed vector potential of a simulated 100 nm magnetite sphere as a function of initial specimen orientation and for a range of defoci, using simulated micrographs (top) and the analytical estimate (bottom). Plots on the left and right show slices at $\gamma' = 0$ and $\gamma = \pi/2$, respectively.
Figure 6.7: A one voxel (3.125 nm) thick slice through the origin of each component of \( \mathbf{A} \) for a 40 nm diameter magnetite sphere reconstructed from simulated micrographs with a defocus of 2 \( \mu \)m. Top row: The exact simulated vector potential. Middle row: Reconstruction with \( \mathbf{m}(\gamma, \gamma') \) chosen to minimise \( \langle \text{tot} E \rangle \). Bottom row: Reconstruction with \( \mathbf{m}(\gamma, \gamma') \) chosen to maximise \( \langle \text{tot} E \rangle \).
Table 6.2: Choice of constants used to match the analytical results to the results of simulations using the 40 nm sphere. The results, themselves, can be seen in Fig. 6.8.

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<td>C</td>
<td>minE</td>
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<td>−3.5 × 10¹³ mm⁻⁸</td>
<td>2.8 × 10⁻²</td>
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Table 6.3: Choice of constants used to match the analytical results to the results of simulations using the 40 nm cube. The results, themselves, can be seen in Fig. 6.11.

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<td>C</td>
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</tr>
<tr>
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<td>−2 × 10² mm⁻⁴</td>
<td>−7 × 10⁵ mm⁻⁸</td>
<td>1.3 × 10⁻¹</td>
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shown in Fig. 6.8. For some additional insight, the mean scaling factors $F_s$ (defined in Sec. 2.6) for the 40 nm and the 100 nm particle are shown in Figs. 6.9 and 6.10, highlighting that $F_s$ is a function of both orientation and defocus.

The results for for the 40 nm magnetite cube, and the corresponding values chosen for $A$, $B$, and $C$, are shown in Fig. 6.11 and Table 6.3, respectively. The appearance of the reconstructed vector potential of the cube differs greatly between different slices. For this reason, we show two sets of slices—one through the origin (Fig. 6.12), and one offset by 16 pixels/18.75 nm (Fig. 6.13)—for this simulated specimen.
Figure 6.8: Scatter plots showing the total RMS errors in the reconstructed vector potential of a simulated 40 nm magnetite sphere as a function of initial specimen orientation and for a range of defoci, using simulated micrographs (top) and the analytical estimate (bottom). Plots on the left and right show slices at $\gamma' = 0$ and $\gamma = \pi/2$, respectively.
6.4. NUMERICAL SIMULATIONS

Figure 6.9: Mean scaling factor $F_s$ as a function of initial magnetisation orientation for the 40 nm sphere. Plots on the left and right show slices at $\gamma' = 0$ and $\gamma = \pi/2$, respectively.

Figure 6.10: Mean scaling factor $F_s$ as a function of initial magnetisation orientation for the 100 nm sphere. Plots on the left and right show slices at $\gamma' = 0$ and $\gamma = \pi/2$, respectively.
Figure 6.11: Scatter plots showing the total RMS errors in the reconstructed vector potential of a simulated 40 nm magnetite cube as a function of initial specimen orientation and for a range of defoci, using simulated micrographs (top) and the analytical estimate (bottom). Plots on the left and right show slices at $\gamma' = 0$ and $\gamma = \pi/2$, respectively.
Figure 6.12: A one voxel (1.17 nm) slice through the origin of each component of $\mathbf{A}$ for a 40 nm cube reconstructed from simulated micrographs with a defocus of 5 $\mu$m. Top row: The exact simulated vector potential. Middle row: Reconstruction with $\mathbf{m}(\gamma, \gamma')$ chosen to maximise $\langle \text{tot} E \rangle$. Bottom row: Reconstruction with $\mathbf{m}(\gamma, \gamma')$ chosen to minimise $\langle \text{dir} E \rangle$. 
Figure 6.13: A one voxel (1.17 nm) slice, 18.75 nm from the origin, of each component of $\mathbf{A}$ for a 40 nm cube reconstructed from simulated micrographs with a defocus of 5 $\mu$m. Top row: The exact simulated vector potential. Middle row: Reconstruction with $\mathbf{m}(\gamma, \gamma')$ chosen to maximise $\langle \text{tot} E \rangle$. Bottom row: Reconstruction with $\mathbf{m}(\gamma, \gamma')$ chosen to minimise $\langle \text{dir} E \rangle$. 

$\times 10^{-9}$ V s m$^{-1}$
6.5 Discussion

Provided that a small number of unknowns (i.e., the constants $A$, $B$, $C$, and $\min E$) are appropriately chosen, the analytical results for the RMS error agree well with the simulations. For the 100 nm sphere, as well as the 40 nm cube, the optimal value for $\gamma$ is always $(\gamma \mod \pi) = 0$. Using the smaller $(d = 40 \text{ nm})$ sphere, the optimum orientation is $\gamma = \pi/2$ for small defocus. In cases such as this, a large enough increase in defocus can increase the error in the retrieved phases to such an extent that the optimal orientation switches from $\gamma = \pi/2$ to $\gamma = 0$, but changes to $(\gamma \mod \pi) = 0$ above some critical defocus value. For large defocus, our results indicate that the optimal value is always $(\gamma \mod \pi) = 0$. These results are consistent with our assertion that improved contrast reduces $\text{rms} E$ when the error in the retrieved phase is low. For a larger particle such as the $d = 100 \text{ nm}$ sphere, additional errors in the phase, due to increased attenuation, result in an increase in error with increasing contrast, even for small $\Delta f$. The results for the 40 nm cube are similar (i.e., the constant $A$ has the same sign) to those for the 100 nm sphere. This is most likely due to errors in the retrieved phase arising from the extremely high phase gradient that can occur when faces of the cube are parallel to the electron beam. Note that the results of the 40 nm particle simulations may not be physically accurate due to the omission of superparamagnetic effects in any of our simulations, which can occur in particles of this size at room temperature [163].

The optimal azimuthal angle is difficult to predict, as it can be highly sensitive to small differences in the errors arising from each tilt series. However, in most cases the reconstruction error is approximately constant with respect to $\gamma'$, and can be treated as such for the purposes of determining the ideal initial orientation of the specimen. Given the small change in error as $\gamma'$ is changed, technical concerns associated with preparing a specimen with an off-axis magnetisation may take precedence over minimising this error.

6.6 Conclusion

The accuracy of VFET reconstructions exhibits a strong dependence on the initial orientation of the sample relative to the reconstruction geometry. The form of this dependence is easily estimated for the case where the object is uniformly magnetised. However, for many materials, there is no such straightforward way of calculating these errors. Nevertheless, many materials of interest exhibit a strong net magnetisation and exhibit similar properties to a magnetic dipole, or uniformly magnetised particle.
CHAPTER 6. EFFECT OF SPECIMEN ORIENTATION

When errors in the phase are large, as is the case when using a propagation based phase retrieval method with large defocus, or when there is significant attenuation, reconstruction errors can be reduced by orienting the particle such that the average magnetic phase contrast in the recorded micrographs is minimised. Conversely, if errors in the retrieved phase can be kept small, maximising the average contrast reduces the total reconstruction error.

We have shown that the ideal specimen orientation can depend on the error metric used to quantify the errors. This means that the orientation may be chosen depending on what quantity is of most interest. For our 40 nm particle simulations, the ideal orientation for reconstructing the direction of the vector potential at each point typically occurs when the magnetisation vector points towards the intersection of the two tilt series. This is also the worst orientation for reconstructing the length of these vectors. The ideal orientation for minimising total errors, or errors in magnitude, is achieved by aligning the magnetisation vector in the plane orthogonal to the intersection of the two tilt series. The error in direction typically has a local minimum here (as a function of polar angle) and this is a desirable choice of orientation if both directional and magnitude errors are to be minimised.

The ideal azimuthal angle in the magnetisation direction may be more difficult to ascertain, because it is highly sensitive to the differences in error due to each tilt series. However, the errors typically vary much less with changes in the azimuthal angle than with changes in the polar angle, so in most cases it may be sufficient to address only the latter.

Where sample preparation techniques allow for the choice of magnetisation orientation, reconstruction errors can be reduced by taking the effect of initial orientation into consideration. This may be achieved using methods designed to produce a desired magnetisation in a given specimen; however, a desired magnetisation direction can also be obtained by selecting an appropriate particle from an agglomeration. The results we have presented here show that these efforts can reduce the reconstruction errors by up to $\sim 50\%$. 

Chapter 7

Using additional tilt series to improve vector field electron tomography accuracy

7.1 Introduction

The use of more than two tilt series in VFET has been considered by Yu et al. [85]. Their method utilises three mutually orthogonal tilt series to reconstruct the vector potential, and circumvents singular surfaces in the FBP algorithm by reconstructing each component of the vector potential from each respective orthogonal pair of tilt series. Simulations using three tilt series are promising, but the method has not been utilised experimentally because of technical limitations making it difficult to acquire a third orthogonal tilt series. In this chapter, we propose a method to utilise additional tilt series that does not pose such limitations. Although this method does not avoid the singular surfaces, it is capable of producing more accurate reconstructions by improving spatial sampling.

The practice of utilising additional tilt series for the reconstruction of scalar fields, in the form of dual-axis electron tomography, is well established [54,164–171]. These methods reconstruct the object function from each tilt series separately, and combine the resulting reconstructions—typically using a selective combination aimed at avoiding errors caused by the missing wedge [165, 168]. Our polyaxial method for VFET reconstruction uses a similar approach. This method employs multiple orthogonal pairs of tilt series, with the axes of rotation (for all tilt series) being coplanar. This geometry is shown in Fig. 7.1. We acquire the additional pairs of tilt series at equiangular intervals, with the angle of rotation of the \( p^{th} \) pair, relative to the first pair, being given by
Figure 7.1: Geometry used for the acquisition of two pairs of tilt series. In the $\alpha_1$ series, the object $O$ is rotated about the $x$-axis and, in the $\theta_1$ series, it is rotated about the $z$-axis. The $\alpha_2$ and $\theta_2$ series are generated by rotating the specimen by $45^\circ$ about the $y$-axis before repeating the measurements. The axis of rotation for each tilt series lies in the $x$-$z$ plane.

$$\psi_p = \frac{(p - 1)\pi}{n_s},$$  \hfill (7.1)

where $n_s$, which must be even, is the total number of tilt series, and $1 \leq p \leq n_s/2$.

After acquiring multiple tilt series, we perform a reconstruction for each pair using the FBP algorithm given by Eq. (2.10), and align the reconstructions by rotating each according to Eq. (7.1) using trilinear interpolation. We then combine the reconstructions by calculating their arithmetic mean:

$$\text{av} A_{i,j,k}^{\text{rec}} = \frac{2}{n_s} \sum_{p=1}^{n_s/2} (p A_{i,j,k}^{\text{rec}}),$$  \hfill (7.2)

where the preceding superscript $p$ distinguishes the separate constituent reconstructions.

In the remainder of this chapter we explore the feasibility of this method. In Sec. 7.2 we present a systematic study of the accuracy of this method using simulations of several different specimens, using a variety of simulation and reconstruction parameter sets. We intend these to be usable—to some extent—as lookup tables, from which the ideal number of tilt series, for example, can be inferred by looking at the results obtained for a similar specimen. In addition to this, the variety of nanostructures considered in this chapter and the range of microscope and recon-
7.2 Simulations

In this section, we present numerical results of the reconstruction of six different simulated specimens. We begin with uniformly magnetised specimens: a nanosphere, an isolated nanocube, and a cluster of nanocubes. We then introduce non-uniform magnetisations, examining a torus with the magnetisation at each point oriented along the toroidal direction, an antidot lattice, and a thin film with domains of varying, in-plane, magnetisations. We utilise different sets of microscope and reconstruction parameters throughout this section, in order to test our polyaxial VFET method under a range of conditions. Most of the parameters used for these simulations are those given in Table 4.1. Each of the following sections (7.2.1–7.2.6) contains a table showing parameters whose values are unique to that section. Where multiple values are used throughout a section, these are comma separated.

In Secs. 7.2.1–7.2.6, we present results of our three RMS error metrics \( \langle \text{tot} E \rangle \), \( \langle \text{mag} E \rangle \), and \( \langle \text{dir} E \rangle \) as well as the mean scaling factor \( F_s \), described in Sec. 2.6, as a function of \( n_s \). For these results, we hold the total number of images \( n_s n_t \) approximately constant, so any changes in error cannot be attributed to including additional data in the reconstructions. Note that this means that a reconstruction using \( n_s = 2 \), and a constituent reconstruction of a corresponding \( n_s > 2 \) case, utilise a different number of images in their respective tilt series and, consequently, result in errors of different magnitudes. We also show the errors in the constituent vector potentials \( pA^{\text{rec}} \), as bar plots, for at least one \( n_s \) in each section. Slices of each component of the exact vector potential are shown for each specimen, as well as the difference between the exact and reconstructed image, for at least two values of \( n_s \). The reconstructed vector potentials can often contain artefacts in planes through the origin that are not representative of the overall quality of the reconstruction. For this reason, we usually choose to display slices that are offset from the origin by a small number of voxels, to avoid these artefacts.

7.2.1 Single, isolated nanosphere

In this section, we explore the use of our polyaxial VFET method for the reconstruction of a uniformly magnetised sphere. As we noted in Ch. 6, this shape decouples the magnetisation direction from the geometry of the specimen. This makes it an interesting object to study in the context of polyaxial VFET, in which it is the na-
ture of the orientation-dependent errors that determines whether the technique can reduce errors or not. The parameters used in this section are shown in Table 7.1.

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<td>$M$</td>
<td>$M_0\hat{z}, M_0\hat{y}$</td>
<td>$a$</td>
<td>150 nm</td>
</tr>
<tr>
<td>$n_s,n_t$</td>
<td>~128</td>
<td>$\Delta f$</td>
<td>5 $\mu$m, 50 $\mu$m</td>
</tr>
<tr>
<td>multislice</td>
<td>✓, ×</td>
<td>$d$</td>
<td>40 nm</td>
</tr>
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</table>

Table 7.1: Parameters used for the nanosphere specimen. The tick and cross in the “multislice” row indicates that this section includes simulations that do, and do not, utilise the multislice method, respectively.

**Sphere with magnetisation oriented in the $z$-direction**

Figure 7.2 shows each of our error metrics, for the reconstruction of the nanosphere, as a function of the number of tilt series. For this specimen and the microscope/reconstruction parameters used here, utilising four tilt series provides a slight reduction in the total and directional RMS errors, over using two tilt series, but worsens (slightly) the reconstruction of the length of the vectors. Increasing the number of tilt series above four causes all RMS errors to increase. The mean scaling factor deviates further from unity as soon as the number of tilt series is increased above two.

![Figure 7.2: Errors as a function of the number of tilt series, for a nanosphere with the magnetisation oriented in the z-direction. (a) Total, magnitude, and directional RMS errors. (b) Mean scaling factor.](image)

(a) (b)
In Fig. 7.3, we break the four tilt series reconstruction into its constituent reconstructions $pA^{\text{rec}}$, and display the errors in each. Here we see that averaging reconstructions reduces all three RMS errors, but leaves the mean scaling factor approximately constant.

Figure 7.3: (a) Total, magnitude, and directional error in reconstructions with $\psi = 0$ and $\psi = \pi/4$, from left to right, respectively (blue). Errors in the vector potential produced by averaging the other two reconstructions (green). These errors correspond to a magnetisation with initial orientation in the z-direction. (b) Mean scaling factor for these reconstructions.

Despite the small quantitative difference between the errors for the two and four tilt series reconstructions, the difference can be seen as a smoothing of high frequency errors in the reconstructed vector potential. Slices in the $x$-$y$ plane, of the exact vector potential of the 40 nm uniformly magnetised sphere, are shown in Fig. 7.4. The error in the vector potentials reconstructed using two and four tilt series, respectively, are also shown. The $y$-component is fairly well reconstructed even in the $n_s = 2$ case, while the $x$- and $z$-components are not. This is expected based on our results in Ch. 6, where we found that the 40 nm sphere results in the largest RMS reconstruction error when the magnetisation is oriented in the $y$-direction. It is interesting to note that utilising four tilt series results in more consistent errors across the three components.
Figure 7.4: A one voxel (1.17 nm) slice, approximately 4.7 nm from the origin, of each component of $\mathbf{A}$ for a 40 nm diameter sphere with $\mathbf{M} = M_0 \hat{z}$. Top row: The exact simulated vector potential. Middle row: Error in each component of the vector potential reconstructed from simulated micrographs with a defocus of 5 μm using two tilt series. Bottom row: Error in each component of the vector potential reconstructed using four tilt series.
Sphere with magnetisation oriented in the $y$-direction

The next set of reconstructions is identical to the preceding one, but this time the initial magnetisation is pointing in the $y$-direction. The results for the three RMS errors, as well as the mean scaling factor, are shown in Fig. 7.5.

![Figure 7.5](image_url)

Figure 7.5: Errors as a function of the number of tilt series, for a nanosphere with the magnetisation oriented in the $y$-direction. (a) Total, magnitude, and directional RMS errors. (b) Mean scaling factor.

![Figure 7.6](image_url)

Figure 7.6: (a) Total, magnitude, and directional error in reconstructions with $\psi = 0$ and $\psi = \pi/4$, from left to right, respectively (blue). Errors in the vector potential produced by averaging the other two reconstructions (green). These errors correspond to a magnetisation with initial orientation in the $y$-direction. (b) Mean scaling factor for these reconstructions.
In these reconstructions, we see that increasing the number of tilt series above two immediately begins increasing all RMS errors and causes $F_s$ to deviate further from unity. Interestingly, the change in initial magnetisation orientation causes $F_s$ to start below unity and decrease, rather than start above and increase. For all four metrics, the point at $n_s = 8$ appears to be an outlier; the reason for this is unknown, but it may be the case that certain values of $n_s$ are favourable for minimising angular interpolation errors.

In Fig. 7.6, we break the four tilt series reconstruction into its constituent reconstructions and display the errors in each. In this case, the reduction in errors as the two reconstructions are averaged is very small.

**Multislice simulations using the sphere**

In this section, we examine the effect of diffraction artefacts that arise from scattering of the electron beam from the atomic potentials, and other deviations from the projection approximation, on our polyaxial vfet method. For these simulations, we return to using a magnetisation of $M = M_0 \hat{z}$.

We employ a multislice method with a slice thickness of 0.5 nm, and a resolution of $M_{ms} = 1024$. The resulting micrographs are downsampled to $M = 128$ to match our projection approximation simulations.

![Figure 7.7: Errors as a function of the number of tilt series, for a nanosphere with the magnetisation oriented in the $z$-direction. This simulation uses the multislice method to produce the micrographs. (a) Total, magnitude, and directional RMS errors. (b) Mean scaling factor.](image)

Figure 7.7 shows our three RMS errors, and the mean scaling factor, as a function
of \( n_s \). The errors for a single pair of tilt series are significantly higher than those in Fig. 7.2, where the projection approximation was used. Unlike those results, increasing the number of tilt series above two, here, only worsens the errors. This indicates that utilising additional tilt series may be ineffective when diffraction artefacts in the micrographs are significant.

Figure 7.8 shows the errors in the constituent reconstructions for the \( n_s = 4 \) case. Even with the reduced number of images per tilt series, relative to the \( n_s = 2 \) case (because we have held the total number of images constant as we alter \( n_s \)), the errors are slightly higher than in the best constituent reconstruction; that is, acquiring additional data here has increased the errors. However, for the total and directional errors, the total reconstruction is close, in accuracy, to the better of the two individual reconstructions.

Figure 7.8: Averaging reconstructions with multislice simulated micrographs. (a) Total, magnitude, and directional error in reconstructions with \( \psi = 0 \) and \( \psi = \pi/4 \), from left to right, respectively (blue). Errors in the vector potential produced by averaging the other two reconstructions (green). (b) Mean scaling factor for each of these reconstructions.

Slices of each component of the exact vector potential, and slices of the errors in each component for both the two and the ten tilt series cases, are shown in Fig. 7.9. Streaking artefacts appear in the two tilt series reconstruction along the \( y \)-direction. The ten tilt series reconstruction reduces these artefacts, but introduces additional streaking along other directions.
Figure 7.9: A one voxel (7.8 nm) slice, approximately 4.7 nm from the origin, of each component of $\mathbf{A}$ for a 40 nm sphere with $\mathbf{M} = M_0 \hat{z}$. Top row: The exact simulated vector potential. Middle row: Error in each component of the vector potential reconstructed from micrographs, simulated using the multislice method, with a defocus of 100 µm using two tilt series. Bottom row: Error in each component of the vector potential reconstructed using ten tilt series.
7.2.2 Single, isolated nanocube

We now add a level of complexity to our simulations by using a 40 nm diameter uniformly magnetised nanocube as the specimen. This gives rise to additional orientation-dependent errors caused by the reduced rotational symmetry. The parameters used for these simulations are shown in Table 7.2

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</tr>
<tr>
<td>( n_s n_t )</td>
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<td>( \Delta f )</td>
<td>5 µm/50 µm</td>
</tr>
<tr>
<td>multislice</td>
<td>✓, ×</td>
<td>( d )</td>
<td>40 nm, 20 nm</td>
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Table 7.2: Parameters used for the nanocube specimen.

**Cube with magnetisation oriented in the \( z \)-direction, with 5 µm defocus**

The RMS errors in the reconstruction, and the mean scaling factor, are shown in Fig. 7.10 as a function of \( n_s \), for the \( \Delta f = 5 \) µm case. Unlike the results for the sphere, in the previous section, the reconstructions continue to improve as the number of tilt series increases, up to \( n_s = 6 \). In addition to this, the reduction in error is significant, with the total RMS error falling from \( \sim 55\% \) to \( \sim 35\% \) as \( n_s \) is increased from two to four. Figure 7.11 shows the errors in the constituent reconstructions, alongside those in the total, polyaxial reconstruction, for the \( n_s = 6 \) case.

Slices of the exact vector potential of the 40 nm cube are shown in Fig. 7.12, along with those of the reconstructed vector potentials using two and six tilt series. The reduction in the magnitude of the errors is clearly visible in these images, particularly in the \( y \)- and \( z \)-components. Some artefacts appear in the \( n_s = 6 \) case that are not present in the \( n_s = 2 \) case. Most notable are streaks in the \( z \)-component, along the \( x \)-direction. This can be understood as \( y \)-oriented streaking, and \( x \)-oriented streaking, being artefacts that result from reconstructions with different initial specimen orientations. Averaging these reconstructions reduces the maximum magnitude of the errors, but adds additional artefacts that are not present in a single reconstruction.
Figure 7.10: Errors in the reconstruction of a 40 nm cube oriented in the z-direction, with $\Delta f = 5 \mu m$, as a function of the total number of tilt series used. (a) Total, magnitude, and directional RMS errors. (b) Mean scaling factor.

Figure 7.11: (a) Total, magnitude, and directional error in reconstructions with $\psi = 0$, $\psi = \pi/6$, and $\psi = 2\pi/6$, from left to right, respectively (blue). Errors in the vector potential produced by averaging the other three reconstructions (green). (b) Mean scaling factor for each of these reconstructions.
Figure 7.12: A one voxel (1.17 nm) slice, approximately 4.7 nm from the origin, of each component of $\mathbf{A}$ for a 40 nm wide cube with $\mathbf{M} = M_0 \hat{z}$. Top row: The exact simulated vector potential. Middle row: Error in each component of the vector potential reconstructed from simulated micrographs with a defocus of 5 µm using two tilt series. Bottom row: Error in each component of the vector potential reconstructed using six tilt series.
CHAPTER 7. USING ADDITIONAL TILT SERIES

Cube with magnetisation oriented in the $z$-direction, with 50 $\mu$m defocus

Next, we increase the defocus to 50 nm and repeat the previous simulations. The four metrics are shown as a function of $n_s$ in Fig. 7.13. For these simulations, the ideal number of tilt series is four. The breakdown of the $n_s = 4$ case is shown in Fig. 7.14. Here we see significant improvement in the total and magnitude RMS errors as the two reconstructions are averaged, but only a slight improvement in the directional error.

![Graph](image)

Figure 7.13: Errors in the reconstruction of the 40 nm cube with the magnetisation oriented in the $z$-direction, using 50 $\mu$m defocus and $n_s n_t = 240$. (a) Total, magnitude, and directional RMS errors. (b) Mean scaling factor.

Slices of the vector potential, as well as errors in the two tilt series and four tilt series reconstructions, are shown in Fig. 7.15. As with the smaller defocus simulations, the improvement in the reconstruction is visible in these slices, though the effect is arguably less pronounced.
7.2. SIMULATIONS

Figure 7.14: (a) Total, magnitude, and directional error in reconstructions with $\psi = 0$ and $\psi = \pi/4$, from left to right, respectively (blue), for the 40 nm cube with magnetisation oriented in the $z$-direction, using a defocus of 50 $\mu$m. Errors in the vector potential produced by averaging the other two reconstructions (green). (b) Mean scaling factor for each reconstruction.
CHAPTER 7. USING ADDITIONAL TILT SERIES

Figure 7.15: A one voxel (1.17 nm) slice, approximately 4.7 nm from the origin, of each component of $\mathbf{A}$ for a 40 nm wide cube with $\mathbf{M} = M_0 \hat{z}$. Top row: The exact simulated vector potential. Middle row: Error in each component of the vector potential reconstructed from simulated micrographs with a defocus of 5 µm using two tilt series. Bottom row: Error in each component of the vector potential reconstructed using four tilt series.
Cube with magnetisation oriented in the \( y \)-direction, with 50\( \mu \)m defocus

We now orient the particle in the \( y \)-direction, again using a defocus of 50 nm. The RMS errors (shown in Fig. 7.16) are lower in this orientation, regardless of the number of tilt series, and we also see a significant improvement with an increase in \( n_s \). For the reconstruction of the length of the vectors in these simulations, the ideal number of tilt series is six, though two may be preferable if accuracy in the vector direction is important. The mean scaling factor also improves significantly as the number of tilt series is increased from two to four.

![Graph](image)

(a) (b)

Figure 7.16: Errors in the reconstruction of the 40 nm cube with the magnetisation oriented in the \( y \)-direction, using 50\( \mu \)m defocus and \( n_s n_t = 240 \). (a) Total, magnitude, and directional RMS errors. (b) Mean scaling factor.

The errors in each two tilt series reconstruction, as well as the errors in the total, polyaxial reconstruction are shown in Fig. 7.17 for the \( n_s = 6 \) case. The averaged reconstruction is significantly better than any of the two tilt series reconstructions it comprises. Slices of the exact vector potential—as well as slices of the error, for both two and six tilt series—are shown in Fig. 7.18. For these images, the slices are taken in the \( x-z \) plane, to facilitate a direct comparison with previous images of the vector potential in this chapter; that is, for these figures, the slice is in the plane orthogonal to the magnetisation direction.

The reduction in error for the \( n_s = 6 \) case, relative to the \( n_s = 2 \) case, is clearly visible. It is also interesting to note the qualitative differences between the appearance of the errors when viewed in the \( x-z \) plane rather than the \( x-y \) plane. Here the errors appear more as ripples than the streaking we are familiar with from the earlier figures.
Figure 7.17: (a) Total, magnitude, and directional error in reconstructions with $\psi = 0$, $\psi = \pi/6$, and $\psi = 2\pi/6$, from left to right, respectively (blue). Errors in the vector potential produced by averaging the other three reconstructions (green). (b) Mean scaling factor for each reconstruction.
Figure 7.18: A one voxel (1.17 nm) slice, approximately 4.7 nm from the origin, of each component of $\mathbf{A}$ for a 40 nm wide cube with $\mathbf{M} = M_0 \hat{y}$. Top row: The exact simulated vector potential. Middle row: Error in each component of the vector potential reconstructed from simulated micrographs with a defocus of 5 µm using two tilt series. Bottom row: Error in each component of the vector potential reconstructed using six tilt series.
Cube with magnetisation oriented in the $y$-direction, with $50\,\mu m$ defocus, for a $20\,nm$ particle

We now repeat the previous set of simulations, but with a reduced particle size. The errors are shown, as a function of $n_s$, in Fig. 7.19. The errors in each two tilt series reconstruction, and those in the combined reconstruction are shown in Fig. 7.20 for the $n_s = 4$ case.

Figure 7.19: Errors in the reconstruction of the $20\,nm$ cube using $50\,\mu m$ defocus and $n_s n_t = 240$. (a) Total, magnitude, and directional RMS errors. (b) Mean scaling factor.

Figure 7.20: (a) Total, magnitude, and directional error in reconstructions with $\psi = 0$ and $\psi = \pi/4$, from left to right, respectively (blue). Errors in the vector potential produced by averaging the other three reconstructions (green). (b) Mean scaling factor for each reconstruction.
Four tilt series produces an improvement in the reconstruction, with additional tilt series increasing the error again. The four tilt series case shows most improvement in the directional error, with the other metrics indicating that the combined reconstruction is worse than the best constituent reconstruction.

**Multislice simulations using the cube**

In this section, we report on the effect of using multislice simulations, on the reconstruction errors for the uniformly magnetised cube. We use a smaller number of images \( n_s n_t = 120 \), and continue using a \( d = 20 \text{ nm} \) wide specimen. To compare the results using the multislice method with those using the projection approximation, we first present the results of the latter in Figs. 7.21 and 7.22. The errors here are very close in magnitude to those in Figs. 7.19 and 7.20, indicating that using twice the number of images does not significantly impact the error under the conditions that apply here.

![Figure 7.21](image)

(a) Total, magnitude, and directional error in reconstructions with \( \psi = 0 \) and \( \psi = \pi/4 \), from left to right, respectively (blue). Errors in the vector potential produced by averaging the other two reconstructions (green). (b) Mean scaling factor for each reconstruction.

We repeat the simulations, this time using the multislice method to produce the micrographs. The errors are shown in Fig. 7.23 as a function of \( n_s \). The errors are much higher when the multislice method is used, but unlike the results for the sphere, increasing the number of tilt series can significantly improve the reconstruction. Here we find that the ideal number of tilt series is \( n_s = 8 \).

The errors of the constituent reconstructions, for the six and eight tilt series
Figure 7.22: Errors in the reconstruction of the 20 nm cube using 50 µm defocus and $n_sn_t = 120$. (a) Total, magnitude, and directional RMS errors. (b) Mean scaling factor.

Cases, are shown in Figs. 7.24 and 7.25, respectively. These results indicate that the RMS errors are mostly independent of their orientation relative to the particle, but the systematic errors in these separate reconstructions are different. Interestingly, the $n_s = 8$ case shows lower errors for the constituent reconstructions than the $n_s = 6$ case (which use a larger number of images). This may be a result of using an odd number of images (fifteen) in each tilt series, which causes a phase map in the $\alpha$-tilt series to occur precisely at the singular surface which, as discussed in Sec. 2.5, occurs at $\alpha = \pi/2$.

Slices of the vector potential, as well as those of the errors for the two, six, and eight tilt series cases, in the $x$-$z$ and $x$-$y$ planes, are shown in Figs. 7.26 and 7.27, respectively.
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Figure 7.23: Errors in the reconstruction of the 20 nm cube using 50 μm defocus and \( n_s n_t = 240 \), utilising the multislice method for simulation of the micrographs. (a) Total, magnitude, and directional RMS errors. (b) Mean scaling factor.

Figure 7.24: (a) Total, magnitude, and directional error in reconstructions using micrographs simulated using the multislice method, with \( \psi = 0 \), \( \psi = \pi/6 \), and \( \psi = 2\pi/6 \), from left to right, respectively (blue). Errors in the vector potential produced by averaging the other three reconstructions (green). (b) Mean scaling factor for each reconstruction.
Figure 7.25: (a) Total, magnitude, and directional error in reconstructions using micrographs simulated using the multislice method, with $\psi = 0$, $\psi = \pi/8$, $\psi = \pi/4$, and $\psi = 3\pi/8$, from left to right, respectively (blue). Errors in the vector potential produced by averaging the other four reconstructions (green). (b) Mean scaling factor for each reconstruction.
Figure 7.26: A one voxel (1.17 nm) slice, at $y \approx 4.6875$ nm, of each component of $\mathbf{A}$ for a 20 nm sphere with $\mathbf{M} = M_0 \hat{y}$. Top row: The exact simulated vector potential. Second row: Error in each component of the vector potential reconstructed from micrographs, simulated using the multislice method, with a defocus of 50 µm using two tilt series. Third row: Error in each component of the vector potential reconstructed using six tilt series. Bottom row: Error in each component of the vector potential reconstructed using eight tilt series.
Figure 7.27: A one voxel (1.17 nm) slice, at $z \approx 4.6875$ nm, of each component of $\mathbf{A}$ for a 20 nm sphere with $\mathbf{M} = M_0 \hat{y}$. Top row: The exact simulated vector potential. Second row: Error in each component of the vector potential reconstructed from micrographs, simulated using the multislice method, with a defocus of 50 µm using two tilt series. Third row: Error in each component of the vector potential reconstructed using six tilt series. Bottom row: Error in each component of the vector potential reconstructed using eight tilt series.
7.2.3 Cluster of nanocubes

In this section, we perform reconstructions using a simulated cluster of nanocubes. This specimen (shown in Fig. 7.28) comprises twenty nanocubes of various sizes and orientations. A transmission electron micrograph of a cluster of polyhedral magnetite nanoparticles is shown in Fig. 7.29 for comparison. The magnetisation in our simulations is globally uniform; i.e., the magnetisation direction of every individual cube is the same and doesn’t depend on the orientation of that cube. The parameters used for these reconstructions are shown in Table 7.3.

Figure 7.28: Cluster of nanocubes used as the specimen shape for simulations in this section.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\hat{m}$</td>
<td>$\hat{z}$</td>
<td>$a$</td>
<td>1 $\mu$m</td>
</tr>
<tr>
<td>$n_xn_t$</td>
<td>$\sim 256$</td>
<td>$\Delta f$</td>
<td>100 $\mu$m</td>
</tr>
<tr>
<td>multislice</td>
<td>$\times$</td>
<td>$d$</td>
<td>$\sim 400$ nm</td>
</tr>
</tbody>
</table>

Table 7.3: Parameters used for the nanocube cluster specimen.
CHAPTER 7. USING ADDITIONAL TILT SERIES

Figure 7.29: Simulated micrograph of the cluster of nanocubes (left). Cluster of polyhedral magnetite nanoparticles, prepared by K. Spiers [150] observed, in this work, using an FEI Tecnai T20 TEM, equipped with an LaB$_6$ source, and operating at 200 kV (right).

**Cluster of cubes with magnetisation oriented in the z-direction**

Figure 7.30 shows our three RMS error metrics, and the mean scaling factor, as a function of $n_s$. All RMS errors improve as $n_s$ increases up until $n_s = 6$. The mean scaling factor follows a similar trend.

We examine the six and ten tilt series cases more closely in Figs. 7.32 and 7.33, respectively, where we show the RMS errors, and the mean scaling factor, in the constituent reconstructions, for the $n_s = 6$ and $n_s = 10$ cases, respectively. In both cases, the errors in $\text{av} A_{\text{rec}}$ are significantly lower than the errors in any of the $p A_{\text{rec}}$. The results for $F_s$ are all greater than unity, with the mean scaling factor for $\text{av} A_{\text{rec}}$ being closer to unity than any of the $p A_{\text{rec}}$. It is theoretically possible to see results like this from localised systematic scaling errors, but these results indicate that global scaling errors are not the source of the systematic errors.

The exact vector potential, for the cluster of nanocubes with the magnetisation oriented in the $z$-direction, is shown in Fig. 7.31, along with errors for the two and six tilt series cases.
Figure 7.30: Errors in the reconstruction of the cluster of nanocubes with the magnetisation oriented in the z-direction. (a) Total, magnitude, and directional RMS errors. (b) Mean scaling factor.
Figure 7.31: A one voxel (7.8 nm) slice, approximately 23.5 nm from the origin, of each component of $\mathbf{A}$ for a cluster of nanocubes with $\mathbf{M} = M_0 \hat{z}$. Top row: The exact simulated vector potential. Middle row: Error in each component of the vector potential reconstructed from simulated micrographs with a defocus of 100 µm using two tilt series. Bottom row: Error in each component of the vector potential reconstructed using six tilt series.
Figure 7.32: (a) Total, magnitude, and directional error, and (b) mean scaling factor, in reconstructions with $\psi = 0$, $\pi/6$, and $2\pi/6$, from left to right, respectively (blue). Errors in the vector potential produced by averaging the other three reconstructions (green). These results use the cluster of nanocubes with $\mathbf{M} = M_0 \hat{z}$.

Figure 7.33: (a) Total, magnitude, and directional error, and (b) mean scaling factor, in reconstructions with $\psi = 0$, $\pi/10$, ..., $4\pi/10$, from left to right, respectively (blue). Errors in the vector potential produced by averaging the other five reconstructions (green). These results use the cluster of nanocubes with $\mathbf{M} = M_0 \hat{z}$. 
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Cluster of cubes with magnetisation oriented in the \( y \)-direction

Figure 7.34 shows the RMS errors, and the mean scaling factor, for reconstructions of the cluster of nanocubes with \( \mathbf{M} = M_0 \hat{y} \). The reduction of errors with an increase in \( n_s \) is not as significant as for the \( \mathbf{M} = M_0 \hat{z} \) simulations, and the errors increase after \( n_s = 4 \). This may partly be a result of the much smaller overall errors, resulting in fewer systematic errors in each \( \hat{p} \mathbf{A}^{\text{rec}} \). By \( n_s = 10 \), the RMS errors are as bad as—or worse than—the \( n_s = 2 \) results. This illustrates the previously mentioned fact that we cannot always make improvements to our reconstruction, by increasing the number of tilt series used. As with our results for the \( z \)-oriented magnetisation, the trend for \( F_s \) follows that of the RMS errors very closely. However, here \( F_s \) is less than unity for each reconstruction, meaning that the RMS errors in the reconstruction are actually improving as the scaling error gets worse.

![Figure 7.34: Errors in the reconstruction of the cluster of nanocubes with the magnetisation oriented in the \( y \)-direction. (a) Total, magnitude, and directional RMS errors. (b) Mean scaling factor.](image)

In Figs. 7.35 and 7.36, we show the total RMS errors in \( \text{av} \mathbf{A}^{\text{rec}} \), along with the errors in the constituent reconstructions, for the \( n_s = 4 \) and \( n_s = 10 \) cases, respectively. For the former, we see that the total and magnitude RMS errors in \( \text{av} \mathbf{A}^{\text{rec}} \) are only slightly smaller than the errors in \( \text{av} \mathbf{A}^{\text{rec}} \) (the better of the two constituent reconstructions). The RMS error in the \textit{direction} of the vector potential, however, is greatly improved by combining the two reconstructions. This is somewhat of a reversal of the results for the \( z \)-oriented magnetisation, where the total and magnitude RMS errors were significantly improved, but the improvement for the directional RMS error was small.
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Figure 7.35: (a) Total, magnitude, and directional error, and (b) mean scaling factor, in reconstructions with $\psi = 0$, $\psi = \pi/6$, and $\psi = 2\pi/6$, from left to right, respectively (blue). Errors in the vector potential produced by averaging the other three reconstructions (green).

Figure 7.36: (a) Total, magnitude, and directional error, and (b) mean scaling factor, in reconstructions with $\psi = 0$, $\pi/10$, ..., $4\pi/10$, from left to right, respectively (blue). Errors in the vector potential produced by averaging the other five reconstructions (green). These results use the cluster of nanocubes with $\mathbf{M} = M_0\hat{y}$. 
The results for the ten tilt series case show that the final reconstruction $\text{av}A_{\text{rec}}$ can actually be worse in some ways than some of the constituent reconstructions. However, the total and magnitude RMS errors are still significantly better than those in the worst (and even the median) constituent reconstructions, and the directional RMS error in $\text{av}A_{\text{rec}}$ is slightly better than those in the individual reconstruction with the lowest error. This suggests that it may often be beneficial to utilise multiple pairs of tilt series, even when we expect the result to be no better than the best $pA_{\text{rec}}$, if we do not know which orientation will give us the lowest errors.

### 7.2.4 Torus with solenoidal magnetisation

In this section, we examine the use of polyaxial VFET for the reconstruction of a magnetised torus. We begin by simulating a toroidal specimen with major and minor radii of approximately 10 nm and 5.5 nm, respectively. We then repeat these simulations after scaling the object up to a major radius of approximately 50 nm and a minor radius of approximately 27.5 nm. In both cases, the magnetisation is given by

$$M = M_0 |\vec{y} \times \vec{r}|.$$  

(7.3)

The specimen shape, as well as the magnetisation direction, is shown in Fig. 7.37, and the parameters used for these simulations are shown in Table 7.4. Note that the length scale $d$, given here, is the sum of the major and minor radii.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$M$</td>
<td>$M_0</td>
<td>\vec{y} \times \vec{r}</td>
<td>$</td>
</tr>
<tr>
<td>$n_s n_t$</td>
<td>240</td>
<td>$\Delta f$</td>
<td>1 $\mu$m, 5 $\mu$m</td>
</tr>
<tr>
<td>multislice</td>
<td>$\times$</td>
<td>$d$</td>
<td>31 nm, 155 nm</td>
</tr>
</tbody>
</table>

Table 7.4: Parameters used for the toroidal specimen.

**Torus with diameter of 31 nm**

The errors in the reconstruction of the vector potential of the torus are shown in Fig. 7.38 as a function of the number of tilt series. The RMS error in vector direction reduces slightly as the number of tilt series increases, up to $n_s = 6$, after which it begins to increase again. All other metrics, including the mean scaling factor, indicate that the reconstruction worsens consistently with every increase in $n_s$. The errors in the individual two tilt series reconstructions, for the $n_s = 6$ case, as well as
those in the combined reconstruction, are shown in Fig. 7.39. All of the error metrics, except for the directional RMS error, show the reconstruction of the vector potential to be worse in the averaged result than in the worst of the individual reconstructions. The directional RMS error in the combined reconstruction is slightly worse than that in the best of the constituent reconstructions.

Figure 7.37: Toroidal specimen used for simulations in this section. The red arrow indicates the direction of the magnetisation, which is always orthogonal to the position vector, with the origin at the centre of the torus.
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Figure 7.38: Errors as a function of the number of tilt series for the small torus.

Figure 7.39: Total, magnitude, and directional error in reconstructions of the small torus with $\psi = 0$, $\psi = \pi/6$, and $\psi = 2\pi/6$, from left to right, respectively (blue). Errors in the vector potential produced by averaging the other three reconstructions (green).

Slices of the exact vector potential, and the reconstruction errors using both two and six tilt series, are shown in Fig. 7.40. The six tilt series reconstruction appears to smooth out some of the high frequency streaking artefacts in the $x$- and $z$-components of the two tilt series reconstruction. However, very large errors in the $y$-component are exacerbated by using additional tilt series.
Figure 7.40: A one voxel (0.78 nm) slice, approximately 3.1 nm from the origin, of each component of $\mathbf{A}$ for a torus with $\mathbf{M} = M_0 |\hat{y} \times \mathbf{r}|$. Top row: The exact simulated vector potential. Middle row: Error in each component of the vector potential reconstructed using two tilt series. Bottom row: Error in each component of the vector potential reconstructed using six tilt series.
Torus with diameter of 155 nm

The simulations using the toroidal specimen were repeated, this time with a particle five times the size of the previous one. We also scale the defocus and reconstruction domain by the same magnitude, as indicated in Table 7.4. The results here (shown in Figs. 7.41 and 7.42) are very similar to those from Sec. 7.2.4, although here we see a slight reduction in the total RMS error as the number of tilt series is increased from two to four. The magnitudes of the RMS errors are very similar for the two particle sizes, but the mean scaling factor is consistently smaller for the larger particle.

![Figure 7.41: Errors as a function of the number of tilt series for the large torus.](image)

![Figure 7.42: Total, magnitude, and directional error in reconstructions of the large torus with $\psi = 0$, and $\psi = \pi/4$, from left to right, respectively (blue). Errors in the vector potential produced by averaging the other three reconstructions (green).](image)
7.2.5 Antidot lattice

The study of magnetic antidot nanostructures has attracted significant interest from materials scientists [172–177]. In this section, we utilise a simulated square lattice antidot array, loosely based on objects studied in the literature [178–180]. The specimen shape and its non-uniform magnetisation are shown in Fig. 7.43, and the parameters used in this section are shown in Table 7.5. For these simulations, the magnetic vector potential is calculated from the magnetisation using Eq. (4.2).

![Specimen shape of the antidot lattice, with the local magnetisation direction indicated in red.](image)

Table 7.5: Parameters used for the antidot lattice.

<table>
<thead>
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<th>Value</th>
<th>Parameter</th>
<th>Value</th>
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<td>M</td>
<td>See Fig. 7.43</td>
<td>a</td>
<td>150 nm</td>
</tr>
<tr>
<td>$n_x n_y$</td>
<td>240</td>
<td>$\Delta f$</td>
<td>50 $\mu$m</td>
</tr>
<tr>
<td>multislice</td>
<td>$\times$</td>
<td>$d$</td>
<td>$\sim 70$ nm</td>
</tr>
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</table>

Figure 7.43: Specimen shape of the antidot lattice, with the local magnetisation direction indicated in red.
CHAPTER 7. USING ADDITIONAL TILT SERIES

Figure 7.44 shows the errors as a function of $n_s$. Like the those of the torus, the total RMS error increases as the number of tilt series is increased. However, these results show a slight improvement in the reconstruction of the length of the vectors rather than the direction (as was the case for the torus), up to $n_s = 8$.

![Graph](image)

(a) Total, magnitude, and directional RMS errors. (b) Mean scaling factor.

Figures 7.45 and 7.46 show slices, at $y = 18.75$ nm and $y = 14$ nm, respectively of the exact and reconstructed vector potentials using two and eight tilt series.

The errors in the constituent reconstructions are shown as bar plots in Fig. 7.47. These results show only a marginal improvement in the reconstruction of the length of the vectors (as well as the mean vector length), but show a large increase in the total and directional RMS errors.
Figure 7.45: A one voxel (2.34 nm) slice of each component of $\mathbf{A}$ for an antidot array reconstructed from simulated micrographs with a defocus of 50 µm. These slices occur 18.75 nm from the origin, approximately 5 nm from the surface of the film. Shown here are the exact vector potential (top), calculated using Eq. (4.2), and the vector potential reconstructed using two (middle) and eight (bottom) tilt series.
Figure 7.46: A one voxel (2.34 nm) slice of each component of $\mathbf{A}$ for an antidot array reconstructed from simulated micrographs with a defocus of 50 µm. These slices occur approximately 14 nm from the origin, inside the surface of the particle. Shown here are the exact vector potential (top), and the vector potential reconstructed using two (middle) and eight (bottom) tilt series.
7.2. SIMULATIONS

\[ \langle \text{tot } E \rangle, \langle \text{mag } E \rangle, \langle \text{dir } E \rangle \]

\[ 0, 20, 40, 60 \]

\[ 1, 0.5, 0 \]

Figure 7.47: (a) Total, magnitude, and directional error in reconstructions with \( \psi = 0, \psi = \pi/8, \psi = \pi/4 \) and \( \psi = 3\pi/8 \), from left to right, respectively (blue). Errors in the vector potential produced by averaging the other four reconstructions (green). (b) Mean scaling factor for each reconstruction.

7.2.6 Thin film

In this section, we test our polyaxial \textsc{vfet} method using a simulated thin film with six magnetic domains. The parameters used in these simulations are shown in Table 7.6, and the specimen is shown in Fig. 7.48.

<table>
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<th>Parameter</th>
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<td>M</td>
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<td>( a )</td>
<td>1 ( \mu \text{m} )</td>
</tr>
<tr>
<td>( n_s n_t )</td>
<td>48</td>
<td>( \Delta f )</td>
<td>10 ( \mu \text{m} )</td>
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<tr>
<td>multislice</td>
<td>( \times )</td>
<td>( d )</td>
<td>( \sim 40 \text{ nm} )</td>
</tr>
</tbody>
</table>

Table 7.6: Parameters used for the thin film simulations.

The $\text{rms}$ errors, as well as the mean scaling factor, are shown in Fig. 7.49. The errors are high, regardless of the number of tilt series ($> 100\%$ for the total $\text{rms}$ error in most cases), largely because of the small value of $n_t$. All error metrics, except the directional $\text{rms}$ error, show an improvement in the reconstruction for the four tilt series case. Increasing the number of tilt series above $n_s = 4$ worsens the reconstruction according to all metrics.

Figure 7.51 shows the errors in the $n_s = 4$ reconstruction, as well as the two separate constituent reconstructions. Slices of each component of the exact vector potential are shown, along with slices of each component of the errors for the two and four tilt series cases, are shown in Fig. 7.50. The \( x \)-component shows a visible
Figure 7.48: Simulated magnetite thin film with six domains. The red arrows indicate the magnetisation direction, and the black lines indicate the domain boundaries. Note that arrow size does not represent magnetisation strength here.

Figure 7.49: Errors as a function of the number of tilt series for the multi-domain thin film.

reduction in the error with the increase in the number of tilt series, but the other two components show artefacts in the $n_s = 4$ reconstruction that are not present in the $n_s = 2$ reconstruction.
Figure 7.50: A one voxel (7.8 nm) slice of each component of $\mathbf{A}$ for the thin film reconstructed from simulated micrographs with a defocus of 10 µm. These slices occur approximately 3.1 nm from the origin, inside the surface of the particle. Shown here are the exact vector potential (top), and the error in the vector potential reconstructed using two (middle) and eight (bottom) tilt series.
Figure 7.51: Total, magnitude, and directional error in reconstructions of the multiple domain thin film, with $\psi = 0$, and $\psi = \pi/4$, from left to right, respectively (blue). Errors in the vector potential produced by averaging the other two reconstructions (green).

### 7.3 Discussion

In this chapter, we have explored the use of multiple orthogonal pairs of tilt series in vfet. We have shown that the improvement can be significant when the number of tilt series is increased from two, to four or six, but utilising additional tilt series beyond this number can be ineffectual, and can even increase the reconstruction errors.

Our simulations have shown that the reduction in errors from using additional tilt series can be significant if the orientation-dependent errors are large, but provides less of a benefit when the total error is already low for every orientation ($< 20\%$). For our uniformly magnetised specimens, we found this technique to be much more effective at reconstructing the vector potential of the cube (and the cluster of cubes) than the sphere. This suggests that our polyaxial vfet method may not be very useful when the specimen is highly symmetric, but the reason for this is, most likely, that such highly symmetric objects are easily reconstructed with a single pair of tilt series.

While averaging reconstructions in this way can reduce errors arising from magnetisation orientation, this is not the case for all sources of error. An example of this is the errors arising from diffraction artefacts. Comparison of our results for the $z$-oriented sphere using the projection approximation and the multislice methods for micrograph simulation show that deviations from the projection approximation can reduce the effectiveness of our polyaxial technique. However, the results for the
cube show that this method can be highly effective, even in the presence of these diffraction artefacts, provided that the systematic orientation-dependent errors are large.

We have also found that this technique may not be very good at reconstructing the vector potential corresponding to a non-uniform magnetisation. For our toroidal specimen, the directional RMS error improves up until \( n_s = 6 \), but the other three metrics get consistently worse as the number of tilt series is increased. The antidot lattice shows a slight improvement in the RMS error in magnitude (up to \( n_s = 8 \)), with the two other RMS errors getting consistently larger as the number of tilt series is increased from two. The ideal number of tilt series, according to the mean scaling factor, is six. The multi-domain thin film is poorly reconstructed using two tilt series, and utilising an additional pair only improves the reconstruction by approximately 20%.

In our simulations, when the specimen is uniformly magnetised and the projection approximation is used to simulate the micrographs, constructing the final output by selecting the more accurate vector from each reconstruction, for each voxel separately (information that would not be available \textit{a priori} in a real experiment), only results in a very slight increase in accuracy over the result obtained by simply averaging the two reconstructions. For this reason, we assert that this simple averaging method utilising Eq. (7.2) is actually close to the best case scenario of using additional tilt series under the aforementioned conditions.

A significant source of error in VFET is the existence of singular surfaces (discussed in Sec. 2.5). These occur in the components of the vector field that point from the origin to the plane perpendicular to the intersection of the two tilt series; in our case, this is the \( x-z \) plane. A major reason that the use of three orthogonal tilt series reduces errors is that each component can be reconstructed without singular surfaces appearing in the algorithm. Unfortunately, this isn’t the case with our polyaxial method. All pairs of tilt series intersect in the \( y \)-direction, preventing us from avoiding the singular surfaces. However, it may be possible to offset multiple reconstructions by a small polar angle (in addition to the azimuthal angle given in Eq. (7.1)), which would cause the singular surfaces to occur in different planes. These reconstructions could then be combined using a weighting that favours the reconstruction furthest away from a singular surface, for each vector.

For experiments with limited tilt range, the missing wedge in each tilt series results in significant local errors in Fourier space that differ between reconstructions. In dual-axis scalar tomography, this is addressed by averaging points that are contained (in Fourier space) within both tilt series, and selecting data from only one tilt series for points that are inside the missing wedge of the other [181]. An
analogous approach with multiple pairs of tilt series for vector tomography may be capable of addressing the orientation-induced errors and error due to the missing wedge simultaneously. It is important to keep in mind that, in our simulations, data are acquired over the entire $360^\circ$ for each tilt series, so the results presented here do not address this possibility.
Chapter 8

Avenues for future research

8.1 Introduction

In this chapter, we discuss limitations of the work presented in this thesis. We also present some incomplete work, and discuss possible directions for future work on vector field electron tomography. We discuss the effect of slowly decaying vector fields, diffraction artefacts, specimen orientation (as it pertains to directional RMS errors), electron beam attenuation, and regularisation of singularities, respectively.

8.2 Vector potential outside the reconstruction domain

The accuracy of simulated micrographs depends on the size of the sample, and the rate at which the vector potential decays outside the sample, relative to the size of the voxel grid on which the simulation is performed. Ideally, the integration of the vector potential should be performed between the electron source and the detector. In simulations, the vector potential is artificially bounded by the range of the voxel grid. This causes the phase shift to be underestimated for a large sample with a slowly decaying vector potential. The phase shift acquired outside the specimen often dominates the total phase shift [182], so the difference between simulations and experiment may be significant.

This effect also highlights an inherent limitation of the reconstruction process. The micrographs used in the reconstruction are zero-padded outside a radius, \( r_w a \), constraining the reconstructed vector potential to a sphere, \( \Omega \), of the same radius. Any phase shift acquired outside \( \Omega \) will result in errors in the reconstructed vector potential. To investigate this effect, simulations were performed with an array of
magnetic nanospheres. To simulate a best case scenario, the vector potential was set to zero outside $\Omega$ (a spherical aperture function) prior to projecting the phase maps, constraining the vector potential to the region on which the reconstruction was performed. These results were then compared with simulations for which the vector potential was projected along the entire length of the voxel grid (a cylindrical aperture function of radius $r_wa$ and length $a$). While the cylindrical aperture still artificially truncates the vector potential, which is constrained to the voxel grid, it accounts for some of the phase shift that occurs outside $\Omega$, and hence, the projected phases are more realistic. As expected, the more realistic simulation, which includes a phase shift due to the longitudinal component of the vector potential outside $\Omega$, produces a higher error than the idealised simulation that is constrained to $\Omega$. These results are shown in Fig. 8.1.

![Figure 8.1](image)

**Figure 8.1: Comparison of total RMS reconstruction errors for micrographs simulated from a cropped vector potential with (a) cylindrical and (b) spherical aperture functions.**

For these simulations, a uniformly magnetised array of 37 magnetite spheres, each of diameter 40 nm, was used as the sample. This was reconstructed on an $M = 128$ voxel array, with $a = 1 \mu m$, and using $n_t = 101$ micrographs per tilt series. A micrograph of this sample, and the corresponding retrieved magnetic phase, is shown in Fig. 8.2. For these simulations, the imaginary potential $V_i$ was set to zero and Eq. (2.6) was used for the phase retrieval.

This is an important aspect of VFET because the vector potential of any sample
8.2. VECTOR POTENTIAL OUTSIDE THE RECONSTRUCTION

Figure 8.2: (a) Simulated micrograph of the specimen used for comparing the effects of spherical and cylindrical aperture functions. (b) Retrieved magnetic phase.

with a large net magnetic moment will include a significant portion that is large outside Ω, and will be subject to these errors. On the other hand, a sample with a very small net magnetic moment (such as a pair of antiparallel, uniformly magnetised spheres) will be reconstructed more accurately. This may limit the accuracy that can be expected in the reconstruction of certain samples, such as uniformly magnetised specimens. This may seem at odds with the results presented in Ch. 7, where we found that uniformly magnetised specimens can be well reconstructed. However, our error metrics do not distinguish between errors of high, and low, spatial frequencies. Therefore, it is likely that a large proportion of the error in the reconstruction of the non-uniformly magnetised specimens in the high spatial frequencies, and may not occur in lower resolution representations of the vector potential. More work is required to understand this complicated aspect of our results.

A large component of the additional error acquired due to phase shifts outside the reconstruction domain is a uniform scaling of the vector potential, which can be measured using the mean scaling factor $F_s$ defined in Eq. (2.28). The scaling is a result of the electron wave acquiring an additional phase shift outside Ω which is then added to the vector potential during reconstruction. A plot of $F_s$ as a function of defocus, for a uniformly magnetised sample, is shown in Fig. 8.3. Under the hypothesis that the local scaling may vary only slightly over Ω, $F_s$ was used to adjust the magnitude of the reconstructed vector potential at every point. Using simulations, it was found that this method was effective in reducing the RMS error in the reconstruction under some circumstances. An example of this is shown in Fig. 8.4.

While the scaling factor in Fig. 8.3 was obtained using knowledge of the ex-
Figure 8.3: Mean scaling factor $F_s$ for a sample with a large net magnetic moment.

act vector potential, it is possible that, with a better theoretical understanding of
the cause of the scaling, an approximate value could be calculated via a different
method. If the scaling factor can be determined from knowledge of the sample and
the experimental parameters alone, the reconstructed vector potential could be used
to approximate it. This also implies the possibility of a recursive method, where
the scaled reconstruction is then used to produce a better approximation for the scaling
factor. Alternatively, it may be possible to approximate $F_s$ from quantities, such as
characteristic length scales and net magnetic moment, that can be determined using
other experimental methods. For example, the magnetic moment of a magnetised
nanoparticle of arbitrary shape can be determined from a phase map [183].

Future work could include a rigorous analysis of the mathematics behind the oc-
currence of the scaling, as well as more detailed evaluation of simulations to quantify
how the sample and the reconstruction process determine its value. It has also been
observed that the scaling varies from voxel to voxel, and it may be that the scaling
factor at each voxel is actually a function of the magnitude of the vector potential
at that voxel. If so, it may be possible to determine the relationship between the
magnitude of the vector potential and the scaling factor analytically, but it is also
be possible to examine this by plotting the local scaling factor against vector length
for every voxel. This latter approach may give insight into the theory behind the
scaling.

If an accurate value for the scaling factor at each point in $\Omega$ can be derived
from previous knowledge of the sample, obtained using other experimental methods,
or from the results of a reconstruction, it will be possible to greatly increase the
accuracy of the reconstruction without any change to the experimental procedure.
8.3. DIFFRACTION ARTEFACTS

As introduced in Sec. 3.4, and briefly alluded to in Sec. 4.4, experimental micrographs can contain delocalisation artefacts [184–186]. These artefacts result from high angle scattering as the electron beam interacts with the crystal lattice. As the beam is focused by the objective lens, aberrations (spherical aberration and microscope defocus) cause these high angle beams to focus at a different part of the image than the central beams do. These artefacts are sometimes referred to as “ghost images”. While spherical aberration can be reduced significantly in an aberration corrected TEM [145] (which is necessary for accurate phase reconstruction [187]), in our work microscope defocus is a necessary component of the propagation based phase retrieval used to obtain projections of the potentials, and is unavoidable if the TEM is not equipped with an electron biprism for off-axis holography. A simplified explanation of the effect of defocus on the location of the dark field image is shown in Fig. 8.5.

For large defocus, the ghost images in the micrographs do not overlap the bright field image of the particle. This allows these artefacts to be removed by the appropriate selection of defocus and the radius $r_w$ of the apodisation function applied to the micrographs. Because the vector potential is not limited to the particle itself, but also fills the space around it, even when the ghost images do not overlap the bright field images, reducing $r_w$ to remove them also reduces the size of the region.

Figure 8.4: Normalised RMS reconstruction errors for a uniformly magnetised sample (red), and the corresponding errors calculated after correcting the reconstructed vector potential using $F_s$ (shown in Fig. 8.3) for each $\Delta f$ (blue). The RMS errors shown here are errors in the vector magnitude, not total RMS errors.

8.3 Diffraction artefacts

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Figure 8.5: Explanation of ghost images. This simplified visualisation of image formation in the TEM shows how defocus (bottom) causes a shift in the location of the dark field images relative to the bright field image. This shift does not occur at Gaussian focus because the high angle beams carrying the dark field image meet the central (bright field) beam. Spherical aberration, which causes an additional shift, is ignored here.

over which the reconstruction takes place. Depending on the application, important information in the reconstructed vector potential may be lost if $r_w$ is made too small.

Another potential way to avoid the effects of ghost images is to utilise an angle-limiting aperture. This can constrain diffraction contrast to interior of the specimen. The capability to simulate the effect of such an aperture is already included in Kirkland’s multislice code [144], allowing a systematic numerical investigation into the relationship between aperture size and reconstruction error, for a range of specimens, to be performed.

### 8.4 Directional orientation-dependent errors

In Ch. 6, we derived an expression for the total RMS error $\langle \text{tot} E \rangle$ as a function of specimen orientation. Here we address the possibility of obtaining an analytical representation of the corresponding direction RMS error $\langle \text{dir} E \rangle$.

We begin with the assumption that a phase map contains more accurate information about a vector when that vector is oriented in the electron propagation direction at that tilt angle. The consequence of this is that the error due to a given
tilt series has a large component along the axis about which the object rotates in that tilt series. To reduce the complexity of the problem, we assume $E_{i,j,k}$ to be a vector pointing along this axis. Specifically,

$$E_{i,j,k} = |E_{i,j,k}| \hat{z}$$  \hspace{1cm} (8.1)

for the $\theta$ series, and

$$E_{i,j,k} = |E_{i,j,k}| \hat{x}$$  \hspace{1cm} (8.2)

for the $\alpha$ series.

For the time being, we restrict our attention to the $\theta$-tilt series. When the initial magnetisation is oriented in the $z$-direction, every $A_{i,j,k}$ lies within the $x$-$y$ plane. Under the assumption given in Eq. (8.1), when $A_{i,j,k}$ lies in the $x$-$y$ plane, the directional error at that voxel due to phase maps in the $\theta$ series is given by

$$\text{dir} E_{i,j,k} \approx |E_{i,j,k}| / |A_{i,j,k}|,$$  \hspace{1cm} (8.3)

where we have made use of the small angle approximation, which is valid when the error is small. This is illustrated in Fig. 8.6(a), and results in a normalised directional RMS error of

$$\langle \text{dir} E \rangle \approx \frac{\langle \text{tot} E \rangle}{\pi},$$  \hspace{1cm} (8.4)

where the factor of $\pi$ arises from our definition of $\langle \text{dir} E \rangle$ (see Eq. (2.24)). Equation (8.4) relies on the approximation

$$\sum_{i,j,k} \frac{E_{i,j,k}}{|A_{i,j,k}|} \approx \sum_{i,j,k} \frac{E_{i,j,k}}{|A_{i,j,k}|},$$  \hspace{1cm} (8.5)

which is valid when $|A_{i,j,k}|$ is approximately constant for all $i, j, k$ (cf. Eq. (5.23)).

The situation is more complicated when $\hat{m}$ is not oriented along the $z$-axis. Consider an initial magnetisation oriented somewhere in the $x$-$y$ plane. In this case, $A$ contains vectors pointing in every direction within a plane containing $E$. The result is an RMS error in direction proportional to the total RMS error:

$$\langle \text{dir} E \rangle = \Upsilon \frac{\langle \text{tot} E \rangle}{\pi},$$  \hspace{1cm} (8.6)

where $\Upsilon$ is a positive constant smaller than 1, and we have again made use of Eq. (8.5). This is illustrated in Fig. 8.6(b).

The error varies sinusoidally between $\hat{m} = \hat{z}$ and $\hat{m} \cdot \hat{z} = 0$, so Eqs. (8.4) and (8.6) give us the following result:

$$\langle \text{dir} E \rangle_\theta = \frac{1}{\pi} \left[ (\hat{m} \cdot \hat{z})^2 (1 - \Upsilon) + \Upsilon \right] \langle \text{tot} E \rangle,$$  \hspace{1cm} (8.7)
CHAPTER 8. AVENUES FOR FUTURE RESEARCH

\[ \langle \text{dir} E \rangle \propto \langle \text{tot} E \rangle / \pi \]

Figure 8.6: (a) Error \( \text{dir} E_{i,j,k} \) in the reconstructed vector direction, utilising a small angle approximation, when the total error vector \( E_{i,j,k} \) is perpendicular to the exact vector potential \( A_{i,j,k} \) at that voxel. (b) Normalised RMS error in the vector direction \( \langle \text{dir} E \rangle \) when the error vectors \( E_{i,j,k} \) are uniformly random over a plane containing the exact vector potential \( A \).

Figure 8.7 shows surface plots of \( \langle \text{tot} E \rangle \) and \( \langle \text{dir} E \rangle \), for a 40 nm sphere. For completeness, the constants \( A, B, \) and \( C \) (see Ch.6), which are chosen to provide closely matching surface plots of \( \langle \text{tot} E \rangle \), are also provided. The shape of the surface plots in Fig. 8.7(c) is consistent with Eqs. (8.7) and (8.8), which show that the directional RMS error is related to the total RMS error via modulation by a sinusoidal function that is small at \( \gamma = 0 \) and larger at \( \gamma = \pi / 2 \). However, the precise way the errors from these two tilt series combine has not been determined. Additionally, it is not clear how many of the several approximations and assumptions used in this derivation are reasonable. Future work can address these shortcomings to provide a complete, analytical explanation for the orientation dependence of the directional RMS errors.
8.4. DIRECTIONAL ORIENTATION-DEPENDENT ERRORS

\[ \Delta f = 2 \mu m \]
\[ \Delta f = 4 \mu m \]
\[ \Delta f = 6 \mu m \]

\[ \gamma' \]
\[ \gamma \]

\[ A \]
\[ B \]
\[ C \]
\[ \min E \]

\begin{tabular}{|c|c|c|c|}
\hline
A & B & C & \min E \\
\hline
$-5 \times 10^{-3}$ & $-1.0 \times 10^5$ mm$^{-4}$ & $-3.5 \times 10^{13}$ mm$^{-8}$ & $2.8 \times 10^{-2}$ \\
\hline
\end{tabular}

Figure 8.7: Surface plots of (a) $\langle \text{tot} E \rangle$, (b) $\langle \text{tot} \bar{E} \rangle$, and (c) $\langle \text{dir} E \rangle$ for a 40 nm sphere. The table shows the constants used in calculating $\langle \text{tot} \bar{E} \rangle$ via Eq. (6.29).
8.5 Attenuation

Inclusion of attenuation in simulated samples (as described in Ch. 4) revealed that, under some circumstances, the reconstruction is actually improved, despite the attenuation not being accounted for in the phase retrieval process. A comparison of the errors resulting from the reconstruction of an attenuating and non-attenuating sample, respectively, are shown in Fig. 8.8, along with results for the absorbing sample when the in-focus image is used in the phase retrieval algorithm.

![Graphs showing error in reconstruction as a function of image noise and defocus](image)

Figure 8.8: Normalised rms error in VFET reconstructions of simulated micrographs as a function of image noise and defocus for a phase object (green); an attenuating object where the reconstruction assumes a phase object (red); and an attenuating object with the TIE modified by inclusion of the in-focus image to account for the absorption (blue). Here, absorption has been modelled using a complex-valued potential.
An attempt was made to calculate the effect that ignoring attenuation has on the retrieved phase. For simplicity, regularisation and magnetism were neglected in the analysis. See Appendix A for the derivation. In the small defocus limit, the approximate phase, $\varphi'$, obtained using the Tie under the phase object approximation, was found to be given by

$$\varphi' = -\frac{V_i}{2V_i} \left( \exp \left( -\frac{2V_i}{V_i} \varphi \right) - 1 \right),$$  \hspace{1cm} (8.9)

where $\varphi$ is the exact phase, and $V_i$ is the complex part of the potential, which is related to the absorption coefficient, $\rho$, by

$$\rho = 2\pi \frac{V_i}{E\lambda}.$$  \hspace{1cm} (8.10)

Equation (8.9) is equivalent to

$$\varphi' = \int_0^\varphi \exp \left( -\frac{2V_i}{V} \varphi'' \right) d\varphi''$$  \hspace{1cm} (8.11)

This suggests that the phase is more accurately reconstructed, as one would expect, when there is less attenuation$^1$. Since this was found to not always be the case in the simulations described above, there must be an additional effect that is unaccounted for in the derivation of Eq. (8.9).

The reduced error from the addition of absorption to the simulations may be due to inherent regularisation that is not apparent in the formulation of the Tie given by Eq. (2.3). Regularisation of this sort arises in the calculation of the projected thickness of a sample from a single defocussed image. For an object that induces no magnetic phase shift, with uniform mean inner potential, $V$, and uniform attenuation coefficient, $\rho$, the projected thickness can be expressed as [74]

$$T(r_\perp) = -\frac{1}{\rho} \ln \left( \mathcal{F}^{-1} \left\{ \frac{\mathcal{F}\{I(r_\perp, \Delta f)/I^{un}\}}{-\Delta fV|k|^2/2E + \rho} \right\} \right).$$  \hspace{1cm} (8.12)

Thus, regularisation of the singularity at $|k| = 0$ arises naturally due to attenuation induced by the sample, and the singularity is eliminated provided that the chosen defocus is negative. The greater the attenuation coefficient, $\rho$, the greater the suppression of noise amplification near the Fourier space origin, and the natural regularisation can be inferred from experimental images [75]. For the sake of

$^1$While the assumption that, in the limit $V_i \to 0$, the result should be $\varphi' = \varphi$, is used in the derivation of Eq. (8.9), this only accounts for a difference of an additive constant, which does not affect the accuracy of the reconstruction because the phase is only defined up to an additive constant.
simplicity, the above example does not include the effects of magnetism, but it is possible that there is a similar effect at play in the simulations shown in Fig. 8.8. If this is the case, it may be possible, if the object is fairly homogeneous, to choose the regularisation parameter based on measurements, or an a priori assumption about the object, rather than choosing the value arbitrarily. This will also provide higher accuracy and decreased acquisition time compared to using a phase retrieval method that accounts for the in-focus image. Future work regarding errors due to deviations from the phase object approximation will include both regularisation and magnetism in the analysis.

It is presently unknown under what conditions increasing attenuation improves the accuracy of the VFET reconstruction process, and at what magnitude of $\rho$ it becomes necessary to account for attenuation in the phase retrieval process. These are open questions that may be addressed by future work on this topic.

8.6 Regularisation

The methods used for regularisation of singularities in VFET (see Sec. 2.5), as well as the values chosen for the regularisation parameters, for a given regularisation scheme, affect the total reconstruction errors. Increasing the size of the regularisation parameters can reduce errors by improving noise-stability at—and near—singular surfaces. However, increasing these parameters also causes the reconstruction algorithm to deviate from the exact solution and the errors begin to increase. To minimise the reconstruction errors, we seek an optimum value for each regularisation parameter.

Figure 8.9 shows how an increase in the mean scaling factor $F_s$, as the incident noise level $\sigma_{in}$ increases, can be offset by an increase in the regularisation parameters $\delta_{TIE}$ and $\delta_{FBP}$.

The simulations presented in this thesis relied on heuristically chosen values for the regularisation parameters. Future work exploring the relationship between errors and regularisation parameters can be informed by the work presented in Ch. 5. Specifically, that analysis provides a means to accurately predict the noise-induced reconstruction errors for a given regularisation scheme or set of regularisation parameters. Quantifying the errors caused by the regularisation method itself, would enable the determination of optimum values for the regularisation parameters.
Figure 8.9: Effect of regularisation parameters on $F_s$. The mean scaling factor $F_s$ increases with increasing noise $\sigma_{in}$. This scaling error resulting from the image noise can be counteracted by increasing the regularisation parameters $\delta_{TIE}$ and $\delta_{FBP}$, or by uniformly scaling the reconstructed vector potential, provided that $F_s$ can be estimated beforehand.
Chapter 9

Conclusions and experimental recommendations

The purpose of the work presented in this thesis is to enable accurate reconstruction of the magnetic vector potentials of real magnetic nanoparticles. In this brief final chapter, we summarise the key findings of this work, and provide some general recommendations for future experimental work in VFET.

In Ch. 5, we showed that noise can cause significant errors in VFET reconstructions, and that these errors can be reduced by increasing microscope defocus. This is consistent with the literature on phase retrieval. In our simulations, for high noise levels ($\sim 5\mu\text{m}$), the defocus required to reduce noise-induced errors—to the point that they are insignificant in comparison to the total error—is approximately $30\mu\text{m}$. In practice, the image noise is usually closer to 1%, and at this lower noise level, $10\mu\text{m}$ is sufficient to keep the noise-induced error small. These results are unique to the specific specimen and reconstruction method used here; changing only the specimen causes the absolute noise-induced error to remain the same, but as a percentage, the error varies with the magnitude of the vector potential. However, we have presented an accurate model for predicting the RMS errors, in VFET reconstructions, resulting from image noise. This model can be used to quantify the total noise-induced RMS errors, as well as the corresponding RMS errors in vector direction and vector magnitude. Our analysis provides the means to quickly calculate the expected noise-induced errors and adjust the microscope defocus accordingly. These results can help approximate optimal experimental parameters, including defocus, but further research is required before we can use these results to determine optimal parameters precisely.

Chapter 6 explored the dependence of the total RMS error on magnetisation orientation for a uniformly magnetised specimen. We determined the most significant
contributions to the total orientation-dependent error; namely, lack of magnetic phase contrast and errors due to defocus. We then used this understanding to obtain a Taylor series expansion of the error contribution of a single phase map, which was then integrated over all angles used in the tomographic reconstruction. This provided us with a result that describes the functional form of the variation of the total RMS error with changing orientation. These results can assist in the determination of ideal magnetisation orientation in VFET experiments, which we have found can constitute a significant contribution to the total error. We have also supplied some visualisations of the vector potentials, exact and reconstructed, which provides some insight into the qualitative nature of these orientation-dependent errors. Specimen orientation has a complicated effect on reconstruction error, and our analysis is far from exhaustive. Most significantly, we address only uniformly magnetised specimens. We can, however, make some general recommendations.

The ideal orientation for very small particles (<50 nm) appears to be at $\gamma = \pi/2$. In a typical VFET experimental setup, this equates to the magnetisation being in-plane with respect to the support film and TEM grid. It is unclear what effects superparamagnetism may have at this scale, as such effects were not included in our simulations. For larger particles (~100 nm), our simulations indicate that the ideal orientation depends on the shape of the particle, and it may be difficult to predict the exact ideal orientation for a given particle without performing simulations first. For particles larger than 100 nm, the ideal orientation will typically be $(\gamma \mod \pi) = 0$; that is, out-of-plane with respect to the support film and grid.

The RMS error metrics do not, however, tell the whole story, and the type of artefact caused by the magnetisation orientation may be of significance. For these purposes, Figs. 6.4, 6.7, 6.12, and 6.13, may provide the necessary insight into the nature of these orientation-dependent artefacts, which can influence the choice of orientation in experiments.

In Ch. 7, we presented a method to perform VFET using additional pairs of tilt series. Six different specimen shapes were considered in our simulations, and our polyaxial VFET method was tested under a variety of conditions for each. We showed that this method can significantly reduce reconstruction errors, without additional combined acquisition time and the concomitant increase in dose to the specimen. We found this technique to be most successful when the specimen is uniformly magnetised, but further research may improve the utility of this method for the reconstruction of arbitrary magnetisation configurations. Our polyaxial reconstruction method shows that the reconstruction errors can be reduced by more uniformly distributing the acquired images. For specimens with a strong net magnetic moment, utilising additional tilt series in the reconstruction can be advantageous. In
our simulations, with the exception of the spherical specimen, the reconstruction of uniformly magnetised objects is always improved by using four tilt series rather than two, and is sometimes further improved by increasing that number to six. The results for non-uniform magnetisations are much less consistent, and we do not recommend utilising more than two tilt series without additional prior investigation into the use of polyaxial VFET. Additionally, the large number of combinations of specimen geometry and microscope and reconstruction parameter sets examined in Ch. 7 provide a broad range of visual and quantitative results that can be used as a lookup table for the purposes of estimating expected error magnitudes and qualitative reconstruction features based on known properties of the specimen and experimental design.
Appendix A: Two derivations of Eq. (8.9)

Derivation 1

We seek to express the approximate phase $\phi'$, which is obtained when the in-focus intensity is assumed to be constant, in terms of the exact phase $\phi$. The TIE (Eq. (2.3)) can be written in terms of $\phi'$:

$$-k \frac{\partial I_0}{\partial z} = I_{\text{in}} \nabla^2 \phi'. \tag{1}$$

Although, here, we have made use of the phase object approximation, Eq. (1) is an equality because $\phi'$ is defined such that it satisfies this equation. We can now equate the right hand sides of Eqs. (2.3) and (1):

$$I_{\text{in}} \nabla^2 \phi' = \nabla \cdot (I_0 \nabla \phi). \tag{2}$$

Taking the inverse divergence of both sides gives the result

$$\nabla \phi' = \frac{I_0}{I_{\text{in}}} \nabla \phi + \nabla \times F \tag{3}$$

$$= \exp [-\rho t] \nabla \phi + \nabla \times F, \tag{4}$$

where $\rho$ is the attenuation coefficient, assumed constant within the specimen, $t$ is the projected thickness of the specimen, and $F$ is an arbitrary vector function. The decaying exponential in Eq. (4) is a reformulation of $I_0/I_{\text{in}}$ in terms of Beer’s law. The attenuation coefficient is related to the imaginary potential via

$$\rho = 2\pi \frac{V_i}{E\lambda}. \tag{5}$$
Substituting Eq. (5) into Eq. (4) gives the result

$$\nabla \varphi' = \exp \left[ -\frac{2\pi V_i t}{E\lambda} \right] \nabla \varphi + \nabla \times F.$$  

(6)

From Eq. (2.1), the projected thickness can be written in terms of the mean inner potential $V$ which is, by definition, constant inside the specimen:

$$t = \frac{\varphi E\lambda}{V \pi}.$$  

(7)

Here, we are neglecting the effects of magnetism. Substituting Eq. (7) into Eq. (6), we obtain

$$\nabla \varphi' = \exp \left[ -\frac{2V_i}{V} \varphi \right] \nabla \varphi + \nabla \times F$$

(8)

$$= -\frac{V}{2V_i} \nabla \exp \left[ -\frac{2V_i}{V} \varphi \right] + \nabla \times F.$$  

(9)

Taking the inverse gradient of Eq. (9), we get

$$\varphi' = -\frac{V}{2V_i} \left( \exp \left[ -\frac{2V_i}{V} \varphi \right] - 1 \right) + G,$$  

(10)

where $G$ is a harmonic function satisfying $\nabla G = \nabla \times F$, which incorporates the constant of integration arising from the inverse gradient operation. For $V_i \ll V/\varphi$, this expression should produce $\varphi' \approx \varphi$. In this case, a linear approximation for the exponential gives

$$\varphi' = -\frac{V}{2V_i} \left( 1 - \frac{2V_i}{V} \varphi \right) + G$$

$$= \varphi - \frac{V}{2V_i} + G.$$  

(11)

To have agreement between between $\varphi$ and $\varphi'$, $G$ is set to $\frac{V}{2V_i}$. Then Eq. (10) becomes

$$\varphi' = -\frac{V}{2V_i} \left( \exp \left[ -\frac{2V_i}{V} \varphi \right] - 1 \right).$$  

(12)

**Derivation 2**

For an object that induces no magnetic phase shift, with uniform mean inner potential, $V$, and uniform attenuation coefficient, $\rho$, the projected thickness, $t$ can be written as [188]
\[ t(r_\perp) = -\frac{1}{\rho} \ln \left( \mathcal{F}^{-1} \left\{ \rho \frac{\mathcal{F} \{ I(r_\perp, \Delta) \} / I_{\text{in}}}{-\Delta V |k|^2/2E + \rho} \right\} \right). \] (13)

This results in a phase shift of
\[ \varphi(r_\perp) = -\frac{\pi V}{\lambda \rho E} \ln \left( \mathcal{F}^{-1} \left\{ \rho \frac{\mathcal{F} \{ I(r_\perp, \Delta) \} / I_{\text{in}}}{-\Delta V |k|^2/2E + \rho} \right\} \right). \] (14)

Solving for \( I_r \) gives
\[ I(r_\perp, \Delta) = \mathcal{F}^{-1} \left\{ \frac{1}{\rho} \mathcal{F} \left\{ \exp \left( -\frac{E \lambda \rho \varphi}{\pi V} \right) \right\} \left( -\Delta V |k|^2/2E + \rho \right) I_{\text{in}} \right\}. \] (15)

If the phase retrieval is performed using Eq. (2.3), and the approximation \( I_0 \approx I_{\text{in}} \), the retrieved phase is
\[ \varphi' = -\frac{\pi V}{\lambda \rho E} \exp \left( -\frac{E \lambda \rho \varphi}{\pi V} \right) \Rightarrow \varphi' = -\frac{V}{2V_i} \exp \left( -\frac{2V_i}{V} \varphi \right). \] (16)

Because the phase is defined only up to an additive constant, which depends on the algorithm used to retrieve the phase, the transformation \( \varphi' \to \varphi' + C \), where \( C \) is an arbitrary constant, can be made:
\[ \varphi' = -\frac{V}{2V_i} \left( \exp \left( -\frac{2V_i}{V} \varphi \right) \right) - C. \] (17)

As in the first derivation, here we assume that \( \varphi' \approx \varphi \) when \( V_i \ll V/\varphi \), which leads to an expression for \( C \):
\[ C = -\frac{V}{2V_i}. \] (18)

Combining Eqs. (17) and (18) provides the final result for the approximate phase:
\[ \varphi' = -\frac{V}{2V_i} \left( \exp \left( -\frac{2V_i}{V} \varphi \right) \right) - 1. \] (19)

Note that this result is equivalent to
\[ \varphi' = \int_0^\varphi \exp \left( -\frac{2V_i}{V} \varphi'' \right) d\varphi''. \] (20)
Glossary

Accents

¯ A macron above a symbol denotes an analytically predicted value. 44
ˆ A circumflex above a vector denotes a unit vector. 27, 40
˜ A tilde above a function denotes a Fourier transformed quantity. 14, 27

Acronyms

**CCD** charge-coupled device. 9, 20, 22
**DPC** differential phase contrast. 10
**EHT** electron holographic tomography. 4
**FBP** filtered backprojection. i, 2, 4–6, 13, 15, 19, 20, 36, 50, 56, 79, 80
**FFT** fast Fourier transform. 15, 22, 23
**LTEM** Lorentz transmission electron microscopy. vii, 1, 5, 8, 9, 11, 29
**TEM** transmission electron microscope. xi, 2, 4, 7–10, 106, 131, 132, 142, 150

**TIE** transport-of-intensity equation. i, vii, xviii, 3–5, 11–13, 15, 20, 35–37, 61, 63, 68, 136, 137, 145, 151


**VFT** vector field tomography. 3
**2D** two-dimensional. 1, 29
**3D** three-dimensional. i, 1, 2, 4, 26, 29, 46
**CT** computed tomography. 2

Errors

**E** Error in the reconstructed vector potential. This is a vector field equal to $\mathbf{A}^{\text{rec}} - \mathbf{A}_r$. 17

$\langle \text{dir} E \rangle$ Normalised RMS value of the angle between the exact and reconstructed vector potentials. 18

$\langle \text{mag} E \rangle$ Normalised RMS error in the magnitude of the reconstructed vector potential. 18

**A\text{noise}** Noise induced component of the reconstructed vector potential. 37
\( \text{rms} \bar{E}_\varphi \) RMS error in a single phase map. 63

\( \langle \text{tot}E \rangle \) Total normalised RMS error between the exact and reconstructed vector potentials. 17, 63

\( \sigma_{\text{in}} \) Noise level in a micrograph in the absence of a specimen. 20, 31, 33, 42, 44, 138

\( \sigma_{\text{out}} \) Standard deviation of the noise induced component of the reconstruction error. 37

**Image processing**

\( K_{\text{max}} \) Magnetic phase contrast measured when the magnetisation of the specimen (assumed uniform) is perpendicular to the electron beam. 62, 63

\( K \) Magnetic phase contrast. 61–63

\( H \) Microscope transfer function. 28

\( I \) Intensity of the electron beam at a given pixel in the detector. 28, 32

\( M_{\text{ms}} \) Linear resolution (in pixels) used for multislice simulations. 29, 33, 86

\( M \) Width of each micrographs in pixels. Reconstructions are performed on a grid of \( M^3 \) voxels. 14, 29, 33, 37, 53, 86

\( \mathbf{k} \) Two-dimensional wave vector representing the coordinates of the Fourier transform of the image. 11, 28

\( a \) Width of each micrograph in units of length. 15

\( r_w \) Radius of the circular apodisation function applied to micrographs, expressed as a fraction of \( a \). 33, 127, 131, 132

\( I_{\text{ideal}} \) The expected intensity at a given pixel; that is, the intensity in the image in the absence of noise. 46

\( I_0 \) In-focus intensity—the value of \( I \) measured at \( \Delta f = 0 \). 11

\( \varphi \) Phase of the electron beam. 27, 137

\( \varphi_e \) Electrostatic component of the exit phase. 10

\( \varphi_m \) Magnetic component of the exit phase. 10

\( \varphi_0 \) Phase of the electron beam at the image plane. 11

**Mathematical symbols**

\( O \) The object to be reconstructed. 14, 60, 80

\( \Pi \) Function that selects a random variable from a Poisson distribution with mean equal to its argument. 32

\( \mathcal{O} \) Order of error. 20, 21, 40–42

\( \mathcal{F}^{-1} \) Inverse Fourier transform. 11

\( \mathcal{F} \) Fourier transform. 11

\( \nabla_{\perp} \) Two-dimensional gradient operator. 11

\( \mathbf{r}_{\perp} \) Position vector in the plane perpendicular to the electron propagation direction. 10

**Microscope**

\( E \) Accelerating potential of the TEM. 10, 33, 146

\( N \) Mean number of detected electrons per pixel in the absence of a sample. 20, 32
GLOSSARY

\( \Delta f \) Microscope defocus in units of length. xiv, 28, 31, 82, 89, 90, 105, 112, 117, 121, 128

\( \lambda \) Electron wavelength. 10, 28, 146

\( k \) Electron wavenumber or, when accompanied by subscripts, spatial frequency. 11

Regularisation

\( \delta_{\text{FBP}} \) Parameter used to regularise singular surfaces in the filtered backprojection algorithm. 16, 33, 51, 138, 139

\( \delta_{\text{INT}} \) Parameter used to regularise singularities caused by the division by \( I_0 \) in the TIE. 16

\( \delta_{\text{TIE}} \) Parameter used in regularising the transport-of-intensity equation. 15, 33, 51, 138, 139

Specimen

\( M \) Magnetisation vector field. 27

\( \rho \) Attenuation coefficient of the material, related to imaginary potential via \( \rho = \frac{2\pi e}{\hbar} V_i \). 137, 138, 145, 146

\( V_i \) Imaginary potential used for simulating attenuation. 128, 136, 137, 146, 147

\( V \) Mean inner potential. 10, 137, 146, 147

\( A \) Exact vector potential. 10

\( m \) Magnetic dipole moment. 27, 62

\( d \) Length scale (e.g., diameter) of the specimen. 68, 82, 89, 105, 117, 121

\( M_0 \) Magnitude of the magnetisation. xiii–xvi, 27, 84, 86, 88, 91, 94, 97, 103, 104, 108

Tomography

\( T \) Tilt series; a set of phase maps over a range of angles. 14

\( A_{\text{rec}} \) Reconstructed vector potential. 14

\( \Omega \) Spatial domain of validity of the reconstruction in three dimensions, typically a sphere with radius \( r_w \). 127

\( \alpha \) Angle of rotation about the x-axis. 14

\( \psi \) Angle between pairs of tilt series. 80

\( \theta \) Angle of rotation about the z-axis. 14

\( n_s \) Number of tilt series employed. xiv, xv, 80–83, 85–87, 89, 90, 92, 95, 98–101, 105–107, 110, 112, 114, 116–118, 121, 122, 125

\( n_t \) Number of micrographs in \( \pi \) radians of each tilt series. xiv, xv, 14, 53, 81, 82, 89, 92, 95, 98–101, 105, 112, 117, 121

\( t \) Projected thickness of the specimen. 145–147

\( F_s \) Scaling factor given by the average magnitude of the reconstructed vector potential divided by the average magnitude of the exact vector potential. xvii, xviii, 71, 81, 86, 106, 110, 129, 130, 138, 139
Bibliography


Appendix B: Publications

The research presented in Ch. 5 resulted in the publication of a first-author paper, which is included in this appendix. A preprint of a second paper is also included.
Analysis of noise-induced errors in vector-field electron tomography

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Vector-field electron tomography (VFET) reconstructs electromagnetic vector fields of magnetic nanomaterials using transmission electron microscopy. The theory behind this reconstruction process is well established, but the practical implications of experimental errors and how they affect the accuracy of the reconstructed vector fields is not well understood, hindering progress in the use of these techniques for routine magnetic characterization of nanomaterials. Here, we present an analysis of the propagation of stochastic errors through a VFET algorithm. A method for determining the contribution of image noise to errors in a reconstructed vector potential is derived. Simulations are performed to test the validity of this method when applied to shot noise, which shows good agreement with theory.

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I. INTRODUCTION

Vector-field electron tomography (VFET) is a relatively new technique for three-dimensional (3D) magnetic imaging, combining phase retrieval with vector tomography to reconstruct electromagnetic vector fields from transmission electron micrographs [1–3]. The characterization of magnetic nanomaterials is a crucial aspect of a wide range of topical areas of research, including spintronics [4], biomedical nanotechnology [5], magnetic recording media [6], and the development of nanomotors [7].

There are many experimental techniques for the characterization of magnetic nanomaterials, such as Kerr microscopy [8], magnetic exchange force microscopy [9], and spin-polarized scanning tunneling microscopy [10], which can be employed to examine components of the magnetization on the surface of the sample. Lorentz transmission electron microscopy (LTEM) has significant advantages over these techniques in that it can be used to probe the internal magnetic structure of a sample [11]. Using LTEM, the phase of the electron wave function at the exit surface of the magnetic specimen can be measured from out-of-focus micrographs or electron holograms, but this provides only two-dimensional projections of the potentials. VFET combines LTEM with tomography (the reconstruction of an object function from its projections) to enable a complete, 3D magnetic characterization of the sample. For example, VFET can be used to reconstruct all three components of the vector potential of a magnetic nanoparticle at all points inside, and around, a sample [12].

Conventional scalar tomography has been employed for decades as a means to probe the internal structure of 3D objects. X-ray computed tomography and positron emission tomography are routinely used as medical diagnostic tools, but scalar tomography has also proved extremely useful in a diverse range of other fields. Examples include the imaging of microscopic biological samples using synchrotron radiation [13] and transmission electron microscopy [14], structural characterization of semiconductor devices using scanning transmission electron microscopy [15], 3D imaging of binary stars using Doppler measurements [16], delineation of magma bodies using seismic tomography [17], and 3D imaging of macroscopic mechanical parts using neutron tomography [18].

The theory of scalar tomography has been extended to the reconstruction of vector fields [19]. The majority of the work on vector tomography relates to the use of acoustic time-of-flight or Doppler measurements to reconstruct fluid flows. Such applications include the imaging of blood flow [20], flue gas velocity in coal-fired power stations [21], and water velocity in lakes [22]. VFET is the application of vector tomography to electromagnetic vector fields, such as magnetic fields, magnetic vector potentials, and current densities [12], using the transmission electron microscope (TEM). VFET requires information of the phase shift induced in the electron beam as it traverses an electromagnetic sample. The exit phase of the beam is measured over two or more tilt series and is used to tomographically reconstruct the vector fields in three dimensions. This requires the phase to be reconstructed from intensity measurements, which can be performed using off-axis electron holography [23] or propagation-based phase retrieval [24].

The theoretical foundations of VFET are well established, with the process having been employed to experimentally reconstruct magnetic nanomaterials [2,25]. However, there are many known sources of error, including image noise, image misalignment, and technical limitations that result in a missing wedge of information in each tilt series, the propagation of these errors through the reconstruction process is not well understood.

In this work, we consider one significant source of error, namely, shot noise in the recorded micrographs. The effect of image noise on the reconstructed vector field has been briefly investigated by Yu et al. [3]. Their work tested the stability of various VFET algorithms when white Poisson noise was added to simulated micrographs, noting qualitative features of the noise-induced artifacts in the reconstructed vector fields.

A significant source of error in VFET reconstructions is the amplification of noise near Fourier-space singularities in both the phase retrieval and vector tomography algorithms. The
effects of noise can be ameliorated by regularization of these singularities. The results obtained by Yu et al. [3] provide a means to identify certain noise-induced artifacts, which they found can occur in the form of streaking in the reconstructed vector potential, which result from the presence of the singularities in the reconstruction algorithms. However, their work did not provide a quantitative analysis of the propagation of noise-induced errors through the reconstruction. Indeed, a quantitative analysis of the propagation of noise through these algorithms has not been presented in the literature. In this work, we address the limitations of previous work by providing a thorough, quantitative, and analytical study of the propagation of image noise through a VFET algorithm.

Here, we derive expressions to calculate expected VFET reconstruction errors as a function of image noise. We use three different root-mean-square (rms) error metrics to quantify the total reconstruction error, and the errors in the direction and magnitude of the reconstructed vectors, separately. We compare the errors predicted using these analytical models with those measured from VFET reconstructions performed using simulated micrographs.

Owing to the large amount of data required and technical considerations, such as alignment of the tilt series, VFET can be a time-consuming and computationally demanding process; consequently, it is important to have a thorough understanding of how variables such as spatial resolution and image acquisition time affect accuracy. This is a key motivation for this work.

We close this introduction with a brief overview of the remainder of the paper. Section II presents the theoretical foundations of VFET, from phase retrieval to tomographic reconstruction, including relevant considerations such as the nature of shot noise. In Sec. III, we define the rms error metrics used to quantify the accuracy of our reconstructions, and derive analytical expressions for these errors as a function of image noise. In Sec. IV, we present errors calculated from tomographically reconstructed vector fields using simulated micrographs, and compare these with our analytical results. We make some concluding remarks in Sec. V.

II. THEORETICAL BACKGROUND

In the TEM, electromagnetic information about the sample is encoded in the phase of the electron wave function at the exit surface via the Aharonov-Bohm effect. The contribution to the phase shift due to the electrostatic potential \( V(r_{\perp}, z) \) is given by [26]

\[
\phi_e(r_{\perp}) = \frac{\pi}{\lambda} \int V(r_{\perp}, z) dz, \tag{1}
\]

where \( z \) is the distance along the optical axis, \( r_{\perp} \) is a position vector in the plane normal to the optical axis, \( E \) is the electron accelerating potential, and \( \lambda \) is the electron wavelength. The contribution to the phase shift due to the magnetic vector potential \( A(r_{\perp}, z) \) is given by [27]

\[
\phi_m(r_{\perp}) = -\frac{e}{\hbar} \int A(r_{\perp}, z) \cdot dz, \tag{2}
\]

where \( e \) is the magnitude of the electron’s charge, and \( \hbar \) is the reduced Planck constant. Under the projection approximation, these two components of the phase shift can be interpreted as projections of the electrostatic potential and the magnetic vector potential, respectively. Here, we consider projections to be straight line integrals of the vector potential in the electron propagation direction. This definition is consistent with the projection approximation of electron holography, which ensures applicability of the projection-slice theorem (which underpins the reconstruction algorithm). Measuring \( \phi_m \) from micrographs at multiple angles over two tilt series provides a way to reconstruct the vector potential. A schematic of the experimental setup is shown in Fig. 1. In the remainder of this section we discuss the theory of the processes used to perform the reconstruction, from phase retrieval to vector tomography, as well as regularization and shot noise.

A. Phase retrieval using the transport of intensity equation (TIE)

In order to obtain projections of the vector potential for its reconstruction, the phase must be inferred from recorded micrographs. This can be achieved using off-axis holography [23] or propagation-based phase retrieval [24]. We use the latter for our simulations. Specifically, a transport of intensity equation (TIE) algorithm is used to compute the phase from simulated out-of-focus micrographs. In addition to being computationally efficient, this method is linear under the phase object approximation. This simplifies the analysis of the propagation of errors from the micrographs to the phase, relative to iterative methods such as the Gerchberg-Saxton algorithm [28]. The TIE relates transverse phase gradients to

![Figure 1](image-url)
the longitudinal derivative of the intensity [27]:

\[ -\frac{k}{\partial z} I_0 = \nabla_\perp \cdot (I_0 \nabla \phi_0), \]

where \( k \) is the wave number, \( I_0 \) and \( \phi_0 \) are the intensity and phase at the image plane \( (z = 0) \), respectively, and

\[ \frac{\partial I_0}{\partial z} \bigg|_{z=0} = 0. \]

(4)

To obtain the phase, given measurements of both \( I_0 \) and \( \frac{\partial I}{\partial z} \), Eq. (3) can be solved using a Fourier-transform method [24]

\[ \psi(r_\perp) = \frac{k}{4\pi^2} \mathcal{F}^{-1} \left\{ \frac{k}{|k|^2} \cdot \mathcal{F} \left[ \frac{1}{I_0} \mathcal{F}^{-1} \left( \frac{k}{|k|^2} \right) \right] \right\}, \]

(5)

where \( k \) is the spatial frequency, and \( \mathcal{F} \) and \( \mathcal{F}^{-1} \) denote the two-dimensional Fourier transform and its inverse, respectively. Under the phase object approximation, when the object is very weakly attenuating, the in-focus intensity \( I_0 \) can be replaced with the incident intensity \( I^0 \), hence [29]

\[ \psi(r_\perp) = \frac{k}{4\pi^2 I^0} \mathcal{F}^{-1} \left\{ \frac{k}{|k|^2} \cdot \mathcal{F} \left[ \frac{1}{I^0} \mathcal{F}^{-1} \left( \frac{k}{|k|^2} \right) \right] \right\}. \]

(6)

This approximation is sufficient for the purposes of noise analysis [30]. For the simulations performed here, both Eqs. (5) and (6) are used separately to compare the results of each with analytical results derived using Eq. (6). For the former, the equation

\[ I_0 \approx \frac{I(r_\perp, \Delta f) + I(r_\perp, -\Delta f)}{2\Delta f} \]

(7)

is used, which reduces the number of images required from three to two. The longitudinal derivative of the intensity can be approximated from out-of-focus micrographs. For this work, due to its simplicity and computational efficiency, a two-image central difference method is employed, i.e.,

\[ \frac{\partial I_0}{\partial z} = \frac{I(r_\perp, \Delta f) - I(r_\perp, -\Delta f)}{2\Delta f}. \]

(8)

The phase obtained using the TIE contains information about both the magnetic and electrostatic properties of the sample. For the purposes of reconstructing the vector potential, it is only the magnetic component that is of interest. There are multiple methods to obtain this. The dependence of the electrostatic phase shift on the electron wavelength can be exploited to obtain the magnetic phase by varying the accelerating potential [31]. Another method assumes that the electrostatic phase shift is proportional to the sample thickness, and uses the in-focus image to calculate the electrostatic contribution and remove it from the total retrieved phase [32]. The separation of the magnetic and electrostatic phases can also be achieved by flipping the sample and recording additional micrographs with the electron beam now traveling in the opposite direction relative to the sample. This allows the magnetic component of the phase to be extracted by exploiting the different time-reversal symmetries of the electrostatic and magnetic Aharonov-Bohm shifts [33]. For phase retrieval using the TIE, this separation can be applied to the micrographs themselves [34], or to the retrieved phases [35]. We use the latter method, for which

\[ \psi_{\text{mag}} = \frac{\psi_f - \psi_r}{2}, \]

(9)

where \( \psi_f \) and \( \psi_r \) are the phase shifts imparted by the sample before and after flipping, respectively. Experimentally, \( \psi_r \) is obtained by rotating the sample by 180° about an axis orthogonal to \( z \), and reversing the resulting phase maps about the axis of rotation.

**B. Vector tomography**

Tomography is the reconstruction of an object function from its projections. Scalar tomography typically takes a series of projections about a single tilt axis and reconstructs a scalar function from these projections. Vector tomography requires the acquisition of additional tilt series to obtain enough information about the specimen to reconstruct a vector field in three dimensions.

For this work, the vector potential \( A(x, y, z) \) is reconstructed using a filtered back projection (FBP) algorithm, which uses the projection-slice theorem in cylindrical coordinates, in combination with the Coulomb gauge condition. This algorithm is given by [3]

\[ A(x, y, z) = \int_0^\pi \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \mathcal{T}^0 \left| \frac{k^0_x}{k^0_y} \frac{k^0_y}{k^0_z} \right| \frac{k^0_z}{k^0_x} \frac{k^0_x}{k^0_y} \left| k^0_z \right| \left| k^0_y \right| \left| k^0_x \right| \]

\[ \times e^{2\pi i (k^0_x x \cos \theta + y \sin \theta + z \alpha)} d\theta dk_d, da, \]

(10)

where \( \theta \) and \( \alpha \) are positive angles of rotation about the \( z \) and \( x \) axes, respectively, \( T^0 \) and \( T^\alpha \) are the tilt series associated with these directions, \( \alpha \) represents Fourier-transformed quantities, and \( k^0_x = k / \sin \theta \) and \( k^0_x = k / \cos \alpha \) are radial spatial frequencies. \( T^0 \) and \( T^\alpha \) are given by [3]

\[ T^0(x, z) = -\frac{\hbar}{e} \psi_{\text{mag}}(x, z) \]

(11)

and

\[ T^\alpha(x, y) = -\frac{\hbar}{e} \psi_{\text{mag}}(y, x). \]

(12)

The geometry for tilt series acquisition is shown in Fig. 2.

In practice, a discrete form of Eq. (10) is used for the reconstruction. For this work, we perform the reconstructions on a cubic voxel grid, and it is convenient to make a change of variables which enables the algorithm to be expressed as three summations. Given this, the algorithm we use can be expressed as

\[ A_{m,n,p} = \frac{\pi}{n_f - 1} \sum_{\mu=0}^{n_f-1} \sum_{l,j,m=-M/2}^{M/2-1} \frac{M^2}{M^2} \frac{M^2}{M^2} \]

\[ \times \left( \mathcal{T}^0(n_f, n_f) e^{2\pi i (n_f + j + m)} + \mathcal{T}^\alpha(n_f, n_f, n_f) e^{2\pi i (n_f + j + m)} \right), \]

(13)
where $M$ is the number of pixels along each direction of the input images; $n_t$ is the number of magnetic phase maps in each tilt series; $m$, $n$, and $p$ are the voxel indices in real space; $n' = m \cos \theta + n \sin \theta$; and $n'' = n \cos \alpha + p \sin \alpha$. The coefficients $\eta_{i,j,\mu}$ and $\eta_{i,j,\mu}$ are given by

$$\eta_{i,j,\mu} = \frac{l^2}{aM(l^2 + j^2)} \begin{bmatrix} \frac{l^2 \sin^2 \alpha + j^2}{l \sin \theta} \\ -l \cos \theta \\ -j \cot \theta \end{bmatrix}$$

and

$$\eta_{i,j,\mu} = \frac{l^2}{aM(l^2 + j^2)} \begin{bmatrix} j \tan \alpha \\ l \sin \alpha \\ -j \cot \alpha \end{bmatrix}$$

Here, $a$ is the width of the input micrographs, and $\theta$ and $\alpha$ are both given by $\pi \mu/(n_t - 1)$. Bilinear interpolation is used to transform from radial to Cartesian coordinates. The inverse discrete Fourier transforms implicit in Eq. (13) can be computed using a fast-Fourier-transform algorithm (see, for example, Ref. [36]).

C. Regularization of singularities

The phase retrieval and FBP algorithms used here contain Fourier-space singularities, resulting in numerical instability and amplification of noise. In order to overcome these problems, solutions can be forced to be finite at the singularities by modifying the algorithms using regularization. In previous work [37], Tikhonov regularization [38] has been employed to deal with the singularity in Eq. (5) at $|k| = 0$, and we follow this approach. This is achieved by applying the following transformation to Eq. (6):

$$|k|^2 \rightarrow \frac{|k|^2 + \delta_{\text{TIE}}^2}{|k|^2}$$

where $\delta_{\text{TIE}}$ is a regularization parameter having dimensions of a reciprocal length. We obtain

$$\varphi(r, \omega) = \frac{k}{4\pi^2l^2m} \begin{bmatrix} |k|^2 F \left[ \frac{\omega}{k} \right] \\ \frac{1}{k} |k|^2 + \delta_{\text{TIE}}^2 \end{bmatrix}$$

This makes the solution finite at $|k| = 0$, allowing numerical computation, and also suppresses amplification of noise near $|k| = 0$. For $|k|/\delta_{\text{TIE}} \gg 1$, Eq. (17) reduces to Eq. (6), but at (and near) $|k| = 0$, the regularized TIE deviates from the exact solution. Because the regularization does not arise naturally from the theory, it induces errors in the reconstruction. Finding an appropriate choice for a regularization parameter is a compromise between choosing a large enough value to adequately suppress errors, while keeping it small enough to remain consistent with the theory and avoiding unnecessarily introducing artifacts from the regularization itself. Tikhonov regularization is employed wherever singularities occur in the algorithms. For Eq. (5), the transformation used is

$$|k|^2 \rightarrow |k|^2 + \frac{\delta_{\text{TIE}}^2}{|k|^2}$$

We have introduced the factor of 2 for consistency in the scale of the regularization parameters. That is, we require that the results obtained using Eq. (5) match those of Eq. (6) when $I_0 = I_m$. The use of Eq. (18) for the former and Eq. (16) for the latter ensures this, provided that $\delta_{\text{TIE}}$ is small.

Equation (10) contains singular surfaces at $k_x = 0$ for $A_x$ and $A_y$. In terms of the tilt angles, these singular surfaces are at $\theta = 0$ and $\alpha = \pi/2$. Due to the practical limitations of recording tilt series over the entire range of $\pi$ radians, it is natural to remove the singular surface at $\theta = 0$ by simply omitting this image from the reconstruction process. For the singular surface at $\alpha = \pi/2$, we again use Tikhonov regularization:

$$\frac{1}{\cos \alpha} \rightarrow \frac{\cos \alpha}{\cos^2 \alpha + \delta_{\text{FBP}}^2}$$

where $\delta_{\text{FBP}}$ is the regularization parameter.

The division by $I_0$ in Eq. (5) can also cause numerical instability in the presence of noise. We address this by regularizing with

$$I_0 \rightarrow I_0^2 + \delta_{\text{INT}}^2 \frac{I_0}{I_0}$$

where $\delta_{\text{INT}}$ is the regularization parameter. Because noise in $I_0$ does not impact greatly on the errors in the reconstruction, the choice of $\delta_{\text{INT}}$ does not affect the noise-induced errors examined in this work. We arbitrarily set $\delta_{\text{INT}} = 0.08I_m$, which is small enough to avoid introducing significant systematic errors in the retrieved phases.

The recorded micrographs are subject to shot noise due to the finite number of electrons recorded at the detector. Shot noise is dependent on beam current and acquisition time [39], and propagates through the phase retrieval process. Singularities in the phase retrieval algorithm can amplify the noise, and the methods of regularization, as well as the parameters used, can be chosen for optimal suppression of this amplification.

D. Shot-noise model

Previous work on the effects of noise in VFET used a white Gaussian [1] or Poisson [3] noise model. These models treat the noise level as a constant, with no variation across the image. In our work, we use a more realistic shot-noise model that takes into account the change in noise level with changing intensity and provides a more accurate distribution of noise over the simulated images, particularly for large defocus and/or when there is significant attenuation.
The scale used for the intensity is arbitrary, and is proportional to the mean number of detected electrons per pixel. Thus, we can define a constant of proportionality $\kappa$ such that

$$I^\text{in} = \kappa N,$$  \hspace{1cm} (21)

where $N$ is the mean number of detected electrons per pixel in the absence of a sample. The noise-free intensity $I^\text{ideal}_{i,j}$ at a given pixel is related to the mean number of detected electrons $N_{i,j}$ at that pixel by

$$I^\text{ideal}_{i,j} = \kappa N_{i,j}.$$  \hspace{1cm} (22)

The actual intensity, taking shot noise into account, is then

$$I_{i,j} = \kappa \Pi(N_{i,j}),$$  \hspace{1cm} (23)

where $\Pi(N_{i,j})$ produces a random integer selected from a Poisson distribution with mean $N_{i,j}$. This leads to the relation

$$\frac{I_{i,j}}{I^\text{ideal}_{i,j}} = \frac{\Pi(N_{i,j})}{N_{i,j}}.$$  \hspace{1cm} (24)

From Eqs. (21) and (22), the mean number of detected electrons is

$$N_{i,j} = \frac{I^\text{ideal}_{i,j}}{\kappa N}.$$  \hspace{1cm} (25)

Substituting this into Eq. (24), the intensity is then given by

$$I_{i,j} = \Pi\left(\frac{I^\text{ideal}_{i,j}}{\kappa N}\right) \frac{I^\text{in}}{N}.$$  \hspace{1cm} (26)

We define an incident noise level

$$\sigma_{\text{in}} = \frac{\sqrt{N}}{N},$$  \hspace{1cm} (27)

which is the fractional noise level in a micrograph recorded in the absence of a sample. This results in slightly underestimated noise levels when the sample is highly attenuating, but more closely matches an experimental scenario where the beam current, acquisition time, and detector behavior is typically the same for every image. The intensity in the noisy image is then

$$I_{i,j} = \Pi\left(\frac{I^\text{ideal}_{i,j}}{\sigma_{\text{in}}^2 I^\text{in}}\right) \sigma_{\text{in}}^2 I^\text{in}.$$  \hspace{1cm} (28)

III. PROPAGATION OF SHOT NOISE THROUGH A VECTOR-FIELD ELECTRON TOMOGRAPHY ALGORITHM

Quantifying errors in reconstructed vector fields differs from quantifying those of scalar reconstructions in that the total error involves errors due to differences in the magnitude of the input and output vector at each voxel, and differences in the orientation of the vectors. What makes a reconstruction accurate depends on the application. For example, for some applications, the magnitude of the vector potential may be of little interest, and accuracy is only required for the direction of the reconstructed vectors. For other applications, it may be important to know the accuracy of the magnitude of the vectors. Other quantities that may be of interest include the curl and divergence of the vector field, and metrics can also be chosen to highlight the accuracy of a reconstructed field in terms of these quantities.

For this work, we use three different metrics to quantify the errors. The first is designed to quantify the total difference between the exact and reconstructed vector fields, and the other two are motivated by two attributes that may be of particular interest, namely, direction and magnitude, respectively. The total normalized rms error metric is defined by

$$E^\text{tot} = \sqrt{\frac{\sum_{i,j,k} |A^\text{rec}_{i,j,k} - A_{i,j,k}|^2}{\sum_{i,j,k} |A_{i,j,k}|^2}},$$  \hspace{1cm} (29)

where $A^\text{rec}$ is the reconstructed vector potential. Our directional rms error is defined as the rms value of the angle between the exact and reconstructed vectors at each voxel and is given by

$$E^\text{dir} = \frac{1}{M^3 \pi^2} \sum_{i,j,k} \cos^{-1}\left(\frac{A_{i,j,k} \cdot A^\text{rec}_{i,j,k}}{|A_{i,j,k}| |A^\text{rec}_{i,j,k}|}\right)^2,$$  \hspace{1cm} (30)

where the factor of $\pi$ is included so that $E^\text{dir}$ is expressed as a fractional error rather than an angle. The normalized rms error in the magnitude of the vector potential is given by

$$E^\text{mag} = \sqrt{\frac{\sum_{i,j,k} (|A^\text{rec}_{i,j,k}| - |A_{i,j,k}|)^2}{\sum_{i,j,k} |A_{i,j,k}|^2}}.$$  \hspace{1cm} (31)

In simulations, the noise-induced component $E^\text{tot}_{\text{noise}}$ of the normalized total rms error can be extracted using

$$E^\text{tot}_{\text{noise}} = \sqrt{E^\text{tot}(\sigma_{\text{m}}^2 - E^\text{tot}(0)^2)}.$$  \hspace{1cm} (32)

The noise-induced components of the directional and magnitude errors, $E^\text{dir}_{\text{noise}}$ and $E^\text{mag}_{\text{noise}}$, respectively, can be obtained in the same manner. In the remainder of this section, we derive analytical expressions for $E^\text{tot}_{\text{noise}}$, $E^\text{dir}_{\text{noise}}$, and $E^\text{mag}_{\text{noise}}$.

A. Effect of shot noise on total RMS errors

Ignoring other sources of error, we can express the reconstructed vector field at a given voxel as

$$A^\text{rec}_{i,j,k} = A_{i,j,k} + A^\text{noise}_{i,j,k},$$  \hspace{1cm} (33)

where $A^\text{noise}$ is the noise-induced component of the vector field.

Substituting Eq. (33) into Eq. (29), $E^\text{tot}_{\text{noise}}$ can be obtained by calculating

$$E^\text{tot}_{\text{noise}} = \sqrt{\frac{\sum_{i,j,k} |A^\text{noise}_{i,j,k}|^2}{\sum_{i,j,k} |A_{i,j,k}|^2}},$$  \hspace{1cm} (34)

where $\sigma_{\text{out}}$ is the standard deviation of the noise-induced reconstruction error. Equation (34) is valid for large $M$. In deriving an analytical model for $\sigma_{\text{out}}$, we utilize the noise variance analysis of McDowell et al. [40]. In the case of phase retrieval using the TIE, the input signal is the longitudinal derivative $\partial_z I$ of the intensity across the image plane. The error in $\partial_z I$ due to noise, when two defocused micrographs are
used to approximate the derivative, is given by [30]
\[ \sigma'_m = \frac{\sigma_{in}}{\sqrt{2\Delta f}}. \] (35)

To determine \( \sigma_{out} \), we express the noise in the Fourier transform of the through focal derivatives as a random complex number \( c_{i,j} \) with variance
\[ \langle c_{i,j}c_{i,j}^* \rangle = I_0^2 \sigma_{in}^2, \] (36)
where \( \langle . . . \rangle \) denotes the expectation value. It is worth noting that, by virtue of being the Fourier transform of a real function (the noise in the through focal derivatives), \( c_{i,j} \) has the Hermitian property
\[ c_{i,j} = c_{-i,-j}^*, \] (37)
but this does not affect our derivation. In practice, a white noise model will not be sufficient to accurately predict errors from all sources of noise that may be present. For this reason, we introduce the filter functions \( H^\theta \) and \( H^\alpha \) to account for the spectrum of the image noise in each tilt series; if this is independent of defocus, these filters applied to the approximated through focal derivatives are identical to those applied to the images used to calculate them. These functions can also incorporate any additional filtering that is performed in an effort to reduce the effects of noise on the reconstruction. Considering only errors due to noise, the Fourier-transformed through focal derivatives constructed from experimental micrographs are then given by
\[ \partial_i \hat{I}_{i,j,\mu}^\theta = \partial_i \hat{I}_{i,j,\mu}^{\text{ideal,}\theta} + \frac{\hbar}{e} H_{i,j,\mu}^\theta c_{i,j,\mu}^\theta, \] (38)
and
\[ \partial_i \hat{I}_{i,j,\mu}^\mu = \partial_i \hat{I}_{i,j,\mu}^{\text{ideal,}\mu} + \frac{\hbar}{e} H_{i,j,\mu}^\mu c_{i,j,\mu}^\mu, \] (39)
where \( \partial_i \hat{I}_{i,j,\mu}^{\text{ideal,}\theta} \) and \( \partial_i \hat{I}_{i,j,\mu}^{\text{ideal,}\mu} \) are the noise-free components of \( \partial_i \hat{I}_{i,j,\mu}^\theta \) and \( \partial_i \hat{I}_{i,j,\mu}^\mu \), respectively.

Noting the linearity of the reconstruction algorithm [Eq. (13)], and considering Eqs. (33), (38), and (39), we can immediately write an expression for the noise-induced component of the reconstructed vector field:
\[ \hat{A}_{m,n,p} = \frac{\pi}{n_1 - 1} \sum_{\mu=0}^{n_1-1} \frac{1}{M^2} \sum_{l,j=-M/2}^{M/2-1} \frac{\hbar}{2} e^{i(\ln^*+jp)/M} H_{l,j,\mu}^\theta H_{l,j,\mu}^\mu c_{l,j} e^{2\pi i(\ln^*+jm)/M}. \] (40)

The variance \( \sigma_{out}^2 \) in each voxel of the reconstruction is the second moment of \( A_{\text{noise}} \):
\[ \sigma_{out}^2 = \frac{\hbar^2}{e^2 q} \left( \frac{\pi}{n_1 - 1} \sum_{\mu=0}^{n_1-1} \frac{1}{M^2} \sum_{l,j=-M/2}^{M/2-1} \xi_{l,j} \left( H_{l,j,\mu}^\theta H_{l,j,\mu}^\mu c_{l,j} e^{2\pi i(\ln^*+jm)/M} + H_{l,j,\mu}^\mu H_{l,j,\mu}^\theta c_{l,j} e^{2\pi i(\ln^*+jm)/M} \right) c_{l,j} \right)^2 \] (41)

where \( q \) is the number of phase maps used to obtain a single magnetic phase map (typically \( q = 2 \)) and
\[ \xi_{l,j} = a^2 M^2 k^2 \] (42)
is the spatial frequency response of the phase retrieval algorithm under the phase object approximation. The noise is uncorrelated between phase maps both within, and between, tilt series, which leads to the result
\[ \sigma_{out}^2 = \frac{I_0^2 \sigma_{in}^2 \pi^2 h^2}{q M^4 (n_1 - 1)^2 e^2} \times \sum_{\mu=0}^{n_1-1} \frac{1}{M^2} \sum_{l,j=-M/2}^{M/2-1} H_{l,j,\mu}^\theta H_{l,j,\mu}^\mu \left( |H_{l,j,\mu}^\theta|^2 + |H_{l,j,\mu}^\mu|^2 \right). \] (43)

where we have made use of Eq. (36). In practice, the spatial frequency spectrum of the noise may be a function of tilt series and angle, but for simplicity, we assume that it is a function only of spatial frequencies, and have removed these dependencies in \( H \). We treat the Poisson noise in our simulations as being approximately uniform, and set \( H = 1 \) in our analytical calculations.

The summations in Eq. (43) can be computed very quickly, and require no knowledge of the properties of the sample. Any

regularization scheme can also be easily incorporated, which enables the effects of regularization on errors due to noise to be calculated quickly.

B. Effect of shot noise on directional rms errors

In this section, we derive approximations for the contribution to \( E_{\text{dir}} \) due to noise. Substituting Eq. (33) into (30), the directional error is given by
\[ E_{\text{noise}} = \frac{1}{M^2 \pi^2} \sum_{i,j,k} \cos^{-1} \left( \frac{|A_{i,j,k} + \hat{A}_{i,j,k}^{\text{noise}}|}{|A_{i,j,k} + \hat{A}_{i,j,k}^{\text{noise}}|} \right)^2, \] (44)
where the circumspace is used to denote a unit vector. The magnitude of the reconstructed vector can be determined using the cosine rule, and is given by
\[ |A_{i,j,k} + A_{i,j,k}^{\text{noise}}| = |A_{i,j,k}| \sqrt{1 + \frac{|A_{i,j,k}^{\text{noise}}|^2}{|A_{i,j,k}|^2} + 2 \frac{\hat{A}_{i,j,k} \cdot A_{i,j,k}^{\text{noise}}}{|A_{i,j,k}|^2}}. \] (45)
Substituting this into Eq. (44), and using the binomial approximation, gives

$$E_{\text{dir}}^n \approx \sqrt{\frac{1}{M^3 \pi^2} \sum_{i,j,k} \left( \cos^{-1} \left(1 - \frac{R_{i,j,k}^2 + 1}{2 |A_{i,j,k}|^2} \right) \right)^2 \left(1 - \frac{|A_{i,j,k}^{\text{noise}}|^2}{|A_{i,j,k}|^2} - \frac{\hat{A}_{i,j,k} \cdot A_{i,j,k}^{\text{noise}}}{|A_{i,j,k}|} \right)^2}. \tag{46}$$

We now introduce the notation

$$R_{i,j,k} = \frac{A_{i,j,k} \cdot A_{i,j,k}^{\text{noise}}}{|A_{i,j,k}| |A_{i,j,k}^{\text{noise}}|}. \tag{47}$$

Here, $R_{i,j,k}$ is a random variable on the interval $[-1,1]$ which, if the orientation of $A_{i,j,k}^{\text{noise}}$ is uniformly random, has a probability density given by

$$p(R_{i,j,k}) = \frac{1}{\pi} \frac{d}{dx} \cos^{-1} \left| x \right|_{x=R_{i,j,k}} = \frac{1}{\sqrt{1 - R_{i,j,k}^2}}, \quad -1 < R_{i,j,k} < 1$$

$$= 0, \quad \text{elsewhere.} \tag{48}$$

Discarding terms of $O(|A_{i,j,k}^{\text{noise}}|^3)$ in Eq. (46) gives

$$E_{\text{dir}}^n = \sqrt{\frac{1}{M^3 \pi^2} \sum_{i,j,k} \left( \cos^{-1} \left(1 - \frac{R_{i,j,k}^2 + 1}{2 |A_{i,j,k}|^2} \right) \right)^2 \left(1 - \frac{|A_{i,j,k}^{\text{noise}}|^2}{|A_{i,j,k}|^2} - \frac{\hat{A}_{i,j,k} \cdot A_{i,j,k}^{\text{noise}}}{|A_{i,j,k}|} \right)^2} \tag{49}$$

and the approximation

$$\cos^{-1}(1 - x) \approx \sqrt{2x}, \tag{50}$$

valid for small $x$ (i.e., low-noise levels), simplifies this to

$$E_{\text{dir}}^n = \sqrt{\frac{1}{M^3 \pi^2} \sum_{i,j,k} \left( \frac{\sqrt{2 R_{i,j,k}^2 + 1}}{|A_{i,j,k}|^2} \right)^2}. \tag{51}$$

Using the binomial approximation, and discarding terms of $O(|A_{i,j,k}^{\text{noise}}|^3)$, gives

$$E_{\text{mag}} = \sqrt{\frac{\sum_{i,j,k} |A_{i,j,k}|^2 \left(1 + |A_{i,j,k}^{\text{noise}}|^2 / |A_{i,j,k}|^2 + 2 R_{i,j,k} A_{i,j,k}^{\text{noise}} / |A_{i,j,k}| \right)^2}{\sum_{i,j,k} |A_{i,j,k}|^2}}. \tag{55}$$

The rms value of $|A_{i,j,k}^{\text{noise}}|$ is $\sigma_{\text{out}}$, while that of $\sqrt{2 R_{i,j,k}^2 + 1}$ is $\sqrt{2}$, and because these are assumed to be independent, they can be evaluated separately to give

$$E_{\text{dir}}^n = \sqrt{\frac{2}{M^3 \pi^2} \sum_{i,j,k} \frac{\sigma_{\text{out}}^2}{|A_{i,j,k}|^2} \approx \frac{\sqrt{2}}{\pi} E_{\text{dir}}^n \sigma_{\text{out}}} \tag{52}$$

where we have made use of the approximation

$$\frac{1}{M^3} \sum_{i,j,k} \frac{\sigma_{\text{out}}^n}{|A_{i,j,k}|^n} \approx (E_{\text{dir}}^n)^n, \tag{53}$$

which is valid when $|A_{i,j,k}|$ is approximately constant for all $i,j,k$. This requirement implies that Eq. (52) may be inaccurate for geometries where the magnitude of the vector potential varies significantly over the domain of reconstruction. For example, a pair of uniformly magnetized particles with antiparallel moments has large $|A_{i,j,k}|$ near each particle, but decays quickly outside the specimen, and directional errors calculated in this way may be significantly less accurate than for a single dipole.

Because we have dropped higher-order terms in the derivation of $E_{\text{dir}}^n$, the accuracy of Eq. (52) in approximating noise-induced errors is expected to be poor for large $\sigma_{\text{in}}/\Delta f$. For accurate results in this region, the $O(|A_{i,j,k}^{\text{noise}}|^3)$ terms in Eq. (46) can be retained, to give the improved approximation

$$E_{\text{dir}}^n = \frac{E_{\text{dir}}^n}{\pi} \sqrt{2 + E_{\text{dir}}^n / 2}, \tag{54}$$

where we have again made use of Eq. (53).

**C. Effect of shot noise on rms errors in magnitude**

Substituting Eq. (45) into (31), we get

$$E_{\text{mag}} = \frac{\sum_{i,j,k} R_{i,j,k}^2 |A_{i,j,k}^{\text{noise}}|^2}{\sum_{i,j,k} |A_{i,j,k}|^2} \sigma_{\text{out}} \sqrt{\frac{\sum_{i,j,k} R_{i,j,k}^2}{\sum_{i,j,k} |A_{i,j,k}|^2}} = \frac{E_{\text{tot}}}{\sqrt{2}}. \tag{56}$$

**IV. COMPARISON OF RECONSTRUCTION ERRORS: SIMULATIONS AND ANALYTICAL RESULTS**

In this section, we present analytical estimates for reconstruction errors, based on the analysis presented in Sec. III, and compare these with the errors in reconstructions performed on simulated TEM data. There are a multitude of parameters that affect reconstruction errors, and these are too numerous to address comprehensively here. For this reason, we restrict our attention to the variation of two parameters of significance, namely, defocus and image noise.
For our simulations, the sample shape was measured from experimental scalar tomograms of an octahedral magnetite nanoparticle. The nanoparticles were produced by dissolving FeSO$_4$·7H$_2$O in water and initiating precipitation using potassium hydroxide and potassium nitrate, according to the procedure described in Ref. [41]. The resulting dry magnetite powder was mixed with ethanol, placed in an ultrasonic bath, and then dispersed onto a holey carbon TEM support using a pipette. Specimen charging was observed, so the nanoparticles were covered with approximately 5 nm of carbon in a Gatan 682 precision etching and coating system, which noticeably reduced spurious charging effects.

The specimen was loaded into an FEI Titan 80–300 kV TEM, operated at 300 kV in the bright field Lorentz imaging modality. Using 5° angular increments over ±65°, a tilt series of bright field images was acquired at a nominal defocus of −10 μm, using a dedicated Fischione 2040 dual-axis tomography holder. With the same angular sampling and range, another tilt series was collected after first rotating the specimen by 90° in plane at a nominal defocus of +10 μm. All images were aligned using cross-correlation algorithms. Defocus-induced rotations between the overfocused and underfocused images were corrected by optimizing the height of cross-correlation peaks, in response to induced relative rotations between image pairs using bilinear interpolation. For each of the two mutually orthogonal tilt series, defocus derivatives were computed and used as input for surface tomography reconstruction of the nanoparticle morphology. The implicit Laplacian-based contrast in the defocus derivatives served as ideal input for image-processing ridge detection since the straight edges of the nanoparticle polyhedron were clearly highlighted in the experimental data. The unstructured point cloud data from both tomograms was combined to partly fill the missing wedge. For details of the reconstruction algorithm used, see Ref. [42].

The simulated sample was constructed by tracing the surface of the particle in three dimensions using 3D modeling software (Blender 2.67). The shape of the simulated sample, along with the experimental tomograms from which it was constructed, is shown in Fig. 3.

A mean inner potential of 17 V [44], mass magnetization of 80 emu/g [45], and density of 5.18 g/cm$^3$ [46] were chosen to simulate a magnetite sample. An imaginary potential of 0.8i V, inferred from the experimental micrographs of the sample being simulated, was added to the mean inner potential to simulate attenuation. These parameters were used to calculate total phase shifts using the projection approximation, and defocused images were then obtained by means of a transfer function [47]. Shot noise of the form given by Eq. (28) was then added to the simulated micrographs.

Figure 4 shows a through focal derivative computed from simulated micrographs, along with one computed from experimental micrographs of the actual sample. Underfocus and overfocus micrographs were simulated for every θ and α, with $M = 64$ and $n_t = 31$. After the addition

![FIG. 3. (Color online) Shape of sample mask used for our simulations (gray) and surface point cloud obtained using scalar tomography [42] applied separately to two tilt series [red (dark gray) and blue (light gray)]. For an animated version of this figure, see Supplemental Material [43].](image)

![FIG. 4. Example of through focal derivatives computed from micrographs with $\Delta f = -10 \text{ μm}$ and $\sigma_{in} = 1\%$ using (a) simulated micrographs and (b) experimental micrographs.](image)
FIG. 5. Slice of the $x$ component of the exact vector potential at $z = 0$ (top), and the same slice taken from reconstructions using various values of $\Delta f$ and $\sigma_{in}$. 

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<th>$\Delta f$ (µm)</th>
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of noise, the micrographs were used to retrieve the phases that comprise $T^\theta$ and $T^\nu$. $\Delta \mathbf{r}$ was reconstructed using Eq. (13), and then used to calculate $E_{\text{tot}}^{\text{noise}}$, $E_{\text{dir}}^{\text{noise}}$, and $E_{\text{mag}}^{\text{noise}}$. These simulations were performed for a range of $\sigma_\text{in}$ and $\Delta f$. Each simulation was performed 10 times for each $\sigma_\text{in}$ and $\Delta f$, with the results averaged, to suppress fluctuations about the mean rms error for large $\sigma_\text{in}$. The $x$ component of the exact vector potential is shown alongside that of the reconstructed vector potentials, for various combinations of $\Delta f$ and $\sigma_\text{in}$, in Fig. 5.

The predictive power of our analysis is quantified using a normalized residual metric defined by

$$\chi_{\text{tot}} = \frac{E_{\text{noise}}^{\text{tot}} - E_{\text{noise}}^{\text{tot}}}{E_{\text{noise}}^{\text{tot}}}$$

where the macron denotes the analytically derived estimate. For the accuracy of $E_{\text{noise}}^{\text{dir}}$ and $E_{\text{noise}}^{\text{mag}}$, the metrics $\chi_{\text{dir}}$ and $\chi_{\text{mag}}$ are similarly defined.

Figure 6 shows $E_{\text{noise}}^{\text{tot}}$ and $E_{\text{noise}}^{\text{tot}}$, respectively, as contour plots over $\sigma_\text{in}$ and $\Delta f$. It is evident that increasing the image noise will result in increased reconstruction errors, as the errors due to noise in the micrographs propagate through the reconstruction algorithm and translate to noisy reconstruction data. The reduction in error as the defocus is increased is less intuitive. This occurs because phase contrast in the micrographs increases with increasing defocus, provided that the defocus is not too large in magnitude. When $\sigma_\text{in}$ remains constant, the ratio of the noise level to the phase contrast signal drops as the defocus is increased, and this results in a reduced noise-induced error in the phase [30]. It is this error that then propagates to the reconstructed vector field.

The contour plots in Fig. 6 are presented to provide an overview of how the total reconstruction error depends on image noise and defocus, in both the simulated and analytical results. For the results that follow, we use 3D surface plots to represent the error data because they provide a clearer comparison between simulated and analytical results, and serve to better highlight the regions in which these results deviate from each other.

Figure 7 shows surface plots comparing the analytically derived estimates $E_{\text{noise}}^{\text{tot}}$ obtained using Eq. (43) in conjunction with Eq. (34), with $E_{\text{noise}}^{\text{tot}}$ obtained from the reconstruction of simulated micrographs by applying Eqs. (29) and (32). These represent the same data presented in Fig. 6, with the addition of simulated results where the reconstruction relied on the phase object approximation.

Because $I_{\text{ideal}}^{\text{dir}}$ varies across the image, the noise level is not, in practice, constant. However, for the analytical calculations here, $H_{i,j}$ is set to unity for simplicity, approximating the noise as uniformly distributed.

The strong agreement between analytical results and simulations shows that Eq. (43) can be used to accurately predict rms errors from the signal-to-noise ratio of transmission electron micrographs, and can be used to determine
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FIG. 9. (Color online) (a) Analytically derived values $E_{\text{noise}}$ obtained using Eq. (56) [red (dark gray)], along with the results of simulations $E_{\text{noise}}$ using Eq. (5) for the phase retrieval [blue (black)], and Eq. (6) [green (light gray)]. (b) Normalized residual $\chi_{\text{abs}}$ plotted against $\sigma_{\text{et}}/\Delta f$ for each method (blue circles and green crosses, respectively).

minimum experimental acquisition times required to keep the reconstruction errors due to noise below a desired limit. It can also be used to quickly compute the noise-induced errors for a variety of experimental parameters. These results also show that, under the conditions used in these simulations, shot noise is adequately modeled as white noise.

Figures 8(a) and 8(c) show a comparison of $E_{\text{noise}}$ and $E_{\text{noise}}^*$ calculated using Eq. (52). The analytical results agree well with the simulations for small $\sigma_{\text{et}}/\Delta f$ but, as expected, deviate for larger $\sigma_{\text{et}}/\Delta f$. Figures 8(b) and 8(d) show that the analytical estimates can be significantly improved by employing Eq. (54).

The results for $E_{\text{noise}}$ are shown in Fig. 9, again highlighting that there is good agreement between the analytical results and those obtained by reconstructing the vector potential from simulated micrographs.

FIG. 10. (Color online) Surface plots showing reconstruction errors (a) $E_{\text{tot}}$, (b) $E_{\text{dir}}$, and (c) $E_{\text{mag}}$. Each subfigure shows both the results obtained using Eq. (5) for the phase retrieval, in blue (black), and Eq. (6), in green (gray).

$E_{\text{tot}}$, $E_{\text{dir}}$, and $E_{\text{mag}}$ are shown as surface plots in Fig. 10. These can be compared with the extracted noise induced errors shown in Figs. 7–9 to see how significant the noise-induced component of the total error is in relation to the combined contribution of all other sources of error.

V. CONCLUSION

Shot noise is an unavoidable source of error in any imaging process. In VFET, shot noise in the micrographs results in errors in both the magnitude and direction of the reconstructed vector potential. An understanding of the relationship between image noise and the accuracy of VFET reconstructions provides various benefits. For a particular application, there may be a maximum tolerable error in a tomographic reconstruction. Understanding the propagation of noise through the reconstruction algorithm, combined with an understanding of other sources of error, will provide a maximum allowable noise level in the micrographs. This in turn will enable determination of a minimum acquisition time per image, allowing for minimal sample degradation and microscope time.

The results presented in Sec. IV show that the propagation of noise in VFET can be accurately quantified for a given reconstruction algorithm, including any regularization scheme, and a known noise level and spatial frequency spectrum. For our simulations, we have used a shot-noise model in which the variance of the noise at each pixel in each image used in the reconstruction is dependent on the intensity of the electron beam. For this reason, the spatial dependence of the noise level is also a function of tilt angle, so the power spectrum of the noise is, in practice, a complicated function of spatial frequencies and tilt angle. However, we have shown shot noise to be adequately modeled as white noise for the purposes of analyzing propagation of errors in the reconstruction. There are other factors that can be taken into account in determining a value for the noise filters used for analytical calculations of VFET reconstruction errors, in cases where setting it to unity is not sufficient. One possible example is the blurring of shot noise in the detector, which is commonly modeled using a modulation transfer function [48] or noise transfer function [49], which could be incorporated into these filter functions. A filtered noise model may be useful for determining the contribution of other stochastic error sources, such as thermal detector noise, and the power spectral density of these noise sources would typically be independent of tilt series and tilt angle, allowing a simple two-dimensional filter to be used in the noise analysis.

With modification, the noise analysis presented here can be used for many other calculations of noise-induced errors in reconstructions where linear, Fourier-transform methods have been employed. This includes, but is not limited to, those arising from other VFET algorithms, as well as scalar tomography and phase retrieval algorithms.

We have derived expressions for the total, directional, and magnitude rms noise-induced errors in vector fields reconstructed from transmission electron micrographs using an FBP algorithm. The results of simulations presented here show that noise-induced errors can be quickly computed to accurately estimate their contribution to the total rms errors in a VFET reconstruction.
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Effect of specimen orientation on the accuracy of vector field electron tomography

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Abstract: Vector field electron tomography (VFET) reconstructs vector fields based on phase maps recorded from two or more orthogonal tilt series. The tomographic reconstruction of vector fields involves considerations beyond those involved in the reconstruction of scalar fields. Here we examine the effect of initial magnetization orientation on reconstruction errors. The orientation of a magnetic particle affects the contrast in the phase maps. This, in turn, affects the accuracy of the reconstructed vector fields. We derive expressions that model the dependence of reconstruction errors on initial specimen orientation when using a filtered backprojection algorithm to reconstruct a vector potential from two tilt series. We compare these analytical results with those from numerical simulations. Our results can inform experimental procedures, such as sample preparation techniques and the choice of tilt series orientations. Specimen orientation can be a significant source of error in VFET, and our results can provide the means to minimize these errors.

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OCIS codes: (110.6960) Tomography, (100.5070) Phase retrieval.

References and links
1. **Introduction**

Magnetic nanomaterials are of interest in many areas of materials science and condensed matter physics. Examples exist in such diverse fields as spintronics [1], magnetic information storage [2], biomedical engineering [3], and the study of nanoscale grain boundaries in hard magnets [4]. In order to fully characterize these materials, it is essential to have techniques to accurately measure their magnetic properties at the nanoscale. Vector field electron tomography (VFET) is a method for the characterization of such magnetic nanomaterials [5, 6, 7]. It has significant advantages over other methods used to characterize magnetic nanomaterials. Unlike magnetic force microscopy [8], it can reconstruct vector fields inside the specimen as well as on the surface, and unlike conventional Lorentz transmission electron microscopy [9], it reconstructs the vectors in three dimensions rather than only projections.

Tomography is the process of reconstructing an object function from its projections. These projections are recorded at multiple angles, as one or more tilt series, by rotating the specimen relative to the source and detector. The original function is then reconstructed from these tilt series, which can be achieved using a backprojection algorithm [6] or iterative reconstruction techniques [10]. Reconstruction of a three-dimensional vector field requires at least two tilt...
series [5]. In VFET, the tilt series are composed of phase maps which can be obtained using propagation based phase retrieval [11] or off-axis electron holography [12].

VFET can be used to reconstruct a variety of vector fields, such as the magnetic induction field, electric current density, and magnetic vector potential [11]. For this work, we consider only the reconstruction of the magnetic vector potential.

The defining feature of a vector field is that, in addition to its magnitude, it has a direction associated with each point in space. The magnitude of the projected vector potential, and hence the magnetic phase contrast, changes as the particle is rotated through at least one of the tilt series, regardless of the geometry of the specimen. Maximizing the average contrast can improve the reconstruction if errors in the retrieved phases are low, but can worsen its accuracy if errors in the retrieved phases are high. This results in errors in the reconstructed vector potential that depend on the initial orientation of the specimen relative to each tilt series. Additionally, the optimum initial orientation can depend on factors such as attenuation and defocus, which affect the accuracy of the retrieved phases.

In this work, we quantify the effect that specimen orientation has on the resulting errors for a reconstruction algorithm utilizing two orthogonal tilt series. In Sec. 2 we review the theoretical basis for the VFET reconstruction process and our error analysis. In Sec. 3 we demonstrate how the concept of contrast can be used to quantify the error contribution from a single phase map due to specimen orientation. In Sec. 4 we use these results to derive equations that explain the changes in reconstruction error as a function of initial specimen orientation, and in Sec. 5 we present results of simulations which we compare with the analytical results. Some concluding remarks are given in Sec. 6.

2. Theoretical background

In the transmission electron microscope (TEM), the electron beam may be modeled as a complex scalar field (beam-like unbound wavefunction) that acquires a phase shift over planes perpendicular to the beam axis due to the Aharonov-Bohm effect as it traverses an electromagnetic specimen [13]. The phase, which is composed of an electrostatic part and a magnetic part, can be measured using electron holography, either in-line or off-axis, and additional measurements can be used to separate the two components [14, 15]. The principle of the magnetic Aharonov-Bohm effect is illustrated in Fig. 1.

VFET reconstructs vector fields from phase maps recorded for multiple angles over at least two orthogonal tilt series [5]. The phase maps, which correspond to the exit phase of the electron wavefunction, can be obtained via any of a number of phase reconstruction methods, including linear [16] and iterative [17] propagation based (in-line) methods, as well as off-axis [18] holographic techniques. We use a propagation based method, reconstructing the exit phase from out-of-focus micrographs using the transport of intensity equation (TIE) [19, 16]. In our numerical work, we simulate one over- and one underfocused micrograph for each angle in each tilt series, and approximate the in-focus micrograph from the two out-of-focus micrographs. After retrieving the phase from the simulated micrographs and separating the magnetic and electrostatic components, we reconstruct the vector potential from the magnetic phase maps using a filtered backprojection algorithm. The remainder of this section will address these processes in greater detail.

The phase shift induced in the electron wavefunction due to the magnetic vector potential \( \mathbf{A}(\mathbf{r}_\perp, z) \) is given by [19]

\[
\phi_m(\mathbf{r}_\perp) = -\frac{e}{\hbar} \int \mathbf{A}(\mathbf{r}_\perp, z) \cdot d\mathbf{z},
\]

where \( z \) is the electron propagation direction, \( \mathbf{r}_\perp \) is the position vector in the plane orthogonal
to $z$, $e$ is the magnitude of the electron’s charge, and $\hbar$ is the reduced Planck constant. There is a corresponding electrostatic phase shift given by

$$\phi_e(r_\perp) = \frac{\pi}{\lambda} \int V(r_\perp, z) dz,$$

(2)

where $E$ is the accelerating potential of the TEM, $\lambda$ is the electron wavelength, and $V$ is the electrostatic potential of the specimen. The total phase shift is then given by

$$\phi = \phi_e + \phi_m.$$

(3)

To obtain this phase from defocused non-interferometric intensity measurements alone, we employ a phase retrieval method based on the TIE, which relates transverse derivatives of the phase to the longitudinal derivative of the intensity [19, 20, 16]:

$$-k \frac{\partial I_0}{\partial z} = \nabla_\perp \cdot (I_0 \nabla_\perp \phi_0).$$

(4)

Here $k$ is the electron wave number, $I_0$ and $\phi_0$ are the intensity and phase at the image plane, respectively, $\nabla_\perp$ is the two-dimensional gradient operator over $r_\perp$, and

$$\frac{\partial I_0}{\partial z} \equiv \frac{\partial I(r_\perp, z)}{\partial z} \bigg|_{z=0}.$$

(5)

In our reconstructions, the derivative of the intensity is obtained using a two-image central difference approximation given by [21]

$$\frac{\partial I_0}{\partial z} \approx \frac{I^+ - I^-}{2\Delta f},$$

(6)

where $\Delta f$ is the defocus, and $I^+$ and $I^-$ are the over- and under-focus intensities, respectively. We calculate the in-focus micrograph using the approximation

$$I_0 \approx \frac{I^+ + I^-}{2},$$

(7)

and solve Eq. (4) for $\phi_0$ using a Fourier transform method [16, 22]:

$$\phi_0 = -\frac{k}{4\pi^2} \mathcal{F}^{-1} \left\{ \frac{k}{|k|^2} \mathcal{F} \left\{ \frac{1}{I_0} \mathcal{F}^{-1} \left\{ \mathcal{F} \left\{ \frac{\partial I_0}{\partial z} \right\} \frac{k}{|k|^2} \right\} \right\} \right\}.$$

(8)
where \( \mathbf{k} \) is the wave vector.

The magnetic component of the phase can be obtained via any of several methods [23]. In this work, \( \varphi_m(r_\perp) \) is obtained from the exit phase \( \varphi(r_\perp) \) using the time-reversal property [7]

\[
\varphi_m = \frac{\varphi - \varphi'}{2},
\]

where \( \varphi' \) is the phase obtained by rotating the specimen by 180° about an axis orthogonal to \( z \) and reflecting the resulting phase map in this axis. By retrieving \( \varphi_m \) for multiple orientations, the tilt series [5]

\[
T^\theta(x, z) = -\hat{h} \varphi_m(x, z, \theta),
\]

and

\[
T^\alpha(y, x) = -\hat{h} \varphi_m(y, x, \alpha),
\]

are constructed. Here, \( \theta \) and \( \alpha \) indicate positive angles of rotation about the \( z \) and \( x \) axis, respectively. We then reconstruct the vector potential using a filtered backprojection algorithm given by [5]

\[
\begin{align*}
\mathbf{A}(x, y, z) &= \int_0^\pi \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \tilde{T}^\theta(|\mathbf{k}| |\mathbf{k}| \theta \left[ \begin{array}{c}
\frac{k_x^2 + k_y^2}{k_z^2 + k_x^2 + k_y^2} \\
\frac{k_y}{k_z}
\end{array} \right] e^{2\pi i k_x^\theta (x \cos \theta + y \sin \theta) + k_z^\theta} \, dk_x \, dk_y \, d\theta \\
&\quad + \int_0^\pi \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \tilde{T}^\alpha(|\mathbf{k}| |\mathbf{k}| \alpha \left[ \begin{array}{c}
\frac{k_x}{k_y}
\frac{k_z}{k_y}
\frac{k_z}{k_x}
\end{array} \right] e^{2\pi i k_x^\alpha (y \cos \alpha + z \sin \alpha) + k_x^\alpha} \, dk_x \, dk_y \, d\alpha.
\end{align*}
\]

Here, \( k_x, k_y, \) and \( k_z \) are the Fourier space coordinates corresponding to the real space coordinates \( x, y, \) and \( z, \) respectively. Fourier transformed quantities are indicated with tildes, and \( k_x^\theta = k_x / \sin \theta \) and \( k_x^\alpha = k_x / \cos \alpha \) are radial spatial frequencies.

3. Orientation dependent errors

In VFET, magnetic phase contrast in the micrographs is the source of the information that is used to reconstruct the vector field. Because the projection of the magnetic vector potential changes with specimen orientation, the magnetic phase contrast varies as the specimen is rotated through each tilt series. The phase maps used in the filtered backprojection reconstruction algorithm (Eq. (12)) are not uniformly distributed over the sphere. Because of this, the average magnetic phase contrast can vary with initial orientation of the specimen.

If the errors in the phase maps are small, maximizing the average contrast in the micrographs minimizes the error in the reconstruction. However, when the errors are significant (e.g., for large defocus), the increased contrast results in an increase in the reconstruction error.

In the remainder of this section, we describe the choice of coordinate system used for the error analysis, and discuss these competing sources of orientation dependent errors.

3.1. Coordinate system used in the error analysis

In our simulations, the two tilt series are acquired as the specimen is rotated around the \( x \)- and \( z \)-axes, respectively. Consequently, it is natural to use spherical coordinates with the pole aligned
Fig. 2. (a) Geometry used for the acquisition of tilt series. In the $\alpha$ series, the object $O$ is rotated about the $x$-axis and, in the $\theta$ series, it is rotated about the $z$-axis. (b) Spherical polar coordinate system used to describe magnetization directions. Here $\mathbf{m}(\gamma, \gamma')$ is the magnetization vector of the particle, which is assumed to be uniformly magnetized, $\gamma$ is the polar angle measured from the negative $y$-axis, and $\gamma'$ is the azimuthal angle, measured from the $x$-axis to the projection of $\mathbf{m}$ onto the $x-z$ plane.

with the $-y$ direction for describing the magnetization direction. In this work, we define our coordinate system such that the polar angle given by

$$\gamma = \arccos \left( \frac{-y}{\sqrt{x^2 + y^2 + z^2}} \right),$$

and the azimuthal angle given by

$$\gamma' = \arctan \left( \frac{z}{x} \right).$$

The geometry of the tilt series acquisition, as well as the corresponding coordinate system given by Eqs. (13) and (14), is shown in Fig. 2. For consistency with previous work [5, 24], the reconstruction algorithm we use involves a different electron propagation direction for each of the tilt series, but for visual clarity, we show only one direction in our diagrams. The two methods are equivalent, amounting only to a different start and end point in the $\alpha$ tilt series. Note, also, that the chosen coordinate system for the tilt series acquisition is arbitrary, and our analysis is valid for any reconstruction using two orthogonal tilt series, provided that $\gamma$ and $\gamma'$ are defined relative to the orientation of the tilt series in the given coordinate system.

3.2. Image contrast as a source of error

In vector tomography, for each vector in the field to be reconstructed, there are orientations for which this vector contributes no contrast in the projection. Specifically, for a dipole, the image contrast vanishes if the moment direction is parallel to the optic axis of the TEM, and contributes the maximum possible contrast if it is orthogonal. In the remainder of this section, we derive an expression for the magnetic phase contrast in terms of moment orientation for a dipole, which we use in Sec. 4 to obtain an expression for the root-mean-square (rms) reconstruction error in terms of the magnetization direction.
The phase object approximation is valid when the electron wavelength is significantly shorter than the characteristic length scale of the sample [5]. Under this approximation, $I_0$ is constant, and Eq. (4) becomes [25]

$$-k \frac{\partial I_0}{\partial z} = I_0 \nabla^2 \varphi_0.$$  \hspace{1cm} (15)

Utilizing a forward difference approximation for $\partial I / \partial z$, the TIE under the phase object approximation can be written

$$\frac{I^+ - I_0}{\Delta f} = \frac{-I_0}{k} \nabla^2 \varphi_0.$$  \hspace{1cm} (16)

This gives us an expression for the overfocused intensity:

$$I^+ = \frac{-I_0\Delta f}{k} \nabla^2 \varphi_0 + I_0.$$  \hspace{1cm} (17)

For our purposes, we use the rms definition of contrast [26, 27]. In the context of VFET, we are concerned with the rms contrast in the magnetic component of the defocused micrographs, which is given by

$$K = \sqrt{\frac{1}{I_0^2(M^2-1)} \sum_{i,j} \left( I^+_{i,j} - I_0 \right)^2}$$

$$= \sqrt{\frac{1}{(M^2-1)} \sum_{i,j} \left[ \frac{\Delta f}{k} \nabla^2 \left( \varphi_m(r_{i,j}) \right) \right]^2},$$  \hspace{1cm} (18)

where $M$ is the width of the micrograph in pixels. Here, we have made use of the fact that, under the phase object approximation, $I_0$ is a constant, and is equal to the mean value of $I^+$. We have also discarded the electrostatic component of $\varphi_0$ because we are only interested in modeling the behavior of the magnetic contrast. In our simulations, we separate the two components using the time-reversal property (Eq. (9)); for the purposes of this analysis, we assume this separation to be perfect.

The phase shift induced in the electron wavefunction by a magnetic dipole is given by

$$\varphi_m = -\frac{e}{\hbar} \int A \cdot d\mathbf{z} = -\frac{\mu_0 e}{2\pi \hbar} \frac{(\mathbf{m} \times \mathbf{r} \cdot \hat{\mathbf{z}}) x^2 + y^2}{x^2 + y^2},$$  \hspace{1cm} (20)

where $\mu_0$ is the permeability of free space, and $\mathbf{m}$ is the magnetic dipole moment. Eq. (20) describes the phase shift of an infinitesimal magnetization density, and the Laplacian of $\varphi_m$ vanishes. To model the magnetic phase contrast arising from a small, but finite, uniformly magnetized particle, we add a regularization parameter $\tau$ to the analytic form of the phase, giving

$$\nabla^2 (\varphi_m) = -\frac{\mu_0 e}{2\pi \hbar} \nabla \frac{(\hat{\mathbf{z}} \times \mathbf{m} \cdot \mathbf{r})}{x^2 + y^2 + \tau^2}$$

$$= \frac{4\mu_0 e}{\pi \hbar} \frac{(\hat{\mathbf{z}} \times \mathbf{m} \cdot \mathbf{r})}{(x^2 + y^2 + \tau^2)^3},$$  \hspace{1cm} (21)
where \( r \equiv (x, y, z) \) is the position vector. In terms of the magnetization, utilizing Eqs. (19) and (22), the contrast is given by

\[
K = \frac{4 \mu_0 e \Delta f |m|}{\pi \hbar k \sqrt{M^2 - 1}} \left[ \sum_{i, j} \left( \frac{\tau^2 (\mathbf{z} \times \hat{m}) \cdot \mathbf{r}}{(x^2 + y^2 + \tau^2)^3} \right)^2 \right]^{1/2},
\]

(23)

\[
= \frac{4 \mu_0 e \tau^2 \Delta f |m| |\mathbf{z} \times \hat{m}|}{\pi \hbar k \sqrt{M^2 - 1}} \left[ \sum_{i, j} \left( \frac{\cos \zeta}{(x^2 + y^2 + \tau^2)^3} \right)^2 \right]^{1/2},
\]

(24)

where \( \zeta \) is the angle between \( \mathbf{z} \times \hat{m} \) and \( r_\perp \). Because the contrast is low far away from the dipole, we can approximate Eq. (24) by constraining the summation to a circular region of radius \( M/2 \) pixels. Under this approximation, \( \cos^2 \zeta \) takes on all values in the range \([0, 1]\), irrespective of the direction of \( m \), and we can write

\[
K \propto |\mathbf{z} \times \hat{m}|.
\]

(25)

In this work, we use a normalized error metric to quantify the accuracy of the reconstructed vector potential, and expect this to be a function of the fractional rms contrast \( K/K_{\text{max}} \), where \( K_{\text{max}} \) is the maximum value \( K \) can take as we vary \( \hat{m} \), i.e., the value of \( K \) with \( |\mathbf{z} \times \hat{m}| = 1 \). Using Eq. (25), we can then express the fractional mean square contrast as

\[
\frac{K^2}{K_{\text{max}}^2} = |\mathbf{z} \times \hat{m}|^2
\]

(26)

\[
= 1 - (\hat{m} \cdot \mathbf{z})^2.
\]

(27)

### 3.3. Truncation error due to the finite difference approximation

In order to calculate the longitudinal derivative of the intensity for use in the TIE, a finite difference approximation must be employed. This results in a truncation error in the derivative, which contributes errors to the retrieved phases and, ultimately, to the reconstructed vector fields. To first order, the truncation error arising from the use of Eq. (6) is [28]

\[
\text{trunc} E \approx \frac{(\Delta f)^2}{6} \frac{\partial^3 I_0}{\partial z^3}.
\]

(28)

This implies that the mean square error in the phase map due to this contribution will be approximately a function of \((\Delta f)^4\).

### 4. Derivation of error estimates

In determining the vector potential of a specimen, the accuracy of a reconstruction method can be quantified in different ways depending on what properties of the specimen are of most interest. For this work, we use a normalized rms error defined by [5]

\[
\text{rms} E = \sqrt{\frac{\sum_{i,j,k} [A_{i,j,k}^{\text{rec}} - A_{i,j,k}]^2}{\sum_{i,j,k} |A_{i,j,k}|^2}}.
\]

(29)
Under the assumption that the rms error contribution \( \text{rms} \hat{E}_\phi \) of a single phase map is a function of contrast, we make use of Eqs. (27) and (28), and expand \( \text{rms} \hat{E}_\phi \) as a Taylor series about \( 1 - (\hat{m} \cdot \hat{z})^2 = 0 \) and \( (\Delta f)^4 = 0 \):

\[
\text{rms} \hat{E}_\phi^2 ((\hat{m} \cdot \hat{z})^2, (\Delta f)^4) = E^2(1,0) + \frac{1}{2} \left[ \partial^2 (\text{rms} \hat{E}_\phi^2) (\hat{m} \cdot \hat{z})^2 - 1 \right] \left[ \partial^2 (\text{rms} \hat{E}_\phi^2) (\hat{m} \cdot \hat{z})^2 - 1 \right] (\Delta f)^4 + \frac{1}{2} \partial^2 (\text{rms} \hat{E}_\phi^2) (\hat{m} \cdot \hat{z})^2 (\Delta f)^8 - \pi B_\alpha (\Delta f)^4 + \frac{3\pi A_\alpha}{16} (m_y^2 + m_z^2)^2 + \pi \text{min}E^2 \alpha (37)
\]

where \( p \equiv (\hat{m} \cdot \hat{z})^2 \) and \( q \equiv (\Delta f)^4 \). Here, we have set the partial derivatives of first order to zero, because we expect extrema in the error function at \( (\hat{m} \cdot \hat{z})^2 = 1 \) and \( (\Delta f)^4 = 0 \). We replace the partial derivatives in Eq. (31) with the constants

\[
A = \frac{\partial^2 (\text{rms} \hat{E}_\phi^2)}{\partial p^2}, \quad B = \frac{\partial^2 (\text{rms} \hat{E}_\phi^2)}{\partial p \partial q}, \quad \text{and} \quad C = \frac{1}{2} \frac{\partial^2 (\text{rms} \hat{E}_\phi^2)}{\partial q^2}.
\]

The mean square error in the phase is then given by

\[
\text{rms} \hat{E}_\phi^2 ((\hat{m} \cdot \hat{z})^2, (\Delta f)^4) = \text{min}E^2 + (B(\Delta f)^4 - A) (\hat{m} \cdot \hat{z})^2 - B(\Delta f)^4 + \frac{3\pi A_\alpha}{16} (m_y^2 + m_z^2)^2 + \pi \text{min}E^2 \alpha (33)
\]

where we have combined the constant terms via \( \text{min}E^2 = E^2(1,0) + \frac{1}{2} A \). In general, the error contribution of the respective tilt series can be different. We use the subscripts \( \alpha \) and \( \theta \) to denote the tilt series to which each error and undetermined constant belongs. Equation (33) then evaluates to

\[
\text{rms} \hat{E}_\phi^2 (\alpha, \gamma, \gamma') = (m_y \sin(\alpha) + m_z \cos(\alpha))^2 (B_\alpha (\Delta f)^4 - A_\alpha) - B_\alpha (\Delta f)^4 + \frac{A_\alpha}{2} (m_y \sin(\alpha) + m_z \cos(\alpha))^2 + C_\alpha (\Delta f)^8 + \text{min}E^2 \alpha (34)
\]

and

\[
\text{rms} \hat{E}_\phi^2 (\theta, \gamma, \gamma') = (m_y \sin(\theta) + m_z \cos(\theta))^2 (B_\theta (\Delta f)^4 - A_\theta) - B_\theta (\Delta f)^4 + \frac{A_\theta}{2} (m_y \sin(\theta) + m_z \cos(\theta))^2 + C_\theta (\Delta f)^8 + \text{min}E^2 \theta (35)
\]

for the respective tilt series. Here we have made use of the fact that the direction of the magnetic moment, in terms of our spherical polar coordinates, \( \gamma \) and \( \gamma' \), is given by

\[
\hat{m} = \sin \gamma \cos \gamma' \hat{x} + \cos \gamma' \hat{y} + \sin \gamma \sin \gamma' \hat{z}. (36)
\]

Integrating Eqs. (34) and (35) over the respective tilt series gives the mean square errors

\[
\text{rms} \hat{E}_\alpha^2 = \frac{\pi}{2} (m_y^2 + m_z^2) (B_\alpha (\Delta f)^4 - A_\alpha) - \pi B_\alpha (\Delta f)^4 + \frac{3\pi A_\alpha}{16} (m_y^2 + m_z^2)^2 + \pi C_\alpha (\Delta f)^8 + \pi \text{min}E^2 \alpha (37)
\]
and

\[
\bar{E}_\theta^2 = \frac{\pi}{2} (m_y^2 + m_x^2)(B_\theta(\Delta f)^4 - A_\theta) - \pi B_\theta(\Delta f)^4 \\
+ \frac{3\pi A_\theta}{16} (m_y^2 + m_x^2)^2 + \pi C_\theta(\Delta f)^8 + \pi \min E_\theta^2.
\]  

(38)

The total reconstruction error due to this alignment is obtained by summing the square-roots of Eqs. (37) and (38):

\[
\bar{E} = \bar{E}_\alpha + \bar{E}_\theta
\]  

(39)

In our simulations, the error contributions from the two tilt series are similar, so we drop the \(\alpha\) and \(\theta\) subscripts to reduce the number of unknown constants to a total of four.

5. Numerical simulations

The error estimate derived in Sec. 4 assumes the object to be dipole-like (small and uniformly magnetized). To test our analytical predictions for a realistic specimen, we compute micrographs from a simulated, uniformly magnetized magnetite specimen. The simulations begin with a sample mask representing a region of constant magnetization and electrostatic potential. For our simulations, we use a spherical sample mask with a diameter of \(d = 100\) nm, centered at the origin. This shape is chosen to decouple the magnetization orientation from the specimen geometry. A Fourier transform method [29] is used to construct the vector potential from the sample mask. The projected potentials are calculated by integrating the magnetic vector potential and the electrostatic potential in the electron propagation direction, and attenuation is modelled in the simulations by including an imaginary component in the electrostatic potential. The exit phase is a linear combination of these projected potentials due to the Aharonov-Bohm phase shifts given in Eqs. (1) and (2). The out-of-focus micrographs are then computed by employing a transfer function formalism (see, for example, Ref. [30]).

A Fourier transform solution to the TIE [16] is used to retrieve the phase from the simulated micrographs, and a time-reversal method [14] is employed to recover its magnetic component. We reconstruct the magnetic vector potential of the simulated object using a filtered backprojection algorithm given by Eq. (12). Slices of the vector potential—exact, and reconstructed from samples with \(\gamma = 0\) and \(\gamma = \pi/2\), respectively—are shown in Fig. 3. The reconstruction error is then calculated using the rms metric defined in Eq. (29). For a given defocus, we vary the magnetization direction over 11 polar angles and 21 azimuthal angles, a total of 191 orientations of the magnetization, and perform the entire simulation and reconstruction process once for each orientation. This gives us the error as a function of specimen orientation. We compare these simulations with our analytical results based on Eqs. (37)–(39). The results for a single defocus are shown as grayscale plots in Fig. 4.

There are various numerical errors that affect the simulations for some parameter combinations. This includes sampling errors and wraparound artefacts. These errors are difficult to quantify, and the sampling would typically be high in experimental tomography. The wraparound effects are a numerical artefact, and do not occur in experimental micrographs. For these reasons we avoid these errors in our simulations, via an appropriate choice of image resolution and domain size, and focus on the error sources that are addressed by our analysis; namely, contrast dependent errors and truncation errors in the finite difference approximation (Eq. (6)) used for the TIE.

Our results, both simulated and analytical, for four different defoci, are presented as two dimensional scatter plots in Fig. 5.

Provided that a small number of unknowns is appropriately chosen, the analytical results for the rms error agree well with the simulations. In the results we present here, the optimal
Fig. 3. A 1 voxel (3.125 nm) thick slice through the origin of each component of $A$ for a 100 nm diameter magnetite sphere reconstructed from simulated micrographs with a defocus of 100 µm. Top row: The exact simulated vector potential. Middle row: Reconstruction with $m(\gamma, \gamma')$ chosen to maximize $\text{rms} E$. Bottom row: Reconstruction with $m(\gamma, \gamma')$ chosen to minimize $\text{rms} E$. 
Fig. 4. (a) Grayscale plots of error as a function of specimen orientation for the $\Delta f = 25 \mu m$ case, using simulations (left) and analytical results (right). The errors here are shown as a function of $\gamma$ and $\gamma'$, and the table shows the values chosen for each constant in the analytical model. (b) The spheres show the results mapped back onto spherical coordinates, with the axes underneath indicating the orientation of these spheres in Cartesian coordinates.

value for $\gamma$ is always $(\gamma \mod \pi) = 0$. However, we have also performed simulations using smaller particles ($d = 40 \text{ nm}$) and found that the optimum orientation can be $\gamma = \pi / 2$ for small defocus, but changes to $(\gamma \mod \pi) = 0$ above some critical defocus value. For large defocus, our results indicate that the optimal value is always $(\gamma \mod \pi) = 0$. These results are consistent with our assertion that improved contrast reduces $\text{rms} E$ when the error in the retrieved phase is low. For a larger particle such as the $d = 100 \text{ nm}$ sphere, additional errors in the phase, due to increased attenuation, result in an increase in error with increasing contrast, even for small $\Delta f$. We have not included the results of the 40 nm particle simulations because of concerns that they may not be physically accurate due to the omission of superparamagnetic effects in any of our simulations.

The optimal azimuthal angle is difficult to predict, as it can be highly sensitive to small differences in the errors arising from each tilt series. However, in most cases the reconstruction error is approximately constant with respect to $\gamma'$, and can be treated as such for the purposes of determining the ideal initial orientation of the specimen.

<table>
<thead>
<tr>
<th>$A$</th>
<th>$B$</th>
<th>$C$</th>
<th>$\text{rms} E$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$5.00 \times 10^{-3}$</td>
<td>$-450 \text{ mm}^{-4}$</td>
<td>$-1.4 \times 10^6 \text{ mm}^{-3}$</td>
<td>$1.33 \times 10^{-3}$</td>
</tr>
</tbody>
</table>
Fig. 5. (Color online) Scatter plots showing the rms errors in the reconstructed vector potential as a function of initial specimen orientation and for a range of defoci, using simulated micrographs (top) and the analytical estimate (bottom). Plots on the left and right show slices at $\gamma' = 0$ and $\gamma = \pi/2$, respectively.
6. Conclusion

The accuracy of VFET reconstructions exhibits a strong dependence on the initial orientation of the sample relative to the reconstruction geometry. The form of this dependence is easily estimated for the case where the object is uniformly magnetized. For many real-world materials, there is no such straightforward way of calculating these errors. However, many materials of interest exhibit a strong net magnetization and exhibit similar properties to a magnetic dipole, or uniformly magnetized particle.

When errors in the phase are high, as is the case when using a propagation based phase retrieval method with large defocus, or when there is significant attenuation, reconstruction errors can be reduced by orienting the particle such that the average magnetic phase contrast in the recorded micrographs is minimized. Conversely, if errors in the retrieved phase can be kept small, maximizing the average contrast reduces the total reconstruction error.

Where sample preparation techniques allow for the choice of magnetization orientation, reconstruction errors can be reduced by taking the effect of initial orientation into consideration.

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