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Magnetization Reversal and Giant Coercivity in Sm-Co/Cu-Ti Particulate Films

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Magnetization Reversal and Giant Coercivity in Sm-Co/Cu-Ti Particulate Films

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***Abstract*—Highly coercive granular SmCo/CuTi films were produced by thermal processing of sputtered $SmCo_5/(CuTi)$ multilayers and investigated experimentally and theoretically. Electron microscopy shows that the processed films consist of spherical grains that have diameters of 5–10 nm and are embedded in a matrix. Both the grains and the matrix phase exhibit the $CaCu_5$ structure, but the matrix is probably Cu-rich. The films have high coercivities of up to 50.4 kOe at 300 K. Analytical calculations and micromagnetic simulations yield a transition from a nucleation-type weak-interaction regime to a discrete-pinning-type strong-interaction regime. The transition occurs at packing fractions similar to those encountered in the investigated films and is accompanied by a coercivity maximum.**

***Index Terms*—Coercivity, discrete domain-wall pinning, granular films, micromagnetic simulations, samarium–cobalt films.**

I. Introduction

CREATING coercivity in magnetic materials with moderate or high magnetization has been a challenge for many decades. For optimized materials, the order of magnitude of the coercivity H_c is given by the anisotropy field $H_a = 2K/\mu_o M_s$, where K is the first uniaxial anisotropy constant and M_s is the net spontaneous magnetization. However, real-structure features such as secondary phases and defects tend to strongly reduce the coercivity [1]–[3]. Past research has lead to the realization of various coercivity mechanisms, including nucleation [4] in cast magnets such as Nd-Fe-B, domain-wall pinning in strongly inhomogeneous magnets [5], and Stoner–Wohlfarth behavior in small particles. A particularly interesting system is SmCo where depending on additives and processing a variety of highly coercive regimes occur [4]–[8].

Industrial pinning-type SmCo magnets [5], as well as some titanium-containing magnets, exhibit a cellular microstructure, where 2:17 grains are surrounded by Cu-rich 1:5 boundaries. However, many systems, including granular magnets [9], are intermediate between pinning, coherent rotation and nucleation, and little work has focused on this regime. Here we focus on highly coercive granular SmCo/CuTi films obtained by thermal processing of sputtered $SmCo_5$/(CuTi) multilayers and investigate the origin of the coercivity.

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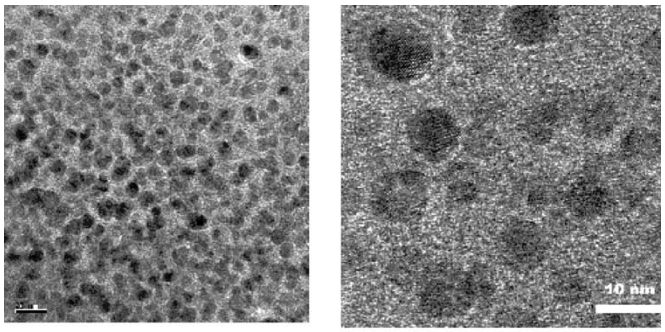

(a) Scale bar: 20 nm (b) Scale bar: 10 nm

Fig. 1. Electron micrographs of a granular $SmCo_5$/CuTi sample: (a) TEM and (b) HRTEM.

II. Structure and Magnetic Properties

The SmCo/CuTi films were sputtered on Si (100) substrates with a Cr underlayer of 90 nm and coverlayer of 18 nm. The structure of the multilayer is $(SmCo_5\ 45\ \text{Å}/\text{CuTi}\ X\ \text{Å}) \times n$, where $X = 2$–10 and $n = 38$–44, corresponding to a total thickness of the SmCo/CuTi layers of about 200 nm. X-ray diffraction, transmission electron microscopy (TEM), and high-resolution TEM were used to determine the crystal and microstructure of the films. Details of the processing and characterization of the films have been published elsewhere [10].

X-ray diffraction and TEM (Fig. 1) show that 30-min annealing at 525 °C yields the 1:5 structure with an average grain size of about 8 nm. The spherical grains with diameters of about 5–10 nm are surrounded by grain boundaries (matrix). Considering the microstructure of SmCo/CuTi permanent magnets [6], it is likely that a small amount of Ti facilitates the precipitation of a Cu-rich 1:5 phase in grain boundary. The grains in Fig. 1 probably have a structure and composition close to $SmCo_5$ and are embedded in a Cu-rich grain boundary phase. The X-ray diffraction patterns have been discussed in [10]; they correspond to the hexagonal $CaCu_5$ structure and indicate that both the grains and the matrix are in the $SmCo_5$ solid-solution range. Other annealed $SmCo_5$/CuTi multilayers show similar structure for CuTi layer thickness ranging from 2 to 12 Å. For the sample shown in Fig. 1, the estimated nominal composition is $SmCo_5(CuTi)_{0.6}$. The small amount of Ti plays an important role in realizing the coercivity. Annealed titanium-free films [10] do not exhibit the granular structure and their coercivity is comparatively low. Note that the granular structures do not exhibit the cellular structure encountered in other SmCo magnets.

The magnetic properties were measured using a SQUID magnetometer having a maximum field of 7 tesla. Fig. 2 shows in-plane hysteresis loops of a

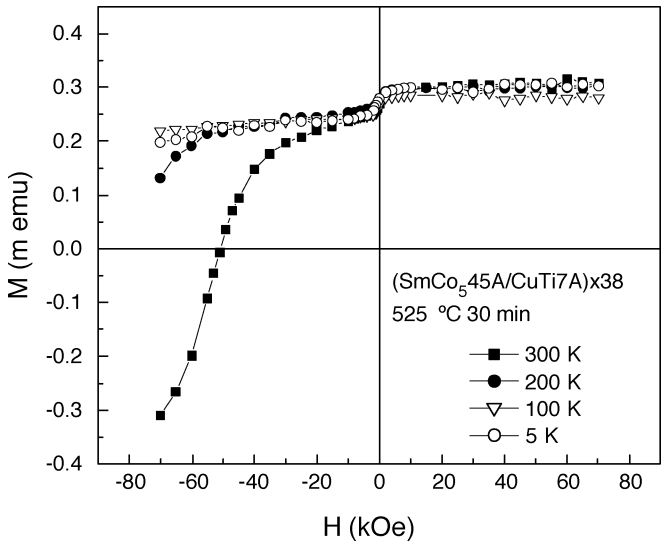

Fig. 2. Hysteresis loops of $SmCo_5$/CuTi at different temperatures (H_c = 50.4 kOe at 300 K).

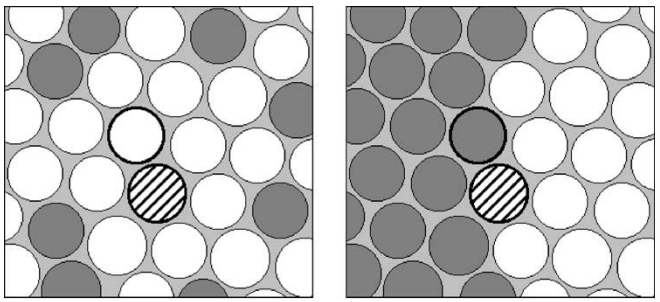

Fig. 3. Schematic spin structure during magnetization reversal in granular systems. (a) Localized nucleation. (b) Discrete domain-wall pinning. The bold-line circles represent the two sublattices taken into account in the analytical model calculation. The reverse field two-grain tries to change the magnetization direction from up (bright) to down (dark).

Cr 900 Å/($SmCo_5$ 45 Å/CuTi 7 Å) × 38/Cr 180 Å sample annealed at 525 °C for 30 min. The room temperature coercivity is 50.4 kOe. Because of the limitation of the 7-tesla field, this value corresponds to a minor loop, and the true coercivity is likely somewhat underestimated. At 200 K and below, the coercivities are larger than 7 tesla. The shape of the loop at 300 K indicates a crystal random distribution.

III. Theoretical Description

To understand the coercivity of granular systems it is necessary to consider the switching of the grains as a function of the external field. Fig. 3 shows a simple analytical model. On a mean-field level, it is assumed that the intergranular interactions can be described by a single coupling parameter [3]. There are two competing mechanisms: nucleation and discrete domain-wall pinning. The two cases are distinguished by local-environment effects. For example, nucleation means that the neighboring particles have a spin-up magnetization while the central particle switches. The switching and discrete-pinning fields are obtained by discretizing the micromagnetic equation

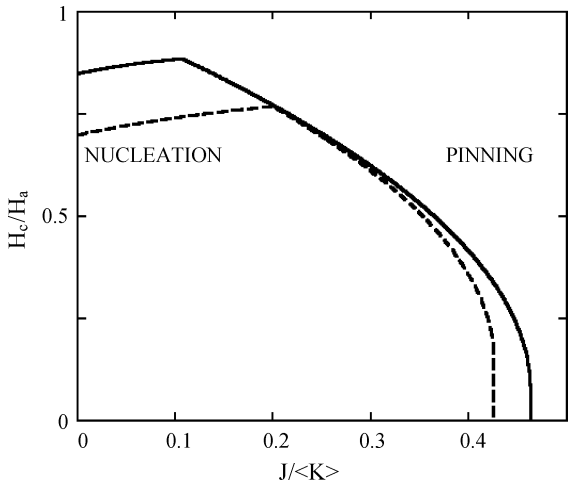

Fig. 4. Exchange dependence of the coercivity for relatively small disorder (solid line) and moderate disorder.

fields and analyzing the stability of the corresponding magnetization configurations. The calculation, which will be published elsewhere, yields the coercivity shown in Fig. 4. For small exchange, the magnetization reversal is realized by nucleation; in the interaction-free limit, this includes ordinary Stoner–Wohlfarth rotation. A characteristic feature is a coercivity maximum at the transition between nucleation and discrete domain-wall pinning.

In a numerical analysis, the material was modeled as an irregular distribution of $SmCo_5$ particles of average size of 8 nm on a hexagonal lattice. We considered two layers, each having 25 particles, embedded in a ferromagnetic medium which is characterized by material parameters differing from those of the particles. To model the disorder, we introduced slight randomness in the size, shape, position and in anisotropy of these particles. Using the OOMMF (Object Oriented MicroMagnetic Framework) NIST code (http://math.nist.gov/oommf/), the Landau–Lifshitz–Gilbert equation is solved for the modeled film at various magnetic field values.

The nucleation field is very high, more than 10 T. By contrast, the discrete pinning field is only about 4 T. This means that the magnetization directions of individual grains remain stable until a domain wall arrives. In a macroscopic thin film, any accidentally created or residual domain wall can sweep through the magnet, and the discrete pinning field is essentially equal to the coercivity. In terms of Fig. 4, this corresponds to the descending branches of the curves. (The nucleation field corresponds to analytic continuations of the ascending branches.)

Fig. 5 shows a snapshot of reversal through domain-wall motion, and Fig. 6 shows the calculated hysteresis for both types of reversal. The parameters taken are $M_s = 1000$ kA/m, $A = 30$ pJ/m, $K = 17 \times 10^6$ J/m^3 for the particles and $M_s = 700$ kA/m, $A = 25$ pJ/m, and $K = 2 \times 10^6$ J/m^3 for the matrix phase. This choice of parameters gives the coercivity of the modeled film to be 5 T which is close to the experimental value. Our calculations suggest that the exchange through the ferromagnetic matrix is quite strong and yields discrete domain-wall pinning type reversal.

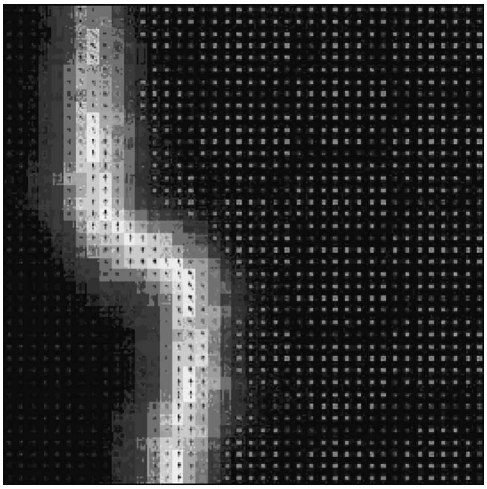

Fig. 5. Snapshot of a domain wall as obtained from micromagnetic simulations. The small dots are the cells of the numerical calculation; due to their small size, 1 nm, intragranular effects are taken into account in this simulation.

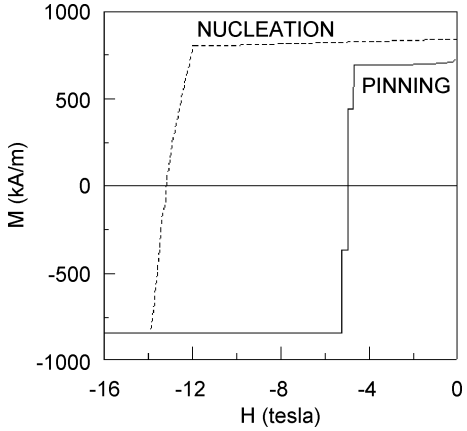

Fig. 6. Numerical hysteresis prediction for a SmCo/CuTi film.

IV. Discussion and Conclusions

A key feature of the coercivity of the granular structures is that the competition between disorder and exchange leads to a transition from nucleation for weak exchange to discrete domain-wall pinning for strong exchange. A very similar mechanism is probably realized in materials for high-density magnetic recording [11], [14]. As discussed in the context of grain-boundary exchange [3], the net interaction between the particles depends on the particle separation and on the chemistry of the matrix phase.

The discrete domain-wall pinning mechanism considered in this paper differs from ordinary strong pinning by the nature of the domain-wall propagation. In the present mechanism, the propagation is realized by jumps over individual grains and keeps some Stoner–Wohlfarth character on a local scale. By contrast, ordinary strong pinning is characterized by the propagation of locally well-defined (though generally distorted) domain walls.

Above the Curie temperature of the boundary phase, the ferromagnetic grains are surrounded by paramagnetic boundaries [7]. In the case of a paramagnetic boundary phase, the exchange between the grains decreases exponentially, as $\exp(-d/\xi)$, where d is the thickness of the grain–boundary region and ξ is the correlation length of the thermodynamical fluctuations [3], [12], [13]. With the exception of the immediate vicinity of T_C, ξ is of the order of about one interatomic distance, so that the propagation of domain walls through the paramagnetic phase is largely suppressed.

In conclusion, we have investigated granular Sm-Co-Cu-Ti with grain sizes of about 8 nm. The Ti plays an important role in realizing the nanostructure and coercivity. The high coercivity of the material, about 5.04 T (50.4 kOe), is explained in terms of a model where highly anisotropic small particles are coupled through a ferromagnetic matrix. The model yields a coercivity maximum corresponding to a transition from a nucleation-type weak-interaction regime to a discrete domain-wall pinning type strong-interaction regime.

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