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FACTORS GOVERNING THE COERCIVITY OF SINTERED ANISOTROPIC M TYPE FERRITE

F. Kools

Philips - ELCOMA, B.G. Materials, Central Development, 5600 MD Eindhoven, The Netherlands

Abstract - A new model for the magnetization reversal of a polycrystalline material is proposed. It is based on the conception that this reversal is governed by the growth of large multicrystal reversed regions. Quantitative elaboration (first approximation) leads to a simple expression for the coercivity \( H_c \) which reflects the influence of the microstructure and also of intrinsic factors. The existence of large multicrystal reversed regions as well as the fact that their growth generally governs the \( H_c \) is demonstrated experimentally. The proposed expression for the \( H_c \) has been tested by some preliminary experimental investigations. Most of the implications are confirmed in first approximation, e.g. the relation of the \( H_c \) with the remanence \( B_r \), the grain size and the anisotropy field \( H_A \).

I - INTRODUCTION

Hexaferrites or "M" ferrites are characterized by the chemical formula \( \text{MeFe}_3\delta \) (\( \text{Me} = \text{Ba}, \text{Sr} \) or \( \text{Pb} \)) and the hexagonal "Magnetoplumbite" or "M" crystal structure. They are widely applied as permanent magnet materials. Most important are the grain oriented or "anisotropic" materials. Large scale production is concentrated on sintered anisotropic M ferrites /1/. The development of sintered anisotropic M ferrites is focused on improving remanence (\( B_r \)) and coercivity (\( H_c \)). In this paper we shall concentrate on the \( H_c \) and investigate theoretically the possibilities to improve the \( H_c \) in particular with respect to the microstructure.

It is generally known that high \( H_c \) requires a small grain size. The influence of other microstructural factors such as porosity and crystal shape, however, is not known in detail. There is also no clear picture about the influence of intrinsic factors such as the anisotropy field (\( H_A \)) and the saturation magnetization (\( J_s \)). This is due to the fact that detailed knowledge about the physical mechanism underlying the reversal of the polycrystalline material is lacking. The available knowledge connected with the magnetization reversal of sintered anisotropic M ferrites can be classified in three categories: (a) theoretical and experimental studies on separate (small) crystals /2-5/, (b) empirical knowledge on polycrystalline materials /6-8/, (c) theoretical and experimental studies which intend to provide a bridge between categories "a" and "b" /9/. In this study we will try to contribute to the knowledge in category "c." We shall develop (sect.2) a semiquantitative model on the reversal of sintered anisotropic M ferrites which uses the knowledge of category "a" and which is compatible with the data from category "b."
II - THEORY

We consider a sintered anisotropic M ferrite having a crystallite size of about 1 μm and a demagnetization curve with a sharp knee in the 2nd quadrant. We shall concentrate on factors governing the irreversible demagnetization. First a set of statements is given (sect 2.1). The first 5 statements underlie the 6th one, where the proposed view on the magnetization reversal of a polycrystalline material is described in qualitative terms. We do not want to suggest that these statements are valid under all circumstances. We shall rather use them as postulates in order to be able to focus on the implications of statement 6. In sect. 2.2, statement 6 is elaborated quantitatively in first approximation, resulting in a rather simple expression for the magnetization coercivity \(H_{cJ}\) of the polycrystalline material.

2.1 Statements

1. In the remanent state the critical diameter for single domain behaviour is > 1 μm; typical crystallites (≈ 1 μm) do not contain stable walls (Fig.1 and ref.10).

2. Magnetization reversal of a typical crystallite proceeds by nucleation and growth of reversed domains, nucleation being the limiting factor /3/.

3. Exchange interaction between neighbouring crystallites is negligible /11/.

4. The internal field needed for nucleation of a wall ("internal nucleation field", \(H_{ni}\)) increases with decreasing size (thickness) of the (unstrained) crystallite and is roughly proportional to \(H_{A}/12,13/.

5. In the first part of the demagnetization curve up to the knee magnetization reversal is limited to some scattered locations characterized by exceptional conditions: low nucleation fields (e.g. giant crystals) and/or high local demagnetizing fields (e.g. sharp edges).

6. The second part of the demagnetization curve, beyond the knee, corresponds to (fast) growth of multicrystal reversed regions. At the boundary of a reversed region, unreversed crystallites are (partly) surrounded by reversed neighbours (Fig. 2). In that case the field of the neighbours (\(H_{n}\)) and the demagnetizing field of the unreversed crystallite (\(H_{D}\)) cooperate with the external counter field (\(H_{e}\)). When the joint counterfield exceeds the internal nucleation field (\(H_{ni}\)) of the concerned crystallite, it will be reversed. When the average joint counterfield exceeds the \(H_{n}\) of the majority of the crystallites, the reversed region can grow without limitation. This condition governs the coercivity (\(H_{cJ}\)). It refers to typical values of \(H_{ni}\), \(H_{D}\) and \(H_{e}\) (in contradistinction to statement 5). The proposed condition for the unlimited growth of a multicrystal region presupposes that crystallites with relative high \(H_{ni}\) are (partly) surrounded by reversed crystals with lower

![Diagram](image)

**Fig. 1.** Stability of walls in an anisotropic polycrystalline material in dependence of the counter field strength. \(H_{cJ}\), \(H_{ni}\) values according to a 1st approx. calculation for typical conditions: \(l = 1 \mu m, d = 0.6 \mu m\), \(J/J_{S} = 0.8\).

**Fig. 2.** Magnetic conditions for an unreversed crystallite in a (ellipsoidal) cavity of reversed material, just at the boundary of a (growing) large multicrystal reversed region (schematically).
The growth of the reversed region around unreversed crystallites is favoured by the fact that walls are not stable near \( H_e = H_{CJ} \) (Fig. 1). When in that case a wall is nucleated in a crystallite, the whole crystallite is reversed. In this way a reversed region can penetrate an unreversed region! Multicrystal reversed regions have been supposed to exist in ferrite magnets /9,14/ and microscopic regions have actually been observed for other materials /15,16/; but no author did base an expression for the \( H_c \) on this phenomenon.

2.2 Expression for the coercivity (elaboration of statement 6, see Fig. 2)

The demagnetizing field \( (H_D) \) is given by the expression

\[
H_D = N \mu_0 J_s^{-1}
\]

(1)

where \( N \) is the demagnetization factor of the concerned crystal, \( J_s \) the saturation magnetization and \( \mu_0 \) the vacuum permeability. Values for \( N \) are taken from ref. 17, assuming the platelike shaped crystallites /1/ to be circular discs. For example, we find \( N = 0.60 \) when the thickness to length ratio \( d/l = 0.3 \) and \( N = 0.36 \) when \( d/l = 0.8 \).

The Lorentz field \( (H_L) \) is in fact the sum of the field contributions of all surrounding crystals. We will apply some strong simplifications:

1. the surrounding reversed material is assumed to be homogeneous and infinitely extended in vertical direction with average magnetization

\[
\langle J \rangle = J_r = B_r = k_2 J_s
\]

(2)

where \( J_r \) is the remanent magnetization and \( B_r \) the remanent induction.

2. the cavity in which the concerned crystal is located corresponds exactly with the room taken up by the crystal itself.

Under these two conditions we may write

\[
H_L = N \mu_0 J_s^{-1} = N k_2 J_s \mu_0^{-1}
\]

(3)

Referring to statement 6 we state that when a typical crystallite is reversed under the conditions as depicted in Fig. 2, the reversed region can grow without limitation. In formula, we write as the condition for polycrystalline reversal:

\[
H_t = H_{ni}
\]

(4)

where \( H_t \) is the (average) counterfield acting upon the crystal and \( H_{ni} \) is the (average) internal nucleation field. The total field \( (H_t) \) is composed of different contributions: the external field \( (H_e) \) the demagnetizing field \( (H_D) \) and the Lorentz field \( (H_L) \). For the present, we restrict ourselves to measurements in a yoke, so that the (external) demagnetizing field from the outer surface of the product can be neglected. In addition we have restricted ourselves to the case \( H_e/H_D/H_L \) (Fig. 2), so we may write

\[
H_t = H_e + H_D + H_L
\]

(5)

The external field \( (H_e) \) for which bulk reversal occurs, is by definition the coercivity \( (H_{CJ}) \). Combining this with eqs. (4) and (5), we get:

\[
H_{CJ} = H_{ni} - H_D - H_L
\]

(6)

Using eqs. (1) (2) and (3) and the approximation

\[
H_{ni} = k_1 H_A \quad \text{(statement 4)}
\]

(7)

where \( k_1 \) is dependent on the crystal size, eq. (6) results finally in

\[
H_{CJ} = k_1 H_A - N(k_2^{-1} + 1) B_r \mu_0^{-1}
\]

(8)

Based on general experience we introduce the empirical formula

\[
H_{CJ} = C - \alpha B_r \mu_0^{-1}
\]

(9)

According to the proposed model, the constants \( C \) and \( \alpha \) are given by:

\[
C = H_{ni} = k_1 H_A
\]

(10)

\[
\alpha = N(k_2^{-1} + 1)
\]

(11)
In these expressions we recognize various $H$ determining factors. Expression (10) implies that the constant $C$ is:

- dependent on particle size (thickness) since $H_{nl}$ increases with decreasing particle thickness.
- independent of the measuring temperature, since $H_{A}$ is nearly independent of the temperature up to about 300°C /1/.
- dependent on substitutions which affect $H_{A}$.

Expression (11) implies that the constant $\alpha$ is:

- dependent on the crystal shape, since $N$ is governed by the crystal shape.
- dependent on $k_2$, i.e. on the density, the degree of alignment and the amount of non magnetic 2nd phase.

Since both $C$ and $\alpha$ are related to the microstructure, they can not be derived experimentally from materials sintered under different conditions. Making use of the implication that $C$ is independent of the measuring temperature they can be determined by measuring $H_{C,j}$ and $B_{T}$ at different measuring temperatures (see sect. 3).

III - EXPERIMENTAL

The existence of multigranular reversed regions was investigated by examining partially demagnetized samples with a small Hall probe and a "magnetic field viewer" foil. The latter can display differently polarized regions on macro scale. Partial reversal was performed by using a yoke equipment. The used samples were flat shaped with a thickness of about 10 mm and a pole area of about 10 square cm. For measurements on the temperature dependence of $B$ and $H$, as well as quantitative microstructural analysis, a group of 12 materials was selected from available samples. These materials are characterized by the same chemical composition but by a different processing and different final properties (Table Ia). The group consists of 4 different powder families (labelled A to D) which have been sintered for 3 different times (labelled 1 to 3).

Demagnetization curves were measured at 26°C, 50°C and 94°C by using a PAR Vibrating Sample Magnometer. Polished and thermally etched cross sections parallel to the preferred axis were examined quantitatively by applying a semi-automatic planimetric method. The number of counted grains was about 700. Finally some available $B$ and $H$ values of Al$_2$O$_3$ substituted materials were selected. These materials differed only in Al$_2$O$_3$ content (0.5 - 2.5 wt.%). Their processing was the same.

Table I Characteristics of the examined material group
(a) Relative density ($d$) and magnetic values (224°C).
(b) Grain size distribution ($d_{50}$, $d_{90}$) and average shape factor $S_{h}$.
\[ d_{50} = \sqrt{S_{h}} \]
\[ d_{90} = \sqrt{S_{h}} \]
\[ S_{h} = 0.5 \pi S_{h}/S_{c} \]
where $S$ and $F$ are the planar grain area and circumference respectively.
(c) Remanence and coercivity at different temperatures.
(d) Values for $C$ and $\alpha$ (formula (9) and Table Ic) and for $N$ (formula (11)), using $k_2$ and $\alpha$ values from Tables Ia and Id.

Table II Remanence and coercivity for a series of Al$_2$O$_3$ substituted materials (from Mr. Jordan, M.M.C. Southport U.K.).
IV - EXPERIMENTAL RESULTS

Multicrystal reversed regions were actually observed. Their size and shape was studied in relation to the degree of reversal \((J/J_0)\), varying from +1 to -1, Fig.3. It appeared that the first reversed regions become visible when \((J/J_0) \geq 0.9\). Continued reversal results first in growing together of the separate regions to one closed region near the outer side of the sample and subsequently in a growth of this region to the centre. Fig. 4 shows as an example of the actual regions when \((J/J_0) \approx -0.2\).

Quantitative examination of the microstructure showed that the distribution of the grain section areas \((S)\) is log normal. Such a distribution is characterized by the median \((S_{50})\) and the spread \((\sigma)\). From the value of \(S_{50}\) the corresponding value of the particle size \((d_{50})\) has been derived, which is used as a measure for the average particle size. Table Ib summarizes the \(d_{50}\) and \(\sigma\) values as well as a measure for the crystal shape (average "shape factor", \(S_n\)).

Table Ic shows the \(B_r\) and the \(H_{cJ}\) values of the 12 samples for the different measuring temperatures, while in table II the \(B_r\) and \(H_{cJ}\) values of some Al\(_2\)O\(_3\) substituted materials are collected.

V - DISCUSSION OF RESULTS

The existence of large multicrystal reversed regions has been proven (Fig. 3,4). It is not only demonstrated that there exist macro regions with different polarity, but also that these regions are extended from the top to the bottom of the sample, like pillars (cf. Fig. 4a and 4b). In addition it is indeed observed that not the initiation but the growth of these regions determines the \(H_{cJ}\), since in a broad \((J/J_0)\) region around \((J/J_0) = 0\), growth is the only observed process. The initiation of reversed regions occurs only at localities with low \(H_{cJ}\). This was verified by cutting parts from the sample and re-examining the first reversed regions. The field at which the first reversed regions appear is not much smaller than the \(H_{cJ}\). So, it is possible that for very homogeneous materials initiation of the first regions determines the \(H_{cJ}\). Compared with the multicrystal regions as observed for co-axial rare earth materials by using Kerr effect /15, 16/ the regions in Mn ferrite are much larger.

Plotting of \(B_r\) versus \(H_{cJ}\) for different measuring temperatures (Table Ic) results in straight lines as also reported earlier/18/. This supports the introduced empirical formula (9). From these straight lines, values for \(C\) and \(\alpha\) have been derived (Table Id). The found \(\alpha\) values are further processed to \(N\) values, making use of eq. (11) and the \(k_{\alpha}\)-values of Table Ia. It can be seen that the resulting \(N\) values are in the right order of magnitude (0.4 - 0.8, see sect. 2.2). This supports the proposed model (statement 6 and formula (8)).
Now we shall try to correlate the found values for C, \( \kappa \) and N (Table Ia,c,d) with the microstructural data (Table Ib). It should be remarked, however, that the variations in microstructures were less pronounced than had been expected. Nevertheless, we have continued the examination of these materials. Table Ic shows that in general C does increase with decreasing \( \delta_{20} \). However, the C-\( \delta_{20} \) relation differs strongly for the different powder families. It is not clear whether this is significant, because we do not exactly know the accuracy of these values. The \( \delta_{20} \) values are left out of the discussion, since the variation in values is smaller than for \( \delta_{20} \) while the inaccuracy is even larger. The expected correlation between the N value and the average shape factor (5h) was not found. It is not clear whether this is due to the small differences between 5h values, or to the fact that actual influence of the crystal shape is less than the model predicts.

With the Al_2O_3 substituted materials (Table II) we would like to determine the dependence of C on the Al_2O_3 content in order to test formula (7). The C values can be derived from the B_n and \( \delta_{20} \) values provided \( \kappa \) is known. Unfortunately the actual materials were no longer available to measure. We took an average value \( \kappa = 1.7 \) (Table IIId) and calculated the C values on this basis. Fig. 5 shows C versus \( \delta/\% \) Al_2O_3. We find indeed a straight line with a slope close to that of \( H_A/19 \).

VI - FINAL CONCLUSIONS

A model for the coercivity of sintered anisotropic M type ferrite is proposed on the basis of the conception that the \( H_c \) is governed by the growth of large multicrystall reversed regions. The resulting expression for the coercivity reads:

\[
H_c = C - \kappa B_n \delta_{10}^{-1/2}
\]

with \( C = k_3 H_A \) and \( \kappa = N/(\kappa + 1) \) where \( k_3 \) is governed by the grain size, \( N \) by the crystal shape and \( k_2 \) by the degree of alignment, the density and amount of non magnetic 2nd phase. The model is compatible with the general experience, in particular with the fact that \( H_c \) decreases when \( B_n \) increases. In addition it provides relations of \( H_c \) with the microstructure (\( k_3 \), \( k_2 \), \( N \)) and the intrinsic properties \( H_A \) and \( I \) (the latter being implicitly given by \( B_n \) and \( k_2 \)). The existence of large multicrystal reversed regions has been demonstrated experimentally, and also the fact that in general their growth determines the \( H > 0 \). The resulting expression for \( H_c \) is verified provisionally by using a selected number of available materials and data. In general the expected dependencies were supported, e.g. the relation of the \( H_c \) with the remanence (\( B_r \)), the crystal size and the anisotropy field (\( H_A \)). The expected influence of the crystal shape was not found. This may be due to the fact that the difference in crystal shape proved to be rather small.

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