Coercivity and remanence in self-assembled FePt nanoparticle arrays[a)]

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FePt-based nanostructured materials are excellent candidates for high density recording beyond 1 Tbit/in^2. We calculate remanence, coercivity, and loop shape of annealed monodisperse FePt nanocrystals, using a modified Stoner–Wohlfarth model. To justify the simplifications of a Stoner–Wohlfarth model detailed finite element micromagnetic simulations were performed. Magnetic measurements on arrays of chemically synthesized FePt nanoparticles show remanence ratios of about 0.6 which is greater than that predicted for a series of noninteracting Stoner–Wohlfarth particles. A small fraction of the particles (5%) is assumed to remain in the disordered fcc phase with low magnetocrystalline anisotropy. Both remanence and coercivity are highly sensitive to the strength of the exchange interactions within a multiple twined nanocrystal. The calculated values are in the range from $J_r/J_s=0.52$, $H_c=0.77$ MA/m to $J_r/J_s=0.61$, $H_c=1.2$ MA/m. The results of the modified Stoner–Wohlfarth model are confirmed by finite element micromagnetic simulations taking into account magnetostatic interactions and allowing nonuniform magnetic structures within a particle. [DOI: 10.1063/1.1557398]

I. INTRODUCTION

Future high density magnetic recording relies on the ability to produce small high coercive particles. Recently, a solution phase chemical procedure has been developed to synthesize monodispersive FePt nanocrystals with controlled size and composition.[1] The magnetocrystalline anisotropy of the particles after annealing was found to be of the order of 10^6 J/m^3.[2] The large anisotropy, the small particle size, and the high packing fraction makes self-assembled FePt superlattices an ideal candidate for future high-density information storage media with an areal density in the Tb/in^2 regime. Synthesized FePt nanoparticles possess disordered fcc structures and are superparamagnetic at room temperature. At 5 K the FePt nanoparticles show ferromagnetic behavior. By raising the temperature the coercivity drops drastically. This is consistent with the small magnetocrystalline anisotropy of the disordered fcc phase. Heat treatment induces the Fe and Pt atoms to rearrange in long range ordered fcc structure that has good hard magnetic properties. Experiments with $Fe_{56}Pt_{44}$ nanoparticles annealing at 500 °C, 550 °C, and 580 °C showed a continuous increase of the coercivity with increasing annealing temperature. Transmission electron microscopy studies show that the phase transformation occurs at 530 °C.[3] The particles are randomly oriented. With increasing annealing temperature, the monodisperse particles coalesce during annealing and form multiple twined nanocrystals. The measured coercivity of optimally annealed $Fe_{55}Pt_{45}$ nanoparticles at 580 °C reaches 716 kA/m. The reduced remanence (J_r/J_s) exceeds 0.5 which indicates either some crystallographic texture or strong interactions between the particles. In summary, the hysteresis properties of annealed FePt nanoparticles show two characteristic features which are clearly different from the Stoner–Wohlfarth behavior for randomly oriented, noninteracting particles: A coercive field which is only about 16% of the Stoner–Wohlfarth value; a remanence ratio of about 0.6.

Harrell and co-workers[4] calculated the viscosity of self-assembled FePt nanoparticles using a noninteracting Stoner–Wohlfarth, Arrhenius–Neel model with a broad distribution of anisotropy fields and randomly oriented easy axes. Chantrell and co-workers[5] used a Monte Carlo method of an interacting fine particle system to study the magnetic properties of FePt nanoparticles. They calculated the temperature dependence of the hysteresis loop in a wide temperature range. The fit to the experimental data requires some nonrandom texture in order to explain the high remanence value of annealed FePt nanoparticles.

In this work we introduce a modified Stoner–Wohlfarth model to explain the properties of annealed FePt nanoparticles. Each multiple twined nanoparticle is represented by a triplet of three exchange coupled Stoner–Wohlfarth particles with mutually orthogonal easy axes. Magnetostatic interactions are neglected. Thus there is no interaction between particles of different triplets. The results are compared with a more general finite element model which takes into account both magnetostatic interactions and nonuniform magnetic states within the nanoparticles. The numerical experiments show that both remanence enhancement and coercivity reduction can be solely explained by exchange interactions between the crystallites of a multiple twined particle. In contrast to the well known result that the remanence increases with larger exchange, we found significant remanence enhancement only when the exchange interactions between the crystallites is reduced with respect to the bulk value of the exchange.

II. MODEL SYSTEMS

The hysteresis loop of the classical Stoner–Wohlfarth theory[6] follow from the subsequent minimization of the micromagnetic energy,

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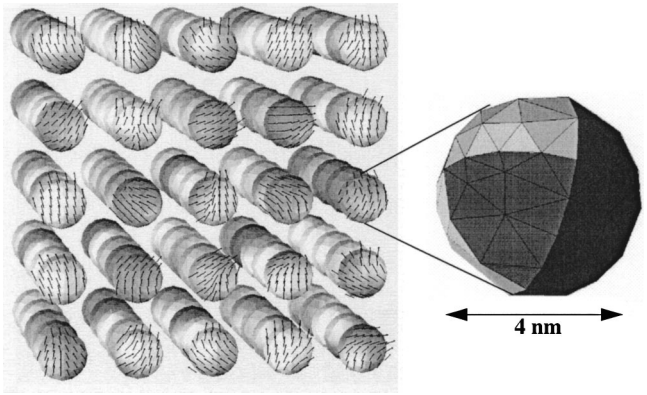

FIG. 1. Finite element model of self-assembled FePt particles. The arrows show the magnetization distribution for zero applied field. Owing to the exchange interactions between the grains which form an FePt sphere, the magnetization arranges nonuniformly. The enlarged sphere on the right-hand side shows the surface of the finite element mesh. The different colors correspond to crystallites with different easy axis direction.

$$E_{\mathrm{SW}}=\sum_i V_i\left(K_u\left(1-\frac{\mathbf{u}_i\cdot\mathbf{M}_i}{M_s}\right)^2-\mu_0\mathbf{H}_{\mathrm{ext}}\cdot\mathbf{M}_i\right) \tag{1}$$

for decreasing external field $\mathbf{H}_{\mathrm{ext}}$. In Eq. (1) the sum is over the particles in the particle ensemble. K_u is the uniaxial anisotropy constant, $\mathbf{u}_i$ is the anisotropy direction of particle i, and $\mathbf{M}_i$ is the magnetization vector of the particle. In addition to the classical energy terms of the Stoner–Wohlfarth theory we have to consider the exchange energy between the particles within a particle agglomerate. We assume that in average three particles agglomerate. According to transmission electron microscopy studies these three particles have mutually orthogonal anisotropy axes. The volumes of each practice are randomly assigned giving a total volume of a one agglomerate of $3(4\pi r^3/3)$, where r is the radius of one particle. Within an agglomerate the particles are exchange coupled to each other. The exchange energy between particle n and particle m is

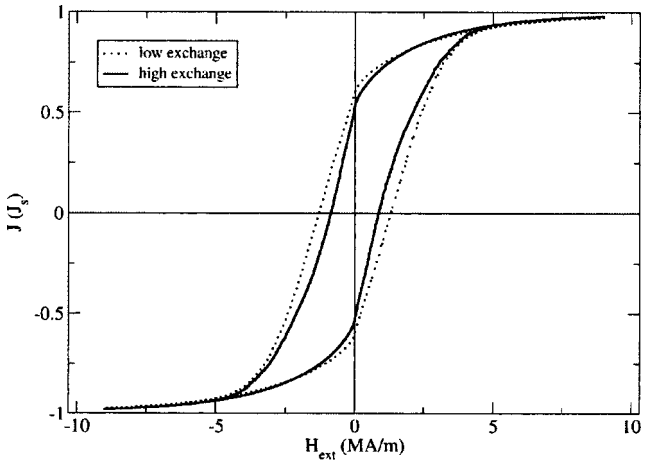

FIG. 2. Hysteresis loops for FePt particles calculated using the modified Stoner–Wohlfarth model for high exchange ($A=1.4\times10^{-11}$ J/m) and low exchange ($A=0.4\times10^{-11}$ J/m).

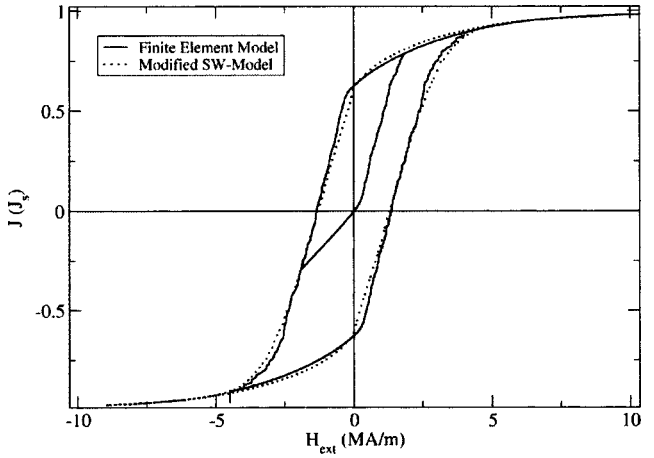

FIG. 3. Comparison of the hysteresis loops calculated from the modified Stoner–Wohlfarth model ($A=0.4\times10^{-11}$ J/m) and the finite element model ($A=0.1\times10^{-11}$ J/m). In addition a recoil curve calculated with the finite element model is shown.

$$E_{\mathrm{ex}}=-\frac{A}{a}F_{nm}\frac{\mathbf{M}_n\cdot\mathbf{M}_m}{M_s^2}. \tag{2}$$

Here F_{nm} is the contact area between the two particles,

$$F_{nm}=r_{nm}^2\pi=\left(\frac{r_n+r_m}{2}\right)^2\pi, \tag{3}$$

and a is the lattice constant which is $a=0.38$ nm for FePt. A small fraction of all particles are assumed to remain in the disordered fcc phase. To represent the fcc phase we simply set $K_u=0$.

The results of the modified Stoner–Wohlfarth energy are compared with a more complex finite element model.[7] A FePt particle is a sphere with a diameter of 4 nm. The sphere is assumed to be composed of three grains with three orthogonal easy directions. In total $5\times5\times5$ spheres form a particle array for the simulations. Figure 1 shows the particle array and the magnetization distribution for zero applied field. The grains are subdivided into tetrahedral finite elements. This finite element models allows to resolve nonuniform magnetic states within each grain. Both magnetostatic interactions between the spheres and exchange interactions between the grains are taken into account. However, for a interparticle distance of 2 nm the magnetostatic interaction were found to be negligible with respect to the anisotropy field. The dipolar interaction field is in the order of 160 kA/m which is only 2% of the anisotropy field. The hysteresis loop is calculated from the solution of the Landau–Lifshitz Gilbert equation for decreasing external field.

The material parameters for the calculations were as follows: $K_u=5.6\times10^6$ J/m^3, $\mu_0M_s=1.31$.[1] The exchange constant was varied in the range of $A=0.1\times10^{-11}$ J/m to $A=2.2\times10^{-11}$ J/m.

III. RESULTS

Figure 2 shows two hysteresis loops calculated with the modified Stoner–Wohlfarth model with high exchange interactions and low exchange interactions between the particles of an agglomerate. The results clearly show that with in-

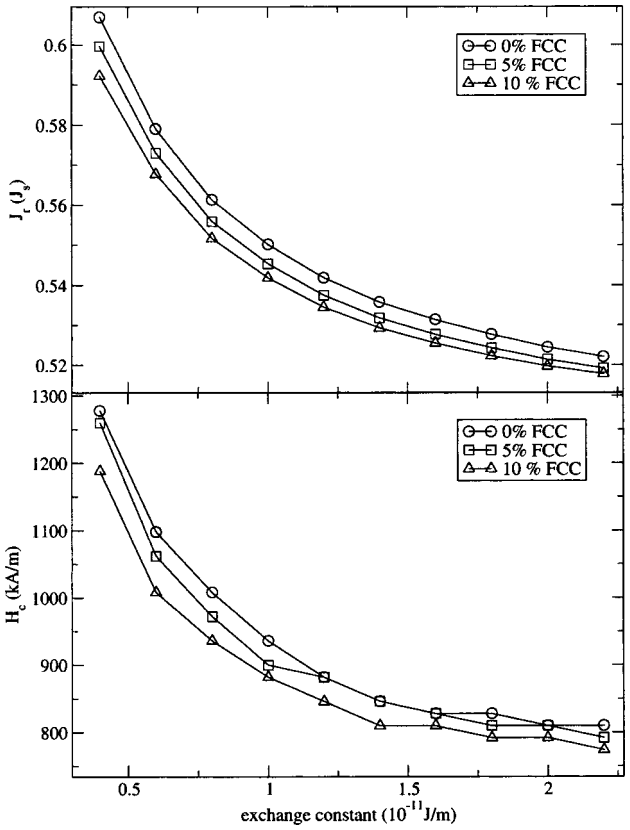

FIG. 4. Hysteresis properties as a function of the strength of the exchange interactions within a particle agglomerate. Top: Reduce remanence as a function of the exchange constant. Bottom: Coercive field as a function of the exchange constant. The error induced by the finite field step is in the size of the symbol.

creasing exchange interactions the coercivity drops reaching about 0.9 MA/m for an exchange of $A=1.4\times10^{-11}$ J/m. This is about 1/10 of the anisotropy field. The reduced remanence decrease from 0.6 to 0.53 when the exchange constant is reduced from $A=1.4\times10^{-11}$ to $A=0.4\times10^{-11}$ J/m. Thus the model qualitatively explains the enhanced remanence found in annealed FePt particle arrays. The increase of the remanence with decreasing exchange has to be attributed to nonuniform magnetic states within an agglomerate. The magnetization of the particles within a cluster are not parallel but point in slightly different directions. In the limit of high exchange interactions the cluster will be uniformly magnetized with a random easy axis. In that case the remanent magnetization will be that of the Stoner–Wohlfarth Model ($M_r/M_s=0.5$). This increase of the remanence with decreasing exchange is in contrast to the expected behavior. This rather peculiar result can be explained as follows: Since the crystallite size is smaller than the exchange length the magnetization becomes arranged uniform within a multiple twined particle for high exchange. Each multiple twined particle behaves like a uniaxial particle with uniaxial anisotropy. The situation is similar to nanocrystalline soft materials where the magnetization averages over several grains.[8] Since the uniaxial anisotropy is random, the reduced remanence is 0.5. Only if the exchange interactions between the crystallites of a triplet are reduced with respect to the bulk exchange the magnetization becomes nonuniform and remanence enhancement can occur. The finite element simulation confirms this behavior. For weak exchange the magnetization becomes arrange nonuniformly within a cluster of three grains as shown in Fig. 1. The particular microstructure of the multiple twined particles plays a minor role, since dipolar interactions are small compared to the magnetocrystalline anisotropy and the exchange interactions. Figure 3 compares the results of the modified Stoner–Wohlfarth model with those of the finite element approach. Loop shape, remanence, and coercive field are in good agreement.

Figure 4 summarizes the magnetic properties of FePt particles as a function of the exchange interactions. Both remanence and coercivity decreases with increasing exchange interactions. The exchange interactions between FePt particles due to agglomeration account for remanence enhancement and the drop of the coercive field with respect to the Stoner–Wohlfarth value. In addition Fig. 4 shows the effect of the remaining fcc phase. Both remanence and coercivity slightly decreases with increasing volume fraction of the fcc-phase.

ACKNOWLEDGMENTS

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