

Proceedings of the Fifth Symposium on
Magnetism and Magnetic Materials

Recent Advances in Magnetism and Magnetic Materials

Taipei, Taiwan

19-20 April 1989

Editors: **H.L. Huang & P.C. Kuo**

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World Scientific

Singapore • New Jersey • London • Hong Kong

Published by

World Scientific Publishing Co. Pte. Ltd.,
P O Box 128, Farrer Road, Singapore 9128

USA office: 687 Hartwell Street, Teaneck, NJ 07666

UK office: 73 Lynton Mead, Totteridge, London N20 8DH

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and Magnetic Materials
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ISBN 981-02-0071-4

Printed in Singapore by Loi Printing Pte. Ltd.

Recent Advances in Magnetism and Magnetic Materials

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PREFACE

The Fifth Symposium on Magnetism and Magnetic Materials was held on the beautiful campus of the National Taiwan University in Taipei on April 19–20, 1989. The purpose of the Symposium is to bring together scientists and engineers and to provide them a forum for presentation, discussion, stimulation and exchange of ideas on the recent experimental and theoretical studies in the field of magnetism and magnetic materials. The scope of the Symposium covers all areas of basic and applied science related to magnetism such as those of the International Conference on Magnetism held in Paris, 1988, but much reduced in scale.

Altogether a total of 49 papers were presented at the Symposium. The papers collected in this volume represent the invited papers of which the full manuscripts have been received by the publication office by the deadline. A good fraction of these invited papers come from oversea scientists. The organization committee was very thankful for their participation which makes this scholarly gathering more enjoyable.

The organization committee wishes to express appreciation to the following institutions for their generous contribution and financial support:

The Ministry of Education, National Science Council, Material Research Lab., ITRI, Material R & D Center, Chung Shan Institute of Science and Technology, Atomic Energy Commission, China Steel Corporation and National Taiwan University.

Finally, special thanks are due to Professor H. C. Yang, V. C. Lee, P. C. kuo, J. H. Hsu, J. W. Chen, C. R. Chang, and Mr. C. S. Wang, among others, and many staff members and graduate students in the department of physics for their fine efforts, corporation and assistance throughout the Symposium.

Huei Li Huang
Chairman, 5M³ Symposium
October, 1989

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COERCIVITY MECHANISMS IN MODERN MAGNETIC MATERIALS

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ABSTRACT: In spite of tremendous activities in the field of high-coercivity and high permeability materials so far it has not been possible to develop materials approaching the theoretical limits of magnetic properties. Usually the real coercive fields amount only 15-25 % of the ideal nucleation field and in high-permeability amorphous materials the actual permeabilities amount almost 1 % of the theoretical limit. It is widely accepted that these discrepancies result from the microstructure of real materials.

On the basis of the theory of micromagnetism the effect of the microstructure on nucleation fields is treated quantitatively.

Examples for nucleation hardened magnets are $Fe_{14}Nd_2B$ and Co_5Sm based sinter magnets as well as melt-spun magnets. The coercive field of these materials is excellently described by $H_c = 2K_1\alpha/M_s - N_{eff} \cdot M_s$ (K_1 = first anisotropy constant, N_{eff} = effective local demagnetization factor). The parameters α and N_{eff} describe the effect of the microstructure on the nucleation field. From an analysis of the temperature and field dependence of H_c it is shown that $\alpha \sim 0.6$ is determined by three effects: 1. Misaligned grains. 2. Imperfect grain surfaces. 3. Incomplete magnetic decoupling of grains. The effective, local demagnetization factor results from strong field singularities at corners and edges of the grains and nonmagnetic precipitates. The improvement of permanent magnets by the elimination of these deteriorating defects will be demonstrated for the $FeNdB$ -type sinter and melt-spun magnets.

1. INTRODUCTION

The most efficient permanent magnets (pms) produced nowadays are based on the compaction or precipitation of small, magnetically saturated particles. Conventional magnets of this type are the $FeBa$ -ferrites and the Alnico magnets. In particular pms produced by sintering have been remarkably developed during the last two decades due to the discovery of a series of rare-earth transition metal intermetallic compounds of hexagonal^{1,2)} or tetragonal symmetry^{3,4)} with large magnetocrystalline anisotropy constants. The most important intermetal-

lic compounds which have become commercially relevant are Co_6Sm , $Co_{17}Sm_2$ and $Re_2Fe_{14}B$, $(Fe, Tm)_{12}RE$. In general these compounds are produced with benefitting additional components of low concentration, as e.g., Al , Cu , Ga , Nb , Zr and Mo . In particular the $Re_2Fe_{14}B$ compounds have become rather important because these are considered as an effective substitute of the rather expensive $CoSm$ alloys since Co and Sm are replaced by the much cheaper elements Fe and Nd . For the $FeNdB$ alloys commercial pms achieve at room temperature energy products of $(BH)_{max} = 30 - 35 KJ/m^3$ which exceed the $(BH)_{max}$ values of the $(CoSm)$ -type magnets. In spite of the large energy products of $FeNdB$ -type magnets, these pms have the disadvantage of a low Curie temperature, $T_c = 583$ K. It is this low Curie temperature which leads to a strong decrease of H_c with increasing temperature thus preventing the application of these magnets above $120^\circ C$. This disadvantage is not only due to the low T_c but also to the deteriorating effects of the microstructure which reduces the coercive field by a factor of 4 to 5 in comparison to the theoretical limit. Fig.1 shows the development of H_c -values of the different types of pms as a function of the first crystal anisotropy constant. It is obvious from Fig.1 that all improvements of H_c are due to the discovery of materials with larger K_1 -values. Nearly no progress has been made during the last two decades concerning the improvement of the microstructure. In order to solve this problem a basic understanding of the influence of the microstructure on the magnetic properties is required. This task has been attacked by experimental and theoretical micromagnetic techniques in a series of papers⁵⁻¹⁰). The results of these investigations will be summarized in the following.

2. NUCLEATION vs. PINNING HARDENING MECHANISMS

2.1 General Remarks

Since the first pms have been developed on the basis of the $Fe_{14}Nd_2B$ intermetallic compound controversial discussions took place concerning the dominant hardening mechanism in these materials. This discussion by no means is academic because in case of the one or the other active hardening mechanism quite different microstructures have to be prepared. Pinning mechanisms require the existence of precipitations of different magnetic properties whereas in the case of a nucleation mechanism the existence of less hard magnetic precipitations leads to a drastic deterioration of the coercive field. I.e., in the latter case the ideal microstructure consists of grains of the hard magnetic phase which are embedded within a planar

three-dimensional network of a nonmagnetic phase (see Fig. 2). In the case of *FeNdB*-magnets this phase corresponds to a *Nd*-rich *NdFe*-alloy for the sintered as well as the melt-spun magnetic materials. In some papers^{11,12}) it has been argued that in melt-spun magnetic materials the magnetic hardening occurs by pinning of domain walls, whereas in sintered pms the nucleation mechanism is considered to dominate¹³⁻¹⁵). As a matter of fact in literature in general different types of hardening mechanisms are discussed oftenly, however, the experimental facts supporting the one or the other mechanism are treated rather superficially. The main hardening mechanisms are the following ones:

1. The nucleation mechanism is characterized by the spontaneous reversion of the magnetization of a grain under the influence of an inversely applied magnetic field, the so-called nucleation field.
2. The pinning mechanism is characterized by the immobilization of domain walls by grain boundaries or precipitations.
3. The combined nucleation and pinning mechanism is characterized by the pinning of the dws of small nuclei which opposes the expansion of the nuclei.

For a decision which one of these mechanisms determines the coercive field we have to consider the microstructure of the different types of pms, melt-spun and sintered ones, as well as the characteristic magnetic properties related to the one or the other hardening mechanisms.

2.2 Magnetic Structure of Sintered and Melt-Spun PMS.

Fig. 3 represents the domain pattern of an aligned, sintered, demagnetized magnet showing grains of diameters up to $10 \mu\text{m}$. The critical diameter for single domain particles is give by $D_c = 1.4\gamma/\mu_0 M_s$ ($\gamma =$ specific wall energy = 240.10^3 J/m^2 , $\mu_0 M_s =$ spontaneous polarization = 1.6 Tesla). In the case of *Fe₁₄Nd₂B* we obtain $D_c = 0.2\mu\text{m}$, i.e., nearly all grains in Fig. 3 are multi-domain particles as observed experimentally. On the other hand in melt-spun materials many grain diameters are below $D_c = 0.2\mu\text{m}$, and consequently here we are mainly dealing with single domain particles. Whereas in sintered magnets the demagnetized state is established by a splitting of each grain into domains, in melt-spun materials the demagnetized state may be established by the random distribution of the easy axes of the single domain grains and a splitting of grains into domains if these have diameters $D > D_c$. In melt-spun materials each hard magnetic grain is found to be magnetically decoupled from the neighbouring grains by a *Nd*-rich phase whereas

in the case of sintered magnets this is only partially true. As may be observed in Fig. 3 many neighbouring grains show domain patterns running through both grains. Furthermore it should be noted that in aligned pms there are always some grains showing a strong misalignment.

2.3 Magnetization Curves And Coercive Field

One of the most convincing experimental result in favour of the nucleation mechanism is the field dependence of the coercive field as measured by means of minor hysteresis loops. Fig. 4 shows H_c as a function of the maximum applied field for three different compositions of $FeNdB$ -based sintered magnets. All three pms show qualitatively the same behaviour: Up to a critical field of $\mu_0 H_{ext} \approx 0.3 - 0.5$ Tesla H_c remains small (< 0.10 T). For larger fields, however, H_c increases rapidly and saturates at far larger values of H_c than the applied field. This behaviour is compatible with a nucleation mechanism where the magnetic saturation is achieved in each grain very easily by domain wall displacements, however, the reversion of \underline{M}_s of the saturated grains requires much larger applied fields. Since obviously the pinning forces are small the dw displacements take place at much smaller fields than the actual coercive field which is determined by the nucleation field of the magnetically saturated grains. The magnetic field, H_{sat} , which is required to obtain saturation of $H_c(H_{ext})$ may be considered as the magnetic field required to obtain magnetic saturation of the grains. This field is mainly determined by the local demagnetizing field acting on each grain. In the case of an isolated grain the average local demagnetizing field is given by $(-N_{||}M_s)$ with $N_{||}$ corresponding to the grains demagnetization factor parallel to the easy axis. For spherical grains with $N_{||} = 1/3$ this gives $\mu_0 H_{sat} = \mu_0 N_{||} M_s = 0.53$ Tesla. This value approximately corresponds to the experimentally determined saturation field. It is therefore selfsuggesting to assume that the local average demagnetization field is nearly independent of the surrounding grains, i.e., the surrounding neighbouring grains produce only a minor strayfield due to the randomness of the magnetization directions within the domains either parallel or antiparallel to the positive c -axis.

These considerations clearly show that in sintered pms the demagnetization process occurs by a nucleation process at a magnetic field much larger than the saturation field H_{sat} . Once the domain walls are pushed out from the grains the magnetic field required for reversion of magnetization abruptly increases up to the value of the nucleation field.

Similar investigations in the case of melt-spun materials so far have not led to an as clear interpretation of results. Since the applied field required for obtaining saturation of H_c corresponds just to the value of H_c itself, it has been seducing to assume that melt-spun pms are governed by a pinning mechanism. For a clarification of this situation we have measured the virgin and the demagnetization curves of melt-spun materials of different grain sizes¹⁶⁾. By applying different quenching rates by varying the wheel frequency melt-spun materials with grain sizes varying between $0.1\mu m$ and several μm s were obtained. These materials therefore contain single domain grains ($< 0.2\mu m$) as well as multi-domain grains. As a consequence the virgin magnetization curve is composed of two regions: At low fields the multi-domain grains are saturated as in the case of sintered pms giving rise to a rather steep magnetization curve as shown in Fig. 5. The following flat increase of the magnetization is due to reversible rotation of M_s within the misaligned grains. At magnetic fields of ~ 2.4 Tesla a second steep increase of the magnetization is observed being due to the reversion of M_s in single domain grains. This process is characterized by the nucleation field of single domain grains. If the magnetic field is reduced it turns out that the critical field of steepest demagnetization just corresponds to the magnetic field of the second step in the virgin magnetization curve. This may be taken as a proof that this field indeed corresponds to a nucleation field. We may consider this critical field as the microscopic coercive field which is somewhat larger than the macroscopic coercive field defined by $J = 0$. Our present interpretation furthermore is supported by the fact that in materials containing a larger amount of single domain particles the second step in the virgin magnetization curve increases (see Fig. 5). In conclusion we may summarize that the behaviour of the virgin and of the demagnetization curve in melt-spun materials supports the nucleation mechanism as the relevant hardening mechanism in these materials.

2.4 Micromagnetic Background Of The Nucleation Field

The temperature dependence of H_c of $FeNdB$ -type magnets is characterized by a monotonous decrease to rather small values of about 0.1 Tesla at $200^\circ C$. Using different compositions and appropriate additives according to Fig. 6 the situation may be improved¹⁷⁾. Best results are obtained for Al and/or Dy doped materials giving coercive fields of 0.4 Tesla at $200^\circ C$. These latter results show up a possibility how to improve the presently reduced applicability of $FeNdB$ -type

pms above 100°C. For a directed improvement of H_c a full understanding of the temperature dependence of H_c is required. In the case of ideal grains with the first anisotropy constant, K_1 , spontaneous magnetization, \underline{M}_s and demagnetization factors N_{\parallel} and N_{\perp} parallel and perpendicular to the easy axis, the nucleation field for an applied field antiparallel to \underline{M}_s is given by

$$H_N^I = \frac{2K_1}{M_s} - (N_{\perp} - N_{\parallel})M_s \quad (1)$$

Here it should be noted that eq. (1) holds for $K_1 > 4K_2$, where K_2 corresponds to the second anisotropy constant. This case applies for $Fe_{14}Nd_2B$ above $T = 272K$. Below this temperature $K_1 < 4K_2$ holds¹⁸⁾ and the nucleation field is given by^{19,20)}

$$H_N^{II} = \frac{4}{3 \cdot \sqrt{6}} \frac{K_2}{M_s} (2 + (K_1 + K_d)/K_2)^{3/2} \quad (2)$$

with $K_d = (N_{\perp} - N_{\parallel})/M_s^2$.

In Fig. 7 the temperature dependence of H_N resulting from eq. (1) and eq. (2) is represented over the whole temperature range below T_c . In the lower part of Fig. 7 some experimental results are shown demonstrating the serious discrepancy between the theoretical predictions and the actual situation. It is the aim of a more realistic micromagnetic theory of the nucleation field to give a quantitative interpretation of the temperature dependence of H_c by taking into accounting the effect of the microstructure on the nucleation field. This indeed seems to be a rather difficult task because the following microstructural effects have to be considered:

1. Inhomogeneities of the anisotropy constant K_1 at the grain surface.
2. Inhomogeneous dipolar strayfields, $H_d(r)$, near edges of the grains.
3. Misaligned grains with misalignment angles ψ .
4. Incompletely magnetically decoupled grains.

For a solution of this complex problem we have to start from the linearized micromagnetic equation^{19,20)} for the angle ψ of \underline{M}_s with respect to the easy axis

$$2A\Delta\psi - \{M_s H_{ext} \cos \psi_0 + M_s H_d(r) - 2K_1(r) \cos 2\psi_0 + 4K_2(r)(3 \sin^2 \psi_0 \cos^2 \psi_0 - \sin^4 \psi_0)\} \psi = 0 \quad (3)$$

Eq. (3) has been solved for the case of a one-dimensional as well as a two-dimensional nucleation process. The nucleation field is defined as the largest

eigenvalue of H_{ext} where a deviation from a homogeneously magnetized state becomes possible. In the special case of a one-dimensional inhomogeneity where the anisotropy constant as shown in Fig. 8 within a region of width, r_0 , is described by a relation

$$K_1(z) = K_1(\infty) - \frac{\Delta K}{ch^2(z/r_0)} \quad (4)$$

($K_1(\infty) = K_1$ within the unperturbed matrix, $\Delta K =$ change of K_1 at the grain surface) the nucleation field or the coercive field writes

$$H_c = \frac{2K_1}{M_s} \alpha_K \alpha_\psi - N_{eff} M_s \quad , \quad (5)$$

where the microstructural parameters have the following meaning:

i) α_K takes care of the magnetocrystalline inhomogeneity and is given by

$$\alpha_K = 1 - \frac{1}{4\pi^2} \frac{\delta'_B{}^2}{r_0^2} \left[-1 + \sqrt{1 + \frac{4\Delta K r_0^2}{A}} \right]^2 \quad , \quad (6)$$

where $\delta'_B = \pi\sqrt{A}/K_1(\infty)$ denotes the fictitious wall width of the uniaxial crystal. Numerical results for α_K are shown in Fig. 9 for varying parameters.

ii) α_ψ takes into account the misalignment of ψ_0 , of the grains and may be written as

$$\alpha_\psi = \frac{1}{\{(\cos \psi_0)^{2/3} + (\sin \psi_0)^{2/3}\}^{3/2}} \cdot \left[1 + \frac{2K_2}{K_1} \frac{(tg \psi_0)^{2/3}}{1 + (tg \psi_0)^{2/3}} \right] \quad . \quad (7)$$

iii) The effective demagnetization factor, N_{eff} in the case of a planar nucleus with a one-dimensional rotation process (see Fig. 8) is given by

$$N_{eff} = 2\pi + N_{loc} \quad , \quad (8)$$

where the term 2π results from the volume charges of the nucleus itself and N_{loc} takes care of the strayfields of the surrounding grains. In the case of a spherical nonmagnetic neighboring grain $N_{loc} = 8\pi/3$ holds, thus giving $N_{eff} = 14\pi/3$.

The complex expression (6) for α_K can be approximated by $\delta'_B/\pi r_0$ over a wide range $2\pi r_0 \leq \delta'_B$ giving

$$H_c = \frac{2K_1}{M_s} \frac{\delta'_B}{\pi r_0} - N_{eff} M_s \quad . \quad (9)$$

With the wall energy $\gamma = 4 \cdot \sqrt{AK_1}$ eq. (9) may be rewritten as

$$H_c = \frac{\gamma}{2\pi r_0 M_s} - N_{eff} M_s \quad (10)$$

Eq. (10) allows another interpretation of the nucleation field which primarily has to be considered as the largest eigenvalue of the micro-magnetic equation. Rewriting eq. (10) in the form $H_c M_s + N_{eff} M_s^2 = \gamma/2\pi r_0$ it becomes evident that nucleation takes place if the field energy of external and stray field just corresponds to the specific wall energy per unit length (or to total wall energy per volume of the nucleus).

Eq. (5) applies for the case where all grains have the same orientation. In reality, however, even in aligned sintered magnets we deal with an appreciable distribution of the alignment angles ψ_0 . A quantitative analysis shows that the standard deviation of ψ_0 is of the order of $\langle \psi_0 \rangle = 20^\circ$. Now we have to consider two cases:

- i) Magnetically decoupled grains behave individually and for a given external field all grains reverse its magnetization for which the condition $H_c(\psi_0) < H_{ext}$ holds. In this case α_ψ in eq. (5) has to be replaced by an average value $\langle \alpha_\psi \rangle$ taking into account the distribution function of ψ_0 .
- ii) If the grains are magnetically coupled because of incompletely formed non-magnetic intergranular phases the grains (Fig. 3) with the smallest α_ψ^{min} determine the bulk coercive field. These are the grains with misalignment angles $\psi_0 \sim 45^\circ$.

In fact according to our present knowledge in general there exists a large number of incompletely decoupled grains giving rise to cascades of demagnetization processes running over many grains once a strongly misoriented grain reverses its magnetization by a nucleation process. The coercive field therefore is given by

$$H_c = \frac{2K_1}{M_s} \alpha_K \alpha_\psi^{min} - N_{eff} M_s \quad (11)$$

Eq. (11) takes care of a large number of microstructural effects deteriorating H_c . The parameter α_K describes the effect of magnetic inhomogeneities at the grain surface where K_1 may have strongly reduced values. α_ψ^{min} takes into account the misalignment of grains as well as the existing incomplete decoupling of grains. The effective local demagnetization factor describes the role of strayfields which results from surface charges of the grain surfaces and from the volume charges of the generating magnetic nucleus.