

Three-dimensional magnetization reconstruction from electron optical phase images with physical constraints

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The ability to characterize three-dimensional (3D) magnetization distributions in nanoscale magnetic materials and devices is essential to fully understand their static and dynamic magnetic properties. Phase contrast techniques in the transmission electron microscope (TEM), such as electron holography and electron ptychography, can be used to record two-dimensional (2D) projections of the in-plane magnetic induction of 3D nanoscale objects. Although the 3D magnetic induction can in principle be reconstructed from one or more tilt series of such 2D projections, conventional tomographic reconstruction algorithms do not recover the 3D magnetization within a sample directly. Here, we use simulations to describe the basis of an improved model-based algorithm for the tomographic reconstruction of a 3D magnetization distribution from one or more tilt series of electron optical phase images recorded in the TEM. The algorithm allows a wide range of physical constraints, including a priori information about the sample geometry and magnetic parameters, to be specified. It also makes use of minimization of the micromagnetic energy in the loss function. We demonstrate the reconstruction of the 3D magnetization of a localized magnetic soliton — a hopfion ring — and discuss the influence of noise, choice of magnetic constants, maximum tilt angle and number of tilt axes on the result. The algorithm can in principle be adapted for other magnetic contrast imaging techniques in the TEM, as well as for other magnetic characterization techniques, such as those based on X-rays or neutrons.

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1 Introduction

Three-dimensional (3D) magnetization distributions link macroscopic magnetic properties of materials and devices, such as hysteresis and topology, to fundamental physical properties, such as exchange stiffness and magnetocrystalline anisotropy. A full understanding of the macroscopic magnetic properties of materials therefore requires a knowledge of their 3D magnetization on the nanoscale. Numerous efforts have been made to develop experimental techniques for the measurement of magnetic fields and magnetization distributions, both as two-dimensional (2D) projections or surfaces and in 3D. Such measurements have included the use of X-rays, neutrons and electrons in both real and reciprocal space, have achieved spatial resolutions ranging from the micrometer to the nanometer scale and have often required the development of new reconstruction algorithms [1-4].

Phase contrast techniques for measuring magnetic information about materials in the transmission electron microscope (TEM) rely on changes to the phase of an incident high energy electron wave arising from the so-called Aharohov-Bohm effect [5, 6]. Techniques such as off-axis electron holography [2, 6] and electron ptychography [7] can be used to record the phase of an electron wave that has passed through a specimen, whereas differential phase contrast imaging [8, 9] and the Fresnel mode of Lorentz microscopy [10] record signals that are approximately proportional to the first and second derivative of the phase, respectively.

In principle, 3D magnetic information can be reconstructed from tilt series of recorded phase (or phase gradient) images [2, 11-13]. Conventional backprojection-based tomographic reconstruction algorithms have been used to measure the 3D magnetic induction field $\mathbf{B}$. Their implementation typically relies on the fact that the gradient of the magnetic contribution to a recorded phase image is proportional to the in-plane component of the magnetic induction projected in the incident electron beam direction [11, 13-15]. Such algorithms are well-established in cryogenic electron tomography of proteins and medical X-ray computed tomography [16] and are commonly implemented in the form of a weighted simultaneous iterative reconstruction technique [17]. They can be used to recover two components of a 3D magnetic induction field from two tilt series of phase or phase gradient images (typically recorded about orthogonal specimen tilt

axes). The third (out-of-plane) component can then be calculated numerically by making use of the criterion that the divergence of the magnetic induction must be zero [13, 14].

The 3D magnetization $\mathbf{M}$ in a material cannot, in general, be recovered directly from recorded phase or phase gradient images. Accordingly, several model-based inversion algorithms have been developed [18-20]. Approaches based on differentiable programming [21] and wavelet regularization [22] have also recently been introduced, with the latter method used to reconstruct the magnetic vector potential $\mathbf{A}$. A model-based algorithm typically involves the iterative use of a forward model to calculate phase (or phase gradient) images from successive guesses for the 3D magnetization distribution in the sample. At each iteration, the algorithm minimizes a loss function and incorporates a priori information, e.g., about the sample geometry. The loss function usually takes the form of the difference between experimental images and simulated images determined from the current guess for the 3D magnetization distribution. A weighting term can be used to include physical or mathematical constraints. For instance, in ref. [18], the L2-norm of the magnetization was used to represent, to a first approximation, the Heisenberg exchange energy in a cubic magnet [23]. In refs. [19, 20], mathematical constraints such as likelihood, sparsity and total variation were introduced into the loss function.

As a result of the vectorial nature of magnetic fields, the number of unknowns in a reconstruction is three times that for scalar tomography. It is therefore typically necessary to record (at least) two independent tilt series of images [24, 25]. However, due to experimental limitations, only one tilt axis can sometimes be recorded. In addition, it may only be possible to tilt a specimen between angles of at most -70° and $+70^\circ$. Such practical limitations impose challenges for the retrieval of 3D magnetic information and may result in non-physical solutions for such an under-determined problem. It is therefore important to develop algorithms that can incorporate additional constraints, such as material parameters and the energy of the system.

Here, we describe a model-based iterative reconstruction algorithm, which incorporates a micromagnetic energy term through an automatic differentiation technique [26]. It also allows fitting to a uniform value for the magnetization defined by the saturation magnetization of the material and two orientation angles specified in spherical coordinates.

We demonstrate the successful application of the algo-

rithm to a challenging 3D localized spin texture — a hopfion ring [27] — in a FeGe nanodisk. We discuss the influence on the reconstruction of several experimental and physical factors, including noise, choice of magnetic parameters, maximum tilt angle and number of tilt axes, as well as the choice of weighting factor between the residual phase difference and the energy term.

2 Methodology

2.1 Forward model

The forward model for magnetization reconstruction in the TEM is based on an analytical formula for the electron optical phase shift ϕ , which includes contributions from the electrostatic potential V and the magnetic vector potential $\mathbf{A}$ within and around the sample, according to the Aharonov-Bohm effect [5]

$$\begin{aligned}\phi &= \phi_e + \phi_m \\ &= C_e \int_L V dz + C_m \int_L A_z dz,\end{aligned}\quad (1)$$

where ϕ_e and ϕ_m are the electrostatic and magnetic contributions to the phase shift, respectively, $C_e = \frac{\pi}{\lambda E}$, $C_m = -\frac{e}{\hbar}$, E is the electron energy, λ is the electron wavelength, e is the elementary charge, $\hbar$ is the reduced Planck constant, A_z is the z component of the magnetic vector potential, L is the path of a line integral and the electron beam direction is along the z axis.

To a first approximation, only the z component, i.e., the out-of-plane component, of the magnetic vector potential contributes to the phase shift. In other words, only the x - y components, i.e., the in-plane components, of the magnetic induction field contribute to the phase shift, due to the nature of the curl operator $\mathbf{B} = \nabla \times \mathbf{A}$.

The magnetic contribution to the phase shift can be separated from the electrostatic contribution by using a number of experimental approaches, such as turning the sample over, performing measurements at different sample temperatures or changing the microscope accelerating voltage [28]. For simplicity, in the text below we use ϕ to refer to the magnetic contribution to the phase shift alone.

The relationship between the magnetic vector potential $\mathbf{A}$ and the magnetization $\mathbf{M}$ can be expressed as a cross-product in k space in the form [29]

$$\tilde{\mathbf{A}}(\mathbf{k}) = -\frac{i\mu_0}{k^2} \tilde{\mathbf{M}}(\mathbf{k}) \times \mathbf{k}, \quad (2)$$

where μ_0 is the vacuum permeability and the tilde represents a Fourier transform. According to the Fourier central slice

theorem, the projection operator in real space is equivalent to a slice operator in k space [16] and the relationship between ϕ and $\mathbf{M}$ can be expressed in k space as follows:

$$\tilde{\phi}(k_x, k_y) = -i\mu_0 C_m [(\tilde{\mathbf{M}}_{\perp}(k_x, k_y) \times \mathbf{k}_{\perp}/k_{\perp}^2)]_z, \quad (3)$$

where $\mathbf{k}_{\perp} = (k_x, k_y, 0)$. In this way, $\mathbf{M}$ can be linked to measurements of ϕ .

A micromagnetic model typically assumes that the magnitude of the magnetization in a material can be represented by the saturation magnetization M_s , where $\mathbf{M} = M_s \mathbf{m}$ and $\mathbf{m}$ refers to the unit magnetization. The geometry of the sample, G , describes its 3D morphology and can be used to define the location of the magnetization, which is zero outside the sample. Experimentally, the geometry of a sample can be determined by using conventional (scalar) electron tomography, e.g., from the amplitude of the recorded electron wavefunction, by creating a binary mask of the outline of the sample at each specimen tilt angle or by performing a separate (e.g., high-angle annular dark-field) tomography experiment.

The unit magnetization $\mathbf{m}$ can be defined in spherical coordinates by the geometry G , the polar angle $\theta(\mathbf{r})$ and the azimuthal angle $\psi(\mathbf{r})$ in the form

$$\mathbf{m} = G \cdot (\sin \theta \cos \psi, \sin \theta \sin \psi, \cos \theta). \quad (4)$$

2.2 Loss function

Here, we include the residual phase between input phase images and reconstructed results and the micromagnetic energy of the system in the loss function. The term in the loss function L_{ϕ} from the residual between the input phase ϕ_0 and the phase ϕ calculated from the reconstructed magnetization is defined by the L2-norm of the difference between the two phase terms

$$L_{\phi} = \sum_r \|\phi_0 - \phi\|_{L2}. \quad (5)$$

The term in the loss function L_m from the magnetic energy is a functional of $\mathbf{m}$ and can include multiple energy terms. Typically, a micromagnetic model includes contributions from the Heisenberg exchange interaction, Dzyaloshinskii-Moriya interaction (DMI) [30, 31], Zeeman interaction, demagnetization and magnetocrystalline anisotropy. Here, we consider only Heisenberg exchange, DMI, Zeeman and demagnetization terms in the algorithm, in order to represent energy terms in cubic chiral magnets [32, 33]. The term in the loss function from the magnetic energy can then be written

$$L_m = A(\nabla \mathbf{m})^2 + D\mathbf{m} \cdot (\nabla \times \mathbf{m}) - \mu_0 M_s \mathbf{m} \cdot (\mathbf{H}_d + \mathbf{H}_{\text{ext}}), \quad (6)$$

where A is the exchange stiffness, D is the DMI constant, $\mathbf{H}_d$ is the demagnetization field calculated using Newell's method [34] and $\mathbf{H}_{\text{ext}}$ is the applied magnetic field. For a material that has significant magnetocrystalline anisotropy, an additional term $-K_u(\mathbf{m} \cdot \mathbf{u})^2$ can be included in the micromagnetic loss function (eq. (6)), where K_u is the anisotropy constant and $\mathbf{u}$ is the anisotropy axis.

In practice, if the external magnetic field is applied using the TEM objective lens, then zero-magnetic-field tomographic experiments are most commonly performed. Tomographic tilting together with an external magnetic field usually requires the use of a dedicated magnetizing stage or specimen holder [15, 35].

The total loss function L can be expressed as a weighted sum of the above two contributions, in the form

$$L = w_\phi L_\phi + w_m L_m. \quad (7)$$

As L_m has units of energy, w_m is expressed in units of eV^{-1} in the present work.

2.3 Reconstruction workflow

Figure 1 illustrates the workflow of the reconstruction algorithm. Input data include experimental measurements (ϕ_0) and the geometry and magnetic parameters of the sample. The geometry and saturation magnetization are used to define the location and magnitude of the magnetization. The magnetization is expressed in terms of its magnitude (M_s) and polar and azimuthal angles (see eq. (4)). A forward model is used to calculate phase images (ϕ) from the magnetization as a function of specimen tilt angle. The residual of the calculated and experimental phase images is used to calculate the phase loss function L_ϕ . Magnetic parameters, including exchange stiffness (A) and DMI constant (D), as well as the external magnetic field, are included in the micromagnetic energy loss function L_m . The two contributions are weighted to create a total loss function L (eq. (7)). The influence of temperature is included simply by selecting magnetic parameters that are appropriate for the chosen specimen temperature.

Here, the initial state of the reconstruction always corresponded to the magnetization pointing in the positive z direction. The algorithm then iteratively minimized the loss function until the magnetization converged to a chosen tolerance value.

We employed an Adam solver [36] with a learning rate of 0.01 for gradient descent optimization with automatic differentiation (AD). AD is used to calculate the partial derivative of a function that is defined by a computer program rather than by a symbolic equation and has been used in other tomo-

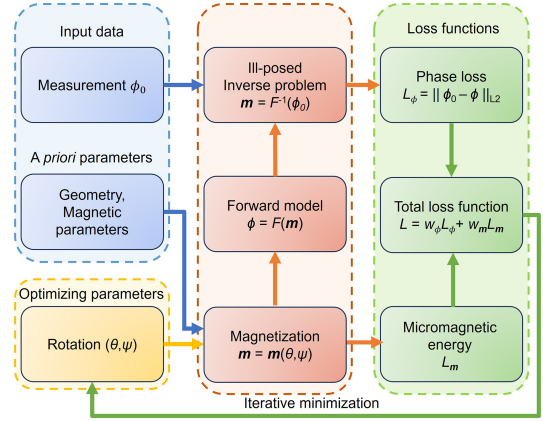


Figure 1 (Color online) Workflow for three-dimensional magnetization reconstruction including physical constraints.

graphic algorithms [21]. It usually involves the decomposition of a function into a sequence of elementary operations and the application of the chain rule to calculate their derivatives iteratively. This feature makes AD effective for optimizing loss functions that involve complex transformations. The project was implemented in the Pytorch [37] framework.

It should be noted that there are typically multiple local minima in the loss function (eq. (7)). The Adam Solver used here is a local, rather than a global, optimization algorithm. Therefore, in principle, the reconstruction can end up in a local minimum of the loss function and may depend on the choice of initial magnetization. In the future, this difficulty can be overcome by using global optimization techniques, such as multi-start methods [38].

3 Results and discussion

In the present work, a 3D magnetic soliton — a hopfion ring [27] — was used to evaluate the magnetization reconstruction algorithm. The hopfion ring, which comprises a hopfion coupled with a skyrmion string, was stabilized in a FeGe nanodisk of diameter 360 nm and thickness 180 nm, as shown in Figure 2(a). The volume was meshed in the form of cubes of side 3 nm. The material parameters used for FeGe [39] were: exchange stiffness constant $A_0 = 4.75 \times 10^{-12} \text{ J m}^{-2}$; DMI constant $D_0 = 0.853 \times 10^{-3} \text{ J m}^{-1}$; saturation magnetization $M_s = 3.84 \times 10^5 \text{ A m}^{-1}$. An external magnetic field of 150 mT was applied in the positive z direction. The magnetic state shown in Figure 2(a), which is displayed as an $m_z = 0$ isosurface, was obtained following the initialization procedure presented in ref. [27], making use of gradient-descent energy minimization. The simulation was performed using the micromagnetic software package Micromagnetic.jl [40]. Figure 2(b) and (c) show the magnetization in central cross-sections of the x - z and x - y planes, respec-

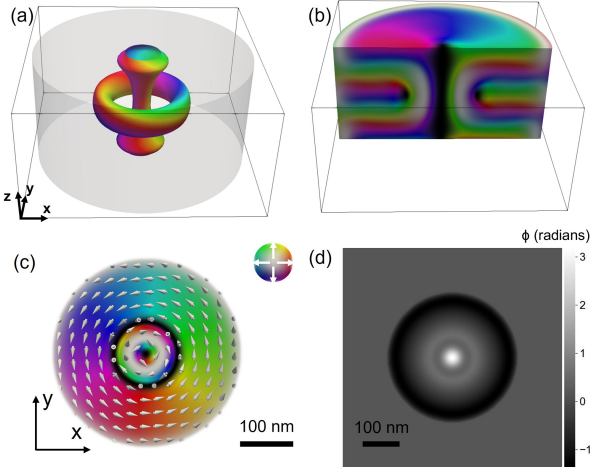


Figure 2 (Color online) Three-dimensional magnetic hopfion ring stabilized in a FeGe nanodisk of diameter 360 nm and thickness 180 nm. The cylindrical axis of the disk is aligned with the axis of symmetry of the hopfion ring. (a) $m_z = 0$ isosurface. (b), (c) Cross-sections of the magnetization in the central x - z and x - y planes, respectively. The color wheel is shown at the upper right of (c). (d) Calculated phase image at 0° specimen tilt angle, i.e., with the incident electron beam parallel to the cylindrical axis of the disk.

tively. In the central x - y cross-section (Figure 2(c)), three vortices are visible from the center to the edge of the disk. For simplicity, we use the magnetization distribution in this plane to assess the quality of the reconstruction below.

Input phase images were calculated from the micromagnetic simulation as a function of specimen tilt angle about the x axis. A representative phase image is shown in Figure 2(d) for a specimen tilt angle of zero. In order to generate input phase images that are representative of experimental data, Gaussian noise was added to each calculated phase image. The standard deviation of the noise was set to 10% of the image contrast $\phi_{0,\max} - \phi_{0,\min}$. This noise level is representative of that in typical experimental data. It can be reduced in the future by using direct electron detectors and/or hologram averaging methods to levels as low as $\frac{2\pi}{1000}$ radians [41,42].

The relative, rather than absolute, contributions of the micromagnetic energy and residual phase to the total loss function affect the reconstruction. Here, for simplicity, w_m was fixed to a value of 1 eV^{-1} and only w_ϕ was varied in the reconstruction. Mini-batch gradient descent was used for iterative minimization of the loss function. The batch size was kept to 4. The reconstruction was assessed by using the mean-square-root error of the reconstructed magnetization $\mathbf{m}'$ and the ground truth $\mathbf{m}$ in the form

$$\begin{aligned} \text{Error} &= \|\mathbf{m} - \mathbf{m}'\|_{L2} \\ &= \frac{1}{N} \sum_{i=1}^N \sqrt{(\mathbf{m}_i - \mathbf{m}'_i) \cdot (\mathbf{m}_i - \mathbf{m}'_i)}, \end{aligned} \quad (8)$$

where N is the number of meshed cells. This criterion is used

in other similar algorithms [19,43].

3.1 Optimal weighting factor

We first evaluated how the micromagnetic energy affects the reconstruction by varying the weighting factor w_ϕ in the loss function. A large value of w_ϕ corresponds to the algorithm favoring a small residual phase between the input images and the results of the reconstruction. In contrast, a small value of w_ϕ corresponds to the reconstruction favoring results with a small micromagnetic energy. A compromise has to be made between the two terms in the loss function, i.e., the reconstructed result has to achieve a reasonably small value of both the residual phase and the micromagnetic energy. Objective criteria for determining this compromise quantitatively are likely to depend on the specimen under examination and on the details of the recorded phases images and their definition is beyond the scope of the present paper. Here, reconstructions were performed as a function of w_ϕ and the quality of the reconstruction was evaluated both from the error and from the magnetization distribution in the central x - y plane. Tilt series of magnetic phase images were used, with specimen tilt angles ranging from -90 to $+90^\circ$ with a 5° step, using orthogonal (x and y) tilt axes. The parameters in the micromagnetic energy term in the loss function were identical to those used in the micromagnetic simulation.

Figure 3 shows the error plotted as a function of weighting factor w_ϕ . The error first decreases with increasing w_ϕ , reach-

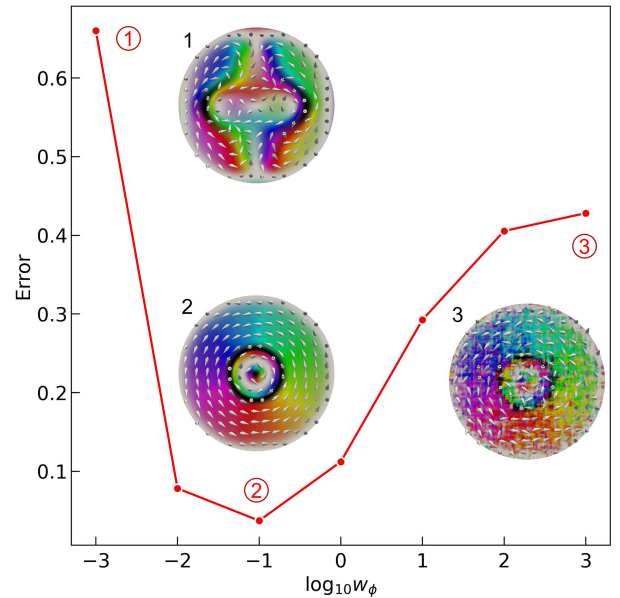


Figure 3 (Color online) Reconstruction error plotted as a function of weighting factor w_ϕ . Magnetization distributions in the central x - y plane are shown for different values of w_ϕ and labeled “1”, “2” and “3”. The optimal weighting factor is determined to be 10^{-1} .

ing a minimum when $w_\phi = 10^{-1}$. It then increases slowly, reaching a plateau for larger values of w_ϕ . The insets show three representative magnetization distributions in the central x - y plane.

For $w_\phi = 10^{-3}$ (labeled “1”), the magnetization distribution is smooth, but differs from the ground truth (Figure 2(c)), resulting in a large error of 0.66. This behavior is expected, as a small value of w_ϕ corresponds to a minimum in micromagnetic energy without favoring consistency with the input phase images. In this way, the loss function for the micromagnetic energy acts as a smoothing kernel, in agreement with regularization techniques used in ref. [18]. In addition, due to the presence of multiple micromagnetic minimum states of the system, a smaller value of w_ϕ may lead to an alternative minimum in the micromagnetic energy, e.g., state “1”. When $w_\phi = 10^3$ (state “3”), the result shows a more noisy magnetization distribution. In this case, the algorithm converged to a minimum value of the residual phase difference between the input phase images and the reconstruction without imposing any physical law, i.e., without considering the micromagnetic energy of the system. For larger values of w_ϕ , the magnetization distribution shows a noisy triple-vortex pattern, similar to the ground truth (Figure 2(c)). As a compromise, an optimal weighting factor $w_\phi = 10^{-1}$ (state “2”) delivers a high quality reconstruction with an almost identical magnetization distribution to the ground truth and a small error of 0.04. In practice, there may not be access to a full series of data from orthogonal tilt axes or a knowledge of magnetic parameters. The choice of optimal weighting factor may depend on the availability of this information.

3.2 Magnetic parameters

The micromagnetic energy term in the loss function requires the magnetic parameters of the material as input (eq. (6)). In ref. [18], only the ferromagnetic exchange energy (the first term in eq. (6)) was implemented in the loss function. In practice, the exchange stiffness A , DMI constant D and saturation magnetization M_s can often be determined independently, for example from macroscopic measurements on bulk samples. The external magnetic field $\mathbf{H}_{\text{ext}}$ is usually a known value in experiments. The demagnetization field $\mathbf{H}_d$ depends on the magnetization distribution, sample geometry and saturation magnetization. It is calculated automatically by the algorithm.

It has been reported in refs. [15, 33, 44, 45], that in thin TEM samples, such as those prepared using focused ion beam milling, there may be a damaged or implanted surface layer. Based on comparisons with micromagnetic simulations, it has been reported that some magnetic parameters can be altered in this region. For example, in B20-type

FeGe TEM samples prepared by focused ion beam milling, the DMI constant in a thin surface layer had to be set to 0 to provide agreement between micromagnetic simulations and experimental measurements [33, 45]. Accordingly, we investigated how the value of the DMI constant in the micromagnetic energy term affects the reconstruction.

Tilt series of magnetic phase images were considered, with tilt angles ranging from -60° to $+60^\circ$ and a 5° step, using two orthogonal tilt axes. The choice of 60° mimics experimental limitations and is discussed below. The weighting factor was chosen to be $w_\phi = 2 \times 10^{-1}$.

Figure 4 shows the reconstruction error plotted as a function of the DMI constant, expressed as D_{rc}/D_0 , where D_{rc} is the value used in the reconstruction and D_0 is the true value. The sign of the DMI was also considered by assigning negative values to D_{rc} . Such a change in sign corresponds to a change in the chirality of the magnetization texture. The insets show four representative magnetization distributions in the central x - y plane. The reconstruction error exhibits a quasi-symmetrical profile centered on $D_{\text{rc}}/D_0 = 1$.

When the accurate DMI value was used, the reconstruction produced a small error and an almost identical pattern of the magnetization distribution in the central x - y plane (state “3”), highlighting the robustness of the algorithm despite the limited range of tilt angles. When the DMI value was set to zero, the chirality of the magnetization in the central x - y plane switched from counter-clockwise to clockwise, result-

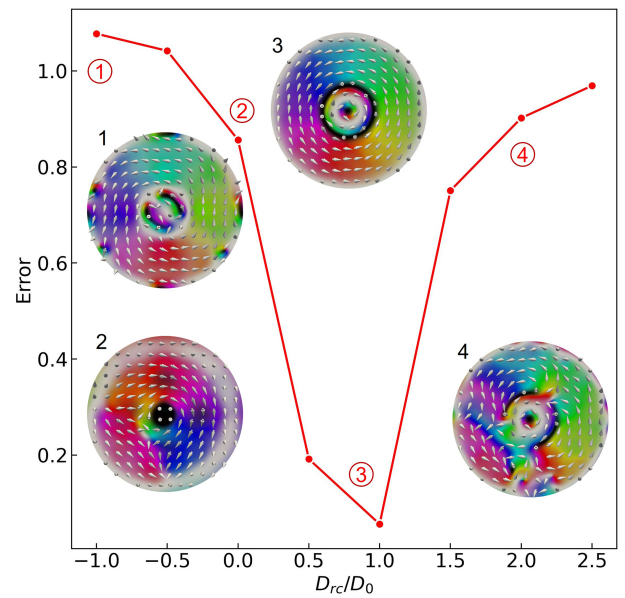


Figure 4 (Color online) Reconstruction error plotted as a function of DMI constant D_{rc}/D_0 . Four representative magnetization distributions in the central x - y plane are shown for different values of D_{rc}/D_0 labeled “1”, “2”, “3” and “4”. The reconstruction corresponding to the accurate DMI constant (“3”) provides an almost identical result to the ground truth shown in Figure 2(c).

ing in a similar “anti-vortex” pattern (state “2”), but without the triple-vortex structure of the ground truth (Figure 2(c)). When the DMI was switched to have a negative value of the same magnitude, i.e., $D_{\text{rc}}/D_0 = -1$, the polarity of the outer vortex reversed to form a counter-clockwise vortex and the hopfion was not retrieved (state “1”). For $D_{\text{rc}}/D_0 = 2$, the reconstruction delivered a random magnetization distribution (state “4”). These results indicate that the reconstruction is very sensitive to the value of the DMI constant, i.e., to the $\mathbf{m} \cdot (\nabla \times \mathbf{m})$ term. In the present example, an accurate value of the DMI constant is essential to deliver a reliable reconstruction. In the reconstructions shown below, D_{rc}/D_0 was set to be 1.

Separate calculations (not included here) show that the values of other magnetic parameters, such as the exchange stiffness A and saturation magnetization M_s , can have a similar effect on the reconstruction. It is assumed that macroscopic magnetic measurements can often be used to determine these values accurately and that TEM sample preparation does not necessarily alter them. In the future, it may be important to define magnetic parameters locally in the reconstruction volume, in particular close to the specimen surfaces, due to effects such as sample preparation damage and oxidation.

3.3 Maximum tilt angle

In practical applications, there is often a limit to the maximum tilt angle of the sample. In cryogenic electron tomography of biological samples using a dedicated single tilt specimen holder, the maximum tilt angle is typically 70° . Dedicated full-tilt-angle specimen holders (see, e.g., refs. [46,47]) are not yet widely available. In scalar tomography, e.g., conventional X-ray computerized tomography and tomography for structural biology, single-tilt-axis acquisition is often sufficient. In contrast, for vector field tomography at least two independent tilt axes are typically required [24, 25, 48]. Therefore, we investigated the influence of maximum tilt angle on magnetization reconstruction.

Tilt series of magnetic phase images were used with different maximum tilt angles and 5° steps, for both a single tilt axis and two orthogonal tilt axes. The weighting factor was set to $w_\phi = 2 \times 10^{-1}$. Figure 5 shows the reconstruction error plotted as a function of the maximum tilt angle for both a single tilt axis (blue) and two tilt axes (red). The insets show four representative magnetization distributions in the central x - y plane.

For the symmetrical object studied here (Figure 2), a perfect reconstruction can be seen to require a maximum tilt angle of at least 75° for a single tilt axis and 30° for two (orthogonal) tilt axes. For the double-tilt series, maximum tilt

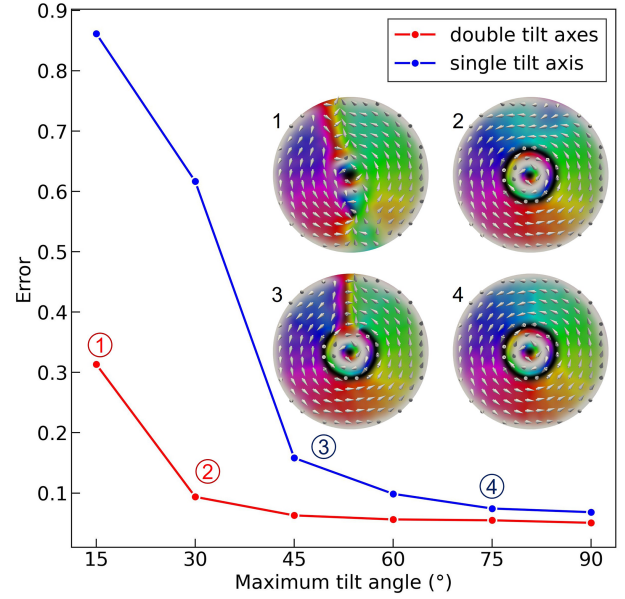


Figure 5 (Color online) Reconstruction error plotted as a function of maximum tilt angle for phase images calculated using single and double tilt axes (blue and red, respectively). Four representative magnetization distributions in the central x - y plane are labeled “1”, “2”, “3” and “4”. Reconstructions for maximum tilt angles of 30° using double tilt axes (“2”) and 75° using a single tilt axis (“4”) match the ground truth shown in Figure 2(c).

angles for two axes of 30° can be achieved in experiments performed using e.g., a Gatan double-tilt specimen holder in a microscope with a conventional pole piece gap. For a larger pole piece gap, one of the tilt axes is likely to reach an angle of at least 60° . Reconstructions with a maximum tilt angle of 60° from a single tilt axis and a maximum tilt angle of 30° from a single tilt axis deliver similar errors. However, in the former case, the 3D magnetization distribution exhibits a considerable difference from the ground truth, indicating that the magnitude of the error alone can be insufficient to evaluate the quality of the reconstruction. For 3D vector fields, several criteria should therefore be used to evaluate the reconstruction. These conclusions may also depend on the symmetry of the object.

3.4 Full reconstruction

We now provide a detailed comparison between the ground truth and the reconstruction obtained from tilt series of magnetic phase images generated about both x and y tilt axes for maximum tilt angles of 30° with 5° steps. Each phase image contained Gaussian noise. The weighting factor was set to 2×10^{-1} . The magnetic parameters used for the reconstruction were identical to those used to generate the ground truth.

Figure 6 shows a comparison between the phase ϕ_0 calculated from the ground truth with noise added and the phase ϕ

calculated from the reconstructed magnetization distribution as a function of tilt angle. Three additional phase images at higher tilt angles, which were not used for reconstruction, are also provided for comparison. The phase images are visually identical at each tilt angle. The fact that the difference ($\phi_0 - \phi$) between the phase images at each tilt angle has a Gaussian distribution (see [Supplementary Figure 1](#)), suggests that the noise in the input phase images was removed successfully by the reconstruction algorithm.

Figure 7 shows a comparison between magnetization distributions for the ground truth and the reconstruction. In the central y - z plane (Figure 7(a)), the magnetic structures of the hopfion and skyrmion were reconstructed successfully.

Three representative cross-sections in different x - y planes were chosen for comparison, as shown in Figure 7(b)-(d). In the central x - y plane ($z = 0$, Figure 7(b)), the three-vortex pattern is well reconstructed. In the x - y plane in which the top edge of the hopfion is located ($z = 20$ nm, Figure 7(c)), a two-vortex pattern is visible in both the ground truth and the reconstruction. In the x - y plane located 5 nm beneath the top surface ($z = 85$ nm, Figure 7(d)), a single-vortex structure is also retrieved reliably in the reconstruction.

For a quantitative comparison of the magnetization distribution between the ground truth and the reconstruction, representative line profiles are plotted in Figure 8. Line 1 in Figure 7(a) is a vertical line that crosses the midpoint be-

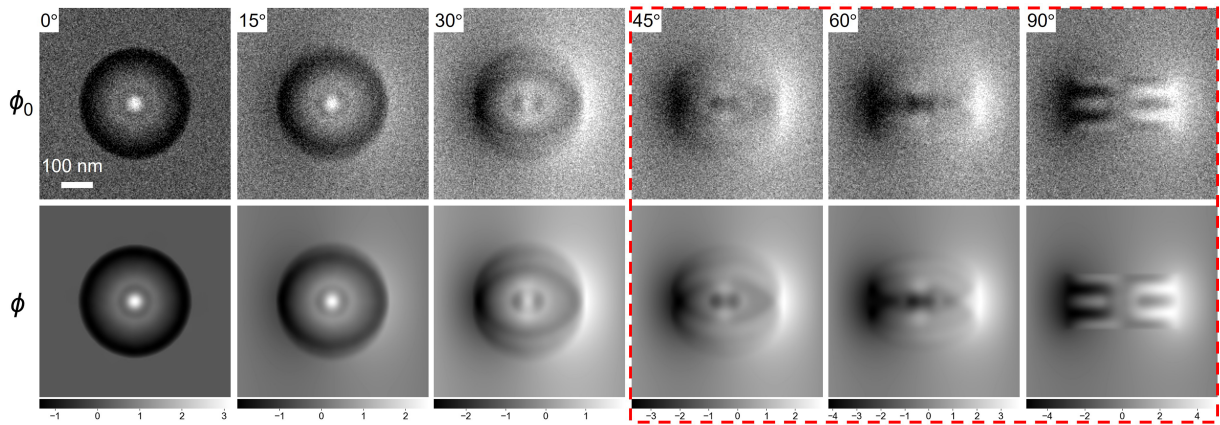


Figure 6 (Color online) Comparison between input phase images with added Gaussian noise (ϕ_0 , upper row) simulated using the forward model and calculated phase images (ϕ , lower row) determined from the reconstructed magnetization distribution. The reconstruction was performed using double tilt series data for a maximum tilt angle of 30° with a 5° step. See text for reconstruction details. Representative phase images are shown as a function of tilt angle about the x axis (horizontal). From left to right, the tilt angles are 0° , 15° and 30° . The tilt angle is labeled at the upper left corner of each image. The color bar for the phase shift is shown at the lower edge of each column. Phase images for higher tilt angles are included for additional comparison. However, these images were not used for reconstruction.

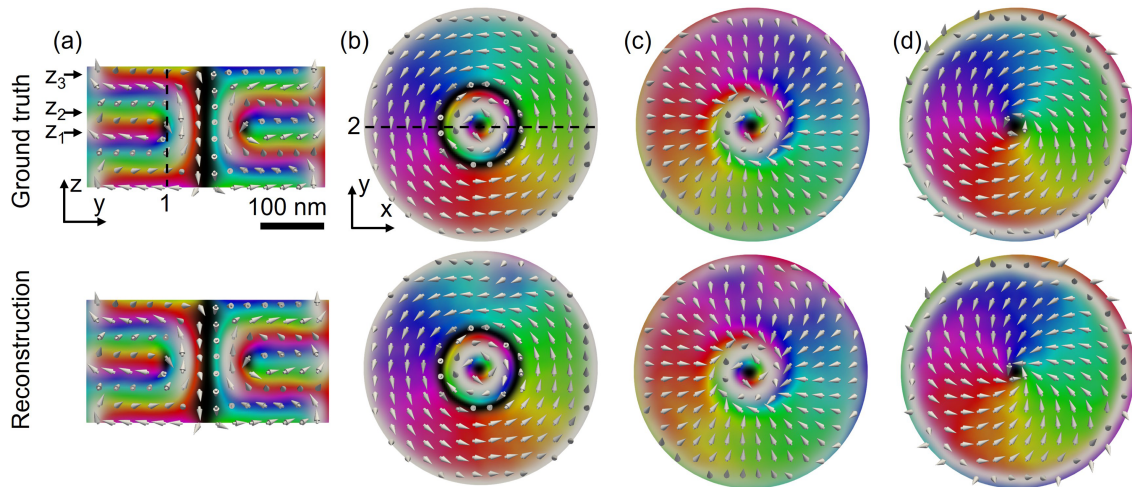


Figure 7 (Color online) Comparison between magnetization distributions for the ground truth (upper row) and reconstruction (lower row). The reconstruction was performed using double tilt series data and a maximum tilt angle of 30° with a 5° step. See text for reconstruction details. Four representative cross-sections of the magnetization distributions are shown: (a) central y - z plane ($x = 0$), (b) $z_1 = 0$, i.e., central x - y plane, (c) $z_2 = 20$ nm, and (d) $z_3 = 85$ nm in x - y planes. z_2 corresponds to the upper edge of the hopfion. z_3 corresponds to 5 nm beneath the upper surface. The positions of these planes are marked in (a). Two additional lines are marked: line “1” at $x = 0$, $y = -60$ nm is marked in (a) and corresponds to the midpoint between the inner and outer edges of the hopfion ring; line “2” at $y = 0$, $z = 0$ is marked in (b) and passes through the centers of the hopfion and skyrmion.

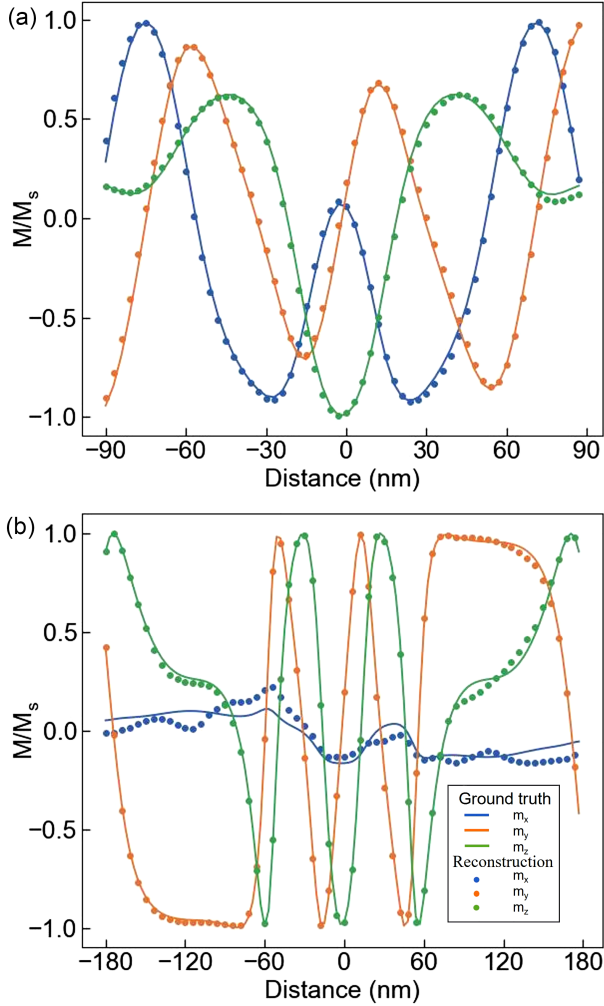


Figure 8 (Color online) Line profiles extracted from the magnetization distributions. Comparisons are shown for each component (x , y , z) of the magnetization between the ground truth (lines) and reconstruction (circles). The magnetization is scaled to the saturation magnetization. The line profiles were extracted from the positions marked in Figure 7. (a) shows line “1” which is at $x = 0$, $y = -60$ nm and corresponds to the midpoint between the inner and outer edges of the hopfion ring. (b) shows line “2” which is at $y = 0$, $z = 0$ and passes through the centers of the hopfion and skyrmion.

tween the inner and outer edges of the hopfion ring in the central y - z plane. Line 2 in Figure 7(b) is a horizontal line that crosses the centers of the hopfion and skyrmion in the central x - y plane. At these positions, each component (x , y and z) of the magnetization is in quantitative agreement between the ground truth (lines) and the reconstruction (circles). In contrast to the phase images, the difference between the magnetization distributions does not have a Gaussian form (see Supplementary Figure 2). This point deserves further investigation in the future.

In standard micromagnetic simulations, the core task is to minimize the micromagnetic energy of the system, with inputs of the geometry and magnetic parameters. Energy-based minimization usually involves the use of conjugate gradient

or gradient descent methods and a Landau-Lifshitz-Gilbert-based approach, with nonzero dissipation continuously reducing the micromagnetic energy of the system [49].

The algorithm presented here takes the same inputs and implements the micromagnetic energy in the loss function, with minimization of the loss function corresponding to minimization of the micromagnetic energy. It also automatically compares calculated phase images from the simulated magnetization with experimental phase images by integrating the residual phase between the reconstruction and the experimental data. The automatic differentiation method used in the iterative algorithm only requires the formulation of the loss function. The gradient directions are then calculated automatically. Other physical and mathematical constraints can be implemented in the loss function, for example by adding a new weighting term. Multi-objective optimization algorithms [50] can also be used to optimize the loss functions L_m and L_ϕ simultaneously in the future.

The advantages of the proposed algorithm are twofold. First, it reconstructs the magnetization $\mathbf{M}$, which is a fundamental property of a material, rather than the magnetic induction field $\mathbf{B}$. By reconstructing $\mathbf{M}$, one can directly calculate $\mathbf{B}$. Second, it incorporates physical constraints, which can include uniformity of the magnetization and micromagnetic energy, in the reconstruction. These constraints improve the quality and fidelity of the reconstruction for such an under-determined problem, especially when dealing with incomplete data.

It should be noted that micromagnetic calculations search for a minimum energy state, which can be either a ground state or a metastable state. The magnetization state that is reconstructed depends on the quality of the recorded experimental images, subsequent analysis steps and choice of weighting factor.

From a practical perspective, as a result of the temporal resolution and signal-to-noise of magnetic imaging in the TEM, it is currently challenging to record tomographic data for mapping dynamical changes in magnetization states during switching processes experimentally. Time-resolved three-dimensional magnetization reconstruction may become possible using dedicated electron microscopes in the future.

4 Conclusions

In this work, a tomographic reconstruction algorithm based on automatic differentiable programming that is able to recover a three-dimensional magnetization distribution from one or more tilt series of electron optical phase images recorded in the TEM has been described. The algorithm allows a priori information to be specified, includ-

ing the geometry of the sample and magnetic parameters such as saturation magnetization, exchange stiffness and the Dzyaloshinskii-Moriya interaction. Physical constraints are included by minimizing a weighted loss function that includes the micromagnetic energy. The robustness of the algorithm has been assessed by applying it to the reconstruction of a three-dimensional magnetic soliton — a hopfion ring. The influence of noise, weighting factor, magnetic parameters and maximum tilt angle has been discussed. Quantitative agreement in magnetization reconstruction has been achieved for two simulated orthogonal tilt series of phase images for a maximum tilt angle of 30°, a 5° step and with Gaussian noise included. The algorithm can be adapted in the future for other magnetic imaging techniques in the TEM, as well as for other magnetic characterization techniques based on X-rays and neutrons. Future directions should also focus on the applicability of the algorithm to real experimental data by considering additional artifacts, local variations in magnetic parameters and complicated (e.g., Néel-type) magnetization textures.

Data availability

Data and codes are made available here <https://github.com/MagRecon/maglab>.

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Conflict of interest The authors declare that they have no conflict of interest.

Supporting Information

The supporting information is available online at <http://phys.scichina.com> and <https://link.springer.com>. The supporting materials are published as submitted, without typesetting or editing. The responsibility for scientific accuracy and content remains entirely with the authors.

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