Effects of exchange bias on magnetotransport in permalloy kagome artificial spin ice

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Keywords: artificial spin ice, exchange bias, magnetotransport

Abstract

We investigate the magnetotransport properties of connected kagome artificial spin ice networks composed of permalloy nanowires. Our data show clear evidence of magnetic switching among the wires, both in the longitudinal and transverse magnetoresistance. An unusual asymmetry with field sweep direction appears at temperatures below about 20 K that appears to be associated with exchange bias resulting from surface oxidation of permalloy, and which disappears in alumina-capped samples. These results demonstrate that exchange bias is a phenomenon that must be considered in understanding the physics of such artificial spin ice systems, and that opens up new possibilities for their control.

Artificial spin ice is a class of magnetic nanostructure that emulates spin ice [1] in a system of single-domain ferromagnetic (FM) islands or connected wires arranged in geometries such that their magnetostatic interactions are frustrated [2–4]. Particular attention has been paid to the kagome artificial spin ice, in which the islands or nanowires comprise the links of a honeycomb lattice [5–18]. The shape anisotropy of the structure results in a single magnetic domain on each island or link with magnetization oriented along the long axis—towards one vertex and away from another—so that the moments form a kagome lattice. Kagome artificial spin ices have been observed to obey a modified version of the ice rules: each vertex is the convergence of three islands or nanowires, with the energetically preferred state found to be either two magnetizations pointing towards the vertex and one pointing away (2-in/1-out) or one pointing towards the vertex and two pointing away (1-in/2-out) [5, 6]. The effective magnetic charges at each vertex order under thermalization in this and other artificial spin ice structures, and have even been shown to exhibit charge screening [18, 19], demonstrating this structure as a model system for investigating collective effects in magnetic frustration. As such, artificial spin ice is an example of a material-by-design, where the properties of the network may be tuned at will. The flexibility in both the tailoring and measurement of such an array has already provided considerable insight into the physics of frustration, and mechanisms which may be used to manipulate the array further can enhance its functionality. In principle, one such mechanism for affecting both magnetic switching and ordering is exchange bias, in particular from the formation of antiferromagnetically-ordered native oxides. The effects of exchange bias from native oxides have been investigated extensively in magnetic thin films, and other nanostructures, but have not yet been considered in manipulating artificial spin ice.

Although experimentally investigated artificial spin ices are typically formed from individual magnetic islands, a number of studies have been performed by measuring the magnetoresistance (MR) of connected elements. In connected honeycomb artificial spin ice networks composed of permalloy (Ni$_{81}$Fe$_{19}$) [5] and cobalt (Co) [17] the MR is dominated by anisotropic magnetoresistance (AMR), through which the resistance of each link changes as a function of the angle between the magnetic moment and the local electric current density. In principle, therefore, the MR provides an electrical probe of the micromagnetic configuration of the network, which may be used to probe the switching and ground state of the lattice. This gives a facile means to observe the...
evolution of such behavior as temperature is varied, for example, and allows valuable insight into the ordering of such arrays. Monte Carlo studies have predicted four distinct phases in kagome artificial spin ice: a high energy paramagnetic phase; an intermediate ice-I phase in which the modified ice rules are obeyed; a lower ice-II phase exhibiting magnetic charge ordering; and a lowest energy loop phase where chiral ordering of individual hexagons occurs [7]. As the ordering of each phase is heavily dependent on the micromagnetics of the lattice, MR studies can provide valuable insight into switching behavior. Recently, in Co kagome networks, the formation of the predicted loop phase was associated with an observation of field reversal asymmetry in the MR at low temperature [17].

In this work, we examine the temperature-dependent magnetotransport properties of connected kagome artificial spin ice networks made from permalloy, both with and without a 2 nm aluminum capping layer. Near room temperature, the uncapped and capped networks exhibit similar behavior, including MR data that are symmetric under field inversion. At sufficiently low temperatures however, the behavior becomes qualitatively different, with asymmetric behavior appearing only in the uncapped networks. This asymmetry in the uncapped samples appears to be associated with exchange bias that sets in at the same temperature. These results demonstrate that exchange bias in artificial spin ice can result in qualitatively new behavior, highlighting the care that must be taken in interpreting MR data when exchange bias may be present. Exchange bias must therefore be accounted for in analysis of asymmetry, and opens the door to the use of exchange bias as a tool for manipulating artificial spin ice phenomena.

The Hall bar geometry honeycomb networks pictured in figure 1 were fabricated by electron-beam lithography on a Si$_3$N$_4$ coated silicon substrate (Si$_3$N$_4$ layer approximately 200 nm thick). Permalloy and aluminum depositions were done by ultra-high vacuum electron beam evaporation at room temperature, with a base pressure of $1 \times 10^{-10}$ Torr, and growth rates of 0.5 and 0.2 Å s$^{-1}$ respectively. On removal to atmosphere, the Al oxidizes in air, forming a self-limited ~2 nm thick AlO$_x$ native oxide layer, thereby protecting the underlying permalloy. Each individual permalloy arm is approximately 800 nm long, 75 nm wide, and 25 nm thick, with 29 hexagons spanning the long axis and 10 hexagons across the short axis of the network. At either end of the long axis, current injection pads spanning the width of the sample are connected to large contact pads far from the sample for ease of electrical connectivity. Similarly, six pads are strategically placed along the sample sides for voltage detection.

Transport measurements were performed in commercial helium-4 cryostats with the ability to vary the temperature ($T$) from 1.8 to 400 K and an in-plane magnetic field that could be applied along any angle through 360° of in-plane rotation. The measurements were made with a 17 Hz ac excitation ($I = 66.7 \mu A$) supplied along the long axis of the sample. Voltage measurements were taken with lock-in amplifiers along the nominal current direction ($R_{1L}$; voltage measured between leads 1 and 3, as indicated in figure 1(c)) and perpendicular to the current direction ($R_{1L}$; leads 2 and 5). Variations in the frequency and the applied current amplitude did not

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**Figure 1.** (a) Scanning electron microscopy image of a representative connected kagome artificial spin ice sample. (b) Magnetic force microscopy image confirming single domain behavior of nanowire links as evidenced by a lack of magnetic contrast outside of the vertices. (c) Graphic illustrating the numeric designation of various leads for voltage measurement. The current $I$ flows across the long axis of the sample. (d) The external field $H$ is applied at an angle $\theta$ measured with respect to the nominal current direction.
significantly affect the results. When field cooling, the external field is applied along the direction of expected field variation for MR measurements. The shape anisotropy of each nanowire link results in a single domain state with the magnetization preferentially aligned along the link’s long axis, as verified by magnetic force microscopy. A typical image is shown in figure 1(b), where the bright and dark spots at the vertices correspond to 1-in/2-out or 2-in/1-out states. Measurements of a similar network with much smaller contact pads (∼400 nm wide) show the same qualitative behavior, indicating that the measured MR features originate from the array itself, not from effects in the contact pads.

We first present $R_{||}(H)$ and $R_{\perp}(H)$ for both the uncapped and capped networks at $T = 2$ K, for an $I$–$H$ angle of $\theta = 0^\circ$ (figure 2). $R_{||}(H)$ exhibits the symmetric behavior expected in a system exhibiting AMR. As is common in magnetic systems reversing via domain wall nucleation and propagation, the sharp dips in $R_{||}(H)$ may be attributed to magnetization reversal events, with the majority of the array appearing to switch at a similar applied magnetic field. Also found is an approximately quadratic high-field AMR dependence at high fields (±10 kOe, not shown), due to the rotation of the magnetization in the ±60° arms out of their respective easy axes (and hence away from the current direction) by the applied field, as expected from Stoner–Wohlfarth-like coherent rotation of a FM towards a hard axis. While the details of the switching events varied with angle, the symmetry seen in the left panels of figure 2 was observed for all orientations of the magnetic field. The qualitative features above the background in $R_{\perp}(H)$ are strikingly different from those in $R_{||}(H)$, although they occur in the same field regime and are thus attributable to the same domain-switching. Unlike $R_{||}(H)$, we find that $R_{\perp}(H)$ displays a ‘low field’ dependence ($|H| < 500$ Oe) that has both the shape and field inversion symmetry of the planar Hall effect (PHE). The shape of the low field signal is reminiscent of the evolution of a uniform, coherently rotating magnetization through a hard axis in a linearly changing field: the PHE signal oscillating about zero as the magnetization passes from an angle of zero to $p$ through $p/2$ with respect to the applied current.

The most surprising feature of the data in figure 2 is that $R_{\perp}(H)$ is not symmetric under field inversion at low temperatures in the uncapped samples, an effect that is almost completely absent in the capped samples. The asymmetry is most stark in the field region between ±500 Oe, with the larger ±750 Oe peaks remaining unaffected. This effect is also observed at other applied magnetic field angles, but is most pronounced at $\theta = 0^\circ$. In a similar network, where the hexagons are rotated 90° with respect to the current direction and with the magnetic field applied perpendicular to the current direction (i.e., the same field-lattice orientation, different four-point measurement geometry), we observe the same asymmetric $R_{\perp}(H)$ behavior, indicating that the asymmetry is an intrinsic property of the arrays.

We find that the asymmetry in $R_{\perp}(H)$ is a low temperature effect, observed only below $T \sim 20$ K. Figure 3 shows $R_{\perp}$ versus $H$ data for the uncapped network at various temperatures after field cooling ($H_{\text{cool}} = 10$ kOe)

Figure 2. Longitudinal and transverse resistance measurements of a permalloy (Py) nanowire network and an Al-capped Py nanowire network at $T = 2$ K and $\theta = 0^\circ$. $\Delta R$ is defined as $\Delta R (H) = R (H) - R (H = 0)$. The $R_{||}$ data of the uncapped network exhibits a more extreme asymmetry than that of the capped network. The uncapped sample was field cooled ($H_{\text{cool}} = 10$ kOe) and the capped sample was cooled in zero field.
from $T = 300$ to $2$ K. These measurements were taken sequentially starting at $2$ K and then at progressively higher temperatures; reversal of the order of measurements has no significant effect on the results. Temperatures below $10$ K appear qualitatively similar in their asymmetries, while above $20$ K, the data appear to be symmetric under field inversion. The $15$ K data show an intermediate state highlighting the evolution of the asymmetry; the asymmetry predominantly affects only one of the field sweeps (the upward sweep for positive field cooling) with that sweep flattening out and then inverting as the temperature is varied. As a measure of the asymmetry, in figure 4 we plot the sum of the slopes of $\Delta R(\perp)$ near $H = 0$ for sweeps increasing and decreasing in magnetic field. This sum of the slopes should be zero for a symmetric MR loop, and indeed we find it to be zero at high temperatures. Below $20$ K, however, the sum of the slopes rises almost monotonically on cooling, demonstrating the onset of asymmetry. The temperature dependence of the asymmetry in $R_\perp(H)$, and its absence in the capped samples, suggests that it may be associated with a native oxide. Permalloy has been found to have a variety of such native oxides, including NiO, Fe$_2$O$_3$, and Fe$_{x}$Ni$_{1-x}$O [20–24]. These antiferromagnetic (AFM) oxides, when interfaced with the FM permalloy, have been shown to result in exchange shifts in the hysteresis loop of permalloy devices below $20$ K [21] and high magnetic damping below $40$ K [23]. The relevance of exchange bias is further evidenced by the behavior of the capped network, where the addition of the aluminum capping layer, whose native oxide is not magnetically ordered, prevents the asymmetry from appearing [25]. We note that in the capped network, sidewall formation of permalloy oxides is still possible, as the capping layer protects only the top of the permalloy, but the area of permalloy exposed to air is significantly reduced. The resulting data show a suppression of the asymmetry due to the addition of the aluminum layer. A small degree of residual asymmetry can be attributed to the oxidation at the sidewalls.

To test the hypothesis of exchange bias as the root cause of the asymmetry in $R_\perp(H)$, we applied an external magnetic field during the initial cooling procedure, and we find that it indeed changes the polarity of the asymmetry (figure 5). When a negative field of $10$ kOe is applied while cooling, the $100$ Oe peaks for both sweeps are ‘concave up’ and the $-100$ Oe peaks are ‘concave down’. Conversely, when a positive field is applied while cooling, the concavities of the ±$100$ Oe peaks are reversed (figure 5). Note that cooling with smaller fields (i.e.,
300 Oe) did not substantially change the resultant data. This dependence on field cooling is reminiscent of the effects of exchange bias, wherein the direction of the applied field during cooling affects the direction in which the hysteresis loop is shifted [20, 26].

To further validate the attribution of the asymmetry to exchange bias, we also used SQUID magnetometry (Quantum Design MPMS) to measure magnetization hysteresis loops of unpatterned permalloy films deposited in the same manner as the networks (without capping). Measurements following field cooling at ±300 Oe show shifted hysteresis loops, with the exchange field $H_E$ measured as the difference in field magnitude between the center of the loop and zero field (inset of figure 4). The onset temperature of exchange bias (the blocking temperature) is consistent with reports of low temperature exchange biasing effects in devices with permalloy

Figure 4. Measurements of the exchange field $H_E$ (distance from center of the $M$–$H$ loop to zero field) as a function of temperature for a Py film are plotted on the left $y$–axis (red line). The film was field cooled ($H_{cool} = 300$ Oe) to 2 K before measurements were taken. The exchange bias appears to take effect between $T = 15$ and 20 K. An asymmetry measure of the magnetoresistance curves plotted in figure 5 is displayed on the right axis for both a non-capped sample (solid blue line) and a capped sample (dashed blue line); this measure is calculated as the sum of the slopes of both sweeps near $H = 0$, for any given temperature. Slopes are calculated by taking the difference between the resistance values at ±50 Oe and dividing by 100 Oe. Inset: Magnetization hysteresis loop of the Py film at 4 K, exhibiting a nonzero $H_E$.

Figure 5. When cooling with an applied external field ($|H_{cool}| = 10$ kOe) through the transition temperature, where the asymmetry appears, the direction of the field influences the asymmetry that develops. The data shown above ($R_{\perp}$) are from the uncapped network at 2 K with the field applied at $\theta = 0^\circ$. The large peaks at $\sim\pm 750$ Oe are shifted about 25 Oe from one sweep to the other, consistent with exchange biasing.
The temperature dependence of the exchange field closely follows the sum of the slopes also shown in figure 4, further confirming exchange bias as the likely cause. Further evidence for the presence of FM/AFM exchange coupling may be seen when considering the AMR signal in $R_\perp(H)$ from the switching of the array, i.e., the sharp dips discussed earlier (figure 2, left panel). The well-known biasing of the underlying ferromagnet, and so shifting of the switching fields (due to the induced unidirectional anisotropy), is both measurable and may be set by the direction of cooling field (as in figure 5). Note also that both $R_\parallel(H)$ and $R_\perp(H)$ (figure 2) show an increase in the switching fields for uncapped versus capped samples, as might be expected of an exchange bias phenomenon.

We now discuss how exchange bias could lead to the observed asymmetry in $R_\perp(H)$. The simplest effect of interfacial exchange in thin film AFM–FM heterostructures—i.e., a FM layer with only unidirectional (in this case arising from exchange bias) and uniaxial anisotropy—is a horizontal field shift in the hysteresis loop, mathematically equivalent to an external bias field in simple models. Beyond this biasing, no asymmetry should develop in the magnetization reversal path with respect to the applied field direction; other than a shift, no field reversal asymmetry should be observed in AMR. However, there are a number of potential explanations for the observed breaking of field reversal symmetry in $R_\perp(H)$ at low $T$, arising from AFM/FM interfacial exchange; two of these we outline below.

First, previous studies have demonstrated that, along with the known unidirectional anisotropy associated with AFM–FM interfacial exchange, other higher-order anisotropies may also be induced in the FM. Primary among the effects of these higher-order anisotropies is the onset of asymmetric magnetization reversal with respect to the applied field direction. For example, in FeF$_2$/Fe and MnF$_2$/Fe bilayers, magnetization reversal has been found to occur via coherent rotation when $H$ is applied antiparallel to the unidirectional anisotropy whereas, when $H$ is parallel, reversal proceeds by domain wall nucleation and propagation [27, 28]. This asymmetric reversal can be more difficult to observe in thin film magnetometry, but is readily apparent in MR

In this case, the origin of the asymmetric AMR curve is attributed to an induced three-fold anisotropy [29] in the polycrystalline ferromagnet. In essence, when interfacial exchange coupling is combined with pre-existing even anisotropies within the FM, odd anisotropies can be generated, with the first order term producing the well-known unidirectional anisotropy. A three-fold anisotropy also arises however, which has been shown to directly induce reversal asymmetry [29]. It is therefore conceivable that such an induced anisotropy from the AFM–FM interface could alter the reversal mechanism with respect to the applied field direction of the permalloy arrays considered here, e.g., by altering the domain wall vertex-depinning symmetry, thereby breaking the field reversal symmetry of the AMR curve.

Alternatively, if PHE is measured in $R_\perp$ (which is sensitive to not only the angle, but the direction of the magnetization with respect to the current flow), reversal symmetry in $R_\perp$ may be broken by exchange bias. In this case, any induced unidirectional anisotropy (via FM/AFM interfacial exchange) may force magnetization reversal to occur through the same reversal path for both applied field directions i.e., giving the same approximate field-current angle for a given magnetic field, regardless of history. This in turn would produce a PHE measurement that is no longer field-reversal symmetric, instead giving an identical curve for fields applied in either the positive or negative directions. In this case, any AMR effects (such as those measured in $R_\parallel$) would remain field-reversal symmetric: any magnetization–current angle $\phi$ provides the same AMR signal as $-\phi$.

The combination of the magnetization hysteresis loops and the MR data demonstrate that exchange bias effects appear below 20 K and substantially affect the reversal of the network. This correlation is further supported by the suppression of the asymmetry by the addition of an aluminum capping layer that reduces the surface area of permalloy exposed to air. It is worth noting that the exchange bias blocking $T$ for Co/CoO heterostructures is strongly dependent on the thickness of the CoO [30], but is likely to be significantly higher than the 50 K value at which recent results using Co artificial spin ice networks begin to reveal asymmetry attributed to a chiral ice-like state [17]. It is therefore unlikely that such asymmetries can be attributed to an exchange bias blocking temperature, even if Co structures are uncapped, and CoO forms on the surface/edges. Our observations nevertheless highlight the care that must be taken in interpreting MR data in connected artificial spin ice networks, given the similarity of the effects observed, and the complex nature of the PHE.

Our work demonstrates that exchange bias can have a significant effect on artificial spin ice; hitherto, artificial spin ice studies have focused almost exclusively on magnetic frustration. By incorporating exchange bias into artificial spin ice, this work opens possibilities of new ways to manipulate the behavior of these model systems. Future artificial spin ice transport studies could incorporate exchange bias to probe the effects of geometric frustration.

The authors acknowledge useful discussions with Dan Ralph, Paul Lammert, Vin Crespi, and Jarrett Moyer. This project was funded by the US Department of Energy, Office of Basic Energy Sciences, Materials Sciences and Engineering Division under grant no. DE-SC0010778. This work was carried out in part in the Frederick Seitz Materials Research Laboratory Central Research Facilities, University of Illinois at Urbana–Champaign. Work at the University of Minnesota was supported by the NSF MRSEC under award DMR-0819885 and a.
Marie Curie International Outgoing Fellowship within the 7th European Community Framework Programme (project no. 299376).

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