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Are there superspin glasses?

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The effect of magnetostatic and exchange interactions on the spin structure of interacting nanoparticles and granular nanomagnets is investigated by model calculations. Effective exchange stiffnesses for inhomogeneous media are defined and determined for some geometries and interactions, and it is argued that typical ensembles of interacting small nanoparticles are micromagnetic systems rather than superspin glasses or superferromagnets. The spin structures of granular magnets often have the character of interaction domains, with far-reaching implications for magnetic phenomena such as hysteresis-loop overskewing. © *2011 American Institute of Physics*. [doi:10.1063/1.3562957]

I. INTRODUCTION

The interplay between atomic and nanoscale degrees of freedom in granular magnets and its thermodynamic implications have remained controversial for many years. One question is the physical nature of spin structures such as superferromagnets and superspin glasses. These structures are defined as ensembles of interacting magnetic particles[1–5] and characterized by deviations from superparamagnetic or "macrospin" behavior.[5,6] They exhibit features such as enhanced relaxation times τ scaling as ξ^z, where ξ is the correlation length and z is a critical exponent. In typical systems, such as permalloy particles embedded in an alumina matrix ($Co_{80}Ni_{20}/Al_2O_3$), the deviations from superparamagnetism are caused by magnetostatic dipole interactions, and exchange is negligible.[6] This leads to the question whether these dipole glasses can be equated with canonical random-exchange spin glasses. Frustration and randomness occur in both classes of materials,[4,6,7] but the long-range character of the magnetostatic interactions may change model predictions quite drastically.[7] Furthermore, dipole interactions are deterministic functions $J(\mathbf{R}_{ij}) = J(R, \theta)$, as contrasted to the truly random exchange bonds in canonical spin glasses $\langle J(\mathbf{R}_{ij})\rangle = 0$. This means that the ground state is a flux-closure state rather than a canonical spin-glass state.

Magnetic dipole interactions are rarely considered in ferromagnetic and spin-glass models,[7] because they destroy the ferromagnetic long-range order and give rise to the paradoxical conclusion that ferromagnetism does not exist in nature. The paradox is solved by separating atomic or intrinsic properties, such as exchange, from micromagnetic or extrinsic phenomena, such as domains. The transition between intrinsic and extrinsic regimes occurs often, but not always, on a length scale of 1 to 2 nm.[8–10]

The complex interplay between micromagnetism and thermodynamics is epitomized by random-anisotropy spin glasses. Originally considered in atomically disordered materials,[11] it has lead to concepts such as correlated spin glasses (CSG),[12] which are easily generalized to nanocrystalline magnets.[8,13] However, these structures are micromagnetic and not associated with an ordering temperature. In fact, some of the hardest magnets ever produced, namely polycrystalline $Sm_2Fe_{17}N_3$ with coercivities in excess of 4 T [40 kOe], are random-anisotropy magnets and therefore nonferromagnetic.[14]

II. THERMODYNAMICS AND MICROMAGNETICS

Thermodynamics is concerned with the free energy $F = E - TS$, where E is the energy and S is the entropy. Phase transitions involve entropies of order E/T_c, where T_c is the phase-transition temperature. In small nanoparticles, both E and S increases with the number N of atoms per particle, but the energy increase is much more pronounced than the entropy increase, $S \sim \ln(N)$. Typical nanoparticles have several thousands of atoms, so that $S \sim 10\ k_B$. Since $T_c = E/S$, phase transitions below room temperature require very small interaction energies E. For the above-mentioned permalloy particles,[6] the dipole-glass transition is achieved by reducing magnetostatic interaction fields to about 10 mT.

As the particles touch, they introduce a strong exchange, and thermodynamic considerations are no longer applicable. The same happens for magnetic nanoparticles embedded in a ferromagnetic matrix, as in the Sm-Co-Cu-Ti system, where the exchange coupling between Sm-Co grains can be tuned by varying the Cu- and Ti-contents.[15,16] The corresponding spin structures have become known as *interaction domains*[17,18] or, more recently, superferromagnets.[19] They are characterized by rough domain-wall boundaries and a 'discrete' pinning mechanism.[15,16] Locally, the ferromagnetic exchange dominates the magnetostatic interaction, but the latter wins on a global scale. Magnetization processes in these structures are of the micromagnetic type, with transitions between noncooperative and cooperative behavior, depending on the strength of structural disorder.[8,15,20]

III. EFFECTIVE EXCHANGE STIFFNESS

Exchange in ferromagnetic materials is described by the exchange stiffness A, which is closely related to the Curie

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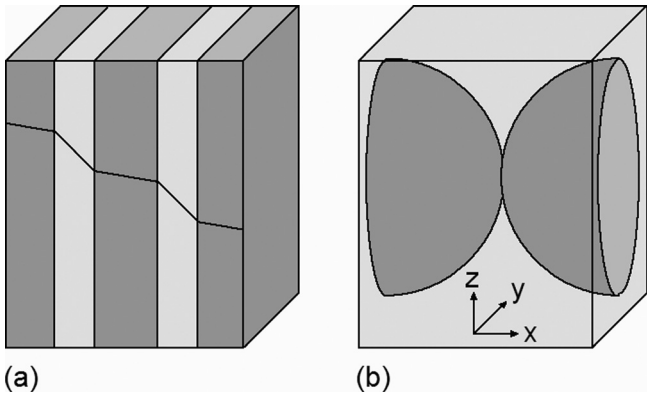

FIG. 1. Exchange coupling of nanoscale regions (dark) through a matrix (bright): (a) layered structure and (b) touching spheres.

temperature, $T_c \sim A\ a/k_B$, determines the spin-wave behavior, and affects domains and domain walls. This section considers and derives effective exchange stiffnesses A_{eff} and effective intergranular exchange constants J_{eff} for granular systems (Fig. 1). The effective exchange interaction is obtained by evaluating the exchange energy

$$E_{ex} = \int A\left(\nabla \mathbf{M}^2\right) dV, \tag{1}$$

whose minimization yields $\nabla(A\nabla\mathbf{M}) = 0$.[21] A_{eff} is determined from Eq. (1) by comparison with a homogenous system.

Assuming that the magnetization rotates coherently in the y-z plane of Fig. 1, one must consider the magnetization angle θ as a function of x. This yields

$$\frac{1}{A_{eff}} = \frac{F_{max}}{L} \int_0^L \frac{1}{A(x)F(x)} dx, \tag{2}$$

where $F(x)$ is the cross-section area and $F_{max} = \max[F(x)]$. First, we consider a two-phase system having exchange stiffnesses A_o (particle) and A' (matrix), volume fractions $1-p$ and p, respectively, and $F(x) = F_{max} = \text{const.}$, as in Fig. 1(a). The corresponding exchange stiffness

$$\frac{1}{A_{eff}} = \frac{1-p}{A_o} + \frac{p}{A'}. \tag{3}$$

For small matrix exchange stiffness A', A_{eff} becomes very small. The magnetocrystalline anisotropy is ignored in these calculations, but their inclusion is straightforward.[22]

For grains that touch each other with a contact area πR_c^2, as in Fig. 1(b), it is reasonable to assume that the magnetization change is localized in a volume of order $4\pi R^3/3$. Equation (1) then yields an effective exchange constant $J_{eff} = J\ R_c/a$ between the spheres, where a is the interatomic distance and R is the particle radius. The effective exchange stiffness $A_{eff} \approx A\ R_c/R$, and taking $R_c = 1$ nm yields ordering temperatures J_{eff}/k_B of the order of a few 1000 K.

Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions between nanoparticles in a nonmagnetic metallic matrix scale as $J_{eff} \sim J_o\ a\ R^2/d^3$, where d is the interparticle distance.[23] Fixing the volume fraction while changing the particle size leads to $J_{eff} \sim a/R$. An effective exchange stiffness is difficult define for this case, because the coupling is oscillatory rather than ferromagnetic. However, on a mean-field level, the spin-glass ordering temperature of N particles is easily estimated as the largest eigenvalue of the $N \times N$ matrix $J_{ij} = J(\mathbf{R}_i - \mathbf{R}_j)$.[24] Using random-matrix theory, similar to the exploitation of Wigner's semicircle law,[7] yields

$$T_c = \frac{1}{k_B}\sqrt{\frac{1}{N}\sum\nolimits_{i \neq j} J_{ij}{}^2}, \tag{4}$$

and $T_c \sim 1/R$. An analogous calculation for magnetostatic interactions between particles of fixed volume fraction yields $T_c \sim R^3$. Magnetostatic interactions dominate the RKKY interaction for $R > 1$ nm, in agreement with earlier findings.[23]

IV. DEMAGNETIZING FIELDS

Surprisingly, the effect of magnetostatic and exchange interactions on the magnetization reversal in granular nanostructures is rather poorly understood. Examples are field cooling (interpretation of ZFC/FC curves) and hysteresis-loop overskewing,[25] an overestimation of demagnetizing-field corrections that is of great importance in permanent magnetism. Dobrynin *et al.*[26] have recently made the controversial suggestion that the overskewing is caused by a previously overlooked magnetostatic 'hard-demag' field contribution of the order of $M/3$. This contribution reduces the overskewing, because it amounts to a replacement of the demagnetizing factor D by D - 1/3. The problem is multifaceted and includes a variety of issues, such as the distinction between the hard-demag field and well-known cavity fields, the striking involvement of self-interaction fields,[27] and the definition of the demagnetizing fields. The last must be done by properly taking into account that demagnetizing fields in extended perfect structures such as thin films and spheres, which are used for comparison, contain a curling-type self-interaction field.[8]

The main role of the demagnetizing fields in permanent magnets is to determine the magnetostatic energy E_{out} stored in free space outside a magnet of volume V or, equivalently, the energy product $(BH) = E_a/2V$. For a toroid of contour length $2\pi R = L$ and gap width g (Fig. 2), this energy is given by the familiar expression

$$E_{out} = \frac{1}{2} D(1-D)\,\mu_0 M^2 V, \tag{5}$$

where $D = g/(L+g)$ is the macroscopic demagnetizing factor. Figure 2 illustrates that the field in free space is essentially determined by the pole density σ of the magnet. Small gap sizes (elongated magnets) yield $D < 1/3$, and replacing this value by $D - 1/3$ yields an unphysical negative magnetostatic energy density in free space.

So where does the hysteresis-loop overskewing come from? A likely explanation is the inhomogeneous character magnetization states encountered in practice. For macroscopic magnets, as illustrated in Fig. 2, the pole density is essentially given by the *average* magnetization, and D is meaningfully defined. This is no longer the case in small-scale

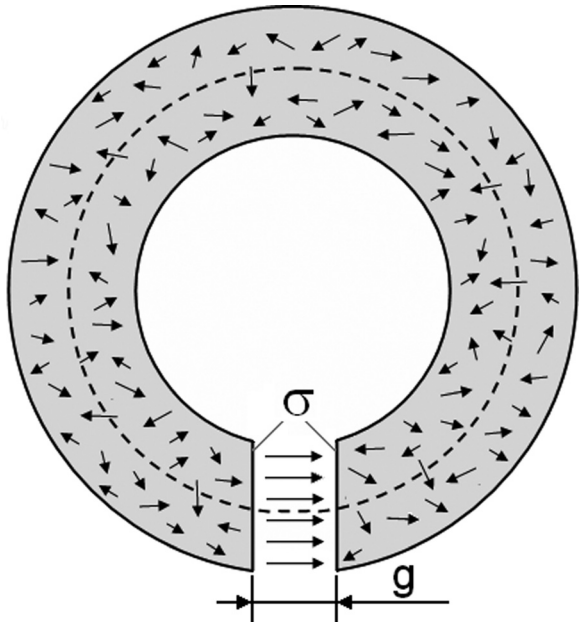

FIG. 2. Field considerations in permanent magnets. Demagnetizing factors of interest in permanent magnetism determine the usable magnetostatic energy in free space (energy product).

nanostructures and, especially, in thin films. Magnetization reversal in small multigrain nanoparticles often involves cooperative magnetization processes (avalanche- type interaction domains), and thin films have inhomogeneous pole distributions both locally (lateral inhomogeneities comparable to film thickness) and globally (domain walls can move without much change in magnetostatic energy, implying $D \sim 0$ rather than $D = 1$).

V. CONCLUDING REMARKS

We have seen that interacting magnetic nanoparticles exhibit a complicated interplay between magnetostatic and exchange interactions, with several challenges for future research. Superspin glasses or, more precisely, dipole glasses, exist in a fairly narrow parameter range, which is determined by the competition between energy and entropy. The terms superspin glasses and especially superferromagnets should not be used as general terms for interacting nanoparticles. In particular, magnetostatic interactions always destroy 'true' ferromagnetism on a macroscopic scale. The distinction between the different types of order is complicated by the experimental situation. Spin-glass-like systems are expected to exhibit a divergence of the nonlinear susceptibility χ_3 at T_c,[7] and this is indeed observed for permalloy particles in Al_2O_3.[2] However, the reverse is not true, and even ordinary ferromagnets show a divergence of χ_3 at T_c. Broadly distributed relaxation times, as epitomized by very flat Cole-Cole plots,[2] are also observed in defect-containing ferromagnets.[28,29] Training and memory effects,[30] frequently associated with superspin glasses, have no specific link to thermodynamics[31] and also occur in micromagnetic systems.

Another phenomenon caused by the competition between exchange and magnetostatic interactions is interaction domains. They violate the homogenity condition for the applicability of traditional demagnetizing-field theory and probably explain the hysteresis-loop overskewing in magnetic nanostructures.

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