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Magnetic reversal in $Sm_2Fe_{17}N_y$ permanent magnets

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Coercivity and nucleation fields for three-dimensional inhomogenities are calculated and discussed. For soft spherical inclusions in a hard magnetic matrix a $(\delta/R)^2$ dependence of the nucleation field on inclusion radius R and domain wall width δ has been obtained. To calculate propagation fields for spherical inhomogenities, a nonlinear model has been used. The corresponding coercive field can be much higher than the nucleation field, particularly if the magnitude of the anisotropy changes very steeply. Soft inclusions with sharp interfaces are ineffective at producing complete magnetic reversal. For partly nitrided particles coercivity and nucleation field are predicted to be identical, which is confirmed by experiments on high-performance $Sm_2Fe_{17}N_{3-\delta}$ magnets.

I. INTRODUCTION

Interstitial introduction of nitrogen into Sm_2Fe_{17} greatly improves the permanent-magnetic properties of the material. Filling the 9*e* octahedral interstices causes a lattice expansion of about 6% and leads to a Curie temperature increase of 350 K. Furthermore, the anisotropy due to the crystal field of the nitrogen atoms changes from moderate easy plane to strong easy axis.[1,2] To obtain magnets with appreciable energy product, the low coercivity of the nitrided powder ($\mu_0 H_c \approx 0.1$ T) must be improved. A possible way is Zn bonding,[3] which at present allows the production of aligned magnets with coercivities up to about 1.9 T.[4,5]

In any case, the observed coercivities are much smaller than expected from the $Sm_2Fe_{17}N_{3-\delta}$ anisotropy field $B_a \approx 22$ T.[6] To explain this behavior, which applies generally to hard magnetic materials, deviations from the ideal crystal structure, such as impurities and surface or grain-boundary phases, have to be taken into account.[7,8] Theoretical treatment of the coercivity problem is complicated, because it leads to nonlinear micromagnetic equations. There are estimations that predict a dependence proportional to δ/R of the coercivity on the domain wall width δ and on the radius R of the inhomogenity,[7–10] but the detailed coercivity mechanisms remain unclear.[7,8] In Ref. 9, the linearized micromagnetic equations have been used to calculate nucleation fields in one and two dimensions. This approach, which has recently begun to be extended to three dimensions,[11–13] allows the determination of magnetic properties directly from the microstructure. The question remains, however, whether the reversed nucleus can propagate and lead to reversal of the whole magnet (cf., e.g., Ref. 7).

Here we calculate nucleation fields for three-dimensional inhomogenities in aligned magnets and then use a nonlinear model to determine the corresponding propagation fields. The theoretical predictions are compared with experimental data on coercivity in homogeneously and inhomogeneously nitrided Zn bonded $Sm_2Fe_{17}N_{3-\delta}$ magnets.

II. THE LINEARIZED THEORY

To describe the magnetic reversal we use the free energy

$$F=\sum_{i=1}^{3}\int\left(A\frac{(\nabla M_i)^2}{M_0^2}-H_iM_i-K_1\frac{(M_in_i)^2}{M_0^2}\cdots\right)d\mathbf{r}, \tag{1}$$

where $H_i(\mathbf{r})$ denotes the local magnetic field, $M_i(\mathbf{r})$ with $M_0=|M_i(\mathbf{r})|$ the magnetization, $K_1(\mathbf{r})$ the second-order anisotropy constant, and $n_i(\mathbf{r})$ the unit vector of the local crystallographic c direction. The exchange parameter A is assumed to be spatially constant (cf., e.g., Ref. 9).[14] $H_i(\mathbf{r})$ includes magnetostatic effects in the form of a macroscopic demagnetization factor.

A magnetic configuration $M_i(\mathbf{r})$ may be realized physically if it corresponds to a local minimum of the free energy, Eq. (1). The nucleation field H_n is the field at which the free-energy minimum of the initial state disappears. At this field the initial configuration $M_i(\mathbf{r})$ becomes unstable, but the state that is reached next may be separated from the final configuration by other free-energy barriers so that complete reversal does not occur. Eventually, at $H_c \geqslant H_n$, all such barriers are removed, and the magnet is aligned in the $-c$ direction.

In the linearized theory, $n_x \ll 1$, $n_y \ll 1$, $M_x \ll M_0$, and $M_y \ll M_0$, and minimization of the free energy Eq. (1) yields

$$\mathbb{B}M_x=K_1(\mathbf{r})n_x(\mathbf{r})M_0+\tfrac{1}{2}H_xM_0^2, \tag{2a}$$

$$\mathbb{B}M_y=K_1(\mathbf{r})n_y(\mathbf{r})M_0+\tfrac{1}{2}H_yM_0^2, \tag{2b}$$

with the operator

$$\mathbb{B}(H_z)=-A\nabla^2+K_1(r)+\tfrac{1}{2}M_0H_z. \tag{3}$$

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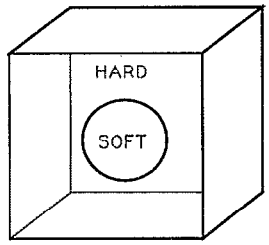

FIG. 1. Spherical soft inclusion in a hard-magnetic matrix.

An anisotropy profile $K_1(\mathbf{r})$ that can be treated exactly is the soft spherical inclusion in a hard-magnetic matrix (Fig. 1) with the anisotropy constants $K_1(r>R)=K_h$ and $K_1(r<R)=K_s \ll K_h$ of the hard and soft phases, respectively.[13] The nucleation field is given by the lowest eigenvalue of $\mathbb{B}$, and using the analogy between $\mathbb{B}$ and the Hamiltonian of the Schrödinger equation we obtain

$$H_n=\frac{2K_s}{M_0}\left(1+\frac{\delta^2}{R^2}\right), \tag{4}$$

with $\delta=\pi(A/K_s)^{1/2}$ being the Bloch wall width[15] of the soft phase. The quadratic term $(\delta/R)^2$ indicates a strong decrease of the nucleation field, compared to a δ/R dependence.

The nucleus can actually propagate in the hard-magnetic phase if the lowest-lying eigenstate $\Psi_0(r)$ of $\mathbb{B}$ is delocalized. Unfortunately, Eq. (4) corresponds to a localized state, and merely describes the switching of the soft inclusion.[13]

III. THE NONLINEAR CASE

To obtain a better description of the magnetic reversal, nonlinear effects have to be taken into account. With

$$M_i(\mathbf{r})=M_0[\sin\Theta(\mathbf{r})\cos\Phi(\mathbf{r})e_{xi}+\sin\Theta(\mathbf{r})\sin\Phi(\mathbf{r})e_{yi} +\cos\Theta(\mathbf{r})e_{zi}] \tag{5}$$

and $n_i=e_{zi}$ we obtain for $H_i=H_z e_{zi}$

$$F=\int [A(\nabla\Theta)^2+A\sin^2\Theta(\nabla\Phi)^2-H_zM_0 \times\cos\Theta-K_1\cos^2\Theta+\cdots]r^2\,dr. \tag{6}$$

In the following we restrict ourselves to spherical inhomogeneities, so minimization with respect to Φ yields $\Phi(\mathbf{r})=\text{const}$. $\Theta(\mathbf{r})$ can be expanded in spherical harmonics

$$\Theta(\mathbf{r})=\sum_{m,n=0}^{\infty} a_m^n(r)\,Y_m^n(\phi,\theta). \tag{7}$$

If we assume that the nucleus has the same symmetry as the inhomogenity (see Fig. 2), only the spherical Y_0^0 term survives, and Eq. (7) becomes

$$F=\int\left[A\left(\frac{\partial\Theta}{\partial r}\right)^2-H_zM_0\cos\Theta+K_1\sin^2\Theta+K_2\sin^4\Theta\right]r^2\,dr, \tag{8}$$

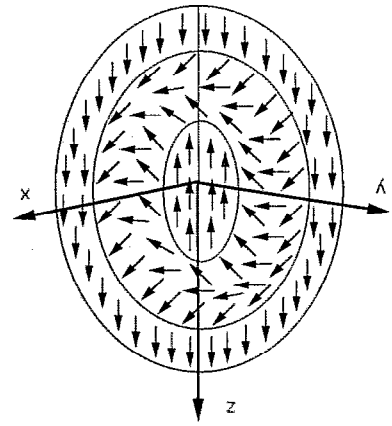

FIG. 2. Nucleus with spherical symmetry.

where the fourth-order anisotropy constant K_2 has been included.[16]

To minimize the free energy, Eq. (8), we use a trial function $\Theta(\mathbf{r})$ with a linear radial dependence between $\Theta(R_0-b)=0$ and $\Theta(R_0+b)=\pi$. Figure 2 shows that $\delta=2b$ is the thickness of a domain wall, which actually represents a cross between Bloch and Néel wall. To investigate the propagation of the nucleus, we assume $b \ll R_0$ and a sufficiently smooth dependence of K_1 and K_2 on r, so we finally obtain the propagation criterion

$$H>\frac{[4K_1(r)+3K_2(r)]\delta}{4M_0r}+\frac{\delta}{4M_0}\left(\frac{dK_1(r)}{dr}+\frac{3}{4}\frac{dK_2(r)}{dr}\right), \tag{9}$$

with

$$\delta(r)=\pi\sqrt{\frac{8A}{4K_1(r)+3K_2(r)}}. \tag{10}$$

To illustrate the result Eq. (9) we use the profile

$$K_i=K_{si}+\Delta K_i\tanh\frac{r-R_0}{B}, \tag{11}$$

where R and $2B$ can be interpreted as radius and interface thickness, respectively, of a spherical soft inclusion (index s) in a hard matrix with the anisotropy constants $K_1=K_{s1}+\Delta K_1$ and $K_2=K_{s2}+\Delta K_2$. In the limit $\delta \ll B \ll R_0$ Eq. (9) yields the coercivity

$$H_c=\frac{\delta}{M_0B}(\Delta K_1+\tfrac{3}{4}\Delta K_2). \tag{12}$$

Equation (12) indicates that the coercivity for large inhomogenities no longer scales as δ/R, which would give extremely small coercivities, but is determined by the "interface quality" δ/B (cf., e.g., Refs. 7 and 8). The sharper the interface, the larger the coercivity.

IV. COMPARISON WITH EXPERIMENT

$Sm_2Fe_{17}N_{3-\delta}$ provides an excellent basis for the study of magnetic reversal processes, because the local nitrogen

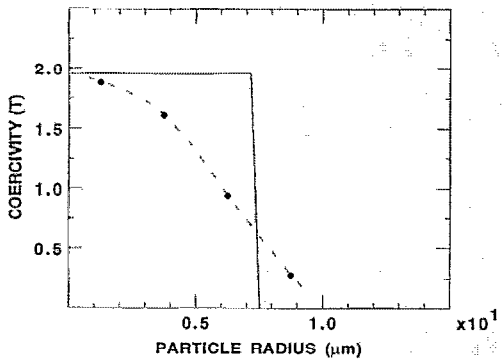

FIG. 3. Experimental (dashed line) and theoretical (solid line) results for the dependence of the coercivity on the radius of the grains at fixed nitrogenation conditions (5 h at 450 °C).

content, and hence the magnetic properties of the magnet, can be continuously varied by controlling temperature and pressure during the gas-solid reaction.[17]

For pure Sm_2Fe_{17}, K_1 is negative (easy plane), but as nitrogen enters the $9e$ sites it creates a strong electric field gradient at the Sm $4f$ shell. The crystal-field coefficients are modified and the anisotropy constants, in particular K_1, increase. At typical nitrogenation conditions (500 °C and 1 bar pressure), the anisotropy at the center of spherical particles changes from easy axis to easy plane at the nominal composition $Sm_2Fe_{17}N_{2.1}$.[17] Here we will use this "soft-core phenomenon" to check the validity of Eq. (9), which can be shown to predict that all grains with less than 2.1 nitrogen atoms per formula unit are soft.

To avoid interference with surface nucleation effects, which are probably responsible for the low coercivity in untreated and polymer bonded magnets,[3] high-coercive Zn-bonded magnets have been used. The experimental results are shown in Fig. 3 (dashed line). After sieving the powder fractions, nitrogenation, Zn bonding, and heat treatment coercivity values up to 1.9 T could be achieved, depending on the particle radius. For 5 h nitrogenation time at 450 °C the calculation predicts a sharp increase of H_c at $R \approx 7.5\ \mu m$ (Fig. 3). The smearing out of the experimental curve (dashed line) is due to the limited quality of the powder fraction. The plateau value $H_c \approx 1.9$ T, which cannot be predicted from the core properties, indicates further, unknown nucleation mechanisms.

V. CONCLUSIONS

The linearized micromagnetic equations can be used to calculate nucleation fields for three-dimensional inhomogeneities. The lowest eigenvalue of the linearized problem yields the nucleation field; but only if the corresponding eigenfunction is delocalized can the nucleation field be interpreted as the coercivity field. Localized eigenfunctions describe magnetization processes that cannot propagate in the bulk magnet.

To obtain more realistic coercivity values, nonlinear equations must be solved. For spherical inhomogeneities a criterion is obtained that predicts, for instance, immediate soft-core propagation in partly nitrided $Sm_2Fe_{17}N_y$ magnets. The corresponding lowering can be observed experimentally and underlines the importance of powder fractionation on properties and processing of the 2–17 nitrides.

ACKNOWLEDGMENTS

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