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Magnetization switching and inverted hysteresis in perpendicular antiferromagnetic superlattices

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The magnetization of antiferromagnetic (AFM) superlattices as a function of applied field was investigated using Monte Carlo simulations. The simulated hysteresis loops of systems with N magnetic layers with AFM coupling between the layers exhibit distinct steps with magnetization that decreases with increasing N. Systems with odd N exhibit 3 steps and inverted hysteresis for $N > 3$, whereas systems with even N exhibit 4 steps, for $N > 2$, and their microscopic switching sequence is non-deterministic and can take two distinct pathways, even though the switching of the global magnetization is exactly reversible. © *2014 AIP Publishing LLC*.
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Magnetic superlattice structures are attractive for the development of meta-materials with unique properties. In antiferromagnetic (AFM) multilayer structures,[1,2] where ferromagnetic (FM) layers are coupled antiferromagnetically and the strength of the inter-layer interaction is on the order of the Zeeman energy,[3,4] meta-magnetic transitions can be induced by moderate magnetic fields. In such systems, the properties can be easily tuned by modifying the inter-layer interactions.

AFM materials are increasingly important in magnetic devices.[5,6] The absence of a strong intrinsic magnetization makes them however difficult to study experimentally. Very large external fields are necessary to induce spin-flop transitions in intrinsic AFM, because the field has to overcome strong exchange interactions, which in AFM with high ordering temperature like CoO and NiO correspond to tens of Tesla. Artificial AFM systems, however, have been fabricated using FM building blocks (Fe[2,7] or Co/Pt multilayers[8,9]). As an example, experimental investigations of [(Co/Pt)/Co/Ru] multilayers have shown that the hysteresis loops exhibit distinct steps, due to switching of individual FM layers.[8,9] These meta-magnetic transitions occur at a critical field H_{cr}, which depends on the interlayer exchange interaction and layer thickness.[10] Meta-magnetic transitions in simple systems with homogeneous exchange interactions are well understood,[11,12] but the hysteretic properties of individual FM building blocks as a function of field in artificial AFM, remain unexplored.

One of the most crucial aspects in the applications of magnetic superlattices is the control and understanding of the global and the local behavior of the system, i.e., the overall magnetization vs. the magnetization of individual FM layers. In this work, we focus on the switching mechanisms of artificial AFM superstructures, and show that the magnetization exhibits metamagnetic behavior with a series of sharp steps, and that in systems with odd number of layers, inverted hysteresis phenomena occur. Moreover, while the global hysteretic behavior is fully predictable, the switching pattern of individual building blocks is non-deterministic with two different characters. These findings show how we can create materials with tailored magnetic properties, key to developing technologies, such as spin-valves, where local behavior plays a central role.

We have simulated perpendicularly magnetized AFM superlattices, using the Monte Carlo method with classical Ising spins. We chose the Ising model, where spins take the values $S = \pm 1$ because it is relevant to the strong perpendicular anisotropy Co-Pt-based structures ($\approx 10^7$ erg/cm^3). Moreover, perpendicular anisotropy is particularly pronounced in ultrathin films,[13–15] which favor a laterally correlated spin structure[8,9,16] due to the competition between anisotropy, dipolar, and exchange interactions,[17] resulting in a single-domain-like state. Therefore, we set the thickness of the FM layers in our systems at 1 atomic plane and focus on single-domain systems.

The structure of the system consists of N identical FM layers, separated by a non-magnetic spacer. Each layer consists of an $L \times L$ square lattice with one spin at each lattice site, where nearest-neighbors interact via a positive exchange coupling J, and the FM layers are AFM coupled. Inter-layer interactions are implemented using a mean-field approach, where each spin in layer n feels the mean magnetic moment m of layers $n-1$ and $n+1$, i.e., m_{n-1} and m_{n+1}, via an inter-layer exchange coupling J_{iec}. We have also implemented the inter-layer exchange coupling as direct spin-spin interactions between nearest neighbors in adjacent layers and have found that the results are identical for the systems we investigated.

Given the above, the Hamiltonian of the system reads

$$\mathcal{H} = \sum_{n=1}^{N} \left(-\frac{1}{2} \sum_{i \neq j} J S_{n,i} S_{n,j} - H_{\mathrm{eff}} \sum_{i} S_{n,i} \right), \qquad (1)$$

where H_{eff} is the effective field, consisting of the external field H, and the inter-layer exchange field $J_{\mathrm{iec}}(m_{n-1} + m_{n+1})$. We express the external field in units of J_{iec}, and set $J = 1$, which is responsible for the long-range order in each FM layer and the Curie temperature T_C.

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The magnetization as a function of H was calculated using the Metropolis algorithm, i.e., by performing single-site updates. $1000 \times N \times L \times L$ Monte Carlo steps were run to allow the system to come to quasi-equilibrium, and additional 10^6 steps were run to obtain averages of the net magnetic moment M, as well as the magnetic moment of each FM layer m. Systems with lateral size $L = 100$, and occasional checks for $L = 250$, were simulated and periodic boundary conditions were used in the x and y dimensions to ensure that our results are not biased by finite system size. H was swept between $-3 \leq H/J_{\rm iec} \leq 3$ with a step of $\delta H/J_{\rm iec} = \pm 0.02$, at $T \approx 0.75 T_{\rm C}$. We chose to simulate at high T to allow the magnetic moments to fluctuate and avoid trapping the system in local energy minima.

Fig. 1 shows M as a function of H, where the direction of the field-sweep is illustrated by different symbols and arrows (red diamonds for decreasing field and blue circles for increasing field). For $N = 2$, the system exhibits two steps, which correspond to a switching from an antiparallel to a parallel configuration of the two FM layers. When H exceeds $J_{\rm iec}$, the two layers are parallel to each other and to the field; and when $H < J_{\rm iec}$, the two FM layers are antiparallel. The steps are hysteretic, with a coercive field of $H_{\rm C}$, which depends on J and T; as $H_{\rm C} \to 0$ the steps would occur at precisely $J_{\rm iec}$. We define $J_{\rm iec} \pm H_{\rm C} = H^{\pm}_{\rm cr1}$ and $2J_{\rm iec} \pm H_{\rm C} = H^{\pm}_{\rm 2cr}$. Note that in real materials the coercivity is equal to, or less than, the anisotropy field $(2K/M)$ of each FM layer (see Brown's paradox[18]).

For $N = 3$, the $M(H)$ loop is drastically changed due to the one uncompensated layer. At high-field, M is saturated, and switches to the AFM state with decreasing H at $H^{-}_{\rm cr1}$, with remanent magnetic moment $M = M_{\rm S}/3$, i.e., corresponding to that of the uncompensated layer ($M_{\rm S} = N\, m_{\rm S}$ is the saturation magnetic moment of the entire system, with $m_{\rm S}$ the saturation moment of a single layer). As the field is further decreased, the system jumps to $M = -M_{\rm S}/3$ at $H = -H^{+}_{\rm cr1}$, and then to $M = -M_{\rm S}$ at $-H^{+}_{\rm cr2}$. When the field-sweep is reversed, the system undergoes the same transitions, from $-M_{\rm S}/3$ to $+M_{\rm S}/3$, and then to $+M_{\rm S}$. The minor loops are centered around $2J_{\rm iec}$ and correspond to switching of the central FM layer, which interacts with the two outer layers, and therefore needs twice as much energy to switch.

Note that at finite T the $m_{1,3}$ of the outer layers is slightly smaller than that of the central layer because they only interact with one FM layer.[19] The simulations show that for $N = 3$ the outer layers at $H = 0$ have $m_{1,3} = 0.98\, m_{\rm S}$, whereas for the central layer $m_2 = -0.99\, m_{\rm S}$. Hence, the net magnetic moment is also reduced $(M = 2m_{1,3} + m_2 = 0.97 m_{\rm S} = 0.97 M_{\rm S}/3)$. Since this effect is small, however, we will not consider it for the rest of the discussion.

When $N = 4$, the system exhibits four steps in the $M(H)$ loop, centered around $\pm J_{\rm iec}$ and $\pm 2J_{\rm iec}$. The switching of the individual FM layers is shown in Fig. 2. At high field, the system is saturated ($M = M_{\rm S}$) and all FM layers are parallel to each other and to the external field. With decreasing field H, at $H^{-}_{\rm cr2}$ either layer 2 or 3 switches to the opposite direction, bringing the system to $M = M_{\rm S}/2$, and then at $H = H^{-}_{\rm cr1}$ another layer (either 4 or 1, depending on whether 2 or 3 switched previously) switches, so that the system is at $M = 0$. When the field is decreased further, the oppositely directed

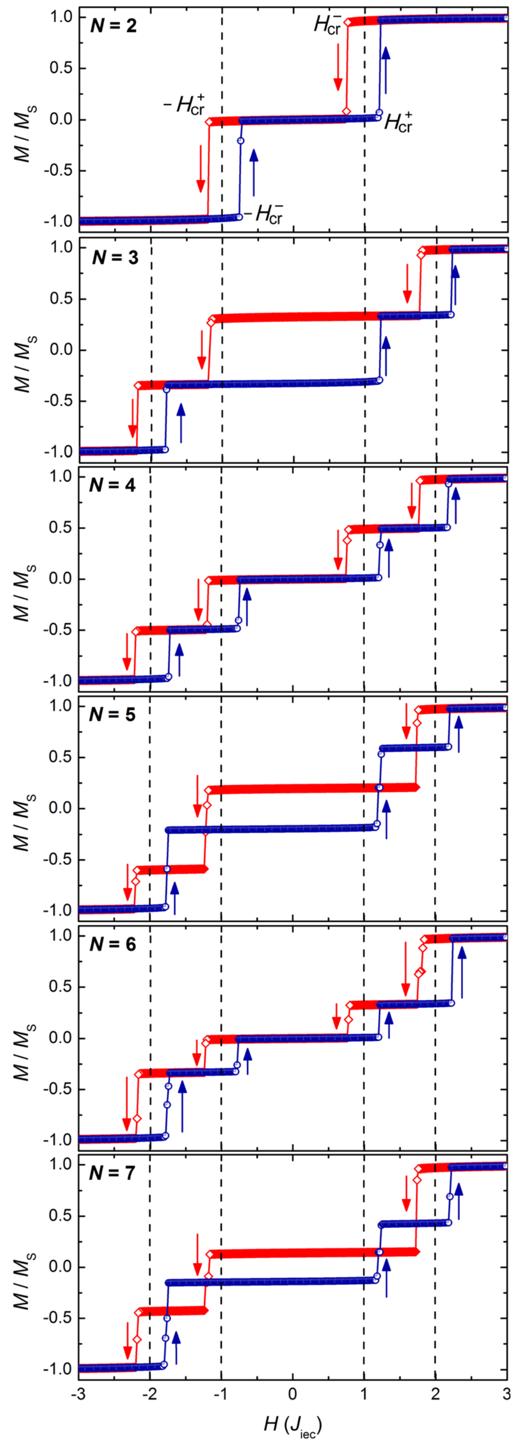

FIG. 1. Net magnetization of AFM superlattices with N FM layers as a function of H. Red diamonds correspond to decreasing field, blue circles to increasing field, also shown by blue and red arrows at each jump in M.

outer layer (either 4 or 1, depending on above sequence) will turn to the direction of the field at $H = -H^{+}_{\rm cr1}$, because it is more susceptible to the external field, having only one interacting layer, and at $H^{+}_{\rm cr2}$ the remaining oppositely directed inner layer will switch to the field direction, bringing the system to $M = -M_{\rm S}$.

During this process, the even-N system thus follows either a sequence of (i) 2–4–1–3 or the reverse (ii) 3–1–4–2. The $M(H)$ of the system does not depend on the sequence of layer switching, but the $m(H)$ of the individual layers changes drastically. If for both decreasing and increasing

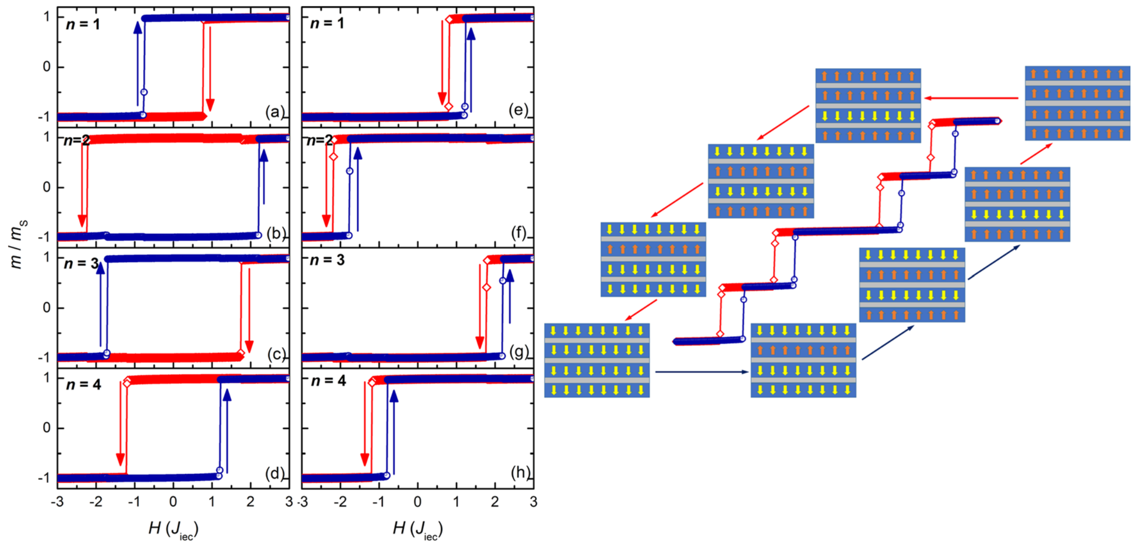

FIG. 2. $m(H)$ of individual layers $n = 1$–4 for $N = 4$ (left); (a) through (d) is one possible sequence of switching (3–1–4–2 on decreasing field, also 3–1–4–2 on increasing field); (e) through (h) another (3–1–4–2 on decreasing field, 2–4–1–3 on increasing field). Right shows the net $M(H)$ and illustration of possible switching process for each layer on going from $M = M_S$ to $M = -M_S$ and back (right). The system can follow two switching sequences: (i) layers 2–4–1–3 (shown in lower figures, increasing field) or (ii) layers 3–1–4–2 (shown in upper figures, decreasing field). When the same sequence occurs with both increasing and decreasing field the hysteresis of the FM layers corresponds to (a)–(d), but when the sequence changes the hysteresis corresponds to (e)–(h). The net $M(H)$ is the same independent of sequence.

field the system switches via (i), then the $m(H)$ of the FM layers is that shown in Figs. 2(a)–2(d), where all loops are centrosymmetric with different coercivities, and half of them have negative hysteresis. If, however, the system follows sequence (i) with decreasing field and then sequence (ii) with increasing field, then the $m(H)$ of the individual layers are those shown in Figs. 2(e)–2(h), where each hysteresis has the same coercivity and is shifted along the field axis, similar to exchange-biased systems.[20,21]

The total energy of the hysteresis loops shown in 2(a)–2(d) and 2(e)–2(h) is exactly the same, because loops in Figs. 2(a) and 2(c) are inverted, and therefore the two modes are energetically equivalent. In the simulations, both switching modes were observed with the same probability, due to the fact that the system has no memory of the specific sequence followed from $M = M_S$ to $-M_S$, and can therefore follow either (i) or (ii) on going from $-M_S$ to M_S.

These two switching processes are the same for any compensated system with $N \geq 4$. A system with $N = 6$ will follow the sequence (2,4)–6–1–(3,5) or the reverse. A system with $N = 8$ will follow (2,4,6)–8–1–(3,5,7) or the reverse. This occurs because the two outer-layers switch at a lower field than all the rest and will remain parallel to the external field, while the switching of the inner layers occurs at H^-_{cr2}, where the system reaches $M = 2M_S/N$.

This finding may be tested experimentally through depth-resolved measurements of the magnetization, such as in magneto-optical Kerr-effect measurements.[9,13] Note that the $M(H)$ for $N = 4$ shown here is nearly identical to the experimental hysteresis of [(Co/Pt)/Co/Ru] multilayers with 4 FM slabs shown in Refs. 8 and 9. In those experimental $M(H)$ loops both local switching modes were observed, but for different samples, which suggests that in real systems, the equiprobability is broken due to defects, which act as pinning centers that influence the magnetization reversal sequence.

The switching sequence can also be affected by an additional field, for example, by biasing the AFM structure with an adjacent FM film. When a local field is acting on one layer of the AFM superlattice, then the second-to-surface layer of the AFM will be the last to switch. When the external field is reversed that layer will be the first to switch back to the other direction. When, however, the local field is reversed, then the second-to-surface layer will be the last to switch. The switching sequence therefore becomes deterministic.

The hysteretic features of even-N systems differ strongly from that of odd-N systems (see Fig. 1), because of the uncompensated layer. Odd-N systems possess a symmetry around the central FM layer, which drastically alters the microscopic switching sequence. A surprising phenomenon occurs for systems with $N \geq 5$, which exhibit inverted steps centered around $\pm 1.5\, J_{iec}$ (see Fig. 1). In Fig. 3, we show the hysteresis of each FM layer for $N = 5$, and a graphical description of the switching sequence in the superlattice.

Starting from the saturated state, $M = M_S$, all layers are parallel to each other and to H. As we decrease H, the second--to-surface layers (2 and 4), switch to the opposite direction and all layers are antiparallel to their adjacent layers. This state has one uncompensated layer, which results in a non-zero net magnetic moment for the entire system ($M = M_S/5$). Then, as the field is decreased further to negative values, the two outer layers switch at $H = -H^+_{cr1}$ and the new state is $M = -3M_S/5$. Then at $H = -H^+_{cr2}$, the central layer also switches and the system reaches $M = -M_S$, where all layers are parallel to each other and to the applied field. Therefore, the switching sequence on going from M_S to $-M_S$ is (2,4)–(1,5)–3.

When the field-sweep direction is reversed, the system follows the same sequence of switching because the central FM layer provides the symmetry of the system. At the first step, at $H = -H^-_{cr2}$, layers 2 and 4 switch and the system reaches the state $M = -M_S/5$, followed by layers 1 and 5 at $H = H^+_{cr1}$ and finally layer 3 at $H = H^+_{cr2}$.

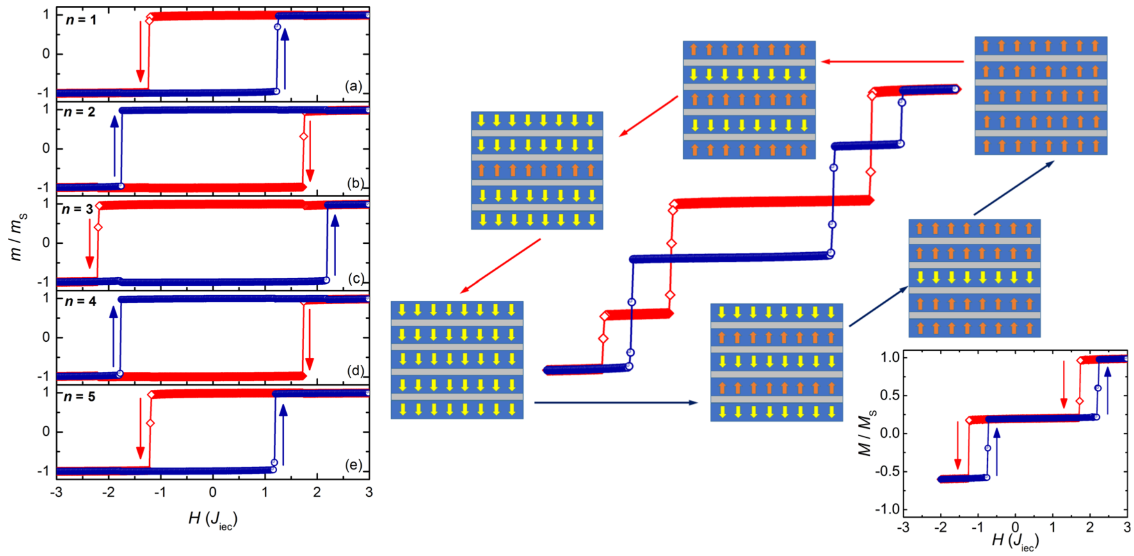

FIG. 3. Magnetization of individual layers for $N = 5$ (left), and illustration of switching process on going from $M = M_S$ to $-M_S$ and back (right). Lower right shows a minor $M(H)$ loop in which the decreasing field-sweep was stopped at $H/J_{iec} = -2$ before the central FM layer switched, and the field sweep was reversed.

We can now explain the occurrence of the inverted $M(H)$ steps in Fig. 1 as the result of the central FM layer, which acts as a pivot for the entire magnetic structure. The inverted hysteresis does not exist if the system does not complete the transition from M_S to $-M_S$. If the field-sweep is reversed before the central layer switches direction, then $M(H)$ only consists of two hysteretic steps, one centered around $-J_{iec}$ and the other around $2J_{iec}$ (Fig. 3). Moreover, this inverted hysteresis only occurs if $H^-_{cr2} - H^+_{cr1} \geq 0$, and therefore $J_{iec} \geq 2H_C$. For a real material, and assuming naively that the coercivity is equal to the anisotropy, the condition for the observation of the inverted hysteresis would be that $J_{iec}M \geq 4K/M$.

Note that in our simulations all of the FM layers were identical to each other, i.e., they had the same m_S. If the layers would have different m_S the main characteristics of the $M(H)$ loops would still be the same, i.e., number of steps and critical field, but the value of M after each step would be different, depending on the contribution of each layer. Moreover, if the layers had different J, the critical fields would be different.

In conclusion, we have found that there are two possible microscopic switching sequences in AFM superlattices with even N, which are not reflected in the measurement of a hysteresis loop of the global magnetization, but which strongly influence the $m(H)$ of individual layers. The two sequences are energetically identical and occur arbitrarily when the system is unbiased. By creating a bias field, however, one can choose which switching mechanism to invoke, thus having absolute control over the global and local magnetization in the superlattice. In superlattices with odd N, we found inverted steps which emerge due to the symmetry of the system and the reversal process. These findings show how to design and fabricate superlattice systems with specific predetermined hysteretic properties and magnetization values, which are vital for technological applications, where a strict control over the magnetization reversal process is required.

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[1]D. Schwenk, F. Fishman, and F. Schwabl, Phys. Rev. B **38**, 11618 (1988).
[2]M. N. Baibich, J. M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Etienne, G. Creuzet, A. Friederich, and J. Chazelas, Phys. Rev. Lett. **61**, 2472 (1988).
[3]S. S. P. Parkin, N. Moore, and K. P. Roche, Phys. Rev. Lett. **64**, 2304 (1990).
[4]M. T. Johnson, S. T. Purcell, N. W. E. McGee, R. Coehoorn, J. aan de Stegge, and W. Hoving, Phys. Rev. Lett. **68**, 2688 (1992).
[5]S. S. P. Parkin, K. P. Roche, M. G. Samant, P. M. Rice, R. B. Beyers, R. E. Scheuerlein, E. J. O'Sullivan, S. L. Brown, J. Bucchigano, D. W. Abraham, *et al.*, J. Appl. Phys. **85**, 5828 (1999).
[6]S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnár, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, Science **294**, 1488 (2001).
[7]R. W. Wang, D. L. Mills, E. E. Fullerton, J. E. Mattson, and S. D. Bader, Phys. Rev. Lett. **72**, 920 (1994).
[8]O. Hellwig, T. L. Kirk, J. B. Kortright, A. Berger, and E. E. Fullerton, Nature Mater. **2**, 112 (2003).
[9]O. Hellwig, A. Berger, J. B. Kortright, and E. E. Fullerton, J. Magn. Magn. Mater. **319**, 13 (2007).
[10]J. Li and D. P. Landau, J. Appl. Phys. **69**, 6158 (1991).
[11]E. Stryjewski and N. Giordano, Adv. Phys. **26**, 487 (1977).
[12]L. L. Hinchey and D. L. Mills, J. Appl. Phys. **57**, 3687 (1985); Phys. Rev. B **33**, 3329 (1986).
[13]P. F. Carcia, A. D. Meinhaldt, and A. Suna, Appl. Phys. Lett. **47**, 178 (1985).
[14]C. Chappert, H. Bernas, J. Ferré, V. Kottler, J.-P. Jamet, Y. Chen, E. Cambril, T. Devolder, F. Rousseaux, V. Mathet, and H. Launois, Science **280**, 1919 (1998).
[15]J. E. Davies, O. Hellwig, E. E. Fullerton, G. Denbeaux, J. B. Kortright, and K. Liu, Phys. Rev. B **70**, 224434 (2004).
[16]O. Hellwig, A. Berger, and E. E. Fullerton, Phys. Rev. Lett. **91**, 197203 (2003).
[17]C. Kittel, Phys. Rev. **70**, 965 (1946); Rev. Mod. Phys. **21**, 541–583 (1949).
[18]W. F. Brown, Rev. Mod. Phys. **17**, 15 (1945).
[19]M. Charilaou and F. Hellman, Phys. Rev. B **87**, 184433 (2013).
[20]J. Nogués and I. K. Schuller, J. Magn. Magn. Mater. **192**, 203 (1999).
[21]A. E. Berkowitz and K. Takano, J. Magn. Magn. Mater. **200**, 552 (1999).