Remanence enhancement in magnetically interacting particles

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In this paper, we report an effective-medium theory on the remanence of magnetically interacting particles to demonstrate the effect of intergranular magnetostatic interactions on the remanence enhancement of materials, which agrees excellently with micromagnetic simulations. A dimensionless parameter λ measuring the competition between anisotropy energy and magnetostatic energy is defined, which completely characterizes the remanence of magnets if the exchange coupling is negligible, appropriate when the grain size is 10 nm or larger. Three distinct regimes were observed: $\lambda<0.1$ for hard magnets, where anisotropy energy dominates and little remanence enhancement is observed; $0.1<\lambda<1$ for intermediate magnets where up to 50% remanence enhancement is observed due to the increased intergranular magnetostatic energy; and $\lambda>1$ for soft magnets, where the dominance of magneto-static energy leads to much reduced remanence in the materials.

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Magnetic materials with large remanence and coercivity are highly desirable for energy storage due to their high-energy product.[1] Over the last 50 years, Stoner and Wohlfarth's celebrated model[2] was widely used to analyze the remanence of magnetic particles, with the remanence given by $M_r = M_s\langle\cos\theta\rangle$, where $\langle\cdot\rangle$ denotes the ensemble averaging, M_s is the saturation magnetization, and θ is the angle between the remanent magnetization of the sample and the easy axis of the particles. While the model provided accurate estimates of remanence for most permanent magnets, the recent development of exchange-spring magnets make the assumption of noninteracting particles invalid,[1,3,4] and new theoretical models considering the magnetic interaction between particles need to be developed in order to understand the remanence enhancement in the exchange-spring magnets and to guide the development and optimization of new material systems.

The exchange-spring magnets are highlighted by the intergranular interaction between magnetically hard and soft phases mixed at nanoscale, which results in dramatic enhancement in the remanence and energy product.[1,5,6] While micromagnetic simulations are able to explain the remanence enhancement due to the intergranular interactions,[7,8] they are often computation extensive and time consuming, and could not offer the deep insight provided by the analytical models. Even more important, while the role of intergranular exchange interaction in remanence enhancement is clearly emphasized by the micromagnetic simulation, the importance of magnetostatic interaction has not been fully appreciated, and has sometimes regarded as unimportant.[7,9] We hope to address these issues in this paper by developing an effective-medium theory of magnetically interacting particles to predict the remanence of a wide range of magnetic materials. It is demonstrated that the variation of remanence with material properties are characterized by a dimensionless parameter $\lambda = M_s^2/6K_1\mu_0$ which measures the competition between anisotropy energy and magnetostatic energy when the exchange coupling is negligible, where K_1 is the anisotropy constant. The analysis agrees excellently with micromagnetic simulation, and highlights the important role played by the magnetostatic interaction in the remanence enhancement when soft magnetic particles are present.

We consider an assemblage of single-phase single-domain magnetic particles, with particle size large enough compared to the exchange length, so that the short-range exchange interaction can be ignored. The model taking into account the exchange energy is currently being developed and will be reported later. As a result, in a remanent state, the potential energy of the assemblage is given by

$$F = \int_\Omega \left(K_1 \sin^2 \alpha[\theta] - \frac{1}{2}\mathbf{M}\cdot\mathbf{H}_d \right) d\mathbf{x}, \qquad (1)$$

where the first term is the uniaxial anisotropy energy at lowest order, with α being the angle between the local magnetization $\mathbf{M}$ and the easy axis of the particles, and the second term is the magnetostatic energy with $\mathbf{H}_d$ being the demagnetizing field induced by the distribution of $\mathbf{M}$ in Ω, the domain occupied by the assemblage. It is emphasized that $\alpha[\theta]$ is a function of θ here which we seek to characterize, while in the Stoner-Wohlfarth model, $\alpha=0$.

We consider the magnetostatic energy first, which can be written as

$$F_d = -\frac{1}{2}\int_\Omega (\overline{\mathbf{M}} + \mathbf{M}')\cdot(\overline{\mathbf{H}}_d + \mathbf{H}'_d)\, d\mathbf{x}, \qquad (2)$$

where the overhead bar is used to represent the volume averaged quantities, and the primed ones are variations from the averages. Clearly, $|\overline{\mathbf{M}}| = M_r$, and the cross products vanish. In addition, $\overline{\mathbf{H}}_d = -(1/\mu_0)\mathbf{N}_\Omega\overline{\mathbf{M}}$ is magnetic field induced by a uniform magnetization $\overline{\mathbf{M}}$ in Ω, where $\mathbf{N}_\Omega$ is the demagnetizing factor depending on the shape of Ω. For in-plane magnetization in an ellipsoidal Ω with dimensions a_1, $a_2 \gg a_3$, $\overline{\mathbf{H}}_d$ is approximately zero. The magnetic field induced by the magnetostatic interaction between particles $\mathbf{H}'_d$ can be determined by solving $\nabla\cdot(\mu_0\mathbf{H}'_d + \mathbf{M}') = 0$ for given distribution of $\mathbf{M}'$ using the Green's-function method. If the two-point correlation function of $\mathbf{M}$ is ellipsoidal,[10] we have

$$\mathbf{H}'_d = -\frac{1}{\mu_0}\mathbf{N}_g\mathbf{M}' = -\frac{1}{\mu_0}\mathbf{N}_g(\mathbf{M}-\overline{\mathbf{M}}), \tag{3}$$

where $\mathbf{N}_g$ is the demagnetizing factor depending on the shape of grains. For spherical distribution, $\mathbf{N}_g = 1/3\mathbf{I}$, which leads to

$$F_d = \frac{V}{6\mu_0}(M_s^2 - M_r^2), \tag{4}$$

where V is the volume of Ω. The treatment for more general grain shape is also possible. The first term of the magnetostatic energy is a constant thus can be ignored in the energy minimization. Combining it with the total anisotropy energy of the system

$$F_a = VK_1\langle \sin^2 \alpha[\theta]\rangle, \tag{5}$$

we obtain the following energy density of the assemblage

$$F = K_1\langle \sin^2 \alpha[\theta]\rangle - \frac{M_s^2}{6\mu_0} f^2,$$

where $f = M_r/M_s$. Clearly the anisotropy energy favors $\alpha = 0$ thus leads to noninteracting model, while the magnetostatic interaction leads the deviation of magnetization from its easy axis. The α, as a result, is a consequence of competition between the anisotropy energy and magnetostatic energy, measured by a dimensionless parameter $\lambda = M_s^2/6K_1\mu_0$. As such, we have

$$F = K_1(\langle \sin^2 \alpha[\theta]\rangle - \lambda f^2). \tag{6}$$

The hard phase is characterized by small λ while soft phase is characterized by large λ. For example, in an exchange-spring magnet consisting of $Fe_{14}Nd_2B$ and α-Fe, λ is approximately 0.1 and 10 for the hard and soft phase, calculated using data from Ref. 7. Clearly for very hard magnets where $\lambda \ll 1$, the dominance of anisotropy energy leads to $\alpha = 0$, recovering the classical results of the Stoner-Wohlfarth model. In general, α need to be determined from energy minimization as a function of θ, $\delta F/\delta\alpha = 0$, leading to the equation

$$\sin 2\alpha[\theta] = 2\lambda f \sin(\theta - \alpha[\theta]), \tag{7}$$

which is coupled with the remanence

$$f = \begin{cases} \int_0^{\pi} \cos(\theta - \alpha[\theta]) W[\theta] \sin\theta d\theta, & \text{3D texture} \\ \int_0^{2\pi} \cos(\theta - \alpha[\theta]) W[\theta] d\theta, & \text{2D texture} \end{cases}, \tag{8}$$

where $W[\theta]$ is the orientation distribution function for the easy axis of particles. For initial random orientation distribution of particles before the application of magnetic field, $W[\theta] = 1$, which is not normalized. In the remanent state, however, the orientation distribution will be changed due to the reversal of magnetic moments, and the extent of this reversal is dependent on the hardness of magnets. Here, we assume that in the remanent state

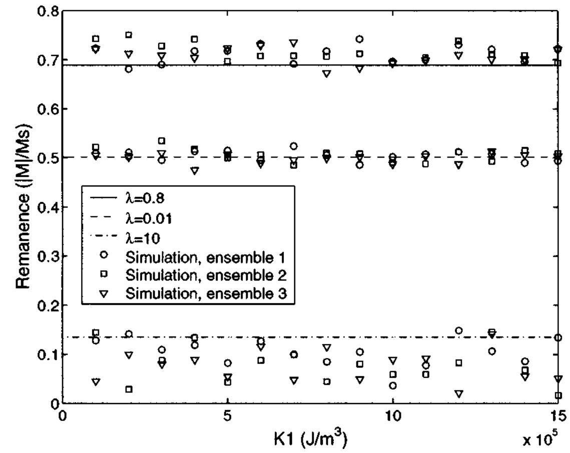

FIG. 1. The remanence of materials as function of K_1 for different λ.

$$W[\theta] = \begin{cases} \frac{1}{2}(1 + e^{-a\lambda}) & \theta \in [0, \theta_c/2] \\ \frac{1}{2}(1 - e^{-a\lambda}) & \theta \in [\theta_c/2, \theta_c] \end{cases}, \tag{9}$$

where θ_c is π and 2π for three-dimensional (3D) and two-dimensional (2D) random texture. We took $a = 0.2$ in all our calculations. Clearly for hard magnets where λ is small little magnetic reversal occurs, while for soft magnets where λ is large, up to 50% of magnetic moments are reversed.

Equations (7) and (8) are coupled and in general need to be solved numerically by iteration. We adopted Gaussian quadrature method for numerical integration in Eq. (8) with known α and bisecting method to solve Eq. (7) for known f.[11] To validate the model, we compared our analysis with the simulation results using micromagnetic code OOMMF developed at National Institute of Standards and Technology.[12] The simulations were conducted on ellipsoidal samples with dimensions $a_1 = a_2 = 300$ nm and $a_3 = 10$ nm, which were divided into cubic cells 10 nm long. For this geometry, the demagnetizing field $\overline{\mathbf{H}}_d$, although very small, was corrected using the demagnetizing factor $\mathbf{N}_\Omega$. 3D or 2D random distribution of easy axis was assumed among all the cells, with magnetization within each cells assumed to be uniform. The samples were saturated by applying a sufficient large magnetic field along the x_1 direction with the initial distribution of magnetization assumed to be random. The exchange constant was assumed to be zero, and the damping coefficient was taken to be 0.5. All the simulations were run on a Pentium IV based desktop computer.

First we demonstrate that our analysis indeed reveals the competition between magnetic-static energy and anisotropy energy, and the magnetic remanence of a wide range of materials is characterized by the dimensionless parameter λ alone when the exchange coupling is negligible, as shown in Fig. 1, where the remanence is plotted as function of K_1 for magnets with fixed λ and 3D random texture. Clearly, we observed that the remanence is independent of K_1 as we claimed regardless of their hardness, which is verified by the micromagnetic simulations. Our calculation agrees very well with the simulation for hard ($\lambda = 0.01$), intermediate ($\lambda = 0.8$), and soft ($\lambda = 10$) magnets, although larger variations

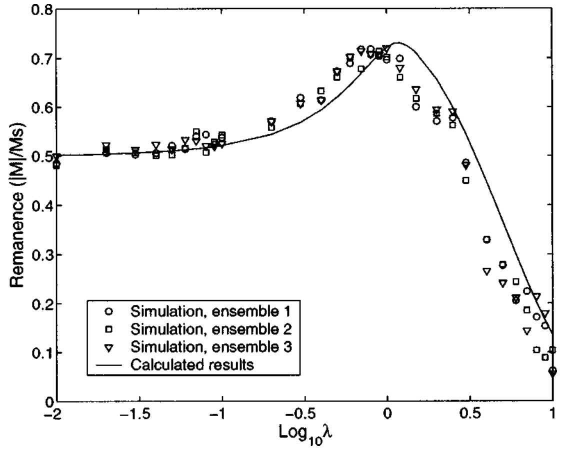

FIG. 2. The remanence of materials of 3D randomly oriented particles as a function of λ.

in simulation results are observed when the magnets getting softer. In addition, we notice that the remanence can be as high as 0.7 for magnets with intermediate hardness, 40% higher than that predicted by the noninteracting model.

To demonstrate the remanence enhancement due to the magnetostatic energy, the variation of remanence as function of λ for magnets with 3D and 2D random texture are demonstrated in Figs. 2 and 3. In both cases, we observe good agreement between our analysis and micromagnetic simulation over a wide range of λ, spanning from 0.01 to 10. Particularly, three regimes with distinct characteristics are observed. For λ increasing from 0 to 0.1, where the magnets are very hard, only slight increase of remanence is observed, suggesting little deviation of magnetic moment from the easy axis of individual particles. In this regime, anisotropy energy dominate and magnetostatic energy is negligible compared to the anisotropy energy, and Stoner and Wohlfarth's noninteracting model applies very well in this regime, which leads to $f=1/2$ for 3D random texture and $f=2/\pi$ for 2D random texture, agreeing with our calculation and magnetic simulation. As a result, we can conclude that for hard magnets, the remanence enhancement is purely due to the exchange coupling, as suggested in Refs. 7 and 9. For intermediate hardness where λ is between 0.1 and 1, marked enhancement of remanence is observed when λ is increased, where remanence as high as 0.75 can be obtained, representing 50% increase for materials with 3D random texture. This enhancement is due to the intergranular magnetostatic interactions, which was ignored by Stoner and Wohlfarth's noninteracting model. In this regime, anisotropic energy still has larger contribution than magnetostatic energy, but the magnetostatic energy is strong enough to lead significant deviation of magnetic moment from the easy axis. Interestingly, the highest remanence appeared to be approximately 0.75 for both 3D and 2D random texture, suggesting that 2D random texture has no obvious advantages over the 3D random texture for intermediate magnets. The highest remanence occurs near $\lambda=1$, where the anisotropy energy and magnetostatic energy have comparable contribution to the overall energy. As such, in this regime, intergranular magnetostatic interaction has a very important role in the remanence enhancement, which cannot be ignored. For soft materials with $\lambda>1$, we observed sharp decrease of remanence with the increase of λ. In this regime as λ increases, magnets become increasingly softer and anisotropy energy becomes more and more irrelevant, and particles tend to form intergranular closure domain to reduce the magnetostatic energy, leading to a sharp decrease in the remanence. In another word, for soft magnets where anisotropic energy is relatively small compared to the magnetostatic energy, intergranular magnetostatic interactions tends to decrease the remanence. In exchange-spring magnets with both hard and soft magnets, this would not be an issue since the magnetic moment of soft phase will be strongly modulated by the easy axis of neighboring hard phase, leading to large remanence enhancement. As a result, the microstructure of exchange-coupling magnets must be carefully tailored to take advantage of both intergranular exchange coupling and magnetostatic interactions for highest remanence enhancement possible, and this part of the work is currently undergoing.

To demonstrate the deviation of magnetic moment from the easy axis, we calculated anisotropy energy density of the assemblage in terms of λ, which measures the angle between the easy axis and the magnetic moment of particles, shown in Fig. 4 for 3D random texture, where K_1 is assumed to be fixed. We observe that for $\lambda<0.1$, anisotropy energy is approximately zero indicated by both our analysis and micromagnetic simulation, suggesting that magnetic moments are aligned along the easy axis of particles. For λ between 0.1 and 1, anisotropy energy increases with λ, suggesting that the magnetic moments start to deviate from the easy axis due to the magnetostatic interactions, and our calculation again agrees very well with the simulation. For soft magnets with λ larger than 1, significant deviation of magnetic moments from the easy axis occurs to reduce the otherwise large magnetostatic energy at the expense of anisotropy energy, leading to much higher anisotropy energy. In this regime, we predicted larger anisotropy energy than micromagnetic simulation. This is because in this regime, especially for very soft magnets, the magnetic moment is no longer modulated by the easy axis and can be regarded as isotropic, so that domain

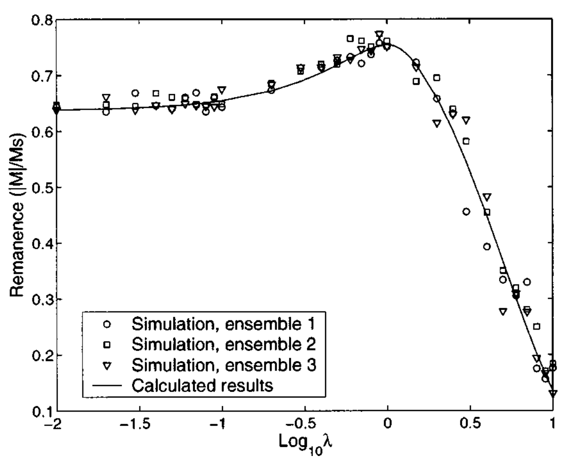

FIG. 3. The remanence of materials of 2D randomly oriented particles as a function of λ.

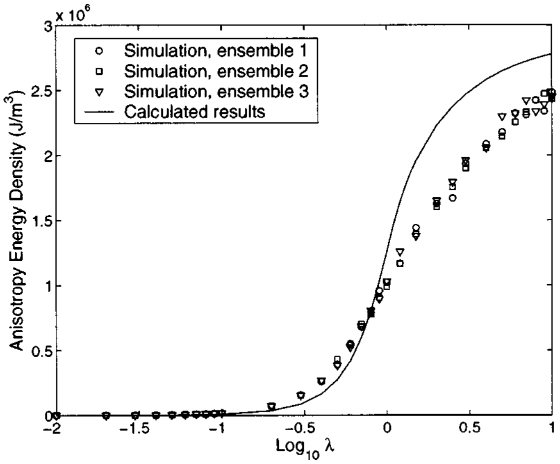

FIG. 4. The anisotropy energy of 3D randomly oriented particles as a function of λ.

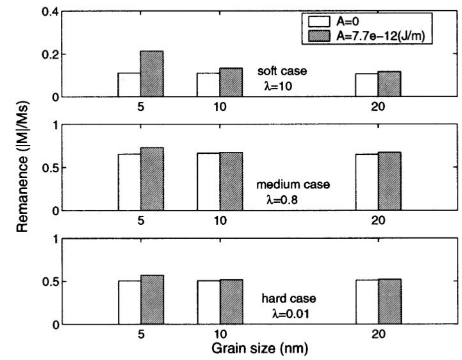

FIG. 5. The remanence of materials of 3D randomly oriented particles as a function of grain size, with and without exchange coupling.

closure occurs to minimize the intergranular magnetostatic energy. Such domain closure is not considered in our analysis. Nevertheless, the domain closure is not a problem in two-phase exchange-spring magnets, since the magnetic moment is strongly modulated by the easy axis of hard phase, and we expect significant deviation of magnetic moment of soft particles from their easy axis, leading to large remanence enhancement.

Finally, in order to justify our ignoring of exchange energy, we present in Fig. 5 the comparison of remanence for materials of different λ and grain sizes with and without exchange coupling, obtained by averaging three ensembles of micromagnetic simulations. The cubic cell size is taken to be the same as grain size. We used $A=7.7\times10^{-12}$ J/m in the simulation obtained from Ref. 7. It is clear that the exchange coupling leads to notable remanence enhancement only at 5 nm grain size, and the enhancement is most significant for soft magnets. Even more interestingly, for intermediate magnets with grain size as small as 5 nm, the remanence enhancement obtained from intergranular magnetostatic interaction is still larger than that obtained from exchange coupling. For materials with 10-nm grain size or larger, there is hardly any difference between magnets with and without exchange coupling, especially for small λ. It demonstrates that the intergranular magnetostatic interaction plays a very important role in the remanence enhancement of materials.

In summary, we have developed a theory to demonstrate that the remanence of magnetic particles can be enhanced by the intergranular magnetostatic energy if it is comparable to the anisotropy energy, but will be sharply reduced if the magnets get increasingly softer.

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