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On the computation of the magnetic phase shift for magnetic nano-particles of arbitrary shape using a spherical projection model

E. Humphrey, M. De Graef*

Carnegie Mellon University, Pittsburgh, PA 15213, USA

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ABSTRACT

The magnetic phase shift of an electron wave traveling through a magnetized object is computed by considering the object to be made up of a collection of uniformly magnetized spheres arranged on the nodes of a cubic grid. In the limit of vanishing grid size, this approach becomes equivalent to other numerical approaches. Update equations are derived for the change of the magnetic phase shift when the magnetization of a single object voxel is modified. Example phase shift calculations are presented for a uniformly magnetized sphere, circular disks with an infinitely sharp vortex core and a smooth core, and an oval disk with a pair of vortices and an antivortex.

1. Introduction

Lorentz transmission electron microscopy (LTEM) relies on the fact that the phase of a high energy electron wave will be modified by any magnetic object in or near its path. The Aharonov–Bohm phase shift [1] consists of two contributions, one from the electrostatic lattice potential $V(\mathbf{r})$, and one from the magnetic vector potential, $\mathbf{A}(\mathbf{r})$

$$\varphi_t(\mathbf{r}_\perp)=\varphi_e(\mathbf{r}_\perp)+\varphi_m(\mathbf{r}_\perp)$$
$$=\frac{\pi}{\lambda E}\int_{-\infty}^{+\infty}V(\mathbf{r}_\perp+\ell\boldsymbol{\omega})\,\mathrm{d}\ell-\frac{e}{\hbar}\int_{-\infty}^{+\infty}\mathbf{A}(\mathbf{r}_\perp+\ell\boldsymbol{\omega})\cdot\boldsymbol{\omega}\,\mathrm{d}\ell, \tag{1}$$

where E is the relativistic electron accelerating voltage, λ is the electron wave length, and $\boldsymbol{\omega}$ is a unit vector along the electron trajectory; ℓ parameterizes the electron position along this path. In this contribution, we will be concerned primarily with the magnetic component φ_m of the total phase shift φ_t.

Over the past two decades, several phase shift computation approaches have been published. For thin-film objects with a non-uniform but periodic magnetization state, Mansuripur [2] suggested an approach based on an explicit computation of the magnetic phase shift using the Fourier transform of the magnetization, $\mathbf{M}(\mathbf{k})$. His approach accounts for the projection effects due to an inclined incident electron beam, but assumes a uniform magnetization profile along the direction normal to the film; as a result of this approximation, the phase shift can be expressed as a 2D inverse Fourier series

$$\phi_m(\mathbf{r}_\perp)=\frac{2e}{\hbar}\sum_{m=0}^{P}\sum_{n=0}^{Q}{}' \mathrm{i}\frac{t}{|\mathbf{q}|}G_{\mathbf{p}}(t|\mathbf{q}|)(\hat{\mathbf{q}}\times\mathbf{e}_z)\cdot(\mathbf{p}\times(\mathbf{p}\times\mathbf{M}_{mn}))e^{2\pi\mathrm{i}\mathbf{r}_\perp\cdot\mathbf{q}}, \tag{2}$$

where the prime indicates that the term $(m,n)=(0,0)$ does not contribute to the summation, $\mathbf{q}=(m/P)\mathbf{e}_x^*+(n/Q)\mathbf{e}_y^*$ is the frequency vector, $\mathbf{e}_x^*$ and $\mathbf{e}_y^*$ are reciprocal unit vectors, $\mathbf{p}$ is the beam direction expressed in the orthonormal reference frame, t is the sample thickness, a hat indicates a unit vector, and the function $G_{\mathbf{p}}(t|\mathbf{q}|)$ is given by

$$G_{\mathbf{p}}(t|\mathbf{q}|)=\frac{1}{(\mathbf{p}\cdot\hat{\mathbf{q}})^2+p_z^2}\,\mathrm{sinc}\left(\pi t|\mathbf{q}|\frac{\mathbf{p}\cdot\hat{\mathbf{q}}}{p_z}\right), \tag{3}$$

where $\mathrm{sinc}(x)\equiv\sin(x)/x$; for normal beam incidence, we have $G_{\mathbf{p}}=1$. This formalism was extended to more complex magnetization configurations (as generated, for instance, by micromagnetic simulations) by Haug et al. [3] and applied to the simulation of interference fringes in coherent Fresnel domain wall images. The influence of sample tilt on the magnetic phase shift was also investigated in this study and is particularly relevant to the algorithm presented in the present paper.

Beleggia and Zhu [4] proposed an expression for the 2D Fourier transform of the magnetic phase shift for the case of uniformly magnetized particles of arbitrary shape. The magnetization state is expressed as $\mathbf{M}(\mathbf{r})=M_0\hat{\mathbf{m}}D(\mathbf{r})$, where $D(\mathbf{r})$ is the shape function (equal to 1 inside and 0 outside the particle) and M_0 is the saturation magnetization. Their expression is given by

$$\varphi_m(\mathbf{k})=\frac{\mathrm{i}\pi B_0}{\phi_0}\frac{D(k_x,k_y,0)}{k_\perp^2}(\hat{\mathbf{m}}\times\mathbf{k})|_z, \tag{4}$$

where $\phi_0=h/2e=2070\ \mathrm{T\,nm}^2$ is the flux quantum, $B_0=\mu_0M_0$ is the saturation induction, and the shape amplitude $D(\mathbf{k})$ (the Fourier transform of $D(\mathbf{r})$) is evaluated in the plane (k_x,k_y) normal to the beam direction. Explicit expressions for the magnetic phase shift for uniformly magnetized spheres, cylinders, rectangular prisms, and arrays of such nano-particles can be found in [4]; this approach was generalized to arbitrary polyhedral shapes as well as combinations of multiple polyhedral shapes in [5].

* Corresponding author. Tel.: +1 412 268 8527; fax: +1 412 268 7596.
E-mail address: degraef@cmu.edu (M. De Graef).

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Recently, we reported on the successful determination of the 3D magnetic vector potential for thin rectangular and elliptical plates [6]; a tomographic approach using four tilt series (two sets with orthogonal tilt axes, and, for each tilt axis, two series with the sample in the usual orientation and flipped upside down) was used in conjunction with a vectorial filtered back-projection algorithm [7,8] to reconstruct the vector potential on a voxel grid with cube edge length of 6.52 nm. While the reconstruction provided a reasonable representation of the spatial variations of the vector potential, the usual tomographic artifacts were also apparent in the reconstruction: a blurring that is typical of the filtered back projection approach, as well as streaking due to the missing wedge in the tilt series.

In x-ray computed tomography experiments, the filtered back-projection result is typically only used as a starting point for subsequent iterative refinements of the reconstructed object. Such refinement algorithms come in many flavors and acronyms, among others: algebraic reconstruction technique (ART) [9]; simultaneous algebraic reconstruction technique (SART) [10]; discrete algebraic reconstruction technique (DART) [11]; dual axis tomography [12], etc. In addition, recent work in the signal processing community has resulted in model-based iterative reconstruction (MBIR) approaches that require not only a back-projection algorithm but also a forward projection algorithm and an efficient numerical method to update the simulated projections when the state of a single object voxel is changed. Such an approach makes use of prior knowledge about the object, it can handle noisy data sets, and allows for the refinement of large numbers of parameters, such as affine transformations to align individual images, gain normalization factors, and so on. A recent example of the application of MBIR principles in the area of high angle annular dark field (HAADF) STEM tomography can be found in [13].

Application of MBIR concepts to vector field electron tomography (VFET) implies the need for an efficient algorithm to compute the magnetic phase shift starting from the object's magnetization state. In addition, the algorithm must be capable of efficiently updating the total phase shift whenever the magnetization of a single object voxel is modified. An additional desirable aspect of MBIR-type methods is that they allow for a multi-grid approach to the iterative reconstruction of the object; in other words, the reconstruction can first be carried out on a coarse object grid, and is subsequently refined as the algorithm converges. Executing the simulation at a coarser resolution level first will typically improve the convergence speed with respect to a reconstruction that uses the finest resolution from the start.

In the remainder of this contribution, we describe a new algorithm that is capable of updating the magnetic phase shift when the magnetization of a single object voxel is changed. The algorithm is also compatible with a multi-grid approach, and the object's voxel resolution can be changed easily throughout the computation. Section 2 describes the forward projection algorithm, and in Section 3 we show that the algorithm provides an accurate multi-grid alternative to the approaches described before. We conclude this paper with a description of the use of the new algorithm for iterative reconstruction of the magnetic vector potential of a magnetized object of arbitrary shape.

2. Theoretical model

2.1. Object sampling grid

A magnetized object of arbitrary shape is described by a magnetization vector field $\mathbf{M}(\mathbf{r})$ and a shape or characteristic function $D(\mathbf{r})$; by definition, $\mathbf{M}(\mathbf{r}) = 0$ wherever $D(\mathbf{r}) = 0$, i.e., outside the object. To discretize the object, we introduce a 3D grid of cubic voxels, with the origin placed at the object's center-of-mass. The cubic voxels have edge length $2a$, so that the 3D cubic lattice has nodes at the locations $\mathbf{r}_{ijk} = 2a(i,j,k)$ where (i,j,k) is a triplet of integers taking on all the values for which $D(\mathbf{r}_{ijk}) = 1$. Since we will be interested in computing the electron wave phase shift as a function of object tilt angle, we define the cartesian reference frame of the object (at zero tilt) to be such that the cubic grid has its $\mathbf{e}_x$ and $\mathbf{e}_y$ directions parallel to the plane of the detector, and the $\mathbf{e}_z$ direction pointing towards the electron gun (opposite the beam direction). Changing the resolution of the cubic grid is accomplished simply by changing the value of the grid lattice parameter $2a$.

To compute the magnetic phase shift as a function of object orientation we will need to define the average magnetization state for each cubic lattice cell, and apply Eq. (4) to determine the corresponding phase shift due to that cell. While such an approach is feasible (and, in fact, has been reported for rectangular prisms in [4]), a computational issue arises in which the projection of a cube is different along different projection directions $\hat{\omega}$. Since the shape amplitude of a cube is given by

$$D_{\mathrm{cube}}(\mathbf{k}) = 8a^3 \,\mathrm{sinc}(k_x a)\,\mathrm{sinc}(k_y a)\,\mathrm{sinc}(k_z a), \tag{5}$$

this function would need to be recomputed for each tilted object orientation, since the rotation mixes the components of the (k_x,k_y,k_z) coordinate arrays.

A computationally more efficient approach is to replace the voxel cubes by equal-volume spheres of radius R

$$V = 8a^3 = \frac{4\pi R^3}{3} \rightarrow R = a\left(\frac{6}{\pi}\right)^{1/3} = 1.2407a. \tag{6}$$

The projection of a sphere does not depend on the sphere's orientation, so the associated arrays (derived explicitly in the next section) need to be computed only once for each grid resolution $2a$ and can be re-used for all projection directions.

The equal volume spheres in neighboring voxels have a small overlap due to the fact that the radius is slightly larger than a but in the limit of decreasing grid lattice parameter this does not pose a problem. Each sphere has associated with it a magnetization vector $\mathbf{M}(\mathbf{r}_{ijk})$. Since magnetization is defined as the total moment per unit volume, in the limit of vanishing a one recovers the atomic moments that make up the entire object. Therefore, the magnetic phase shift of the object is approximated by the superposition of magnetic phase shifts due to uniformly magnetized equal-volume spheres that are substituted on the voxel lattice, as shown schematically in Fig. 1. Changing the grid resolution $2a$ is then simply accomplished by changing the number of spheres along each coordinate direction, as shown schematically in the three consecutive multi-grid generations of Fig. 1.

2.2. Sphere lattice phase shift

According to Eq. (4), the magnetic phase shift due to a single uniformly magnetized sphere of radius R requires knowledge of the shape amplitude for the sphere, which is easily shown to be

$$D_{\mathrm{sphere}}(\mathbf{k}) = 3V\frac{j_1(kR)}{kR}, \tag{7}$$

where $j_1(x) = \sin x/x^2 - \cos x/x$ is the spherical Bessel function of the first order and V is the volume. Substitution in Eq. (4) results in

$$\varphi_m(k_x,k_y) = \frac{3\pi i B_0 V}{R\phi_0}\frac{j_1(k_\perp R)}{k_\perp^3}(\hat{\boldsymbol{\mu}} \times \mathbf{k})|_z, \tag{8}$$

where (k_x,k_y) are frequency components in the plane normal to the projection direction $\hat{\omega}$. Introducing direction cosines (μ_x,μ_y,μ_z) for the magnetization unit vector $\hat{\boldsymbol{\mu}}$ (expressed in the beam reference