Manifestation of higher-order inter-granular exchange in magnetic recording media

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Exchange coupling between magnetic grains is essential for maintaining the stability of stored information in magnetic recording media. Using an atomistic spin model, we have investigated the coupling between neighbouring magnetic grains where magnetic impurity atoms have migrated into the non-magnetic grain boundary. We find that when the impurity density is low, a biquadratic term in addition to the bilinear term is found to better describe the inter-granular exchange coupling. The temperature dependence of both terms is found to follow a power law behaviour with the biquadratic exchange constant decaying faster than the bilinear. For the increasing grain boundary thickness, the inter-granular exchange reduces and also decays more quickly with temperature. Further simulations of a grain at a bit boundary show an unexpected energy minimum for in-plane magnetisation. This feature is reproduced if the biquadratic exchange term is included. *Published by AIP Publishing.*
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Obtaining the optimal level of coupling between magnetic grains in recording media is an important challenge for future high-density magnetic recording devices. To achieve suitably high areal densities, the grain size and pitch distance are being reduced to a few nanometers.[1,2] Inter-granular interactions, both long-ranged magneto-static and short ranged exchange coupling, play an important role in the properties of magnetic recording media, including the dynamic behaviour during the recording process[3] and the long-term thermal stability.[4] However, the origin and nature of this short-ranged exchange coupling until recently were unexplored. As the design of recording media becomes more complex, an understanding of the nature and magnitude of the exchange coupling and the development of a realistic representation of the exchange for use in recording models becomes increasingly important.

Initial experimental measurements of inter-granular exchange by Sokalski *et al.*[5] simplified the system to two thin films of magnetic media separated by a non-magnetic inter-layer giving a well-characterized system to investigate the physics of the exchange coupling. From these and other experimental measurements,[6,7] the inter-granular exchange is observed to decay exponentially with the grain boundary thickness and linearly with temperature.

A model for this interaction is proposed to be magnetic impurities present in the grain boundary which provides a channel for the grains to interact directly.[8] Simulations using the proposed impurity model by Evans *et al.*[8] on a multilayer system similar to Sokalski's showed the same exponential dependence on the grain boundary thickness as observed experimentally and also extracted the temperature dependence of the inter-granular exchange parameter which is important for future heat assisted recording schemes.

In addition to its origin, the particular angular form of the inter-granular exchange is known to have a large impact on the modelled bulk properties of granular media and on the recording performance.[9–11] The inter-granular exchange is typically written in the form of a Heisenberg exchange, often referred to as "bilinear."[12,13] As we show later, the Heisenberg form does not necessarily provide the correct angular dependence of the energy or torque. We find that at high impurity density, the energy is dominated by a micro-magnetic exchange stiffness behaviour, where at the limit with a continuous magnetisation, the exchange energy can be modelled using

$$E_{ex} = \frac{J_{eff}}{2} \phi_{ij}^2,$$ (1)

where $\phi_{ij} = \mathrm{acos}(\mathbf{m}_i \cdot \mathbf{m}_j)$ is the total change in the magnetization angle across the grain boundary that separates the grains. However, in the low impurity density regime more relevant to magnetic recording media, we find that our results, obtained using an atomistic model, are better described by including both a bilinear and biquadratic Heisenberg exchange term of the form

$$E_{ij} = A_{ij}(1 - \mathbf{m}_i \cdot \mathbf{m}_j) + B_{ij}[1 - (\mathbf{m}_i \cdot \mathbf{m}_j)^2],$$ (2)

where A_{ij} and B_{ij} are the bilinear and biquadratic exchange constants between grains i and j and $\mathbf{m}_i$ is the normalised grain magnetisation.

Biquadratic exchange has previously been observed in magnetic multi-layers,[14] such as Fe/Cr/Fe,[15] where a 90° alignment of the layers is observed. Direct inter-layer interactions, such as Ruderman–Kittel–Kasuya–Yosida (RKKY) exchange and dipole fields, do not successfully account for the magnitude of the measured biquadratic exchange or its temperature dependence. Models proposed by Slonczewski determined that the origin of the biquadratic term lies not in the fundamental quantum mechanics, which does permit higher-order exchange terms, but arises from surface roughness[16] or loose spins in the
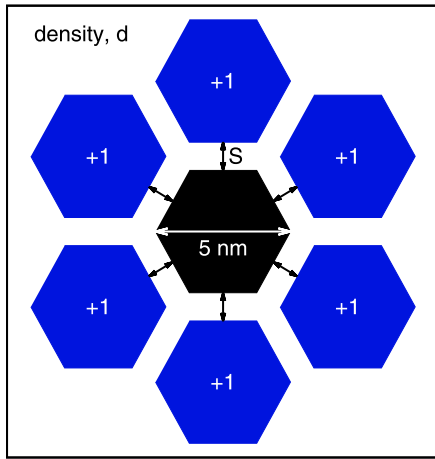
FIG. 1. Schematic showing the set-up used to calculate the inter-granular exchange. Each grain has a diameter of 5 nm, and the grain boundary thickness is given by the separation of the grains S. Initially, the magnetisation of the outer grains is constrained to be $m_z = +1$.

inter-layer.[17] Of particular interest is this second model based on loose spins, which is analogous to the present work and predicts the temperature dependence of the coupling through the thermal effects on the loose inter-layer spins.

    To explore the inter-granular exchange in recording media, we employ an atomistic spin model[18] based on localised magnetic moments in the same manner as employed in Ref. 8, and extra details of the model and method can be found there. In contrast to Ref. 8, where a multi-layer is simulated, we model the perpendicular recording media as a set of hexagonal grains; a central grain surrounded by 6 neighbouring grains of uniform size is shown schematically in Fig. 1. These grains are separated from each other by a non-magnetic grain boundary with a specified density of magnetic impurity atoms. These atoms are assumed to be of the same type as those in the grains, and they provide a direct interaction channel without the need for long range interactions. On the atomic-scale, the system is described by a Heisenberg exchange Hamiltonian $\mathscr{H} = -1/2 \sum_{i \neq j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$, where J_{ij} is the exchange constant in terms of the unit vector spins $\mathbf{S}_i$. The summation for the exchange is carried out over nearest neighbours only. Each atom has a magnetic moment of $\mu_s = 1.44 \ \mu_B$, and the exchange constant is $J_{ij} = 5.6 \times 10^{-21}$ J which gives a $T_C \approx 1280$ K. For the magnetic impurity atoms in the grain boundary, it is assumed that there is no variation in the exchange constant or magnetic moment. Realistically, these will vary depending on each atom's local environment, but we do not consider this here. Each hexagonal grain has a corner-to-corner diameter of 5 nm, while the crystal structure is set as a fcc lattice with a unit cell size of $a = 3.54$ Å. Periodic boundary conditions are employed in the $\hat{\mathbf{z}}$ direction to remove any surface effects, and the grain boundary thickness is varied by locating the grain centres further apart, which is denoted by S in Fig. 1. Measurements by Araki *et al.*[2] show that in high-density perpendicular media, the grain boundary thickness is less than 1 nm. We focus on this region and simulate grain boundary thicknesses of 0.5 nm, 0.75 nm, and 1 nm, which correspond to 3, 4, and 5 monolayers, respectively.

    Calculations are performed using the Constrained Monte Carlo (CMC) method,[19] which fixes the direction of the
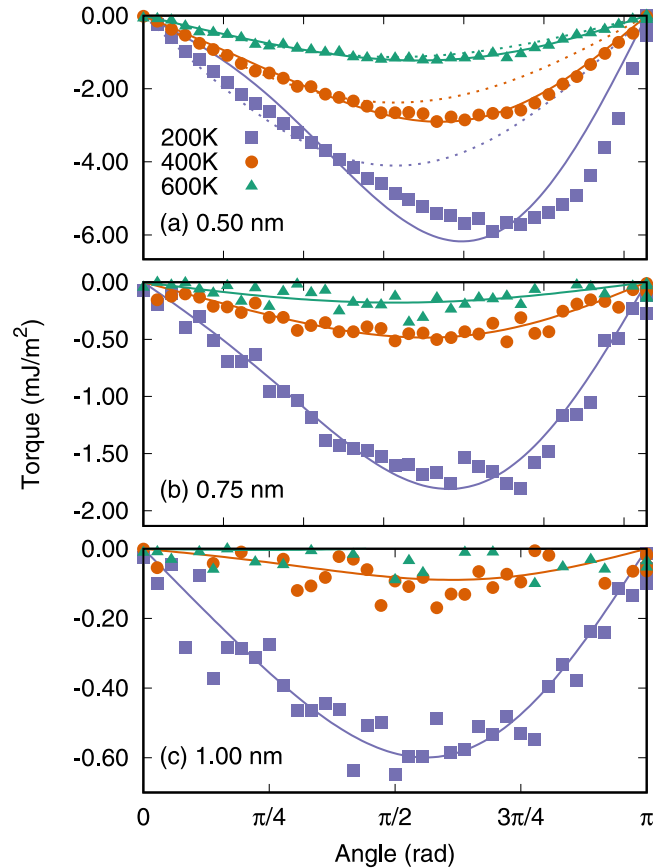

FIG. 2. The torque acting on the central grain for separations: (a) 0.50 nm, (b) 0.75 nm, and (c) 1.00 nm at 20% density for 100 K, 300 K, and 500 K. Solid lines show a fit to the data using Eq. (2), and the dashed lines in (a) show a fit using only the bilinear term.

magnetization but allows its magnitude to fluctuate. The CMC method generates joint moves of two individual spins such that the magnetization direction remains constant. A hybrid constrained/unconstrained Monte-Carlo algorithm has previously been implemented into the VAMPIRE code[18,19] which allows us to fix the magnetisation orientation for the grains, whilst the spins within the grain boundary are unconstrained and follow the usual Metropolis Monte-Carlo algorithm. Initially, the surrounding grains are constrained to be fixed in the $+z$ direction, and the central grain is constrained at different polar angles ϕ ranging from 0° to 180° in steps of 5°, with $m_y = 0.0$. At each angle, the macroscopic torque, τ, is computed for each grain from which the free energy can then be calculated.[19] The Monte-Carlo simulations are performed using 10 000 steps for equilibration and 10 000 steps to gather averages of the properties for each separate constraint angle.

    To illustrate the non-Heisenberg angular variation of the inter-granular exchange energy, Fig. 2 shows the calculated torque acting on the central grain for three different temperatures and grain boundary thicknesses. Graphically, we normalise the torque and energy by the contact area of the grains. In all cases, there is a strong asymmetry in the torque, which cannot be accounted for by using only a bilinear exchange term. Even if one considers differing bilinear exchange magnitudes between the central grain and the surrounding grains, the net torque will still have an angular dependence given by the bilinear exchange since the surrounding grains are all
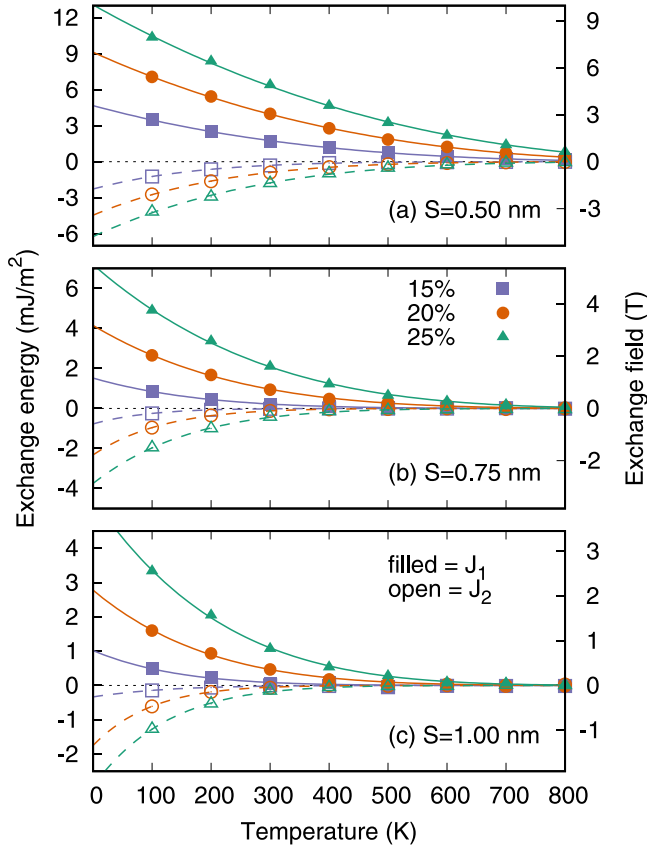
FIG. 3. The temperature dependence of the effective inter-granular exchange constants for (a) 0.50 nm, (b) 0.75 nm, and (c) 1.00 nm at low density. The bilinear exchange constant (A_{ij}) is shown as filled points and the biquadratic (B_{ij}) as open points, and the lines are a fit to the data using Eq. (4).
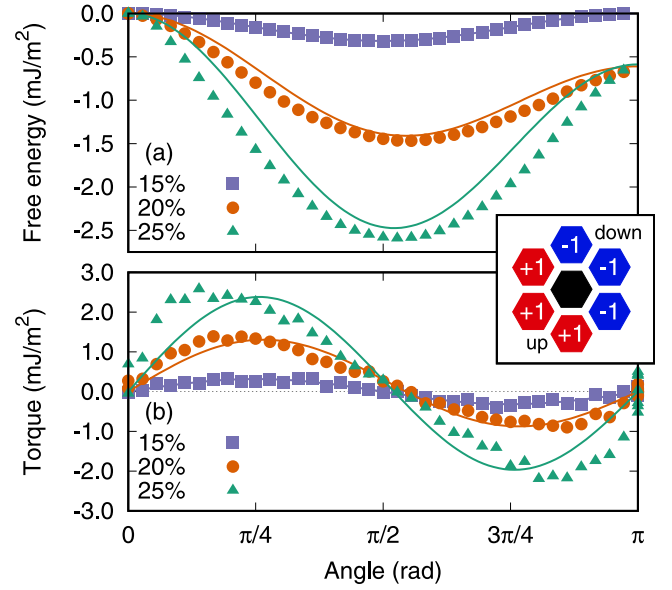

FIG. 4. Free energy (a) and torque (b) when the central grain has 3 neighbours constrained with $m_z = +1$ and 3 neighbours with $m_z = -1$ for 15%, 20%, and 25% density at 300 K. The configuration is shown in the inset of (b). Lines show a fit using both bilinear and biquadratic terms as given in Eq. (5).

constrained in the same direction. An alternative is modelling the energy and torque using a micro-magnetic exchange stiffness approach which would yield a linear torque term: however, this only agrees within a small angle limit. The solution we propose to this dilemma is to introduce a biquadratic term in the low density limit.

Rather than fitting the free energy directly, which can be obtained by integrating the torque, we find that fitting the torque directly is more accurate since it is more sensitive to the angle. The inter-granular exchange is extracted by fitting

$$T(\phi) = -\frac{\partial E}{\partial \phi} = -A\sin\phi - 2B\cos\phi\sin\phi, \quad (3)$$

with A and B being fitting parameters. As shown in Fig. 2, the additional biquadratic exchange term provides an improved fit compared to only the bilinear term, which is shown as dashed lines in Fig. 2. Importantly, the biquadratic term explains the asymmetry in the results of the atomistic model. The combination of bilinear and biquadratic terms describes the numerical results for the low exchange coupling regime but begins to break down with increasing inter-granular exchange corresponding to small grain boundary thicknesses and low temperatures, for example, 200 K in Fig. 2(a). This limits our biquadratic exchange model to low inter-granular exchange coupling, but this is in the regime that is of interest for experimental situations, i.e., low impurity density and temperatures of 300 K and above.

The variation in bilinear and biquadratic exchange constants, extracted from the fitting, with temperature is shown in Fig. 3. The exchange energy can be expressed as an exchange field using $H_{\text{exch}} = A/M_s V$ using our reference saturation magnetisation of $M_s = 6 \times 10^5\,\text{J\,T}^{-1}\,\text{m}^{-3}$ which is calculated from the magnetic moment per atom volume. There is a clear decay with increasing temperature, which is best modelled using a power law expression. For both the bilinear and biquadratic exchange, we fit

$$A(T) = A_0 \left(1 - \frac{T}{T_C}\right)^\alpha, \quad (4)$$

to the data, where A_0 and α are the fitting parameters and T_C is held constant at the approximate Curie temperature (1200 K). This behaviour models the data well, as shown in Fig. 3, and we see that the biquadratic exchange decays faster than the bilinear term. This observation is consistent with previous simulations of the metamagnetic phase transition in FeRh based on a competition of bilinear and higher order (4-spin) terms.[20]

Previous simulations have shown a linear temperature dependence of the exchange coupling[8] in agreement with earlier experimental measurements.[7] In the present case, the exponent, α, scales with density and separation, and so, a situation may arise where the temperature dependence is close to linear. These results agree with Ref. 8 where at the right temperature for Heat Assisted Magnetic Recording (HAMR), the inter-granular exchange will be negligible. However, as the grain boundary thickness is reduced, the exchange persists to increasingly high temperatures. We notice that the biquadratic term is always negative in our system, which shifts the position of the peak torque to higher angles.

Finally, we turn to the situation where the central grain lies at a bit boundary and the bits have opposite orientation. To represent this within our model, we consider 3

neighbouring grains with their magnetisation up ($m_z = 1$) and 3 with their magnetisation down ($m_z = -1$); a schematic of this configuration is shown as an inset in Fig. 4(b). In this configuration, the central grain experiences different torques forcing it to align in both orientations. Due to the symmetry of the bilinear term, the energy contributions act against each other, whilst the biquadratic terms are additive. In our model, the exchange coupling arises from randomly sited impurity spins in the grain boundary, and so, there is a dispersion in the local exchange. This means that realistically, the competing bilinear terms will not cancel completely but will depend on the difference in the bilinear constants. This leads us to a total energy of the form:

$$E = -\Delta A(1 - \cos\phi) - B_{\text{tot}}\sin^2\phi, \qquad (5)$$

where $\Delta A = A_\uparrow - A_\downarrow$ is the difference in the bilinear terms and $B_{\text{tot}} = B_\uparrow + B_\downarrow$ is the sum of the biquadratic terms, while $\uparrow$ and $\downarrow$ indicate the coupling to either neighbouring grain orientation.

Figure 4 shows the free energy and torque calculated for the central grain for this configuration. The torque in panel (b) is fitted directly using Eq. (3), but now, the coefficients represent the parameters in Eq. (5). Using these fit parameters, Eq. (5) is compared with the free energy computed by integrating the torque, which is shown in panel (a). There is a good agreement between the data and the fit for the low densities and exchange values expected for recording media. At the higher densities, the fit does not match the CMC data exactly but nonetheless provides a good qualitative representation of the key behaviour. The energy shows a slight mismatch as it is aligned with zero energy at $\phi = 0$ rather than the true minimum. In these examples, there is a clear difference in the coupling to the different orientations; i.e., $\Delta A \neq 0$, which leads to the asymmetry of the free energy. Without any anisotropy, the central grain will minimise energy by having its magnetisation in-plane, but if anisotropy is included, there would be a preference to align with grains that it most strongly couples to. If only the bilinear exchange is considered, then it is clear that the resulting $\sin\phi$ torque will not accurately represent the actual torque. Alternatively, if a micro-magnetic exchange is considered where the torque can be approximated as $T = -A(\phi - \pi/2)$, this form fits well around $\pi/2$ but overestimates the torque close to 0 and π.

As discussed in the introduction, the origin of biquadratic exchange in multi-layers can be explained through interface roughness and loose spins in the inter-layer. The biquadratic exchange observed here may arise from a similar situation since it is dominant at low impurity density. Consequently, although we have no interface roughness in our model, we expect strong local fluctuations in behaviour with analogous effects similar to those of interface roughness. We note that a biquadratic term was not observed by Evans *et al.*[8] in a multi-layer structure where the torque is only observed to have a linear dependence on the angle. However, the torque presented by Evans *et al.* is for a much

larger grain boundary thickness than considered here. Since the biquadratic exchange occurs at low impurity density, the magnitude of the inter-granular exchange at these thicknesses may be too small to observe.

To summarise, we have observed a manifestation of biquadratic inter-granular exchange in magnetic recording media. We have used an atomistic spin model to simulate a system with uniform hexagonal grains separated by an grain boundary in which magnetic impurities reside. Using a constrained Monte-Carlo method, the torque and free energy are computed as a function of angle for the central grain. The angular dependence of the torque indicates the presence of a biquadratic exchange term. The temperature dependence of both the bilinear and biquadratic exchange follows a power law, with the biquadratic exchange decaying faster than the bilinear. Further simulations of a grain at a bit boundary again show the necessity of the biquadratic term since the atomistic model predicts that the grain will tend to align in-plane, which cannot be described by the bilinear term. The origin of this manifestation may to be similar to Slonczewski's mechanism for biquadratic exchange in magnetic multi-layer structures as it relies upon clusters of random size in the grain boundary.

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