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MICROMAGNETIC ANALYSIS OF THE MAGNETIC HARDENING MECHANISMS IN RE-Fe-B MAGNETS

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Abstract. – The nucleation fields of sintermagnets are determined on the basis of micromagnetism taking into account the deteriorating effects of local stray fields, misaligned grains and reduced anisotropy constants of grain surfaces. It is shown that the nucleation hardening model gives a coherent description of the outstanding properties of RE-Fe-Nd magnets.

1. Introduction

The outstanding magnetic properties of modern High-Tech permanent magnets (pms) are determined by their intrinsic as well as their microstructural properties. Whereas the intrinsic properties as the spontaneous magnetization, $M_s$, the crystal anisotropy constant, $K_1$, or the Curie temperature, $T_c$, for a given intermetallic compound (Co$_8$Sm, Co$_{17}$Sm$_2$, Fe$_{14}$Nd$_2$B) can be improved only moderately by other alloying elements [1] secondary properties as the coercive field, $H_c$, the susceptibility $\chi$ or the remanence, $B_r$, depend sensitively on the microstructure of the materials [2, 3]. Therefore much efforts have been invested recently to analyse the effect of the microstructure on the quality of pms [4, 5]. In principle two lines have been pursued:

(1) improving the intrinsic material parameter as, e.g., the anisotropy constant $K_1$;

(2) optimizing the microstructure in order to suppress deteriorating effects on $H_c$.

During the last four decades in fact much progress has been made in developing materials with large crystal anisotropy constants. This procedure is suggested by the famous result of Brown [6] for the nucleation field for reversion of $M_s$ of a homogeneously magnetized particle

$$H_N = \frac{2K_1}{M_s} - N_{\text{eff}} . M_s,$$  \hspace{1cm} (1)

where $N_{\text{eff}}$ denotes an effective demagnetization factor being given by $N_{\text{eff}} = 4\pi$ for platelets and $N_{\text{eff}} = (-) 2\pi$ for elongated particles. In figure 1 we have represented the experimental coercive fields as a function of $K_1$. Figure 1 also contains the crystal fields $2K_1 / M_s$. Only in the case of Alnico the theoretical nucleation field is governed by the form effect being given by $2\pi M_s$. According to figure 1 the steady increase of $H_c^{\text{exp}}$ during the last four decades is nearly exclusively due to the increase of $K_1$ in the new compounds. In fact equation (1) predicts $H_c$ values being a factor of 4–10 larger than those realized in practice. This drastic discrepancy is known as Brown’s paradoxon [6], because from a theoretical point of view such a drastic reduction of $H_N$ is not expected. Until lately 10 % -20 % of the theoretical nucleation fields could be realized. Only in the new alloys (Co, Fe, Cu)$_{17}$ (Zr, Sm)$_2$ and Fe$_{14}$Nd$_2$B 40 % of the nucleation field could be obtained. Further improvement of pms is not so much a problem of the intrinsic material parameters but of the microstructure. This problem is most urgent in the case of FeNdB-pms because their low Curie temperature of $T_c = 576$ K leads to a strong temperature dependence of $H_c$ above room temperature.
2. The microstructure of sintermagnets

An ideal sintermagnet consists only of two phases: a ferromagnetic phase of high uniaxial anisotropy, e.g., Fe$_{14}$Nd$_2$B, and a nonmagnetic phase (Nd-rich) used for liquid phase sintering, which isolates the ferromagnetic grains perfectly from each other by a thin layer as schematically shown in figure 2. In addition the transition region between the two phases should take place within one atomic layer. Actually these rather strict requirements cannot be realized in technical pms where a large spectrum of “defect structures” contributes to the deterioration of the coercive field. Some of the main types of microstructural defects are the following ones: nonmagnetic phases (e.g., Fe$_4$NdB$_4$), misaligned grains (misalignments with a standard deviation of about 20° have been observed [2, 7]), incompletely magnetically decoupled grains due to a small fraction of the nonmagnetic phase, and finally a finite width of the transition region between the hard magnetic phase and the nonmagnetic phase. Because of the complexity of the problem we consider at first the nucleation fields of ideal grains, and then the effects of the above mentioned “defects”.

![Microstructure of an ideal sintermagnet](image)

**Fig. 2.** Microstructure of an ideal two-phase sintermagnet.

3. Nucleation fields in ideal grains

A comparison of experimental with theoretical results requires a detailed knowledge of the ideal critical fields for the reversion of the spontaneous magnetization. For a brief representation of these problems we consider a uniaxial single domain particle of ellipsoidal shape with the easy axis parallel to the rotational symmetry axis. The particle is characterized by demagnetization factors $N_T$ and $N_L$ for magnetization parallel or perpendicular to the easy axis. Assuming a uniform rotation process of $M_s$ the total magnetic Gibbs free energy density, $\phi'$, is composed of crystal energy, stray-field energy and magnetostatic energy in the external field, $H_{ext}$, which is assumed to be applied under an angle $\psi_0$ with respect to the negative easy axis. $\phi'$ depends on $\psi_0$ and the angle $\varphi$ between $M_s$ and the easy axis, and up to the third order anisotropy constant $K_3$, is given by

$$\phi' = K_1 \sin^2 \varphi + K_2 \sin^4 \varphi + K_3 \sin^6 \varphi + \frac{1}{2} M_s^2 \times (N_T \cos^2 \varphi + N_L \sin^2 \varphi) + M_s H_{ext} \cos (\varphi + \psi_0).$$  

The critical fields where reversal of $M_s$ takes place follow from the minimum condition

$$\frac{d\phi'}{d\varphi} = (K_1 + K_d) \sin 2\varphi + 2K_2 \sin^2 \varphi \sin 2\varphi + 3K_3 \sin^4 \varphi \sin 2\varphi - H_{ext} M_s \sin (\varphi + \psi_0) = 0,$$

and the stability condition

$$\frac{d^2\phi'}{d\varphi^2} = 2 (K_1 + K_d) \cos 2\varphi + 4K_2 (3 \sin^2 \varphi \cos^2 \varphi - \sin^4 \varphi) + 6K_3 \sin^4 \varphi (5 \cos^2 \varphi - \sin^2 \varphi) - H_{ext} M_s \cos (\varphi + \psi_0) \leq 0,$$

where we have substituted

$$K_d = \frac{1}{2} M_s^2 (N_L - N_T).$$

The following explicit results may be derived from equations (3) and (4) if $K_3$ is neglected [8]: for $H_{ext}$ applied antiparallel to $M_s$, $\psi_0 = 0$, from equation (3) it is found that $M_s$ starts to deviate from the easy axis at a first nucleation field of

$$H_{N1}^H = \frac{2K_1}{M_s} - M_s (N_L - N_T).$$

From equation (4) we find that the spontaneous rotation of $M_s$ into the opposite direction takes place at $H_N^H$ if the condition $(K_1 + K_d) > 2K_2$ holds, i.e., $d^2\phi'/d\varphi^2 < 0$. However, for $K_1 + K_d < 4K_2$ the condition $d^2\phi'/d\varphi^2 > 0$ holds and therefore $M_s$ at $H_N^H$ reversibly rotates out of the easy axis. This magnetic state becomes unstable for $d^2\phi'/d\varphi^2 = 0$ at an instability field (secondary nucleation field)

$$H_{N2}^H = \frac{4}{3\sqrt{6}} \frac{K_2}{M_s} \left(2 + \frac{K_1 + K_d}{K_2}\right)^{3/2},$$

and a critical angle of

$$\sin^2 \varphi_N = -\frac{1}{6} (K_1 + K_d - 4K_2) / K_2.$$

Applying equations (6) and (7) to the case of Fe$_{14}$Nd$_2$B it turns out that for $T > 272$ K $H_{N1}^H$ governs the rotation of $M_s$, whereas at lower temperatures $K_2$ increases so strongly that $H_{N2}^H$ determines the spontaneous rotation process. Figure 3 shows the temperature dependence of the nucleation fields over the whole ferromagnetic temperature range as determined from
equations (3) and (4) including the $K_3$-term. The material parameters where those determined on single crystals of $\text{Fe}_{14}\text{Nd}_2\text{B}$ [9].

4. Nucleation field for oblique applied fields

For oblique applied fields we have to solve equations (2) and (4) for arbitrary angles $\psi_0$ of the applied field. For vanishing anisotropy constants $K_2$ and $K_3$ the solution has been given by Stoner and Wohlfarth [10]. Under the oblique field $M_s$ rotates out of the easy direction, and the critical field where $M_s$ reverses spontaneously its direction is smaller than $H_N$. Taking into account $K_2$ we find from first order perturbation theory [11]

$$ H_N(\psi_0) = H_N^0 \alpha_{\psi}^{\text{nuc}} = H_N^0 \cos \frac{1}{\psi_0} \left( 1 + \left( \frac{\psi_0}{\chi} \right)^{2/3} \right)^{3/2} \cdot \left\{ 1 + \frac{2K_2}{K_1} \frac{\left( \frac{\psi_0}{\chi} \right)^{2/3}}{1 + \left( \frac{\psi_0}{\chi} \right)^{2/3}} \right\}. \quad (9) $$

Numerical results for the angular dependence of $H_N$ for different temperatures and including $K_2$- and $K_3$-effects are shown in figure 4. It is of interest to note that a minimum of $H_N$ in general is observed near $\psi_0 = \pi / 4$ and for $K_2 = K_3 = 0$, $H_N$ is reduced by a factor of 2 for $\psi_0 = \pi / 4$. The temperature dependence of the numerically determined minimum values, $H_N^{\text{min}}(\psi)$, is shown in figure 3.

5. Nucleation in inhomogeneous planar regions

If nucleation takes place in an infinitely extending planar region the rotation angle $\varphi$ of the lowest nucleation modes depends only on the $z$-coordinate and the linearized micromagnetic equation may be written as

$$ 2A(z) \frac{d^2 \varphi}{dz^2} - \left\{ 2K_1(z) - M_s \left( H_{\text{ext}} + N_{\text{eff}}M_s \right) \right\} \varphi = 0. \quad (10) $$

The first term of equation (10) results from the exchange energy, $A$ denoting the exchange constant. The strayfield, $N_{\text{eff}}M_s$, may be due to different sources [12], e.g., strayfields of nonmagnetic inclusions, misoriented grains, external surface charges or the selfstray field of the magnetic nucleus. $K_1(z)$ corresponds to the first crystal anisotropy “constant” varying along the $z$-direction. A suitable Ansatz for $K_1(z)$ leading to explicit results is given by [12]

$$ K_1(z) = K_1(\infty) - \frac{\Delta K}{\chi(z / r_0)^2}. \quad (11) $$

$K_1(\infty) = \text{anisotropy constant within the hard-magnetic phase}$, $\Delta K = \text{change of } K_1 \text{ at the centre and } r_0 = \text{width parameter of the inhomogeneity.}$ With equation (11) the eigenfunctions of equation (10) are the hypergeometric functions. These exist only for discrete values of $H_{\text{ext}}$. The lowest eigenvalue corresponds to the coercive field being given by [12]

$$ H_c = \frac{2K_1}{M_s} \alpha_K^{\text{nuc}} - N_{\text{eff}}M_s, \quad (12) $$

$$ \alpha_K^{\text{nuc}} = 1 - \frac{1}{4\pi^2} \frac{r_0^2}{\chi} \left[ -1 + \sqrt{1 + \frac{4\Delta K r_0^2}{A}} \right]^2. \quad (13) $$
with a fictitious wall width $\delta_b = \pi \sqrt{A/\Delta K_1}$. Over a wide range equation (12) writes for $2\pi \tau_0 > \delta_b$

$$H_c = \frac{2K_1(\infty)}{M_s} \left( \frac{\delta_b}{\pi \tau_0} \right) - N_{\text{eff}} M_s. \quad (14)$$

Equation (12) may be generalized to the case of misoriented grains including magnetic inhomogeneities, by replacing in equation (12) $\alpha_{K}^{\text{nuc}}$ by the more general $\alpha$-parameter $\alpha = \alpha_{K}^{\text{nuc}} \cdot \alpha_{\psi}^{\text{nuc}}$ where $\alpha_{\psi}^{\text{nuc}}$ is defined by equation (9). Since in real pms we are dealing with a distribution function of the misalignment angles $\alpha_{\psi}^{\text{nuc}}$ has to be replaced by certain averages $\langle \alpha_{\psi}^{\text{nuc}} \rangle$ the calculation of which depends on the type of magnetic coupling of the grains:

1. magnetically decoupled grains: in this case each grain with $H_c(\psi_0) < H_{\text{ext}}$ reverses its magnetization;
2. strongly coupled grains: if the grains are magnetically coupled because of incomplete nonmagnetic grain boundaries the grains with the smallest nucleation fields, i.e., with the smallest $\alpha_{\psi}^{\text{min}}$, ($\psi_0 \simeq \pi/4$) determine the bulk coercive field.

Since the $\alpha_{\psi}^{\text{nuc}}$-parameter shows a broad minimum (Fig. 4) for all angles $\psi_0$ there exist always enough grains with a small $\alpha_{\psi}^{\text{nuc}}$. According to these considerations the coercive field of a sintermagnet being characterized by misaligned grains and inhomogeneous magnetic regions is given by

$$H_c = \frac{2K_1(\infty)}{M_s} \alpha_{K}^{\text{nuc}} \left\langle \alpha_{\psi}^{\text{nuc}} \right\rangle - N_{\text{eff}} M_s. \quad (15)$$

$\left\langle \alpha_{\psi}^{\text{nuc}} \right\rangle$ corresponds to the minimum value, $\alpha_{\psi}^{\text{min}}$, if the grains are strongly coupled, and in the case of decoupled grains an average over all $\alpha_{\psi}^{\text{nuc}}$-values has to be calculated for which $H_c(\psi_0) \leq H_{\text{ext}}$ holds [7, 11].

6. Micromagnetic analysis and discussion of results

A satisfactory understanding of the magnetic hardening mechanism of pms requires the interpretation of a large spectrum of experimental results: temperature and angular dependence of $H_c$, field dependence of $H_c (H)$ of minor hysteresis loops, nucleation of reversed domains at grain boundaries and the formation of cascades of reversed domains.

Since the first attempts to produce pms on the basis of RE-transition metal alloys by the sinter technique it has been a matter of controversy whether the high coercive fields are due to strong domain wall pinning or to large nucleation fields. One of the most prominent experiments in favor of the nucleation model is the field dependence of the coercive field of minor hysteresis loops shown in figure 5 for three different compositions of FeNdB-type pms. In all three cases the magnetic field required to saturate $H_c (H_{\text{ext}})$ is found to be a factor of 2-4 smaller than the induced coercive field. This result also has been confirmed by Hiroawa and Sagawa [3] and is also supported by theoretical results for the maximum pinning field which should not exceed 1/3 of the nucleation field.

Figure 6 shows the angular dependence of $\alpha_{\psi}$-parameters for the nucleation (Eq. (9)) and the pinning model ($\alpha_{\psi}^{\text{pin}} = 1/ \cos \psi_0$) [11]. Included are experimental results for Fe$_{71}$Nd$_{20}$Al$_2$B$_7$. The average $\langle \alpha_{\psi}^{\text{nuc}} \rangle$ has been determined for a Gaussian distribution of $\psi_0$-values with a standard deviation of $\langle \psi_0 \rangle = 20^\circ$, taking care of the condition that all grains with $H_c(\psi_0) < H_{\text{ext}}$ have reversed their magnetization. Similar values of standard deviations were also used recently by Givord et al. [7, 13]. The rather flat angular dependence of $\langle \alpha_{\psi}^{\text{nuc}} \rangle$ reflects the fact that for all angles $\psi_0$ there exist always an appreciable number of $45^\circ$-misaligned grains with $\alpha_{\psi}^{\text{nuc}} \simeq 1/2$. In fact neither pinning nor nucleation ideally fits the experimental $\psi_0$-dependence of $H_c$. However, the nucleation model deviates not as strong from the experimental result as the pinning model.

A further rather sensitive test of the magnetic hardening models is the temperature dependence of $H_c$. For our analysis we have considered pms as produced by Vacuumschmelze, Fe$_{77}$Nd$_{15}$B$_8$, Sumitomo, Fe$_{77}$Nd$_{15}$B$_8$, and the MPI für Metallforschung (Fe$_{71}$Nd$_{20}$Al$_2$B$_7$). For testing the theoretical predictions of the nucleation model we consider the plot suggested by equation (15):

$$H_c^{\text{exp}} / M_s \text{ vs.} \frac{2K_1}{M_s^2} \alpha_{K}^{\text{nuc}} \left\langle \alpha_{\psi}^{\text{nuc}} \right\rangle. \quad (16)$$
Fig. 6. - Angular dependence of $\alpha^\text{nuc} \psi$ and $\alpha^\text{pin} \psi = 1 / \cos \psi$. The average $\langle \alpha^\text{nuc} \psi \rangle$ has been determined for a Gaussian distribution of the misalignment angles with a standard deviation of $\angle \psi = 20^\circ$. Experimental points ( - - - -) refer to the relative value $H_c (\psi_0) / H_c (\psi_0 = 0)$ of an Fe$_{71}$Nd$_{20}$Al$_2$B$_7$ pm.

where the left hand side corresponds to the experimental results and the right hand side to the theoretical results. For numerical calculations we assume magnetically strongly coupled grains, i.e., $\langle \alpha^\psi \rangle$ is replaced by the minimum value $\alpha^\text{min}$. Further parameters are the change $\Delta K_1$ of $K_1$ and the width $r_0$ of the nucleation region. Assuming a transition to a nonmagnetic grain boundary phase we may use $\Delta K_1 = K_1 (\infty)$. $\alpha^\text{min}$ is determined from equation (9) and the parameter $r_0$ appearing in $\alpha^\text{nuc}$ is chosen in order to obtain an optimum linear relation for equation (16). It should be noted that similar plots have been used previously by putting either $\alpha^\text{nuc} \langle \alpha \psi \rangle = 1$ [4] or $\langle \alpha \psi \rangle = 1$ [2]. From the slopes of these plots average values for $\alpha$ could be determined, however without relation to the microstructure. In the present case the only fitting parameter, $r_0$, turns out to lie in a range of $4 \text{ Å} < r_0 < 14 \text{ Å}$ as shown in figure 7, where the upper and lower bounds of linear behaviour of the plot according to equation (16) are represented. From the slopes of the linear regions in figure 7 we find $\alpha$-parameters of 0.6-0.7 which according to $\alpha = \alpha^\text{nuc} \cdot \alpha^\text{min}$ may be decomposed into $\alpha^\text{min} \approx 0.7$ and $\alpha^\text{nuc} \approx 0.9$. These rather large values of $\alpha^\text{nuc}$ indicate that the grains of the pm behave nearly as ideal single domain particles with nearly perfect surface regions. The deterioration of $H_c$ therefore predominantly results from misaligned grains and the demagnetization field $N_{\text{eff}}M_s$. From an extrapolation of the linear region in figure 7 we obtain the effective demagnetization factor, $N_{\text{eff}}$. For sintermagnets of Vacuumschmelze and of Sumitomo we find $N_{\text{eff}} = 7.2 \pi$ and for the FeAlNdB-pm of MPI, $N_{\text{eff}} = 6.4 \pi$. Previously it has been shown that this quantity in general is composed of three terms [12]

$$ N_{\text{eff}} = N_m + N_{\text{loc}} + 2\pi. \quad (17) $$

Here $N_m$ denotes a demagnetization factor due to the shape of the pm, $N_{\text{loc}}$ takes care of the grain's shape and the arrangement of the surrounding grains and the term $2 \pi$ is due to the self strayfield of the planar nucleus. For a spherical nonmagnetic inclusion with $N_{\text{loc}}(8\pi/3)$ and a platelike pm with $N_m = 4\pi$ we obtain $N_{\text{eff}} = 26\pi/3$ in agreement with the values derived from our analysis.


