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On beating the superparamagnetic limit with exchange bias

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Abstract – In order to investigate the possibility of "beating the superparamagnetic limit with exchange bias" (Skumryev V. *et al.*, *Nature*, **423** (2003) 19) we perform atomistic modeling on Co/CoO magnetic nanoparticles varying the strength of the ferromagnet/antiferromagnet interfacial coupling. Our results show that exchange-biased systems exhibit an increased energy barrier along the bias direction, with a corresponding decrease of the barrier in the opposite direction. For systems with large values of the interfacial coupling, the ferromagnetic core is found to be unconditionally stable in the bias direction. Such a system is ideal for thermal stability since there is no energetically stable reversed state, providing the antiferromagnetic shell is unchanged. In order to permit magnetic recording, which essentially requires a bi-stable system, we propose a heat-assisted recording method, whereby both the ferromagnet (FM) and antiferromagnet (AF) are switched. Upon heating to the Néel temperature of CoO, the AF magnetic order is destroyed allowing switching of the FM core with a small applied field, with thermal stability reappearing on cooling through T_N.

It is widely known that future advances in the areal density of magnetic recording devices are essentially limited by the magnetic recording trilemma —namely that a decrease in the bit size requires smaller particles with an increased magnetocrystalline anisotropy for long-term thermal stability. This then causes an increased coercivity of the material which cannot be written to by the limited head field in a device [1]. The conventional resolution to this problem comes from attempting to reduce the effective write field for the material without compromising the long-term thermal stability. Examples of such approaches include exchange-coupled composite (ECC) media [2,3], synthetic antiferromagnet ECC media [4–6], and Heat-Assisted Magnetic Recording (HAMR) [7–9].

However, it is also possible to increase the thermal stability of a material without increasing the coercivity, namely through the use of exchanged bias media. Skumryiev *et al.* [10] recently suggested to "beat the superparamagnetic limit" using the exchange bias phenomenon, demonstrating an increase of the blocking temperature in a system of Co/CoO nanoparticles. Exchange bias gives rise to a displacement of the hysteresis loop due to the coupling of a ferromagnetic layer (FM) to an antiferromagnetic (AFM) one. However, the shift of the loop also increases the switching field and so in itself is of little use for recording applications. In this letter we propose a scheme where exchange bias is combined with HAMR (which enables the writing process) to achieve an increase in thermal stability of a ferromagnetic nanoparticle without increasing the *effective* write field, thus circumventing the magnetic recording trilemma. The scheme is studied using an atomistic model for Co/CoO nanoparticles to investigate the switching and thermal stability properties as a function of the interfacial FM/AFM exchange coupling. We show that for low interfacial FM/AF coupling the exchange bias does increase the magnetization energy barrier. Unfortunately at the same time the energy barrier for the backward reversal of the opposite magnetization state decreases. For larger interfacial exchange the FM state becomes unconditionally stable. In order to achieve a bistable system in this case the AFM must be reversed, thereby changing the bias

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direction. In order to do this the system is heated above the AFM Néel temperature $T_N \ll T_C$ (the FM Curie temperature) which allows the resetting of the AFM such that the FM is stable in the applied field direction.

Our model consists of a Heisenberg Hamiltonian which takes into account the exchange, anisotropy and Zeeman energy terms, given by

$$\mathcal{H} = -\sum_{i,j} J_{ij}\mathbf{S}_i\mathbf{S}_j - \sum_i K_i S_z^2 - \sum_i \mu_i \mathbf{H}\mathbf{S}_i. \qquad (1)$$

Here the first term is the Heisenberg exchange where the summation over the second index j goes over the nearest neighbors only, the exchange parameter $J_{ij} = J_{\mathrm{FM}} = 11.2 \times 10^{-14}$ erg ($T_C = 1390\,\mathrm{K}$) for FM core spins, $J_{ij} = -J_{\mathrm{AFM}} = -4.2 \times 10^{-14}\,\mathrm{erg}$ ($T_N = 400\,\mathrm{K}$) for AFM shell spins, $J_{ij} = J_{\mathrm{int}}$ for the spins at the interface, which is varied as a parameter. The anisotropy values were chosen as those approximately corresponding to Co: $K_i = K_{\mathrm{FM}} = 4.644 \times 10^{-17}$ erg/atom for the core spins and to CoO $K_i = K_{\mathrm{AFM}} = 50 K_{\mathrm{FM}}$ for shell spins. The magnetic moments of the Co and CoO were chosen to be the same $\mu = 1.4\mu_B$. Note that in these calculations, similarly to a commonly used approach (see, for example, [11]), we have neglected the effects of dipolar interactions. In the case of isolated spheroid nanoparticles this is a safe approximation, since there is very little shape anisotropy in the system, and the particle size is below the limit for a single domain Stoner-Wohlfarth particle. The nanoparticles were cut in a spherical shape from a simple cubic lattice with a core diameter of 9 atomic sites ($N_{\mathrm{core}} = 365$ atoms) and a shell two atomic sites thick ($N_{\mathrm{shell}} = 656$ atoms). Since the particles number an odd number of atoms across, this consequently gives rise to an uncompensated interface between the core and shell. Our investigations with differently prepared nanoparticles and interfaces (including a randomly prepared one [12]) showed that the qualitative character of the behavior in these cases is the same as the one presented in the present letter, however, with a large dispersion of the properties. For the system dynamics we used the integration of the Landau-Lifshitz-Gilbert (LLG) equation with Langevin dynamics. First, the hysteresis loops were calculated at 0 K in order to define the intrinsic coercivity and energy barriers in the system. To prepare the nanoparticle in a state with exchange bias we initialized the magnetization state with all magnetic moments in the $S_z = 1$ direction and minimized the magnetic energy in the presence of a large (positive) applied field. As a result, the antiferromagnetic part of the interface had a net interface magnetic moment responsible for the exchange bias.

Figure 1 presents a set of hysteresis cycles for different values of the interfacial exchange parameter J_{int} ranging from 0.3% to 5% of J_{FM}. The inset of this figure shows the behavior of the coercive, switching and bias fields as a function of the interfacial exchange. The coercive field is understood to be the width of the hysteresis cycle and the switching field to be that corresponding to

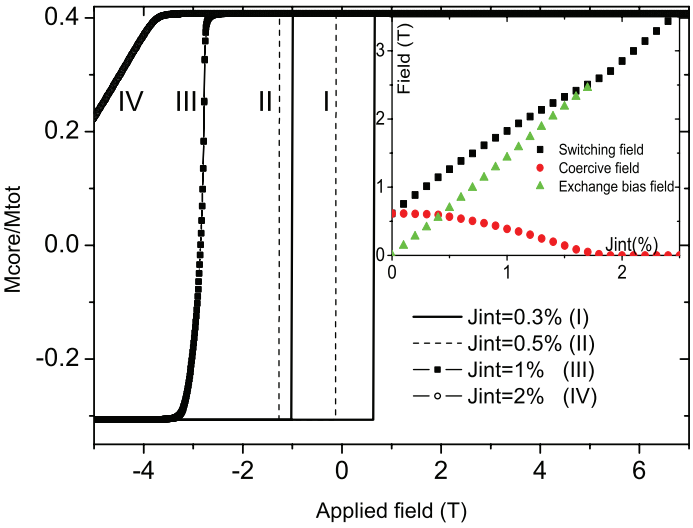

Fig. 1: (Colour on-line) Modeled hysteresis cycles for Co/CoO nanoparticle for various exchange coupling strengths J_{int} (% of J_{FM}). The core magnetization is normalized to the value when both FM and AFM are aligned with the applied field. The inset shows the switching, exchange bias and coercive fields as a function of J_{int}.

the irreversible magnetization change. Different regimes can be distinguished: I) the hysteresis cycles with small interfacial exchange are displaced to the left showing conventional exchange bias. This regime corresponds to the results presented in ref. [10], although as always in a real situation there is rounding of the hysteresis cycle due to the dispersion of nanoparticle sizes and properties. II) Further increase of the interfacial exchange leads to a situation in which the hysteresis cycle occurs completely in negative fields. III) For larger coupling strength (2%) we observe that the core magnetization undergoes a completely reversible ("exchange-spring"-like) rotation, since it is strongly pinned by the antiferromagnetic shell with large anisotropy. The coercive field strictly speaking is zero. IV) For even larger exchange coupling strength (5%) the shell magnetization becomes strongly coupled to the core and the AFM participates in the magnetization reversal. Consequently, the effective anisotropy of the whole system is averaged between the AFM and FM. Since reversal of the antiferromagnet requires a large external field this region is associated with a strong increase of the switching field.

In fig. 2 we present energy barriers of the nanoparticle as a function of the interfacial exchange parameter in regime I, corresponding to the remanent states in fig. 1. They were calculated using the Lagrangian multiplier technique [13], *i.e.* performing a minimization with a constrained average magnetization direction. This method allows the evaluation of energy barriers of nanoparticles using multi-spin atomistic models. The regime I is indeed characterized by an increase of the energy barrier separating the magnetization states with positive and negative magnetization directions. However, it is

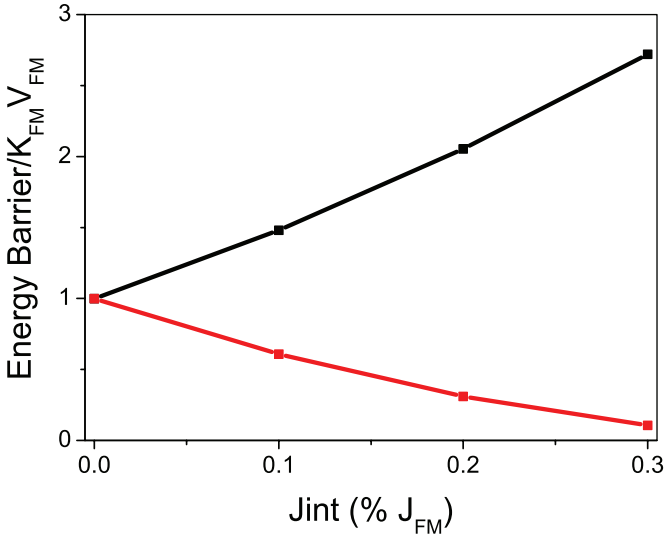

Fig. 2: (Colour on-line) Energy barriers at the remanence for Co/CoO nanoparticle as a function of $J_{\rm int}$ for the descending (upper curve) and ascending (lower curve) branches of the hysteresis loop.

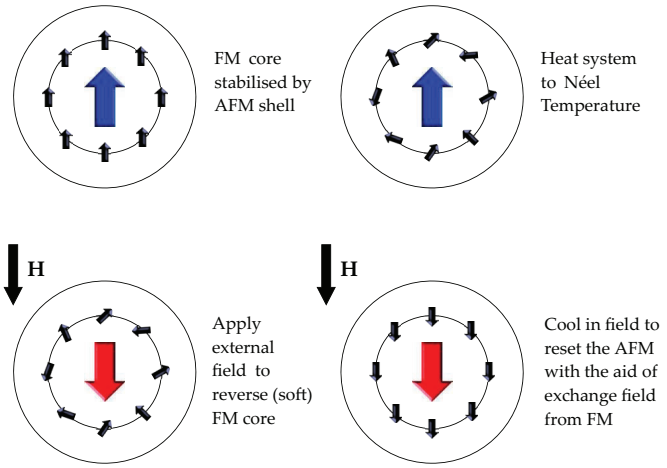

Fig. 3: (Colour on-line) The proposed heat-assisted scheme to reverse the FM magnetization upon heating over T_N of the AFM.

important to note that the opposite energy barrier, separating the negative and positive magnetization directions, is decreased. The blocking temperature would reflect only the largest energy barrier and therefore, the result is consistent with the data reported in ref. [10]. However, since for magnetic recording applications both energy barriers must be high, it is evident that this situation is not suitable for this purpose.

The regime corresponding to larger FM/AFM coupling deserves further analysis, which we address in the following. In these cases the ferromagnetic state is unconditionally stable; by which we mean that only one stable state exists in zero field. The opposite magnetization state is unstable in zero field without resetting the exchange bias direction. This means that it is not possible to reverse the magnetization of the ferromagnet alone without changing the antiferromagnetic state. Consequently, in order to change the magnetization state an extremely large field must be applied. Thus, although the energy barrier of the stable state is sufficiently large to guarantee thermal stability, we nevertheless return to the "magnetic recording trilemma", *i.e.* the gain in the energy barrier is accompanied by an impossibly large increase in the write field.

To overcome the problem, we suggest the utilization of HAMR. Our proposed recording scheme is sketched in fig. 3. Namely, if $T_N \ll T_C$ as in Co/CoO case, one can heat the magnetic system up to a temperature T slightly higher than T_N, destroying the antiferromagnetic order. At this state the ferromagnetic medium could be switched by fields comparable with its coercivity at temperature T and therefore the magnetization is easily reversed. To illustrate this possibility, we perform HAMR modeling on a 5.5 nm diameter nanoparticle with a 4 nm diameter Co core. The interfacial exchange coupling was set to $J_{\rm int} = 5\%$ of $J_{\rm FM}$, to give an unconditionally stable state for the core and an entirely reversible hysteresis loop. The system is first equilibrated at room temperature in a large external field to force the exchange bias in the desired direction, so that the stable state of the core is along the positive z-direction. The initialization field is then deactivated and the system equilibrated at $T = 300\,{\rm K}$. A static negative field of 1 T was then applied, which is insufficient to reverse the core magnetization at 300 K. The temperature was then instantaneously raised to 450 K, simulating the effect of a laser pulse. After that the temperature is assumed to follow a gaussian cooling profile given by: $T = 300\,{\rm K} + 150\,{\rm K}\exp(-t^2/t_0^2)$, where t is the time and $t_0 = 500\,{\rm ps}$ is the characteristic cooling time. Note that this model assumes that the electrons and lattice are in thermal equilibrium at all times, which, due to the relatively long cooling timescale, is a valid approximation. We use the value of the damping parameter $\alpha = 0.1$, reflecting the well-known increase of the damping at high temperatures. The results of the magnetization dynamics for the system are presented in fig. 4. On application of the temperature pulse, the AFM shell is demagnetized, allowing the reversal of the FM core in the applied field. As the system is cooled the AFM recovers its order but with significant superparamagnetic fluctuations. The exchange field from the reversed FM core ensures that the AFM recovers along the correct direction, thus completing the reversal process. A visualization of the system after reversal is shown in fig. 5. The core magnetization is represented by a single macrospin, while one sublattice of the AFM shell is displayed. Our simulations confirm the feasibility of this HAMR process showing the possibility to change the FM state in less than 500 ps. The timescale of reversal in the case of the coupled FM/AFM with HAMR is limited by the thermally activated reversal time of the FM, at temperatures much smaller than T_C.

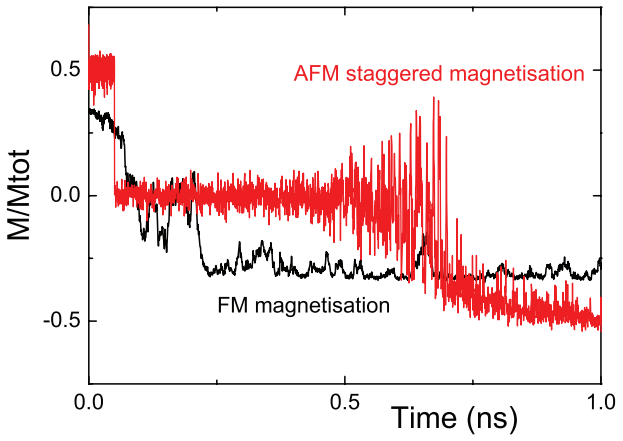

Fig. 4: (Colour on-line) The FM and AFM magnetization dynamics during the heat-assistant magnetization reversal upon heating over T_N of the AFM. The staggered AFM magnetization is plotted to show the degree of the deviation from the ordered AFM state. The magnetization is normalized to the case when both FM and AFM are fully aligned.

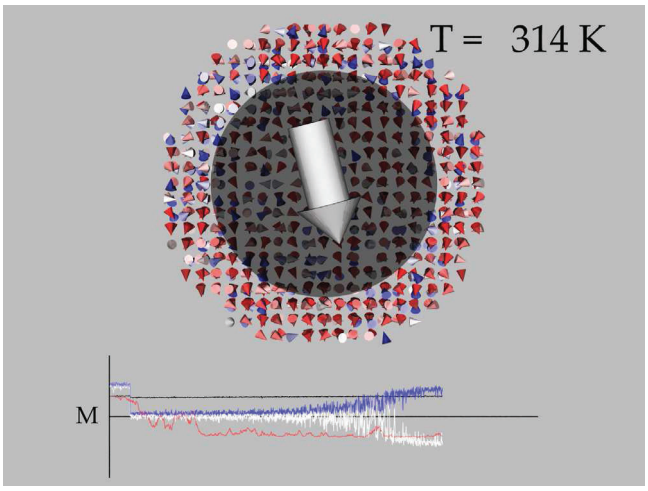

Fig. 5: (Colour on-line) Visualization of the coupled system after the HAMR reversal process (see the supplementary on-line movie (`Movie.wmv`) of the reversal process, showing the demagnetization and recovery of the AFM shell, as well as coherent reversal of the FM core).

In conclusion, we have investigated the possibility of "beating the superparamagnetic limit with exchange bias" as a technique to achieve ultra-high density magnetic recording. The recently developed Lagrangian multiplier method [13] has allowed us to evaluate energy barriers in an exchange-biased system showing that the coupling of the FM core with the AFM shell increases the energy barrier. Unfortunately, our results also show that, for small interfacial coupling, the energy barriers corresponding to the backward switching are reduced. We have also shown that for larger interfacial coupling the FM state is very stable, however, the magnetization reversal in this case involves the AFM and requires a very large applied field. We have proposed a new recording scheme based on the use of HAMR, in which the system is heated above the Néel temperature, destroying the AFM order and allowing to reverse the FM magnetization. Consequently, the possibility of the "beating the superparamagnetic limit with exchange bias" still remains feasible, although not without HAMR. The proposed scheme has an advantage compared to the conventional HAMR because it only requires sufficient heating to destroy the order in the AFM, which for CoO is $T > T_N \approx 400\,\mathrm{K}$. This avoids many problems related to HAMR in high anisotropy (FePt-like) media where heating up to almost $T = 1000\,\mathrm{K}$ is necessary, leading to unwanted problems, for example with lubricants. The desired behavior requires some degree of core/shell coupling, although, fortunately, not large. Whether this coupling could be achieved and controlled experimentally is an open question. Furthermore, the nanoparticle geometry investigated in the present letter is not an absolute requirement. One could envisage, for example, an exchange-biased thin film with subsequent lithography.

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