Switching behaviour of coupled antiferro- and ferromagnetic systems: exchange bias

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Abstract

The switching behaviour, under reversal of an external field, of a simple, ideal magnetic nanoparticle is studied and the interplay between antiferromagnets and ferromagnets elucidated. It is found that the switching between various multi-**q** ordering in fcc antiferromagnets (as found theoretically in NiO nanoparticles (Kodama and Berkowitz 1999 *Phys. Rev.* B **59** 6321 and Lindgård 2003 *J. Magn. Magn. Mater.* **266** 88)) in a field severely limits the exchange biasing potential. The interface between the different magnets is found to be that originally assumed by Meiklejohn and Bean (1956 *Phys. Rev.* **102** 1413).

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Magnetic nanoparticles are supposed to be too small to have internal domain walls and multidomains. Does this mean that the internal structure is co-linear? In particular, is it co-linear during the process of a flip of the total spin of the particle? The answer to both questions is No!

Conventionally, the flipping of the spin of a nanoparticle under a field reversal has been described by considering the behaviour of the total spin. That is, the sum of all spins, assumed collinear, for a ferromagnet [4, 5] and similarly for each sublattice (two) for an antiferromagnet [6]. We shall here test this assumption by Monte Carlo simulations of the switching behaviour of a spherical nanoparticle.

The flipping of an antiferromagnet (AFM) in atomic proximity to a ferromagnet (FM) is a puzzling problem, giving rise to the exchange bias effects (a violation of the time-inversion symmetry of the magnetization: $M(H) = -M(-H)$ under reversal of the magnetic field, H). The effect arises because an external magnetic field interacts much weaker on the antiferromagnet than the ferromagnet, causing the former to be more resistant to flipping. However, the effect is found to be smaller than expected by various theoretical models.

To elucidate the problem it is of interest to eliminate a possible domain wall mechanism [7] by studying the effect of the contact between two hemispheres of an AFM and an FM

nanoparticle (too small to hold a static domain wall). Because of the finite size, the AFM particles will have net moment due to the uncompensated spins S_{tot} in a number, N_U, highly sensitive to the shape and size of the particles. This will, of course, attempt to align with an external field. Although the model is highly idealized, similar conditions might be obtained in a mixture of AFM and FM particles in partly oxidized metallic FM particles.

2. Model: spherical antiferromagnetic or semi-spherical ferro- and antiferromagnetic nanoparticles combined

Let us study a model, which is relevant for NiO, either alone or interacting with an fcc ferromagnet. The Hamiltonian for NiO includes a Heisenberg exchange interaction, J_{ij}, and a local uniaxial anisotropy field D_{ax} in the y direction and a planar anisotropy, D_p, keeping the spins preferentially in the x–y plane:

$$\boldsymbol{H} = -\Sigma_{ij} J_{ij}\mathbf{s}_i.\mathbf{s}_j - \Sigma_i D_{ax,i} s_i^{y2} - \Sigma_i D_{p,i} s_i^{z2} - H \Sigma_i s_i^{y}.$$

For the model ferromagnet we assume a Hamiltonian of the same form, using only a single, strong nearest-neighbour ferromagnetic interaction. The coupling between the two hemispheres is also assumed to be between nearest neighbours. The spin-flip behaviour has been studied using the Monte Carlo

(MC) technique. This also allows the investigation of more complicated systems (for example, the combined FM/AFM hemispheres) than those amenable by analytical theories.

3. Relaxation behaviour for various models

For an Ising model, corresponding essentially to having D_{ax} very large, the flipping of the moment of a ferromagnetic particle is a nucleation phenomenon, which starts at the point of the weakest interaction (or molecular field) acting on a spin. This will typically be at the surface and in particular at corners or defects. By Monte Carlo simulations, we have found [8] that the switching field, $H_s(t)$, at which the magnetization on average switches after a certain waiting time, t, has a maximum as a function of the total number of spins, N. This is an entropy effect showing that for relatively large N there are more places to start the nucleation. This is a mechanism different from the usual domain wall argument.

For Heisenberg systems we expect other more soft switching modes. The standard model is the uniform switching. However, calculations of the internal spin wave modes [9] show that a twisting mode with large surface spin-deviation amplitudes is a close candidate. The uniform mode picture is confirmed as a quantum phenomenon in the very small molecular magnets as $Mn_{12}Ac$ [10]. Nature seems to know about the various excitation states. However, it is hard to imagine how the small spins actually behave during the flip process. For this reason we shall perform (classical) spin-flip studies using the Monte Carlo technique.

4. The Monte Carlo method

The Monte Carlo technique is basically simply a calculation of the energy determined by the Hamiltonian of various configurations of the system. The configurations are found by rotating one randomly selected spin after the other in the cluster by a small angle, and testing the resulting energy change ΔE. The move is accepted if ΔE is less than zero, but accepted, with the probability $\exp(-\Delta E/kT)$, if it is larger than zero; k is the Boltzmann constant. Hence, at finite temperatures, T, the system can escape from being trapped in a local energy minimum. More efficient updating schemes have been devised for a fast approach to the ground state. However, for the transient phenomena of the total moment change of a cluster under field reversal a physically transparent updating is necessary. The time is measured in Monte Carlo steps per spin (MCSS). To avoid nucleation sites a spherical system is optimal. The above single-spin method does give the correct set of configurations for a possible switching path of the moment. However, it is highly unlikely that the method finds the optimal path (i.e. across the minimum energy barrier—in our case the uniform mode rotating the total moment in the basal plane) because that is just one out of infinitely many other paths. Hence, the Monte Carlo updating (for the combined AFM/FM hemispheres) was done as a two-step process as follows: (a) allow the system to seek the best relaxed configuration by single-spin updates for 50 MCSS and (b) calculate the total spin vector, $\mathbf{S}_{tot}$, and propose a

small uniform rotation, which is proportional to the torque exerted on the AFM and the FM part—and a Monte Carlo test was performed. If the move is accepted all the individual spins are rotated by the same amount and the procedure goes back to (a). This speeds up the flipping procedure and gives a preferential set of paths around the uniform modes, but including excursions dictated by thermal fluctuations and hence showing configurations corresponding to internal mode excitations.

5. The switching path of an fcc antiferromagnetic NiO cluster

Let us here review the results of MC simulation for a pure NiO sphere. Kodoma and Berkowitz [1] have found (by a rather different method) that the ground state of an NiO cluster is not the simple 1-$\mathbf{q}$ structure, with $\mathbf{q} = \langle 111 \rangle$, as expected for the bulk, but rather a multi-$\mathbf{q}$ structure. Using the MC method we [2] have tested and confirmed this for a slightly simpler model with respect to the interactions. For an $N = 381$ spin NiO cluster, using $J_1 = -15.9$ K, $J_2 = 221$ K, $D_p = 50$ K and $D_{ax} = 0.1$ K, we find the minimum energy structure as a 4-$\mathbf{q}$ structure (8 sublattices) having equal populations of $\mathbf{q} = \langle 111 \rangle_x, \langle 111 \rangle_y, \langle \bar{1}11 \rangle_x$ and $\langle \bar{1}11 \rangle_y$, with the moments in the directions indicated by subscripts. This structure is also found for other values of N up to at least $N = 5979$. It gives in real space antiferromagnetic rows (in the [111] plane $\perp z$ direction along $\langle 111 \rangle$) of spins pointing in the x and y directions in adjacent rows, respectively (see a small excerpt in figure 1, left). Let us call the structure $S_0(4q)$. The basal plane structure is identical to that of [1]. Further, we found that it has an energy advantage over the 1-$\mathbf{q}$ structure, figure 1, right, of only -0.27 K/spin, and that there are several other multi-$\mathbf{q}$ structures (2, 3 and 4 with other polarizations) of intermediate energies. Interestingly, although the real-space configurations are very different patterns of either co-linear or canted spins, the excess moment of all investigated structures (including the 1-$\mathbf{q}$) is the same, namely $N_U = 18$ spins, when the cluster is spherically cut around a central spin (different cuts may give different excess spin values). These almost-degenerate states play a role during the switching process. Figure 2, left, shows how the $S_0(4q)$, which has the excess moment in the $-y$ direction, switches at $T = 30$ K when subject to a field $H = 10$ T in the $+y$ direction. First (at $t \sim 70t_0$, $t_0 = 500$ MCSS) a rather pure $\langle \bar{1}11 \rangle_x$ state is formed (heavy indigo line) and this component stays with some fluctuations for the time shown. However, up to the switching time $t \sim 200t_0$, where S_{tot}^y passes through 0, the other x components (thin greenish lines) grow systematically and a new 4-$\mathbf{q}$ state is formed with about equal populations of all x components: $\langle 111 \rangle_x, \langle 1\bar{1}1 \rangle_x$, $\langle 1\bar{1}1 \rangle_x$ and $\langle \bar{1}11 \rangle_x$. At the same time a new component $\langle 1\bar{1}1 \rangle_y$, indicated by the heavy orange line, is rapidly growing up, while the x components decrease, and the system finally forms a 2-$\mathbf{q}$ state with $\langle \bar{1}11 \rangle_x$ and $\langle 1\bar{1}1 \rangle_y$ at $t \sim 400t_0$—with small populations of all the others. However, that is not the ground state. It fades away, as shown in figure 2, right, and is systematically taken over for a time period of a rather pure 1-$\mathbf{q}$ y-component structure: $2/3\langle \bar{1}11 \rangle_y + 1/3\langle 1\bar{1}1 \rangle_x$ peaking at
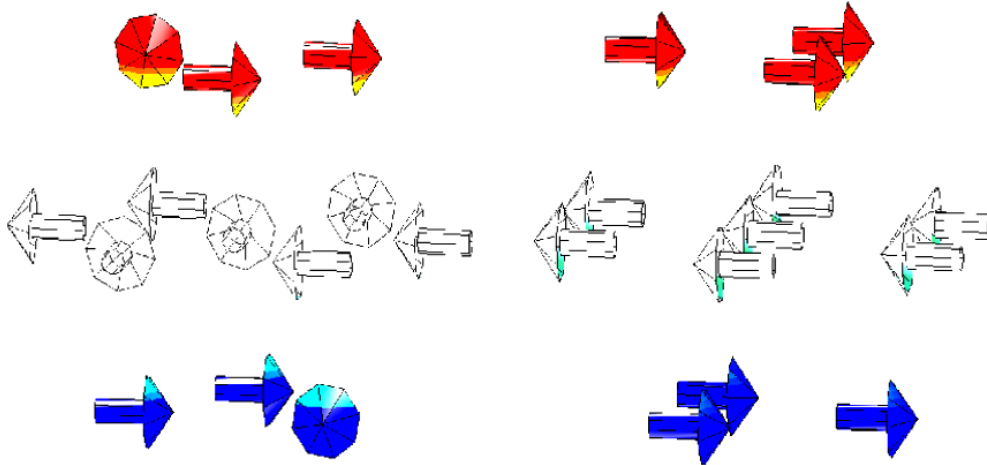
Figure 1. The ground state of an NiO cluster. A very small spherical excerpt of an $N = 381$ fcc spin cluster is shown. The $\langle 111 \rangle$ axis (z) is vertical and the view is almost along the y direction. The simple 1-**q** structure, with alternating co-linear planes, is shown to the right. The energetically more favourable 4-**q** structure, called $S_0(4q)$, left, consists of alternating rows of perpendicular spins in the [111] plane.
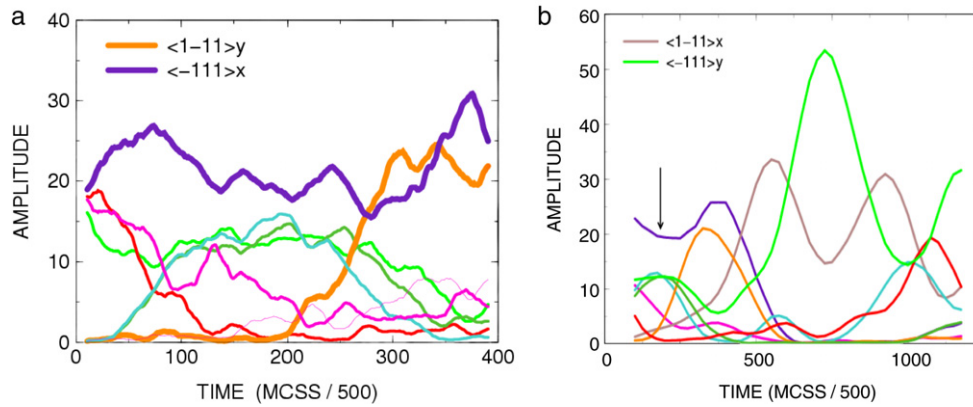
Figure 2. Switching behaviour of an NiO cluster after application of an external field opposite to the excess moment direction, as revealed by the MC simulation at $T = 30$ K. The time development of the AFM Fourier components is shown. The four components $\langle \cdots \rangle_x$ with x polarization are greenish/bluish; the $\langle \cdots \rangle_y$ components are reddish. Details during the switching time interval are shown to the left. At $t = 0$ the structure consists of equal populations of $\langle 111 \rangle_x$, $\langle 111 \rangle_y$, $\langle \bar{1}11 \rangle_x$, and $\langle \bar{1}11 \rangle_y$. The $\langle \bar{1}11 \rangle_x$ component (heavy indigo line) shows an almost constant amplitude in the shown time interval. The switching occurs at $t \sim 200t_0$, where $t_0 = 500$ MCSS. There the structure consists of about equal populations of the equivalent **q** vectors and x polarization. At full reversal (at $t \sim 400t_0$) a 2-**q** structure formed by $\langle \bar{1}11 \rangle_x$ and $\langle 1\bar{1}1 \rangle_y$ (heavy orange line) is found. This, however, does not remain stable at longer times and alternative combinations pop up. This is shown in the right panel, which covers a longer time span with running-time-averaged data. The arrow indicates the switching time.

$t \sim 750t_0$. This again gives way to a 4-**q** $xxyy$ structure at $t \sim 1100t_0$, and so on. The conclusion is that at $T = 30$ K and $H = 10$ T the ground state is degenerate between a large number of multi-**q** states. A 4-**q** structure is described in real space by up to eight sublattices and with moments pointing in seemingly all directions. Clearly, this complicates a calculation of the internal excitation spectrum, as well as opening up possibilities for spin-flip patterns involving transitions between several multi-**q** structures. It should be mentioned that even bulk fcc antiferromagnets show complicated multi-**q** structures in a field [11]. It is interesting that the uncompensated moment (for $N = 381$) in the y direction for all these multi-**q** states is the same: around $S_{\mathrm{tot}}^y = 18$. In addition there is a fluctuating uncompensated moment, S_{tot}^x, in the x direction. Because of the planar anisotropy the moments are mainly in the plane, but components in the z direction also occur. We conclude that the energy barrier between the various states is less than 30 K.

During the switching process the uncompensated moment rotates (with large fluctuations) in the plane from the $-y$ direction ($t = 0$) to the x ($t \sim 200t_0$) and on to the $+y$ direction ($t \sim 400t_0$), just as expected for the uniform rotation mode. However, the internal structure changes simultaneously dramatically, as we have seen. Whereas the system follows a simple systematic path in reciprocal space (mode-space), the corresponding real-space spin configurations change in a seemingly chaotic way over quite large regions. Some snapshots are shown in [2]. This means one must be very careful using simple, local real-space spin configurations for discussing the flipping process. On the other hand, it is very interesting that the simple, single spin-flip MC method used in the above studies does catch the mode switching, which clearly is the relevant mechanism both for a Heisenberg ferro- and antiferromagnet. At $T = 30$ K the switching involves transitions and shifting population of the eight various uniform

AFM modes. Whereas the internal twisting involving internal modes does occur, it plays a minor role at $T = 30$ K.

For larger fields the $S_0(4q)$ structure passes at the switching time the pure 1-**q** $= \langle 111 \rangle_x$ state, then goes through the 4-**q** $= \langle 111 \rangle_x$, $\langle 1\bar{1}1 \rangle_x$, $\langle 1\bar{1}1 \rangle_y$, $\langle \bar{1}11 \rangle_y$ state, and then directly to the 1-**q** $= \langle \bar{1}11 \rangle_x$ state with a small, equal population of all other states. This AFM structure with spins canted (but being almost perpendicular) to the field is the stable state for $T = 30$ K and large fields (here $H = 100$ T). It is that expected for a bulk AFM in a field. For large fields, the uncompensated moment is not sufficient to maintain the alignment along the field. The switching pattern of the fcc antiferromagnet (at least for a cluster) is thus much more complicated than generally assumed. This might have a bearing on the understanding of the exchange bias, where it is the question of a combined switching of adjacent ferro- and antiferromagnetic layers. To avoid the problem of domain walls let us study the phenomenon in a small cluster, which is too small to have domain walls, but where the multi-**q** possibilities may play a role. In fact the domain wall problem [7] (whether parallel or perpendicular to the interface) is replaced by having a superposition of various equivalent domains.

6. Switching behaviour of an AFM/FM sphere

Consider a spherical fcc cluster, where the upper half plus the central plane consist of an antiferromagnet, NiO, and the lower half is a ferromagnet. The nearest-neighbour interaction between them, J_{int}, may be varied. The interactions in the model ferromagnet are, for simplicity, assumed to be nearest neighbour only and 10 times stronger than in NiO: $J_{\text{ferro}} = 10\, J_1$, in order to have a reasonable transition temperature, $T_c(\text{MF}) = 1270$ K relative to $T_N(\text{MF}) = 760$ K for NiO.

The first question to investigate is the nature of the interface spin structure in this pure and highly simplified system. Could the multi-**q** structures play a role and justify the AFM canted arrangement proposed by Koon [12], or is the simple co-linear structure assumed by Meiklejohn and Bean [3] (MB) correct? The $N = 381$ spin cluster has $N_{\text{AFM}} = 221$ spins and the excess number of spins $N_U = 39$, whereas the ferromagnetic hemisphere has $N_{\text{FM}} = 160$ spins. The number of spins at the interface are 60 AFM and 54 FM, giving a number of interface bonds ~ 150. The anisotropy has a larger effect in the AFM, $-D_{\text{ax}}^{\text{AFM}} N_{\text{AFM}}$, than in the FM, $-D_{\text{ax}}^{\text{FM}} N_{\text{FM}}$, whereas the torque on the AFM in a field is much smaller by a factor $N_U/N_{\text{FM}} = 0.24$. Using the MC simulations of the ferromagnet combined with any of the above-discussed multi-**q** structures for the AFM, the ground state (at low T and $H = 0$) invariably switched to the simple MB picture with the AFM in the single **q** $= \langle 111 \rangle_y$ state with the spins parallel and aligned with the FM at the interface—even with a very small interface interaction, $J_{\text{int}} = 0.1 J_1$. Only at very large fields ($H \sim 100$ T) was it possible to introduce a twist by which the AFM spins far away from the interface assumed the perpendicular (canted) structure. Hence this model system lends no support to the Koon picture, which assumed the antiferromagnetic spins to
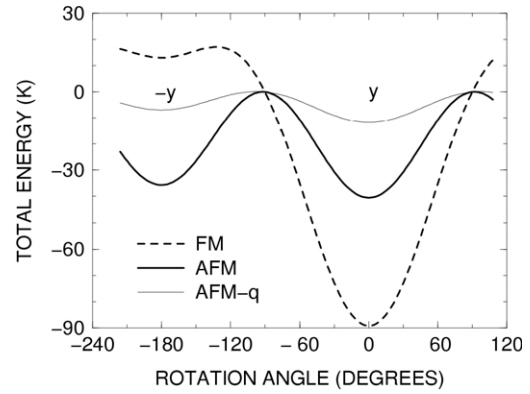
Figure 3. Idealized energy landscape for a hypothetical uniform mode reversal of the FM (dashed) and the AFM (NiO) hemispheres (full line) of an $N = 381$ spin cluster under an applied external field of $H = 0.1$ T. Even assuming the same axial anisotropy, $D_{\text{ax}}^{\text{AFM}} = D_{\text{ax}}^{\text{FM}} = 0.1$ K, the differences between the FM and AFM properties yield very different energy barriers. However, we remark that this picture is not confirmed by the MC simulations. The AFM barrier is much reduced due to multi-**q** structure transformations, shown as the thin line (AFM-q).

be essentially perpendicular to the field and the ferromagnetic ordering.

The next question is: how does the switching occur after reversal of the magnetic field? Since the optimal ordering between the AFM and FM hemispheres is collinear, one may expect that the uniform mode switching would occur, at least at low temperatures. The energy landscape for a uniform turn of the ferro- and antiferromagnetic part is given by $E = -N_i D_{\text{ax}}^i \cos^2(\varphi) + U_i H \cos(\varphi)$, where $i = $ FM or AFM and $D_{\text{ax}}^{\text{AFM}} = D_{\text{ax}}^{\text{FM}} = 0.1$ K. It is shown in figure 3 for $H = 0.1$ T ($g\mu_B s H \sim 1.34$ K for $g = 2$, $s = 1$ and $H = 1$ T). It shows a small energy barrier, $B_{\text{FM}} \sim 2$ K, for the FM, and a much larger, $B_{\text{AFM}} \sim 18$ K, for the AFM hemispheres. Hence, in this picture at low temperatures ($T < 18$ K) a switching of the ferromagnetic part should easily occur without a switching of the AFM part—even in the case of a weak coupling. However, this is not seen in the MC simulations, not even without coupling. The AFM structure dissolves in multi-**q** structures, which facilitates a switching of the AFM part. Figure 4 shows a snapshot during the process for moderate interface couplings $J_{\text{int}} = J_1$. The picture shows the cluster with the $\langle 111 \rangle$ axis being vertical and the ferromagnet (red upper half) has initially been aligned to the left ($-y$ direction); the lowest plane still shows this ordering. The AFM (lower part) was initially in the $\langle 111 \rangle_y$ state with the spins parallel to the FM in the interface; the outermost layers still show that ordering. The temperature is $T = 0.01$ K and a field of 30 T is applied in the left direction ($+y$). After 80 000 MCSS the typical structure shown appears. Evidently the switching starts at the weakest position, namely at the interface plane, and here at the right edge. It is further clear that it occurs as a quasi-static twist mode (a frozen spin wave) extending all over the cluster. The MC technique has thereby demonstrated the low-lying spin wave mode for the combined AFM–FM system, which has large amplitudes at the middle plane both in the FM and the AFM hemispheres. The twist extends into both systems in a similar way (when the
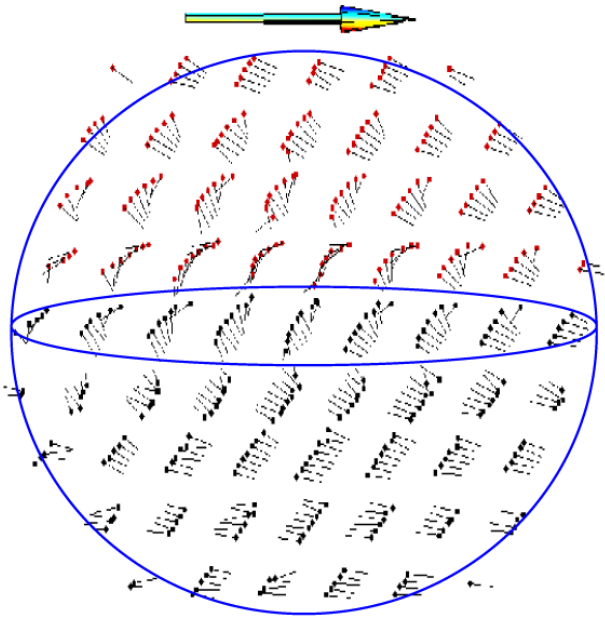
Figure 4. Snapshot of an intermediate step in a reversal process for an $N = 381$ AMF/FM particle. The $\langle 111 \rangle$ direction is vertical and the view is almost along the x direction. The lower five layers are formed by NiO and the upper four layers, red part, by the ferromagnet. This part is initially magnetized along the $-y$ direction (to the left). An external field along the $+y$ direction (large arrow) is applied. The flipping is clearly far from that expected for a uniform mode. The flipping is associated with considerable twisting similar to 'frozen' spin waves. It is initiated at the weakest point, namely at the interface and starting from the surface. Clearly energy considerations taking only small local interface regions into account are not adequate.

AFM alternation of spin direction is ignored). Clearly in this case the switching (i) does not follow a uniform mode rotation and (ii) a local description, considering only a few spins, is inadequate since all spins must be included.

7. Relevance for the exchange bias picture

Magnetization reversal is generally a non-equilibrium phenomenon. In MC simulations the reversal is observed after a certain waiting time, given as a number of MCSS, which is limited by the computer time available. To speed up the switching we have in the following allowed a partial uniform updating using both steps (a) and (b) as described above. In real time the accessible waiting times correspond to very fast reversals and this gives rise to unrealistically large switching fields. However the general behaviour should be similar to that expected for real experimental timescales. In the simple uniform mode picture, which might hold for low temperatures, the total (or excess) spin has to pass from one minimum to another across a barrier B with probability $\exp(-B/kT)$. The energy landscape is shown in figure 3. In the case of strong coupling, $J_{int} = 10J_1$ (this corresponds to a field acting across the interface of over 20 T per spin), both hemispheres switch simultaneously. Hence there is no exchange bias effect. The AFM structure during the flipping process is observed to be twisted along the $\langle 111 \rangle$
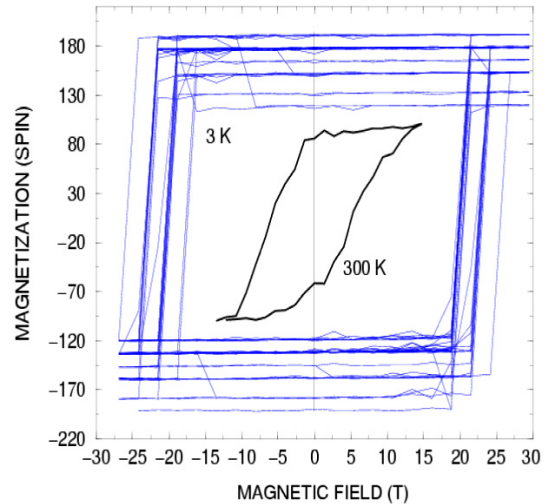
Figure 5. A simulated hysteresis loop at $T = 3$ K and $T = 300$ K for an FM/AFM particle, where the AFM part has a large axial anisotropy, $D_{ax}^{AFM} = 50$ K. The process is started at negative fields and magnetization and then cycled 40 times. At $T = 3$ K all paths are shown separately (thin blue line). A weak exchange bias may be obtained. For $T = 300$ K the average path (black line) is shown for. There is no clear exchange bias.

direction, because the FM is flipping faster. For large temperatures the AFM structure consists of, and fluctuates between, many multi-**q** states. This facilitates the switching. Even for a very large AFM anisotropy, $D_{ax}^{AFM} = 500$ K, $D_{ax}^{FM} = 50$ K and $J_{int} = J_1$ the exchange bias observed in the simulations at $T = 300$ K is very weak, whereas it is clearly present at $T = 30$ and 200 K. This is shown in figure 5, which shows the simulated hysteresis loop for $T = 3$ and 300 K. The system was started with a bias in the $-y$ direction at $H < 0$. H was then gradually cycled to positive and back to negative values 40 times. The average is shown for $T = 300$ K.

8. Conclusion

The ground state of a small, spherical NiO cluster is a 4-**q** multi-**q** state. This is, however, only 0.3 K/spin more stable than the simple 1-**q** state. For a $N = 381$ spin AFM cluster the excess moment due to uncompensated spins is 18 spins for all the nearly degenerate states. These differ substantially with respect to the spin configurations in real space. For moderate external fields the 4-**q** state with the excess moment along the field is the ground state (for large fields the spins turn perpendicular to the field). Under field reversal the AFM structure switches through a multitude of multi-**q** states. For $T = 30$ K and $H = 3$ T the system is degenerate with respect to a number of multi-**q** states. The MC simulation shows slow migrations among such states. During switching the spin system is generally twisted on a large length scale. This invalidates simple local energy arguments.

A spherical model system consisting of a hemisphere of NiO and a hemisphere of a ferromagnet was studied. Here the AFM ground state is at low temperatures the simple 1-**q** state with the moments parallel to the ferromagnet at the interface,

as expected by Meiklejohn and Bean [3]. At low temperatures the switching does not follow the rotating uniform mode model. Although the system has different energy barriers for the FM and AFM hemispheres, the switching occurs simultaneously (even for small couplings). Hence no exchange bias effect is observed when the FM and AFM have similar axial anisotropies, even at low temperatures. At room temperatures a small exchange bias effect is observed when the AFM has a large axial anisotropy. However, it is very much reduced due to fluctuations among the various multi-**q** states, which facilitates the switching of the antiferromagnetic hemisphere. This study was undertaken for the present model system in order to avoid any complications from possible domain switching mechanisms.

We have demonstrated that the multi-**q** nature of the AFM states in small fcc crystals (here spherical) is capable of reducing the expected exchange bias very substantially. We have previously demonstrated [11] stable multi-**q** structures in bulk AFM in a field. Perhaps even bulk NiO orders into a 4-**q** structure, as conjectured by Mukamel and Krinsky [13]. The possibility of having multi-**q** structures may be a contributing reason (and a new model) for the observed, much reduced, exchange bias also in other more realistic systems. If this conjecture is true, a larger exchange bias should be obtainable if antiferromagnets with lower symmetry than that of fcc are used.

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