Numerical micromagnetics of an assembly of (Fe,Co)Pt nanoparticles

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A micromagnetic model has been developed describing the influence of the $K_1$ and the particle size on the magnetization reversal processes in (Co,Fe)Pt nanoparticles on the order of 4 to 80 nm in diameter. The magnetostatic interactions between the particles are calculated using an accelerated boundary element method. From the comparison of the results of the finite-element simulations with "macrospin" calculations in which only precessional rotational magnetization processes are possible, it is obvious that inhomogeneous rotational magnetization and nucleation processes are dominant in larger particles. Depending on the $K_1$ values and the volume fraction of the hard (fct) and soft (fcc) (Co,Fe)Pt nanoparticles, the calculated coercive field values are on the order of $\mu_0H_c=0.1$ to 1.2 T, in good agreement with experimental results. © 2005 American Institute of Physics. [DOI: 10.1063/1.1848452]

I. INTRODUCTION

Knowledge of the magnetization dynamics is of great importance for the understanding of the switching processes and the improvement of magnetic recording media and spin electronic devices. Micromagnetic simulations of switching behavior reveal the details of the magnetization distribution and dynamic magnetization reversal processes. Experimentally, in situ domain observation using Lorentz electron microscopy, magnetic force microscopy, and time-resolved magnetic imaging provides a detailed understanding of domain formation and reversal processes, but these techniques are extremely difficult to use for domain observations in the case of magnetic nanoparticles. $L_1_0$ ordered metallic phases such as FePt and CoPt exhibit very large magnetocrystalline anisotropy, and are regarded as the best candidates for the next generation high density recording media. As-prepared (Fe,Co)Pt nanoparticles synthesized by the chemical processing route exhibit a chemically disordered fcc crystal structure with a low magnetocrystalline anisotropy value $K_1$. Postannealing at temperatures above 500 °C leads to the transformation into the chemically ordered fct $L_1_0$ phase with large $K_1$ values on the order of $10^6$ to $10^7$ J/m$^3$. High coercivities over $\mu_0H_c=1$ T have been reported for particles and films with the magnetic grain size of around 10 nm.1-3 However, typically results in broad distributions of particle sizes, which may be further aggravated by agglomeration during annealing have been found experimentally.

In order to support the interpretation of experimental hysteresis curves of a powder containing a mixture of ordered and disordered nanoparticles, a finite-element (FE) micromagnetic model has been developed. The numerical results clearly show the influence of particle shape and size, the value of magnetocrystalline anisotropy, and the volume fractions of soft and hard magnetic particles on the hysteresis properties for several hundreds of nanoparticles. The numerical (FE) model based on the Landau–Lifshitz–Gilbert equation including thermal activation processes fully takes into account the long-range stray-field coupling and also the direct exchange coupling between the particles in highly packed particle assemblies. We present a three-dimensional micromagnetic simulation using a hybrid finite-element/boundary-element (FE/BE) model and taking into account realistic particle size and phase distributions consisting of a mixture of fcc and fct phases. The magnetostatic interactions between the particles are calculated using an accelerated BE method taking into account the realistic shape of the particles, which are on the order of 4 to 80 nm in diameter. From the comparison of the results of the FE simulations with macrospin calculations, in which only precessional rotational magnetization processes are allowed within the particles, it is obvious that inhomogeneous rotational magnetization and nucleation processes are dominant in larger particles and also depend on the $K_1$ values.

II. MICROMAGNETIC SIMULATION MODEL

In the full three-dimensional treatment, the time evolution of the magnetization follows the Landau–Lifshitz equation of motion, which describes the physical path of the magnetization $\mathbf{M}$ toward equilibrium. The Landau–Lifshitz equation contains an effective field that takes into account the external field, the demagnetizing field, the exchange field, and the anisotropy field. For the calculation of the demagnetizing field, a hybrid FE/BE method is used.8 Particles larger than the exchange length may lead to inhomogeneous reversal modes. In order to resolve the magnetization inhomogeneities, the particles have to be divided into tetrahedral finite elements9,10 with a constant edge length between 3 and 5 nm. In order to keep the discretization error small, the mesh size has to be smaller than the exchange length, which is determined by the Néel or Bloch wall parameters. On every node point of the FE mesh, the Landau–Lifshitz equation has to be solved. A backward differentiation formula is used for the time integration, which leads to a system of nonlinear

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equations. After applying Newton’s method, the resulting system of linear equations is solved using the scaled generalized minimal residual method.9

For particles on the order of the exchange length, the magnetization within each particle will deviate only slightly from a parallel orientation. In that limit, the simulation time can be decreased by applying the so-called “macrospin” approximation. Instead of assigning a magnetic spin to every node point of the FE mesh, only one spin called “macrospin” is related to each particle. The long-range magnetic stray-field coupling between the particles is accurately taken into account using the underlying FE mesh. Under what condition the macrospin approximation can be applied to particles larger than the exchange length is investigated here.

Figure 1 shows the size distribution of a mixture of 70% uniaxial fct phase and 30% fcc phase. Various sets of minimum–maximum values for the smallest diameter of the oblate ellipsoidal particles have been used for the calculations, such as 4–10, 8–20, 10–40, and 20–40 nm.

III. RESULTS AND DISCUSSION

The numerical simulations show that in the case of an assembly of 100% uniaxial (Co,Fe)Pt noninteracting, oblate nanoparticles, the coercive field is on the order of \( \mu_0H_c = 1.2 \) T. The remanence value of \( J_r = 0.5 \) T corresponds to the value predicted by the classical Stoner–Wohlfarth theory. Figure 3 compares the hysteresis curves of an assembly of 125 cubic and uniaxial particles. The numerical simulations using the macrospin model neglect inhomogeneous magnetization reversal processes inside the particles, but show only a slight difference to the full micromagnetic FE simulation in the case of large magnetocrystalline anisotropy. Figure 4 compares the hysteresis curves of macrospin simulations of 125 and 1000 randomly oriented particles with the FE simulations of 125 particles. Inhomogeneous magnetization reversal leads to a smaller coercive field value. A similar effect on the coercive field has been found in the simulations of a mixture of 50% uniaxial \((K_1 = 1.0 \times 10^6 \text{ J/m}^3)\) and 50% cubic \((K_1 = 1.0 \times 10^5 \text{ J/m}^3)\) particles coupled only by the long-range dipolar stray field. The hysteresis curves of Fig. 5 show two regions of reversal processes in the case of 10–40 nm particles. The soft magnetic cubic particles already start to reverse in the first quadrant, whereas the hard
magnetic ones are completely reversed only in the third quadrant. The transient magnetization processes at $H_{\text{ext}}=H_c$ clearly show the expansion of domain walls, especially inside the larger soft magnetic particles (Fig. 6). Increasing the size distribution by a factor of 2 and assuming a mixture of 80% uniaxial particles, the numerical macrospin simulations revealed an increase of the coercive field as shown in the hysteresis curves of Fig. 7. Varying the size distribution and the ratio of the volume fraction of hard and soft phases, the simulations show that in the range of 60% to 90% of the hard phase, the larger particle size distribution reveals an increase of the coercive field (Fig. 8). The reason for this behavior is the fact that in the case of a smaller concentration of soft particles the increased dipolar coupling leads to the increase of the coercive field.

Comparing the numerical results with experimental data, it is clear that the magnetocrystalline anisotropy values of the (Co,Fe)Pt nanoparticles and the size distribution mainly determine hysteresis properties. The numerical data are in good agreement with the shape of experimental hysteresis curves, coercive field, and remanence values. In summary, in the case of (Co,Fe)Pt nanoparticles on the order of 5–80 nm, the macrospin simulations for a large number of particles obtain comparable results with FE micromagnetic simulations that fully take into account inhomogeneous transient magnetization states. The main advantage of the macrospin simulations is the drastic reduction of computational time (Table I).

**ACKNOWLEDGMENTS**

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**TABLE I.** Dependence of the computational time for different numerical micromagnetic simulation methods.

<table>
<thead>
<tr>
<th>Simulation method</th>
<th>Particle size [nm]</th>
<th>Time [min]</th>
</tr>
</thead>
<tbody>
<tr>
<td>125 particles, FE</td>
<td>4–10</td>
<td>2054</td>
</tr>
<tr>
<td>125 particles, FE</td>
<td>8–20</td>
<td>6050</td>
</tr>
<tr>
<td>125 particles, macrospin</td>
<td>4–10</td>
<td>8</td>
</tr>
<tr>
<td>125 particles, macrospin</td>
<td>8–20</td>
<td>20</td>
</tr>
<tr>
<td>1000 particles, macrospin</td>
<td>4–10</td>
<td>390</td>
</tr>
<tr>
<td>1000 particles, macrospin</td>
<td>8–20</td>
<td>482</td>
</tr>
</tbody>
</table>

**FIG. 5.** Comparison of the hysteresis properties of a mixture of 50% fct (uniaxial) and 50% fcc (cubic) particles calculated with the macrospin model (1000 particles) and the FE model (125 particles).

**FIG. 6.** FE micromagnetic simulation shows the transient magnetization states within the mixture of 50% fct (uniaxial) and 50% fcc (cubic) particles during the reversal process at $H_{\text{ext}}=H_c$.

**FIG. 7.** Calculated hysteresis curves showing the dependence of the coercive field on the particle size of a mixture consisting of 80% fct (uniaxial) and 20% fcc (cubic) particles.

**FIG. 8.** Dependence of the coercive field on the ratio between hard (uniaxial fct) and soft (cubic fcc) phases and the size distribution of the particles.