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This Final Report covers the period from July 1, 2001 through June 30, 2016.

1 Research Report

During the period July 1, 2001 to June 30, 2016, the DOE-supported research project covered a significant number of research topics, all of them related to the study of magnetic materials. Part of this work was experimental but the main focus was on theoretical analyses of magnetic materials characterization approaches, such as Lorentz transmission electron microscopy (LTEM) using phase reconstructions; vector field electron tomography (VFET); and in-depth analyses of the demagnetization tensor field for uniformly magnetized particles of arbitrary shape.

A total of 39 papers were published in peer-reviewed journals over the 16 years of this research program. In the following sub-sections, we list the abstracts for all 33 journal papers; the interested reader may find more details in the actual publications. Conference papers are also listed in the list of publications at the end of this report, but are not covered in the following sections due to the fact that these papers typically do not have an abstract.

1.1 Quantitative non-interferometric Lorentz microscopy [1]

Noninterferometric phase reconstruction based on the transport of intensity equation (TIE) is applied to experimental images for a Permalloythin film and to a computational magnetization pattern for a high density magnetic recording medium. An alternative derivation of the TIE is given, based on linear image formation theory. A compact formal solution, suitable for numerical computation, is given.

1.2 Understanding magnetic structures in permanent magnets via in-situ Lorentz microscopy, interferometric and non-interferometric phase reconstruction [2]

We present our observations of field- and orientation-dependence of magnetic domains and their reversal nucleation at grain boundaries in polycrystalline $Nd_2Fe_{14}B$, using Fresnel- and Foucault-Lorentz microscopy. The local magnetization associated with the domain and domain-wall in $Nd_2Fe_{14}B$ and in precipitated Fe particles was mapped using an interferometric holography as well as a novel non-interferometric method based on the "transport of intensity" equation.

1.3 A new symmetrized solution for phase retrieval using the transport of intensity equation [3]

We propose a novel symmetrization method for solving the transport of intensity equation (TIE) using fast Fourier transforms for situations where the input images may or may not exhibit spatial periodicity. The method is derived from the analysis of intensity conservation law and the internal symmetry of the TIE, and is illustrated for both a computational and an experimental data set.

1.4 Electron-optical phase shift of magnetic nanoparticles, part II: Polyhedral particles [4]

A method is presented to compute the electron-optical phase shift for a magnetized polyhedral nanoparticle, with either a uniform magnetization or a closure domain (vortex state). The method relies on an analytical expression for the shape amplitude, combined with a reciprocal-space description of the magnetic vector potential. The model is used to construct two building blocks from which more complex structures can be generated. Phase computations are also presented for the five Platonic and 13 Archimedean solids. Fresnel and Foucault imaging mode simulations are presented for a range of particle shapes and microscope imaging conditions.

1.5 Lorentz study of magnetic domains in Heusler-type ferromagnetic shape memory alloys [5]

We report on magnetic domain observations using in-situ Lorentz microscopy of the ferromagnetic shape memory alloys of the type Ni_2MnGa and Co_2NiGa. The domain configurations are analyzed by means of the Transport-of-Intensity phase reconstruction method.

1.6 On the computation of the demagnetization tensor field for an arbitrary particle shape using a Fourier space approach [6]

A method is presented to compute the demagnetization tensor field for uniformly magnetized particles of arbitrary shape. By means of a Fourier space approach it is possible to compute analytically the Fourier representation of the demagnetization tensor field for a given shape. Then, specifying the direction of the uniform magnetization, the demagnetizing field and the magnetostatic energy associated with the particle can be evaluated. In some particular cases, the real space representation is computable analytically. In general, a numerical inverse fast Fourier transform is required to perform the inversion. As an example, the demagnetization tensor field for the tetrahedron will be given.

1.7 On the computation of the demagnetization tensor for uniformly magnetized particles of arbitrary shape. Part I: Analytical approach [7]

A Fourier space formalism based on the shape amplitude of a particle is used to compute the demagnetization tensor field for uniformly magnetized particles of arbitrary shape. We provide a list of explicit shape amplitudes for important particle shapes, among others: the sphere, the cylindrical tube, an arbitrary polyhedral shape, a truncated paraboloid, and a cone truncated by a spherical cap. In Part I of this two-part paper, an analytical representation of the demagnetization tensor field for particles with cylindrical symmetry is provided, as well as expressions for the magnetostatic energy and the volumetric demagnetization factors.

1.8 On the computation of the demagnetization tensor for uniformly magnetized particles of arbitrary shape. Part II: Numerical approach [8]

In Part I, we described an analytical approach to the computation of the demagnetization tensor field for a uniformly magnetized particle with an arbitrary shape. In this paper, Part II, we introduce two methods for the numerical computation of the demagnetization tensor field. One method uses a Fourier space representation of the particle shape, the other starts from the real space representation. The accuracy of the methods is compared to theoretical results for the demagnetization tensor of the uniformly magnetized cylinder with arbitrary aspect ratio. Example computations are presented for the hexagonal plate, the truncated paraboloid, and a so-called "Pac-Man" shape, recently designed for MRAM applications. Finally, the magnetostatic self-energy of a uniformly magnetized regular polygonal disk of arbitrary order is analyzed. A linear relation is found between the order of the polygon and the critical aspect ratio for in-plane vs. axial magnetization states.

1.9 On the magnetostatic interactions between nanoparticles of arbitrary shape [9]

The general expression for the magnetostatic energy of two magnetized nanoparticles with arbitrary shape and magnetization state is derived within the framework of a Fourier space approach. It is shown how the standard dipoledipole interaction, valid for large interparticle distances, should be modified in order to take into account the shape anisotropy of each particle. Explicit computations are given for a simple system of two interacting cylinders. For magnetic nanowires, i.e., cylinders with a very large aspect ratio, a simple derivation shows that the interaction is of monopolar, rather than dipolar, nature.

1.10 On the computation of the demagnetization tensor for particles of arbitrary shape [10]

A new method is presented to compute the demagnetization tensor of particles of arbitrary shape. By means of a Fourier space approach it is possible to compute analytically the Fourier representation of the demagnetization tensor for a broad class of magnetic nanoparticles. Then, specifying the direction of the uniform magnetization, the demagnetizing field and the magnetostatic energy associated with the particle can be evaluated.

1.11 General magnetostatic shape-shape interactions [11]

The magnetostatic interaction energy between two magnetic elements of arbitrary shape is presented as a convolution between the cross-correlation of the particle shapes and the dipolar tensor field. A generalized dipoledipole interaction is derived, where the magnetic moments associated with the two particles interact through a magnetometric tensor field, carrying all the shape information. Example computations are given in order to verify the correctness of the formalism. The well-known result of the interaction between prisms, employed in most micromagnetic simulations, is correctly retrieved. The numerical accuracy of the method is also compared to a simple analytical result. Finally, one additional example computation, two interlaced interacting rings, is presented to show the generality of the formalism.

1.12 Imaging techniques in magnetoelastic materials [12]

In this Chapter, we will first describe the Lorentz microscopy method from the classical physics point of view, followed by a quantum mechanical description in terms of the phase of the electron wave traveling through the sample. Then, we introduce a phase reconstruction method, based on the transport-of-intensity equation, which permits determination of the total phase shift of the electron wave. In the second half of the Chapter, we will apply the method to selected magnetoelastic compounds, in particular Ni_2MnGa and Co_2NiGa. One of the most important observations is the presence of *magnetoelastic tweed* in the austenitic state of Co_2NiGa.

1.13 Shape-induced ferromagnetic ordering in a triangular array of magnetized disks [13]

A magnetic transition induced by shape anisotropy, geometry, and dipolar interactions has been found in a system of three single domain thin disks. The phase transition occurs only when the disks are in close proximity, and for a narrow range of aspect ratios. Near the transition, the system has an abrupt change from a closure-domain state with zero net magnetization to a magnetized state. The transition can be detected by changes in the hysteresis loops. Micromagnetic simulations with realistic parameters confirm the establishment of ferromagnetic ordering.

1.14 Demagnetization factors for elliptic cylinders [14]

The magnetometric (volume averaged) demagnetization factors for cylinders with elliptical cross section are computed using a Fourier-space approach and compared with similar results obtained with a different treatment. The demagnetization factors are given as a series expansion in the eccentricity ϵ of the elliptical cross section, where the terms up to order ϵ^{10} are given explicitly as a function of the cylinder aspect ratio. Other simplified expressions, valid in restricted regimes, are also given. Two different series expansions, obtained previously and valid in particular combinations of shape parameters, are recalled and compared with the new results. After the computation of the magnetostatic and exchange-energy terms associated with a vortex closure-domain state in the elliptic cylinder, the single-domain limit, or the critical size below which the structure can support quasi-uniform magnetization, is derived and discussed.

1.15 The equivalent ellipsoid of a magnetized shape. [15]

The equivalent ellipsoid for magnetized bodies of arbitrary shape can be determined by imposing the equality between the demagnetization factors of the two shapes of equal volume. It is shown that the "commonsense" criterion for mapping two different shapes by imposing the equality of the demagnetization factors for equal aspect ratios often results in large errors. We propose a general method for the rigorous determination of the equivalent ellipsoid. The cases of the exact equivalent ellipsoids for discs, cylinders with elliptical cross section and prisms are worked out and discussed.

1.16 Phase diagram for magnetic nano-rings [16]

The minimum-energy single-domain magnetization state in a magnetized nano-ring is determined as a function of material and shape parameters. A phase diagram is derived within the framework of a Fourier-space approach for magnetic computations, showing the expected position of the ground state for any given set of external degrees of freedom. A series of micromagnetic simulations for suitably chosen parameters, show excellent agreement with the obtained theoretical results. An electron holography experiment has been carried out as a test on phase diagram reliability. The validity of the treatment, in particular the simplification employed in choosing ideal uniform rather than more physical quasi-uniform single-domain states, is thoroughly discussed in order to establish clear boundaries of applicability of the phase diagram.

1.17 The fluxgate ring-core demagnetization field [17]

The local demagnetization factor for the ring-core flux gate is derived analytically, based on a tangential magnetization model. The results are in good agreement with experimental data for a wide range of ring shape parameters. Approximate expressions in the limit of a narrow, thin ring are obtained, and indicate that the local demagnetization factor scales with the ratio of the cross-sectional area to the total area of the ring. Analytical modeling of the demagnetization factors for a uniform magnetization state results in an underestimate of the local cross-section averaged demagnetization factors by 50% or more.

1.18 Self-energy and demagnetization factors of the general ellipsoid: A self-contained alternative to the Maxwell-Stoner approach [18]

A transparent, exhaustive, and self-contained method for the calculation of the demagnetization tensor of the uniformly magnetized ellipsoid is presented. The method is an alternative to the established Maxwell derivation and is based on a Fourier-space approach to the micromagnetics of magnetized bodies. The key to the success of the procedure lies in the convenient treatment of shape effects through the Fourier representation. The scaled form of the demagnetization factors which depends on two dimensionless aspect ratios is argued to be their natural integral representation. Amongst other advantages, it allows for the immediate implementation of symmetry arguments such that only one of the principal factors needs to be computed. The oblate and prolate ellipsoids of revolution are examined from the same general point of view. The demagnetization factors for these distinct types of spheroid are seen to be related by analytic continuation of well-known Gaussian hypergeometric functions.

1.19 Vector field electron tomography of magnetic materials: Theoretical development [19]

The theory of vector field electron tomography, the reconstruction of the three-dimensional magnetic induction around a magnetized object, is derived within the framework of Lorentz transmission electron microscopy. The tomographic reconstruction method uses as input two orthogonal tilt series of magnetic phase maps and is based on the vector slice theorem. An analytical reconstruction of the magnetic induction of a single magnetic dipole is presented as a proof-of-concept. The method is compared to two previously reported approaches: a reconstruction starting from the gradient of the magnetic phase maps, and a direct reconstruction of the magnetic vector potential. Numerical examples as well as estimates of the reconstruction errors for a range of magnetic particle shapes are reported.

1.20 Demagnetization factors for cylindrical shells and related shapes [20]

Magnetostatic self and interaction energies can be computed via demagnetization factors whenever the magnetic state is close to a uniform state, e.g. in the presence of a strong applied field, or when the dimensions involved are within the single-domain limit. We derive analytical expressions for the demagnetization factors of cylindrical shells and rings with rectangular and square cross-sections. The factors are given either as a combination of elliptic integrals or as a series expansion in powers of the dimensionless ratio between inner and outer radii. Limiting cases are analysed for particular ranges of the shape parameters. We also investigate the ring with a square cross-section, and the elliptic ring, where analytical expressions are provided only for small eccentricity. Finally, we introduce the dipolar coupling integral encoding magnetostatic interactions between a magnetized cylinder and a thin coating on its lateral surface.

1.21 General magnetostatic shape-shape interactions forces and torques [21]

Expressions for the magnetostatic interaction force and torque between two magnetic objects of arbitrary shape are derived within the shape amplitude formalism. A generalized force is derived as the gradient of the magnetometric tensor field, which is the convolution of the cross-correlation of the object shapes with the dipolar tensor field. Expressions for the mechanical and magnetic torques are also derived in terms of the magnetometric tensor field. Expressions suitable for numerical evaluation are given as finite Fourier summations. Example computations are given for the interactions between pairs of uniformly magnetized spheres (for which analytical results are compared to numerical results), cubes, octahedra, tetrahedra, and cuboctahedra. The accuracy of the derived numerical relations for energy, force, and torques is of the order of 0.1% for object spacings smaller than the object dimensions.

1.22 Magnetostatics of the uniformly polarized torus [22]

We provide an exhaustive description of the magnetostatics of the uniformly polarized torus and its derivative self-intersecting (spindle) shapes. In the process, two complementary approaches have been implemented, position-space analysis of the Laplace equation with inhomogeneous boundary conditions and a Fourier-space analysis, starting from the determination of the shape amplitude of this topologically non-trivial body. The stray field and the demagnetization tensor have been determined as rapidly converging series of toroidal functions. The single independent demagnetization-tensor eigenvalue has been determined as a function of the unique aspect ratio α of the torus. Throughout the range of values of the ratio, corresponding to a multiply connected torus proper, the axial demagnetization factor Nz remains close to one half. There is no breach of smoothness of $N_z(\alpha)$ at the topological crossover to a simply connected tight torus ($\alpha = 1$). However, N_z is a non-monotonic function of the aspect ratio, such that substantially different pairs of corresponding tori would still have the same demagnetization factor. This property does not occur in a simply connected body of the same continuous axial symmetry. Several self-suggesting practical applications are outlined, deriving from the acquired quantitative insight.

1.23 Recent progress in Lorentz transmission electron microscopy: applications to multiferroic materials [23]

After a brief review of the basic methods of Lorentz transmission elec- tron microscopy (LTEM), including the Transport-of-Intensity formalism for phase reconstruction, we present a few examples of the application of LTEM to multi- ferroic materials, in this case ferromagnetic shape memory alloys. We discuss ob- servations of magnetic domain walls pinned to anti-phase boundaries in Ni_2MnGa, and domain wall behavior under an in-situ applied magnetic field in Fe-Pd-Co.

1.24 Three-dimensional study of the vector potential of magnetic structures [24]

The vector potential is central to a number of areas of condensed matter physics, such as superconductivity and magnetism. We have used a combination of electron wave phase reconstruction and electron tomographic reconstruction to experimentally measure and visualize the three-dimensional vector potential in and around a magnetic Permalloy structure. The method can probe the vector potential of the patterned structures with a resolution of about 13 nm. A transmission electron microscope operated in the Lorentz mode is used to record four tomographic tilt series. Measurements for a square Permalloy structure with an internal closure domain configuration are presented.

1.25 The influence of magnetostatic interactions in exchange-coupled composite particles [25]

Exchange-coupled composite (ECC) particles are the basic constituents of ECC magnetic recording media. We examine and compare two types of ECC particles: (i) core-shell structures, consisting of a hard-magnetic core and a coaxial soft-magnetic shell and (ii) conventional ECC particles, with a hard-magnetic core topped by a soft cylindrical element. The model we present describes the magnetic response of the two ECC particle types, taking into account all significant magnetic contributions to the energy landscape. Special emphasis is given to the magnetostatic (dipolar) interaction energy. We find that both the switching fields and the zero-field energy barrier depend strongly on the particle geometry. A comparison between the two types reveals that core-shell ECC particles are more effective in switching field reduction, while conventional ECC particles maintain a larger overall figure of merit.

1.26 Nanoscale structure of the magnetic induction at monopole defects in artificial spin-ice lattices [26]

Artificially frustrated spin-ice systems are of considerable interest since they simulate the spin frustration and concomitant rich behavior exhibited by atoms on a crystal lattice in naturally occurring spin-ice systems such as pyrochlores. As a result of the magnetic frustration, these systems can exhibit magnetic monopole type defects, which are an example of an exotic emergent quasiparticle. The local magnetization structure of such monopole defects determines their stability and thus is critical to understanding their behavior. In this paper, we report on the direct observation at room temperature of the nanoscale magnetic structure of individual magnetic monopoles in an artificially frustrated two-dimensional square spin-ice lattice, using high-resolution aberration-corrected Lorentz transmission electron microscopy. By combining the high-resolution microscopy with micromagnetic simulation, we demonstrate how nucleation of defect strings, reminiscent of Dirac strings, connecting monopole defects controls the demagnetization process in these spin-ice lattices.

1.27 On the magnetostatics of chains of magnetic nanoparticles [27]

A novel approach is presented for the computation of the magnetostatic energy of straight and bent chains of identical, uniformly magnetized particles of arbitrary shape. The formalism relies on the concept of the magnetometric tensor field, and allows for closed form expressions for the magnetostatic energy, demagnetization factor, Young's modulus, and bending modulus of chains in terms of the shape amplitude of the particles. Analytical solutions are presented for straight chains of spheres, cubes, and cylinders, and for bent chains of spheres. Numerical results include chains of octahedra, tetrahedra, cuboctahedra, and bi-cones. The axial demagnetization factor for the bi-cone shape is derived in analytical form. An approximate energy expression, using the full shape-dependent interaction formalism for short separation distances, and the standard dipolar interaction expression for larger distances, is introduced.

1.28 Magnetic interactions and reversal of artificial square spin ices [28]

Artificial spin ices are nanoscale geometrically engineered systems that mimic the behavior of bulk spin ices at room temperature. We describe the nanoscale magnetic interactions in a square spin ice lattice by an experimentally verified model that accounts for the correct shape of the magnetic islands. Magnetic force microscopy measurements on lithographically fabricated lattices are compared to Monte Carlo simulations of the reversal process of two lattices with different lattice spacings. Lattice node statistics and correlations show significant differences in the reversal mechanism for lattices with different spacings. The effect of structural variations is also compared for the two lattice reversals.

1.29 Forces between a permament magnet and a soft magnetic plate [29]

Forces between a hard/permanent magnet of arbitrary shape and an ideally soft magnetic plate in close proximity are derived analytically from the image method applied to magnetostatics. We found that the contact force, defined as the force required to detach the hard magnet from the plate, coincides with that between two identical touching permanent magnets. Furthermore, if the hard and the soft magnets are displaced by some amount, their attraction equals that between two identical permanent magnets displaced by twice that amount. Experimental results are presented that validate the theoretical framework and highlight its limits of applicability.

1.30 On the computation of the magnetic phase shift for magnetic nano-particles of arbitrary shape using a spherical projection model [30]

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.

1.31 Separation of electrostatic and magnetic phase shifts using a modified transport-of-intensity equation [31]

We introduce a new approach for the separation of the electrostatic and magnetic components of the electron wave phase shift, based on the transport-of-intensity equation (TIE) formalism. We derive two separate TIE-like equations, one for each of the phase shift components. We use experimental results on FeCoB and Permalloy patterned islands to illustrate how the magnetic and electrostatic longitudinal derivatives can be computed. The main advantage of this new approach is the fact that the differences in the power spectra of the two phase components (electrostatic phase shifts often have significant power in the higher frequencies) can be accommodated by the selection of two different Tikhonov regularization parameters for the two phase reconstructions. The extra computational demands of the method are more than compensated by the improved phase reconstruction results.

1.32 Theoretical study of ferroelectric nanoparticles using phase reconstructed electron microscopy [32]

Ferroelectric nanostructures are important for a variety of applications in electronic and electro-optical devices, including nonvolatile memories and thin-film capacitors. These applications involve stability and switching of polarization using external stimuli, such as electric fields. We present a theoretical model describing how the shape of a nanoparticle affects its polarization in the absence of screening charges, and quantify the electron-optical phase shift for detecting ferroelectric signals with phase-sensitive techniques in a transmission electron microscope. We provide an example phase shift computation for a uniformly polarized prolate ellipsoid with varying aspect ratio in the absence of screening charges.

1.33 Recent advances in Lorentz microscopy [33]

Lorentz transmission electron microscopy (LTEM) has evolved from a qualitative magnetic domain observation technique to a quantitative technique for the determination of the magnetization state of a sample. In this review article, we describe recent developments in techniques and imaging modes, including the use of spherical aberration correction to improve the spatial resolution of LTEM into the single nanometer range, and novel in situ observation modes. We review recent advances in the modeling of the wave optical magnetic phase shift as well as in the area of phase reconstruction by means of the Transport of Intensity Equation (TIE) approach, and discuss vector field electron tomography, which has emerged as a powerful tool for the 3D reconstruction of magnetization configurations. We conclude this review with a brief overview of recent LTEM applications.

2 Publications with DOE support

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