Direct Observation of Unconventional Topological Spin Structure in Coupled Magnetic Discs

C. Phatak,[1] A. K. Petford-Long,[1,2] and O. Heinonen[1,3]

[1]*Argonne National Laboratory, 9700 S. Cass Avenue, Argonne, Illinois 60439, USA*
[2]*Department of Materials Science and Engineering, Northwestern University, 2220 Campus Drive, Evanston, Illinois 60208, USA*
[3]*Department of Physics and Astronomy, Northwestern University, 2145 Sheridan Road, Evanston, Illinois 60208, USA*

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Confined magnetic thin films are known to exhibit a variety of fascinating topological spin states such as Skyrmions, vortices, and antivortices. Such topological excitations are fundamentally important to our understanding of quantum critical phenomenon and related phase transitions. Here we report on the direct observation of an unconventional topological spin state and its behavior in antiferromagnetically coupled NiFe discs at room temperature. The observed spin structure is similar to the theoretically predicted merons which have not yet been observed directly. We have used *in situ* Lorentz microscopy magnetizing experiments combined with micromagnetic simulations to follow the stability and the behavior of the meron state. The work presented in this paper will open new opportunities for direct experimental investigation of various topological states that can provide insights into the fundamental physics of their interactions.

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The concept of topological ground states and excitations originated in the study of exotic field theories but has more recently emerged in all branches of physics [1], particularly strongly correlated systems such as high temperature superconductors [2], Mott insulators [3,4], as well as topological insulators [5,6]. A major hindrance in the direct study of these states is that most of them occur only at very low temperatures in bulk materials and cannot be visualized or probed directly. By artificially changing the degrees of freedom in the system, for example, by patterning, these states can be accessed at room temperatures. For example, Skyrmions have been observed in 2D confined thin films of FeGe at close to room temperature [7]. Similarly, patterning ferromagnetic thin films to submicron sized discs leads to the formation of the vortex state [8,9].

One nontrivial topological spin state is the meron state that originally emerged as a solution to classical Yang-Mills theories [10], in which the spins point radially outward or inward from a core within a plane and point out of the plane at the core [Fig. 1(a)]. In particle physics, merons have been described in the context of quark confinement and can only exist in pairs [11]. There have also been several theoretical predictions regarding the existence of merons in condensed matter systems, e.g., as ground states in quantum Hall droplets [12,13], or thin chiral magnetic films [14], although the experimental verification is still lacking.

In this work, we will show that the combination of confinement and an antiferromagnetic interfacial exchange coupling mediated by a spacer layer gives rise to a topological spin state which is similar to the meron state and that can be directly observed experimentally. The meronlike topological state in AF-coupled Py ($Ni_{80}Fe_{20}$) discs was observed using aberration-corrected Lorentz transmission electron microscopy (LTEM). LTEM offers the ability of imaging domain behavior in all the layers simultaneously with high spatial resolution, with the sample sitting in a field-free state. We also employ the *transport-of-intensity equation* (TIE) based phase reconstruction technique to obtain thickness-integrated magnetic induction maps. The response of the discs to an applied magnetic field is observed using an *in situ* magnetizing holder that applies a controlled magnetic field in the plane of the discs. Our experimental observations combined with micromagnetic modeling lead us to conclude that the observed spin state is similar to the meron state.

Although ferromagnetic (FM) layers coupled antiferromagnetically (AF) via nonmagnetic spacers have been studied previously, there has been relatively little work on the spin states of patterned AF-coupled structures. Previous work on imaging of the domain behavior in such systems has used x-ray photoemission electron

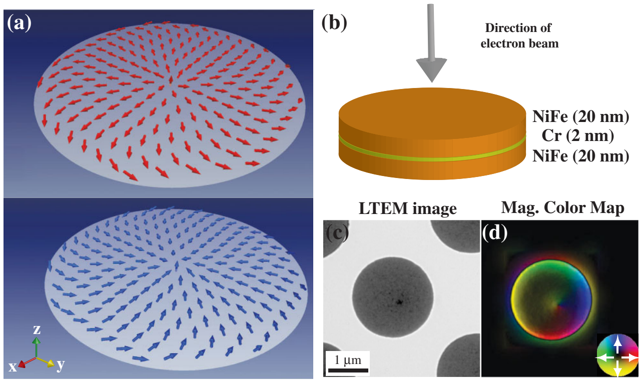

FIG. 1 (color online). (a) Schematic showing the spin structure of a meronlike state with opposite chirality; (b) schematic showing the trilayer discs and direction of the electron beam for imaging, and (c) LTEM under-focus image, and (d) reconstructed magnetic induction color map.

microscopy, as well as magnetic force microscopy [15]. However, these techniques are limited to observing the behavior in the top surface layer only and do not give information about the bottom layer.

Figure 1(b) shows a schematic of the trilayer sample prepared using electron-beam lithography. Py(20 nm)/Cr(2 nm)/Py(20 nm) discs were 2 μm in diameter, sputter deposited on an electron-transparent SiN membrane support. The Cr spacer thickness of 2 nm was chosen because Cr-mediated Ruderman-Kittel-Kasuya-Yosida (RKKY) coupling between the Py layers shows an antiferromagnetic peak for this particular spacer thickness [16]. The sample was observed in LTEM mode in its virgin state prior to exposure to any external field. The schematic also shows the direction of the electron beam indicating that the contrast seen in the LTEM images is a composite from all the layers. Figure 1(c) shows an under-focus LTEM image of one such disc. Using a through-focus series and the TIE technique for phase retrieval we obtained the thickness-integrated magnetization map of the disc as shown in Fig. 1(d) . The out-of-focus LTEM image shows a black dot contrast visible at the center of the disc. This contrast is similar to that observed for the core in a vortex state, for which the black or white dot contrast is related to the chirality of the vortex state [17]. The reconstructed magnetization map shows color contrast only around the core region and near the edge of the disc. There is almost no color contrast seen in between the core and the edge of the disc.

In order to understand the observed contrast and the resulting spin structure in the discs, it should be noted that this contrast results from the combined phase shift imparted to the electrons by each of the discs. If we assume that both discs are in vortex states with the same chirality, the out-of-focus image would show a single black or white dot at the core and the reconstructed magnetization map would show color across the entire disc area. Although we see a single black dot in the experimental image, the color indicating magnetization is not seen across the entire disc, but only near the core and the disc edge. Thus the discs cannot contain vortices with the same chirality. Next, if we assume the discs have vortices with opposite chirality, then the out-of-focus image would show either no contrast since the phase shift resulting from one disc would be identical and opposite to the phase shift resulting from the second disc, or would show a black and a white dot simultaneously if the vortex cores do not exactly line up. However, in the out-of-focus image, we see no such evidence for the simultaneous presence of white and black dots, and the black dot is clearly visible. Thus the discs cannot have a spin structure consisting of vortices with opposite chiralities. These observations indicate the formation of an altogether different spin state.

To gain further insight and understand the resulting experimental contrast, micromagnetic simulations were performed for 2 μm NiFe discs. The coupling between the discs was modeled using an interlayer exchange coupling field, H_{IEC}, where positive values of H_{IEC} indicate FM coupling and negative values indicate AF coupling [18]. A field-free equilibrium magnetization in each disc was obtained for different values of H_{IEC}. LTEM images were then simulated using this magnetization as the input to compare with the experimentally observed LTEM images. Figure 2 shows the simulated out-of-focus LTEM image, magnetization color maps, and the magnetization in each disc close to the interface between the individual disc and the nonmagnetic spacer layer. Figure 2(a) shows this for a FM coupling with $H_{\mathrm{IEC}} = +15.6$ Oe, corresponding to an interlayer exchange energy of $0.025\ \mathrm{erg/cm^2}$. As expected, both discs show vortex states with same chirality, and the simulated LTEM image shows a black dot at the vortex core. The magnetization color map shows contrast all across the disc as mentioned earlier. This is inconsistent with the experimentally observed magnetization color map, which confirms that the observed magnetization state is not a vortex. Figure 2(b) shows the equilibrium magnetization obtained for AF coupling with $H_{\mathrm{IEC}} = -15.6$ Oe, and with an initial state consisting of two vortices with same chirality and polarity.

The resulting magnetization distribution in each disc is very interesting and unexpected. The magnetization points radially outward in the bottom disc and radially inward in the top disc, and curls in plane near the core and edge of the disc. At the core, the magnetization in each disc points out of plane in the same direction. The simulated out-of-focus LTEM image shows a black dot at the core and the magnetization color map shows color contrast only around the core and the edge of the disc. This is consistent with the experimentally observed LTEM image contrast and the magnetization color map, thus confirming that each of the discs is indeed in a spin state similar to the topological meron state. The parts of the disc where the magnetization curls in-plane, i.e., the core and edge of the disc, result in a nonzero phase shift of the electrons. The region for which one disc contains radial inward magnetization, superimposed on radial outward magnetization in the second disc,

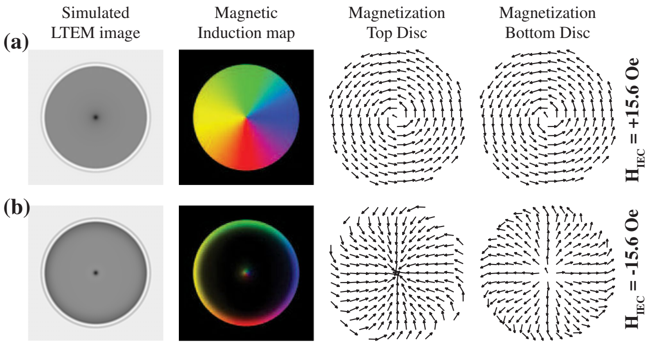

FIG. 2 (color online). Micromagnetic simulation results showing the magnetization in top and bottom disc along with simulated LTEM underfocus image and magnetic induction map for various interlayer coupling strengths (a) +15.6 Oe, (b) and −15.6 Oe.

results in a net zero phase shift of the electrons and no contrast. As a consequence, the magnetization color contrast is seen only around the core and the edge of the disc. It should be noted that such a magnetization state exists only because of the confinement and exchange coupling between the two discs. Such a state is highly improbable in a single disc or a 2D magnetic thin film.

Figure 3 shows the change in modeled magnetization in the bottom disc as the meronlike state evolves from an initial state in which the two layers contain vortices with opposite chiralities. The top right corner indicates the simulation time steps in nanoseconds. As the magnetization relaxes, the spin structure starts to deform and "unwind" from the curling vortex state to the meronlike state. A similar transition occurs in the top disc with the magnetization pointing in the opposite direction as compared to the bottom disc shown in the figure. The deformation is of course topologically invariant and preserves the Chern-Pontryagin topological index. The micromagnetic simulations also indicate that the meronlike state is a stable equilibrium state for negative H_{IEC} in the absence of an external field. In the meronlike state, the circulation of the spin along the disc edge is the same for both discs. A state with two vortices with opposite chiralities has lower total energy, but in order to access this state from the meronlike state, a rather large energy barrier associated with reversal of the magnetization along the disc edge in one disc has to be overcome.

We speculate that the following mechanism leads to the formation of an as-deposited meronlike state. Inhomogeneities in the spacer layer thickness will cause the RKKY coupling to fluctuate spatially. Local ferromagnetic coupling near the disc edge will lead to the spins near the outside of the disc lying preferentially parallel to the disc edge. As the sample thickness increases during deposition, H_{IEC} diminishes but if the ferromagnetic spin circulation around the edge has already formed, it would take a large energy to reverse it and form vortices with opposite chirality, and thus the system forms the meronlike state. Furthermore, spin circulation at the disc edge implies that the nucleation energy in an applied in-plane field is lower for the meronlike state than for vortices with opposite chiralities. This is consistent with experimental observations in which discs initially not in a meronlike state at zero field form a meronlike state after saturation in an in-plane field.

The quasistatic response of the magnetization configuration to an applied external field was studied *in situ* by LTEM using a magnetization holder in which the sample sits in a uniform region of the in-plane field that can be altered in a controlled manner. Figure 4(a) shows a series of out-of-focus LTEM images as the field is varied from 0 to 75 Oe. The direction of the applied field in indicated in the figure. The image at zero field shows the black dot at the core of the meronlike state as observed earlier. At low fields, the black dot contrast moves along a trajectory that is approximately at 45° to the external field. As the field is increased, the single black dot contrast is seen to split into two black dots. The field value at which the initial dot splits into two varies somewhat from sample to sample. With increasing field, the two black dots move away from each other and towards the edge of the disc, with the center-of-mass of the dots continuing along the 45° trajectory.

In a vortex state, the vortex core will move in a direction perpendicular to the applied field [19]. In case of two discs on top of each other, if the vortices have the same chirality, they will move in the same direction, and if they have opposite chirality, they will move in opposite directions. The observed behavior is not consistent with this and points toward a novel emergent behavior, which has not been reported earlier in literature.

Micromagnetic simulations were performed to understand the observed behavior. The magnetization behavior was studied for an AF coupling of $H_{\mathrm{IEC}} = -15.6$ Oe, at which the discs formed a stable meronlike state as shown earlier in Fig. 2(b). A field was then applied in increments of 10 Oe and at each step, the magnetization was allowed to relax for 3–15 ns. Figure 4(b) shows the simulated underfocus LTEM images as well as the magnetization adjacent

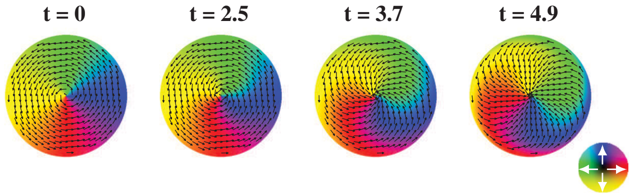

FIG. 3 (color online). Micromagnetic simulations showing the evolution of the meronlike state from a vortex state in the bottom disc for $H_{\mathrm{IEC}} = -15.6$ Oe. The numbers in the top right corner show the simulation time steps in nanoseconds.

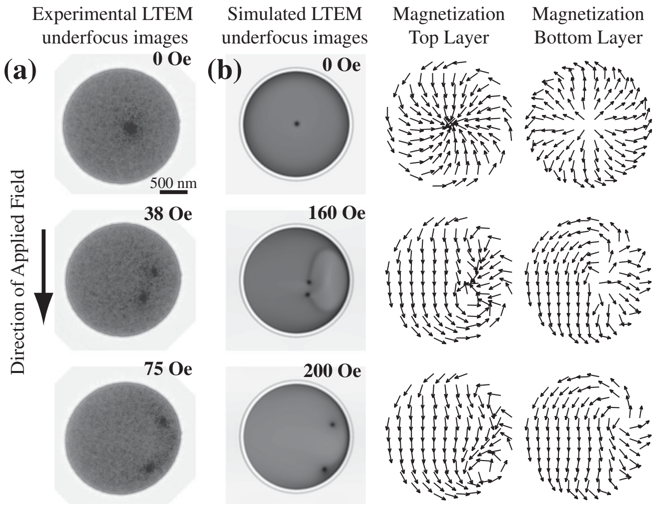

FIG. 4. (a) LTEM underfocus images recorded as a function of applied in-plane magnetic field, and (b) micromagnetic simulations for a disc in meronlike state with interlayer coupling strength of -15.6 Oe.

to the interface from each of the disc as a function of applied field. The simulated LTEM images show the same behavior as seen in the experimental images. The simulations show that the magnetizations in the two layers start to scissor with components along the field at low fields. This drives the formation of a low-angle domain wall at approximately 45° to the applied field [Fig. 4(b)], causing the cores to move downward at 45° to the field. There is a good agreement between the simulations and the experimentally observed behavior. There are some differences, in particular, the field values at which the cores separate and at which they disappear were lower in the experiments than in the simulations. These can be attributed to the differences between the ideal sample (simulations) and the experimental sample. There are no inhomogeneities or pinning sites in the simulations whereas in the experimental sample inhomogeneities such as grain boundaries will pin the cores, similar to what has been observed for vortex cores [20]. In the presence of thermal fluctuations and an external field, the cores will keep moving from shallow pinning sites until they reach sites with pinning strengths that cannot be overcome by thermal fluctuations. In the simulations, the meronlike state is not pinned nor do thermal fluctuations assist its motion, and as a result the state moves slowly on the short time scale of the length of the simulations along a trajectory of degenerate or nearly degenerate states. Therefore, the state observed at the end of a simulation in a particular field depends on the length and history of the simulation. As a result, the experimental response is observed at lower applied fields than what is obtained in the simulations. As a result, the experimental response is observed at lower applied fields than what is obtained in the simulations. For additional information about micromagnetic simulations and a movie showing the *in situ* magnetization results, please refer to the supplementary material [21].

In summary, we have observed meronlike spin structures in antiferromagnetically coupled ferromagnetic discs. While this state is not the minimum-energy state in the absence of an external field, it is a stable equilibrium state. The existence of the meronlike state and the ability to access it easily at room temperature may open a path to interesting dynamical behavior for single Py/Cr/Py discs, as well as interesting static and dynamical behavior of arrays of such discs. This will be important to control and predict the behavior of nanostructured magnetic materials, which are key to the next generation of magnetic storage and memory applications [22]. Additionally, this research also opens new opportunities for the study of interesting physics of critical phenomenon and related topological states directly at room temperature.

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