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Hysteresis of ultrasmall Fe–Pt particles

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The magnetization reversal in very small FePt nanoparticles is investigated by analytical and numerical calculations. The modeling focuses on particles with diameters from 3 to 15 nm, as produced by a salt-matrix annealing technique. Experiment shows that the particles exhibit a certain degree of structural inhomogeneity, which has a far-reaching effect on the magnetic hysteresis. In particles larger than about 10 nm, the magnetization-reversal mode is strongly inhomogeneous, and there are several scenarios that depend on the symmetry of inhomogeneity. Small particles reverse nearly coherently, and the coercivity is essentially equal to the volume-averaged anisotropy. In this case, nonrectangular hysteresis loops reflect factors such as grain misalignment, particle-size distribution, and different degrees of $L_1_0$ order in different particles.

Figure 1 shows high-resolution TEM images of 3 and 8 nm FePt nanoparticles after annealing in a salt matrix at 700 °C for 4 h. It was found that the separating media (salt and surfactants) conserve the particle sizes during annealing. The standard deviation of the nanoparticle diameters is about 10%, based on particle-number averages obtained from the high-resolution TEM images. XRD and SAED patterns confirm the phase transformation from fcc FePt to $L_1_0$-ordered FePt; details about the crystal structure of the alloys have been published elsewhere.8,9

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Figure 2 shows room-temperature hysteresis loops for different particle sizes. The saturation magnetization in-
creases monotonously with increasing particle size, whereas the coercivity reaches a maximum at a particle size of 8 nm. The increase of coercivity for the 3–8 nm particles reflects the improved chemical ordering (\(L1_0\) order parameter), which translates into an enhancement of the magnetocrystalline anisotropy. The decrease of the coercivity for large particle sizes is of micromagnetic origin, associated with real-structure imperfections such as polycrystallinity and reduced anisotropy at the surface (see below).

Since we are primarily interested in the effect of imperfections, as contrasted to the magnitude and direction of the average anisotropy, we assume that the particles are \(c\)-axis aligned along the direction of the field \(H=Hc\). Starting from saturation, \(M=M_c, e\), nucleation involves a small magnetization component \(M_{\text{mol}}=M_m\), where \(m=m_c e_x+m_y e_y\). The components \(m_c\) and \(m_y\) are degenerate in fair approximation, so that we can restrict ourselves to the consideration of \(m=m\). As elaborated elsewhere, the nucleation modes obey the partial differential equation,

\[
-A\nabla^2 m + [K(r) - \mu_0 M_s/H/2]m = 0,
\]

where the lowest-order uniaxial anisotropy constant \(K(r)\) describes the real or defect structure of the magnet. (For homogeneous particles, \(K(r)=\langle K \rangle\), Eq. (1) reproduces coherent rotation, irrespective of domain-wall width and particle size.)

In Eq. (1), the magnetostatic interaction is incorporated into \(K\) (shape anisotropy) and \(H\) (interaction field). We also make the approximation \(H(r)=H_{\text{ext}} - DM\), that is, the magnetostatic self-interaction is described by a demagnetizing factor. This is reasonable because the particles are nearly spherical and magnetically hard. It has also been shown that exchange inhomogeneities \(A(r)\) have a relatively small effect on the hysteresis.\(^{15}\)

There are exact solutions of Eq. (1) for a number of planar and three-dimensional cases, but all calculations require an explicit knowledge of the “defect structure” \(K(r)\). What can we conclude about the nucleation mode from the symmetry of the grain imperfection, without explicit knowledge of \(K(r)\)? Some cases frequently encountered in practice are reduced surface or interface anisotropy, defects localized at grain or particle surfaces, and bulk defects.\(^{15,18}\)

To treat the problem perturbatively, we start from a homogeneous system where Eq. (1) assumes the form \(-\nabla^2 m - k^2 m = 0\) and, in spherical coordinates,
generally some admixture of “excited” states |e⟩, meaning that the reversal mode is only approximately coherent. The symmetry of |e⟩ depends on the defect structure K(r). Asymmetric defects, such as the dark circle shown in Fig. 3(d), involve a superposition of degenerate p-type modes |e⟩=|p⟩, which is then admixed to the uniform mode by ⟨0|K|p⟩=(3/4πR³)∫S₀(r)K(r)p(r)dV. The lowest-lying nonuniform symmetric mode s₁(r) or |e⟩=|1⟩, shown in Fig. 3(e), is more difficult to admix because it has a relatively large k. However, ⟨0|K|1⟩=(3/4πR³)∫S₀(r)K(r)s₁(r)dV may be quite large for inhomogeneities located at the particle surface, while ⟨0|K|p⟩=0 by symmetry.

Perturbation theory yields the nucleation field

$$H_n = \frac{2(K)}{\mu_0 M_s} - \frac{2|⟨e|K|0⟩|^2}{\mu_0 M_A k^2}.$$  (4)

Consider, for example, a thin shell of reduced anisotropy (shell thickness Δ). The reduction in the average ⟨K⟩ is proportional to Δ, whereas the reduction in the second term scales as Δ². The second term is therefore a small correction unless k is small (or R is large).

To investigate the magnetization reversal numerically, we have performed calculations using a C++ spin-relaxation code with built-in monitoring of subsystem magnetizations. Figure 4 shows the results of a typical systems. In the figure, the nanoparticle is divided into 552 numerical cells, as shown in the inset. To model the real structure, we have assumed a shell of reduced anisotropy. The respective shell thicknesses are 1 and 2 nm for the 10 and 20 nm particles, meaning that the shells occupy about half the particle volume in both cases.

The average anisotropy is assumed to be about 3.3 MJ/m³, that is, about 50% of the anisotropy Kmax of fully ordered FePt. However, the anisotropy is unevenly distributed: the core has 0.75Kmax, whereas the shell has only 0.25Kmax. The particle with D=20 nm exhibits a pronounced magnetization inhomogeneity during switching, as can be seen by comparing the shell magnetization (gray) with the full loop. By contrast, for 10 nm particles, the core-shell particle has nearly the same hysteresis loop as a homogeneous particle with K=0.5Kmax. Equation (4) contains a k³ dependence of the magnetic field, meaning that anisotropy-field difference is enhanced by a factor of 4 if the feature size doubles.

In conclusion, we have investigated the origin and realization of hysteresis in small FePt nanoparticles. In lowest-order theory, applicable to very small particles (D=3 to about 10 nm), the coercivity is given by the particles’ average anisotropy, irrespective of the geometry of the imperfection but including the particle-size dependence of the anisotropy. In particles larger than 10 nm, there is a clear distinction between imperfections nearly symmetrically distributed over the particle’s surface and localized defects. The analysis of the involved spherical Bessel functions reveals that the surface modes have a relatively high energy but are often easily excited by typical defects, due to the large fraction of surface atoms, and the same picture is reproduced by numerical simulations.

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