Models for Magnetization Mechanisms in soft polycrystalline ferrites are reviewed. Two main properties are considered: reversible initial magnetic permeability, which appears at low applied fields, and magnetic hysteresis. The Globus model, which assumes a combination of pinning, bulging and displacement of domain wall appears to be consistent in the whole applied field range. This model has recently been refined in order to obtain a better agreement with experiment.

INTRODUCTION

The complexity of magnetic phenomena has not allowed to establish a first-principles model for magnetization processes. The quantum-mechanical nature of the interaction between magnetic atoms was first approximated by a naive (but genial) “molecular field” model by P. Weiss [1], and it was in 1938 that Heisenberg [2] proposed a more physical basis. The currently used expressions for crystalline anisotropy are still phenomenological approximations to the problem of interaction between spins and orbital (crystalline) moments. Magnetic domain wall structures have not been completely understood, and their dynamics, consequently, has to be analyzed in detail.

In the case of ferrimagnetic oxides things seem worse: crystalline structure is far more complex, the magnetic structure is antiparallel in the simplest case (it can be “triangular”, or even “helical”), and preparation technology has been a nightmare for long time. However, magnetic oxides present some advantages: in some cases, stoichiometry is very precisely defined, and once technology has been established, very pure, defect free samples can be obtained. It is true that single crystals are difficult to obtain, but polycrystalline samples have provided a very convenient material for research, with practically only one type of defect easy to characterize, to test simple theories.

Polycrystalline samples are also the normal industrial materials, and hence the impact of these theories on practical applications should be more direct.

In this paper, we analyze the models that have been proposed to explain magnetization phenomena in soft polycrystalline ferrites, at low measuring frequencies. First, we consider the low field irreversible range, and then magnetic hysteresis.

LOW FIELDS

The most interesting property at low applied field is initial magnetic permeability, which is defined as:

$$\mu = \frac{dB}{dH}, \quad H \to 0, \quad B \to 0$$  \hspace{1cm} (1)

In the general case (i.e., anisotropic single crystals), B and H can have different directions and $\mu$ is then a tensor. However, for polycrystals formed by many crystallites oriented in a random fashion, B is parallel to H and $\mu$ is hence a scalar.

Initial magnetic permeability results from small, reversible changes in demagnetized state of the material. This demagnetized structure of Weiss domains separated by Bloch walls provides the basis for initial permeability mechanisms. Two main mechanisms appear: a) A uniform, collective rotation of all the magnetic moments in each magnetic domain, in order to approach the applied field direction; and b) A displacement of the Bloch walls in order to increase the volume of domains whose direction is the same than that of the applied field, by decreasing the volume of domains with an opposite magnetization direction. These two mechanism are schematized in Fig. 1. The rotational mechanism was analyzed by Snoek [3], who proposed the relation:

$$\left(\mu - 1\right)_{\text{ROT}} = \frac{4\pi M_s}{H_A}$$  \hspace{1cm} (2)
Fig. 1. Schematic representation of reversible magnetization mechanisms. a) Wall displacement mechanisms and b) Rotational.

with $M_s =$ saturation magnetization and $H_A =$ anisotropy field. The main feature of the rotational mechanism is that it is controlled by the anisotropy field, which can be very high in ferrites. In polycrystalline samples, measured values were not in agreement with calculation with Eq. (2).

The magnetic wall displacement mechanism has been studied by several authors. Smit and Wijn [4] found:

$$\mu_w \approx 4 \frac{Ms^2D^3}{y_d}$$  \hspace{1cm} (3)

with $D =$ wall length, $y =$ wall energy and $d =$ domain width. A similar treatment led J. Verweel [5] to:

$$\mu \approx \frac{4Ms^2\cos^2\theta}{kL}$$  \hspace{1cm} (4)

where $k =$ stiffness parameter, $L =$ domain width and $\theta =$ angle between magnetization vector and applied field. In general, experimental initial permeability showed a wide variation range which cannot be attributed to the stiffness parameter, or domain width. Globus [6, 7] studied the specific case of polycrystalline ferrites and proposed a model for the wall permeability. In this model, the grain is supposed to have a spherical shape, and to be divided by a diametrical wall into two 180° domains. This Bloch wall is pinned to the grain boundary and the effect of a small applied field is to produce a bowing on the wall. This membrane-like behavior of the wall has a reversible character, Fig. 2.

Globus calculated [8] the bowing $x$ of the wall for minimum energy conditions, and neglecting second order terms, calculated the initial permeability as:

$$\mu - 1 = \frac{3\pi Ms^2D}{4y}$$  \hspace{1cm} (5)

where $y =$ wall energy and $D =$ mean grain diameter. At constant temperature, this relation predicts a linear behavior of $(\mu - 1)$ vs. $D$, for ferrites having same composition, which has been widely verified [9-11]. The solution to the exact equation has shown [12] an error of about 0.4%, and does not change the functional form of $\mu(D)$. However, the experimental results show an independent term, i.e., the initial permeability is not zero, but retains a well defined value, as $D \to 0$. This value is smaller than the value for rotational permeability, by considering that the anisotropy field, $H_A$ in Eq. (2) is due only to magneto-cristalline anisotropy. Globus showed [6, 8] that in mechanically hard, polycrystalline material like oxides, there is an intrinsic magnetoelastion contribution to the total anisotropy. This total anisotropy is composition dependent and can be calculated from monodomain samples [13] or from spin resonance
frequency [14]. An additional verification of both results was made [15] by comparing the extrapolation of the $\mu$ vs D relation for $D \to 0$, against these values, for Ni-Zn ferrites. The quantum-mechanical origin of this anisotropy contribution in terms of a volume anomaly has also been given [16].

One of the basis of Globus model is the existence of a “friction” parameter $f$, a force by unit length, that provides the pinning phenomena of the domain wall on the grain boundaries. This term represents the opposition of the wall to pass through lattice defects, and can be related to the domain wall surface area that is created or destroyed [17] during its displacement. The origin of $f$ is hence closely related to that of $\gamma$. The ratio $f/\gamma$ is a constant, independent of temperature for YIG and Ni-Zn ferrites [12], as determined from initial permeability and critical field measurements, as well as hysteresis measurements [17].

Domain wall pinning in inhomogeneities and dislocations has also been studied [18-20] as related to coercive force in permanent magnets.

Globus model assumes that there is no contribution from magnetostatic energy (i.e., free poles). This could only be fulfilled if magnetic flux shows continuity through grain boundaries. Experimental results [24] show that this continuity of flux can be achieved by a proper sintering technique.

Initial magnetic permeability, in the other hand, is a very sensitive property. Its thermal variations, especially at the Curie temperature allow a very precise determination [21] of the magnetic, and if measured in a continuous mode [22], also an evaluation of the chemical homogeneity of the samples. Problems like non-Stoichiometry [11], technology of preparation [23], wall topography [25], porosity and pressure effects [26], and others, have been analyzed by means of initial magnetic permeability.

**HYSTERESIS**

Ferromagnetic hysteresis is probably the most fundamental expression of magnetic ordering, involving microscopic parameters as exchange interactions, crystalline and magnetoelastic anisotropies, domain wall dynamics, as well as macroscopic parameters as porosity, grain boundaries, shape of the sample, etc.

At the end of the last century, Ewing proposed a model [27] for magnetic hysteresis based on a periodical arrangement of classical magnets. The resulting hysteresis loop exhibited some of the qualitative features of experimental hysteresis loops.

The early workers [28-30] on this subject considered that the basis of hysteresis was frictional resistance.

Again, the magnetic domain and domain wall concepts of Weiss [31] were the basis for hysteresis mechanisms.

Kersten [31, 32] and Becker and Doring [33] introduced the notion that the changes in magnetization could be interpreted in terms of domain wall displacements, and that these movements were troubled by the presence of defects in the solid. The pinning and depinning of walls on defects provide a convenient mechanism for the irreversible nature of hysteresis. Recent theories on coercivity [18-20] are based on the same ideas. Jiles and Atherton [34] have proposed a model for hysteresis on this basis and the mean field approximation. In this model, the mutual interactions of moments is expressed as a coupling coefficient, and the friction term as a pinning coefficient. There is also a scaling parameter which represents the ratio of the coupling to the mean field and the external field. The resulting hysteresis loops reproduce well the general characteristics of experimental hysteresis loops, but so far, probably due to the difficulty for evaluating the several parameters in the model, it has not been directly compared with specific experimental loops.

For the particular case of polycrystalline soft ferrites, the Globus model has been extended to the irreversible range [6, 17]. Schematically, it is shown in Fig. 3.

![Fig. 3. For each point, it is shown the domain wall shape and position within the spherical grain.](image-url)
where relates the critical field $H_{c_1}$ to the friction term (force by unit length of pinned domain wall). This relationship has been verified in Ni-Zn ferrites [36]. Once the magnetic wall has been depinned, it is displaced to a new location within the grain that depends on the value of the applied field, which sets an equilibrium between the magnetic pressure (external field on the domain wall surface area) and the friction term (perimeter of the domain wall pinned to the grain boundary). The path OAB is the magnetization curve. Now, if the applied field is eliminated, the domain wall recovers its plane shape but remains pinned to its new position, leading to a non-zero net magnetization, the remanence. The section BC has a small slope which is due to the debowing of the domain wall. Now, an opposite field is applied, and at first, the domain wall is bowed in the direction imposed by the field; when the critical field for this position of the domain wall is attained, it is then depinned and goes through the center to the position that corresponds to the value of the applied field. The wall is not pinned during this movement because for any position which is near to the center, the critical field is smaller than the applied field, as far as the critical field is related to the perimeter of the wall $H_{c_1} \sim 1/D$. Section DE is hence an abrupt change from an equilibrium position to a new one. In order to complete the hysteresis loop, the applied field is decreased (section EF is symmetrical to BC) and then inversed (section FGB is symmetrical to CDE). This treatment of Globus model led to the recognition of the functional form of microstructure parameters, i.e., grain diameter and anisotropies [37]. For example, plotting the hysteresis loop of several samples of same composition but different grain diameter against the product HD (instead of H) produces a single loop [38]; plotting $M/M_s$ versus $HD/H_T$, where $H_T$ = total anisotropy field, a single (“universal”) hysteresis loop is obtained [39] for several compositions, at different temperatures and with various grain diameters. In the other hand, a calculation of the area within the loop has shown that it can lead to the direct determination of the friction term and the domain wall energy [17]. However, the Globus model was not able to attempt to perform a numerical calculation of the hysteresis loop.

An analytical prediction of the hysteresis loop can be made [40] by a more detailed analysis of Globus ideas; the equilibrium position of the domain wall is calculated by using the force expression that has been derived from the minimum energy conditions; the movement of the domain wall takes into account the fact that it is a bulged wall, and the critical field for the inversion of the domain wall is calculated considering that the bulging is also in the opposite direction than in the case of the initial magnetization process. This leads to a very good agreement between calculated and experimental hysteresis loops, Fig. 4. Some improvements can be achieved by taking into account the grain diameter distribution [41] within the expression. In particular, a rounding of the magnetization curve is obtained, as shown in Fig. 5.

\[ H_{c_1} = \frac{2f}{M_D} \]  \hspace{1cm} (6)

![Fig. 4. Comparison between a) Predicted hysteresis loop from Ref. [40] and b) Experimental results of YIG, from Ref. [38].](image)

![Fig. 5. Effect of the distribution of grain size on the magnetization curve. a) Theoretical magnetization curve with a single grain diameter. b) Calculated magnetization curve by introducing the effect of an experimental distribution of grain diameter, measured [41] on a Ni-Zn ferrite.](image)

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