EXPERIMENTAL AND THEORETICAL STUDIES OF INTERACTIONS IN PARTICULATE RECORDING MEDIA

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In the paper new experimental methods for the investigation of interactions in particulate ferromagnetic materials are presented. The methods are analysed with Preisach-type models. The dynamic interactions in nanoparticulate systems are also discussed.

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1. Introduction

Modern recording media are characterised by high particle densities and the interactions between the magnetic moments of the constitutive ferromagnetic particles are difficult to avoid. In classical recording media, the interactions are modelled usually by means of two types of fields: the static statistical interaction field and the mean interaction field [1]. For systems of very small particles, when the magnetic relaxation phenomenon is important, the interaction field is fluctuating at a high rate and it is much more difficult to experimentally evaluate it and to describe it theoretically [2,3].

2. Experimental evaluation of static interactions

The effect of inter-particle interactions of any particulate magnetic recording medium are not observable in magnetisation cycles such as the Major Hysteresis Loop (MHL) but they may be evident in more complex magnetisation processes. The starting point to identify magnetisation curves sensitive to interactions was the Wohlfarth formula, [4], concerning the relation between the Isothermal Remanent Magnetisation (IRM), and DC Demagnetisation (DCD) curves. The Henkel plot [5] (see Fig.1) and the deltaM plot [6,7] are measuring the deviations from the Wohlfarth relation. One can mention that there are other related methods based on the Wohlfarth relation [8-10].



Fig.1. The Henkel plot.

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New experimental procedures to obtain deltaM-like curves were suggested recently [11-15]. We shall refer to these curves as to *generalised deltaM plots*. All these curves are sensitive to both statistical and mean field interactions between the ferromagnetic particles in the system. The experimental procedure proposed by Bissell consists in measuring the variations of the remanent magnetic moment of the sample after the application of \pm H fields, starting from any reproducible remanent state. The total variation of the remanent magnetic moment is used in the calculation of a deltaM-like curve [16, 17] (see Fig.2). An important inconvenient of the classical deltaM procedure is the difficult experimental procedure to obtain an accurate AC demagnetised state for the IRM process. In the generalized deltaM procedure this problem is avoided.



Fig. 2. A Generalized ΔM plot (ΔM_g).

To provide the experimentalists with a more general view of the all generalized deltaM for a sample the Integral Generalized deltaM (IGDM) was introduced. In the IGDM curve one point is the integral of the generalized deltaM as a function of the remanent moment used in the experiment. The IGDMs are not requiring an AC demagnetized state in order to evaluate the interactions in the sample. Recently, a simplified version of the IGDM procedure was also developed. In this procedure, esentially, the classical IRM is replaced with a IRM curve starting from the DC demagnetized state, much more easy to obtain experimentally [18-20].

Recently the transverse susceptibility method was also used to obtain information about the interactions in both micro and nanoparticulate media [21-25].

3. The Preisach Model

The Preisach model was published by Ferenc Preisach in 1935, [26], and is still considered as the best hysteresis model at our disposal [27] after its profound mathematical analysis performed by Krasnoselskii and Pokrovsky [28]. Following the spirit of this analysis, Mayergoyz, has formulated the mathematical basis for the Preisach model applied to particulate ferromagnetic media [29,30]. The Classical Preisach Model (CPM) is a scalar hysteresis model. CPM is usually dedicated to describe hysteresis in ferromagnetic systems but one may note that the model may be very well applied to any process in which hysteresis is observed (e.g. [31]).

Let's consider an ensemble of ferromagnetic particles in which the interactions are very weak. In this case one may say that the hysteresis loop of the ensemble is the sum of the hysteresis loops of the particles. When the interactions are no more negligible, each particle will experience not only the external applied field but also the field created by the neighbour particles. The effect of this field, the "interaction field", is a modification of the particle hysteresis loop, usually a displacement of the hysteresis loop along the field axis; the switching fields of the hysteresis loops are for this reason different (see Fig.3). The main idea of the Preisach model is to use the switching fields of the particles

as a co-ordinate system.



Fig. 3. The Preisach plane a) How the magnetic moment is associated to a point in the Preisach plane ; b) Coordinates axes in Preisach plane.

Mayergoyz has shown that the wiping-out and congruency are the necessary and sufficient conditions for a system in order to be accurately described by a CPM, [30]. Proofs of the Mayergoyz theorem may be found in [30] and [32].

In the CPM it was assumed that the supplementary field produced by the neighbour particles remains invariable when the magnetic moment of the sample changes. This is referred to as the "statistical stability" of the Preisach distribution and may be also related to the congruency property. In reality, the interaction field changes if the magnetisation state of the sample is different. The most simple way of taking into account this modification, is to consider that the interaction field is the sum of two fields: the statistical interaction field, which is the interaction field when the sample is demagnetised and a mean-field interaction field dependent on the magnetic moment of the sample.

In order to take into account the mean-field interaction field, in the Moving Preisach Model (MPM) [33] the effect of a change in the magnetisation is to move the Preisach function in the Preisach plane along a line of unit slope. A linear mean-field term is usually considered but non-linear moving terms may be considered as well [34-35].

To solve the identification problem (to calculate the model's parameters) in the MPM one has to find both the Preisach distribution and the moving parameter. The most difficult problem related to the identification is to discern between the moving parameter which characterises the mean-field interactions and the dispersion of the interaction field distribution characterising the statistical interactions. This is the motivation for the continuous search for experimental curves not only sensitive to interactions but sensitive to only one type of interaction (statistical or mean field).

5. Dynamic interactions between the particles

The CPM describes static magnetisation processes, that is, processes in which the effect of relaxation is neglected. Visintin [28] defined hysteresis as a rate independent memory effect, so the viscous-type effects are not included. Mayergoyz [30] considers both static and dynamic hysteresis. A system is characterized by static hysteresis if its branches of hysteresis nonlinearities are influenced

only by the past extremes values and not by the speed of input variations.

The most important time effect on magnetisation phenomena is the magnetic viscosity (also named magnetic after effect). It includes all the influence of time without taking into account [37] the influence of the sample inductance, eddy currents, irreversible relaxation of chemical or topological microstructure of the material and relaxation phenomena with characteristic time less than 10^{-5} s. The magnetic aftereffect is essentially due to the energy barriers which may be overcome by thermal energy. One may identify three categories of magnetic viscosity: i) reversible aftereffect (associated with diffusion of impurity atoms), ii) irreversible aftereffect and iii) quantum tunneling of magnetisation [37]; the irreversible after effect is due au fond to the thermal activation [38]. Another time effect is the accommodation which appears when the minor loops are cycled between two fields; this effect, which is in disagreement with the wiping-out property, was included in the Preisach model by E. Della Torre [39] and is explained as the result of the motion of particles within the Preisach distribution.

Mayergoyz developed the Dynamic Preisach Model (DPM) [40] in which the irreversible aftereffect is considered in a purely phenomenological manner. Thermally induced magnetic relaxation has also been included in Preisach-type models in a more physical way by Mayergoyz [41] and by Souletie following the ideas initially developed by Néel [42] which will be referred to as the Preisach-Néel Model (PNM). The PNM was used to explain the effect of the finite temperature T and observation time t on the Henkel plots of ac and thermally demagnetised systems [43] and the maximum in field dependence of the isothermal remanent magnetisation and thermoremanent magnetisation for the spin glasses [44]. A similar approach has been used by Bertotti [45] to deal with the domain wall dynamics in metals. In all the approaches of the PNM, until now, only the linear expression of the energy barrier as a function of the field was used. In [1,3] it is presented a PNM for a Stoner-Wohlfarth [46] (SW) particle system which introduces a non-linearity in the energy barrier expression (see Fig. 4).



Fig. 4. The Preisach plane in a positive applied field H_a , for $h_t=0$ (no relaxation process - like in the static CPM) (a) and for $h_t\neq 0$ in the PNM for a Stoner - Wohlfarth particle system (b).

Sp – superparamagnetic particles region, Bk – blocked particles region.

6. Static and dynamic interactions in the PNM

When one applies an external field to a particulate medium each particle is feeling not only this field but also the field created by the neighbor particles. In classical recording media the relaxation effects may be neglected at the room temperature due to the fact that the energy barriers are sufficiently high in comparison to the thermal energy. When the relaxation is important in the system the statistical interaction field is fluctuating at a high rate. This was proven by Dormann [2] to be equivalent to an

increase of the particle anisotropy. These interactions are qualitatively different from the static ones; they will be referred to as the *dynamic interactions*. In the terms of the Preisach model, that means that the dynamic interactions will influence the coercive field distribution.

In the system one have both blocked and superparamagnetic particles. The blocked particles are creating the static statistical interaction field and the superparamagnetic particles the dynamic interaction field. The mean interaction field is given by all the particles.

In the time and temperature dependent magnetisation processes the weight of the blocked or superparamagnetic particles is changing. The effect of this modification is taken into account in the PNM as a variation of the standard deviation of the interaction field distribution. The variation of the dynamic interactions intensity is inducing a variable displacement of the Preisach distribution in the second bisector direction (see Fig. 5). A similar displacement, but in the opposite direction and with other physical motivation, was considered in [44] to explain the appearance of a maximum in the field dependence of the isothermal remanent magnetisation and the thermoremanent magnetisation in spin glasses.



Fig. 5. The Preisach distribution for two different values of the experimental temperature and/or experimental time: (1) at low temperature the static statistical interactions are important and (2) at high temperature the dynamic interactions produce the displacement of the Preisach distribution and the diminishing of the effect of the static statistical ones.

In [3] the PNM was used in the analysis of the waiting time effect in the relaxation under field of the zero-field-cooled magnetisation of ultrafine particle systems. To the best of our knowledge, this is the first explanation of an ageing effect in fine particle systems.

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