MAGNETIC CORRELATIONS IN SMALL PARTICLE SYSTEMS, STUDIED WITH THE NEUTRON DEPOLARIZATION TECHNIQUE.

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Abstract.
Neutron depolarization theory in particulate media is discussed. A ND experiment yields a correlation matrix ω, describing the micro-magnetic state of the medium. The relations between ω and parameters describing the individual particles, particle interactions, orientational correlations between particles and density variations within the medium are discussed. With these relations the ND technique can successfully be used to study the micro-magnetic state of particulate media. The theory is applied to the results of ND measurements on CrO_2-powder.

1 INTRODUCTION

The three-dimensional neutron depolarization (ND) technique is a powerful method to study static and dynamic properties of magnetic structures in the micron and submicron region (e.g. [1-3]). In this technique the polarization vector of a polarized neutron beam is analyzed after transmission through a magnetic medium. During transmission the polarization vector is affected by magnetic inhomogeneities in the medium: mean magnetic induction ($\langle \underline{B} \rangle$) results in a net rotation of the polarization vector while fluctuations in the local magnetic induction, denoted by $\Delta\underline{B}(\underline{r})=\underline{B}-\underline{B}(\underline{r})$, result in an effective shortening of the polarization vector, called depolarization henceforth. The ND technique yields the mean length along the neutron path over which $\Delta\underline{B}(\underline{r})$ is correlated, $\langle \underline{B} \rangle$ and the local magnetic texture in general.

The ND technique has only recently been applied to the study of magnetic correlations in particulate media [4]. The properties of these media are strongly affected by magnetic particle interactions (e.g [5]), resulting in correlated magnetization orientations of neighbouring particle, and by possible correlations between particle orientations. The ND technique promises to be very useful in studying these media, as it can provide quantitative information about these correlations in a more direct way than other techniques mostly used.

A ND theory suitable for interpreting the results of ND measurements on particulate media has recently been formulated by Rosman and Rekveldt [6]. This paper deals with the latter theory applied to particulate media. According to this theory, a ND experiment yields a correlation matrix ω describing the micro-magnetic state of the medium. The relations between ω and parameters describing the individual particles and different types of superdomains is discussed. The theory is applied to ND measurements on powders of CrO_2 particles.

2 NEUTRON DEPOLARIZATION IN PARTICULATE MEDIA

The depolarization matrix D, expressing the relation between the polarization vector before ($\underline{P}^0$) and after ($\underline{P}^1$) transmission through the medium ($\underline{P}^1= D\,\underline{P}^0$), yields $\langle \underline{B} \rangle$ as well as the correlation matrix ω [6]. The components ω_{ij} (i,j= x,y,z) of ω, related to a particulate medium with a particle volume fraction ε, are defined by

$$\omega_{ij}= \frac{2}{\Delta V}\int_{\Delta V} d^3\underline{r}\int_{z_0}^{z} dz'\,\Delta B_j(x,y,z)\Delta B_i(x,y,z')/[\varepsilon(\mu_0 M_s)^2] \quad (1),$$

with ΔV the volume of a representative part of the medium with thickness ΔL and $\underline{M}_s$ the spontaneous particle magnetization. The propagation direction of the neutron beam ($\underline{e}_0$) is along the z-axis. A relation between ω and $\underline{M}(\underline{r})$ can be derived from eq.(1) by working in Fourier space, resulting in

$$\omega_{ij}= \alpha_{ij} - \frac{4}{9}\Delta L\ \langle M_i\rangle\langle M_j\rangle/(\varepsilon M_s^2)$$

with

$$\alpha_{ij}=\frac{16\pi^4}{\Delta V}\int_{\tilde{S}} d^2\underline{s}\ B_i(\underline{s})B_j(-\underline{s})/[\varepsilon(\mu_0 M_s)^2]$$

$$\underline{B}(\underline{s})=\frac{\mu_0}{(2\pi)^3}\int_{\Delta V} d^3\underline{r}\ [\tilde{\underline{s}}\times(\underline{M}(\underline{r})\times\tilde{\underline{s}})]e^{i\underline{s}\cdot\underline{r}} \quad (2).$$

Here, $\tilde{S}$ is the reciprocal XY-plane, $\underline{s}$ a reciprocal position vector, $\tilde{\underline{s}}= \underline{s}/(\underline{s})$ and m_i the reduced magnetization in the i-direction ($m_i=\langle M_i\rangle/(\varepsilon M_s)$).

Media for which ω is diagonal are considered only, implying that it is assumed that no correlations between ΔB_i and ΔB_j ($i\neq j$) exists along the neutron path. This assumption is generally valid in particulate media provided that $\underline{e}_0$ is along one of the main directions of the magnetization orientation distribution. Then, the quantities ζ=trace(ω) and $\gamma_i=\omega_{ii}/\zeta$ are used to characterize the micro-magnetic state of the medium. Furthermore, ω, ζ or γ_i referring to a state for which m=0 will be denoted by ω^0, ζ^0 and γ_i^0, respectively. The quantity ζ is the correlation length of $\Delta\underline{B}(\underline{r})$ along $\underline{e}_0$. The quantity γ_i, related to the component ΔB_i which is correlated along $\underline{e}_0$, yields the magnetic texture. The exact relation between D on the one hand and ζ and γ_i on the other depends on the orientation of $\langle \underline{B} \rangle$.

2.1 The matrix ω for uncorrelated particles.

The particles discussed in this section are assumed to be ellipsoides with axial dimension b, radial dimension a and volume V. The spontaneous particle magnetization $\underline{M}_s$ is oriented along the b-axis. The correlation matrix ω^s related to such ellipsoides follows from eq.(2) by taking ΔV as the subvolume containing one particle (ΔV=V/ε) and by averaging the obtained relation over the particle orientation:

$$\omega_{ij}=\delta_{ij}\{\frac{V}{4\pi^2}\int_{\tilde{S}} d^2\underline{s}\langle (n^*(\tilde{\underline{s}}))_i^2 F(\underline{s})^2\rangle - \frac{4}{9}\varepsilon^{2/3} m_i m_j V/\langle S\rangle\} \ (3).$$

Here, ⟨S⟩ is the average particle cross-section perpendicular to $\underline{e}_0$, $\underline{n}^*(\underline{s})=\tilde{\underline{s}}\times(\underline{n}\times\tilde{\underline{s}})$ and $\underline{n}$ a unit vector along $\underline{M}_s$. The second term on the r.h.s of eq. (3) corrects for the fact that an increase in $\bar{B}$ trivially results in a decrease in $\Delta B(\underline{r})$ and therewith in ω_{ii}. This factor, which is discussed in [7], can be neglected in the experiments presented in this paper.

The quantity ζ approximates the mean particle size along the neutron path. As an example, fig. 1 gives the quantities ζ^0/a and ζ^0/b versus b/a in case of an isotropic particle orientation distribution. For such a distribution ζ^0 is about proportional to b for b/a<0.1, and to a for b/a>10.

For uncorrelated particles, γ_i^0 is dependent on the particle shape and orientation. In general, the more the particles are oriented along the i-direction the larger the value of γ_i^0 is. In case of an isotropic particle orientation distribution, $\gamma_x^0=\gamma_y^0=1/4$ and $\gamma_z^0= 1/2$,

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independent of the particle shape. This phenomenon, called the intrinsic anisotropy, is extensively discussed in [6]. The strong dependence of γ_i on the particle shape is extremely important when deriving the local particle orientation from a ND experiment.

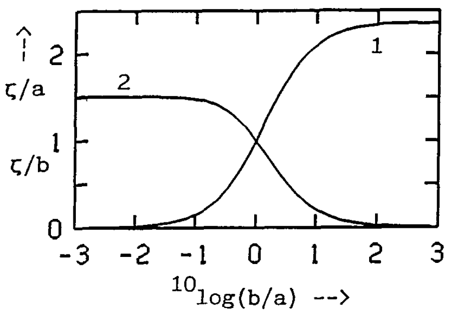

Fig.1 The quantities ζ^o/a (1) and ζ^o/b (2) versus $^{10}\log(b/a)$ in case of an isotropic particle orientation distribution.

2.2 The effect of superdomains on ω.

The effect of the existence of superdomains on ω is considered. A superdomain is defined as a subvolume V_{sd} containing particles of which the components of $\underline{M}_s$ along $\langle\underline{M}\rangle$ have the same sign. Its mean magnetization, $\langle\underline{M}_{sd}\rangle=m_{sd}\varepsilon M_s\underline{n}_{sd}$ ($|\underline{n}_{sd}|=1$), differs from $\langle\underline{M}\rangle$. The contribution of such superdomains to the correlation matrix equals ω of a medium of uncorrelated single-domain particles with a particle volume fraction of 1, a particle size and shape equal to those of the superdomains and a particle spontaneous magnetization of $\langle\underline{M}_{sd}\rangle$, resulting in (see eq.(2) and (3))

$$\omega_{ij}=\delta_{ij}\{\frac{V_{sd}}{4\pi^2}\int_{\tilde{S}}d^2\underline{s}\langle(n^*_{sd}(\tilde{\underline{s}}))_i^2F_{sd}^2(\underline{s})\rangle\varepsilon m_{sd}^2-\frac{4}{9}\varepsilon m_i m_j V_{sd}/\langle S_{sd}\rangle\} \quad (4).$$

The quantities $F_{sd}(\underline{s})$ and $\langle S_{sd}\rangle$ are the form factor and the average cross-section perpendicular to $\underline{e}_o$ of a superdomain, respectively. The quantity $\underline{n}^*_{sd}(\underline{s})=\underline{s}x(\underline{n}_{sd}x\underline{s})$. Following from eq.(4) the contribution of a superdomain with size L_{sd} to ω_{ij} approximates $\varepsilon L_{sd}m_{sd}^2$, a value which may be much smaller than L_{sd}.

Three types of superdomains will be considered: superdomains due to particle interactions, due to orientational correlations between neighbouring particles and due to density variations. It will be assumed that no mixture of these superdomains exist.

Particle interactions may result in switching volumes consisting of several particles and therewith in superdomains. In case of pure particle interactions (so with the exclusion of orientational correlations and density variations) $\langle\underline{M}_{sd}\rangle$ is parallel to $\langle\underline{M}\rangle$ and m_{sd} equals the maximum value of m, a value which is related to the average particle orientation. At $m=m_{sd}$ the superdomains do not exist anymore (by definition!) and hence do not contribute to ω. Note that eq.(4) does not exactly equal zero for $m=m_{sd}$, being due to a small approximation in its derivation. Superdomains due to particle interactions generally result in a γ_k approaching 1, with k (k=x,y,z) denoting the orientation of $\langle\underline{M}\rangle$, as $\langle\underline{M}_{sd}\rangle$ is parallel to $\langle\underline{M}\rangle$. Particle interactions may also result in negative correlations, with obviously a contribution to ω different from eq.(4).

Particle orientational correlations may result in large switching volumes due to the fact that particles with the same orientation reverse magnetization at the same value of the effective field. However, even with the negligence of this effect, orientational correlations may result in superdomains at $m\neq0$, as for $m\neq0$ the mean magnetization of a subvolume containing correlated particles differs from $\langle\underline{M}\rangle$. As the average spread in the particle orientation in these superdomains is smaller than the average spread within the medium, m is always smaller than m_{sd}. As a result, the superdomains may contribute to ω over the total m-range. The contribution of the superdomians to γ_i is related to the average shape of the subvolumes considered and the average orientation of their particles.

Subvolumes with a particle volume fraction equal to $\varepsilon+\Delta\varepsilon$, with ε the average value of ε, act as superdomains with $m_{sd}=m|\Delta\varepsilon|/\varepsilon$ and $\underline{n}_{sd}$ oriented along $\pm\langle\underline{M}\rangle$. Their contribution to ω is given by eq.(4) with the negligence of the second term on its r.h.s. (due to $\langle\Delta\varepsilon\rangle=0$). As a result the contribution of density variations to ω is proportional to $m^2\langle\Delta\varepsilon^2\rangle/\bar{\varepsilon}$. Density variations only seriously affect ω in case of large scale density variations, as $\Delta\varepsilon/\bar{\varepsilon}$ is generally small. As $\langle\underline{M}_{sd}\rangle$ is parallel to $\pm\langle\underline{M}\rangle$, density variations result in a γ_k approaching 1, with k (k=x,y,z) denoting the orientation of $\langle\underline{M}\rangle$. This makes them distinguishable from superdomains due to orientational correlations.

2.3 Discussion.

The ND technique in principal can be applied to those particulate media with values of ζ from 10 nm up to mm's. No other technique yields quantitative information about these correlation length in such a direct way.

Separating the contributions to ω of the individual particles and the different types of superdomains may sometimes be difficult. However, several ways exist which may make this separation possible. At first, the dependence of ω on the magnetic state of the medium can be analyzed. Individual particles and superdomains due to orientational correlations between particles may contribute to ω over the total magnetization range. Density variations may contribute at $m\neq0$ only. Superdomains due to particle interactions do not affect ω in the state of maximum remanence. Secondly, γ_i can be analyzed. If ω is dominated by superdomains due to density fluctuations γ_k should approach 1, with k denoting the orientation of $\langle\underline{M}\rangle$. Thirdly, the analysis of ω for different $\underline{e}_o$ orientations may yield information, based on which this separation can be made. Furthermore, if the particle size, shape and mean orientation is known (e.g. from other techniques), ω can be corrected for the contribution of individual particles.

3 NEUTRON DEPOLARIZATION IN CRO2 POWDERS

This section deals with ND measurements in powders of single-domain CrO_2 particles. The aim is to measure ω versus m during the magnetization reversal process with $\underline{e}_o$ perpendicular to the applied field.

3.1 Experimental.

The CrO_2 particles are elongated with a mean diameter of 20 nm and a mean length of 200 nm [8], with the anisotropy axis along the longitudinal axis. For the ND measurements ($\underline{e}_o$// z-axis) the particles are put in a container surrounded by a coil (850 turns per meter). The particle density is around 1.0 g/cm^3. Pulse shaped currents (up to 400 A) and therewith pulse shaped fields along the y-direction (up to 35 kA/m) are produced in the coil basically by shortcircuiting a charged capacity bank over the coil (typical pulse width 1 ms). During (de)magnetization as well as during measuring the container and coil are positioned in a magnetic yoke made of transformer steel in order to produce a homogeneous field and to shortcircuit any flux from the container. The set up used in the experiments is basically the same as the one used previously [4], however with the single-detector replaced by an array of detectors. The matrix ω is measured versus the heigth H_a of the applied pulsed field, starting from the 'virginal' state (m=0) up to the state of maximum remanence. The direction of the field is subsequently reversed, after which ω is measured during the magnetization reversal process.

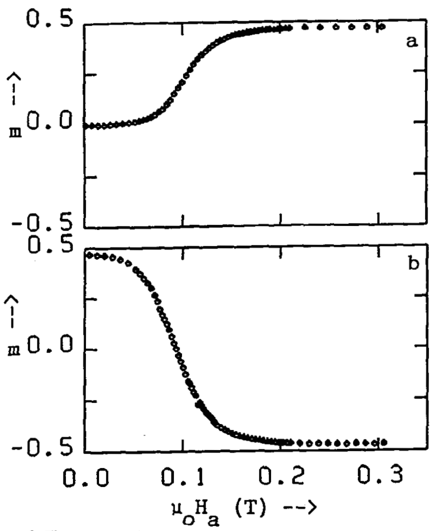

Fig.2 The reduced remanent magnetization m versus H_a, during the magnetization (a) and magnetization reversal (b) process.

3.2 Results.

Fig.2 gives m versus H_a, showing the magnetization (reversal) process. Fig.3 gives the correlation length ζ versus m. Its value (ζ^o=36±3 nm) strongly increases with increasing m during the magnetization process up to values of about ten times the original value. During the magnetization reversal process ζ^f decreases and increases again, with a minimum (ζ^o=100±10 nm) equal to three times the value in the virginal state. The quantity γ_y could not be determined accurately for m≈0, due to experimental circumstances. However, its value appeared to vary between 0.2 and 0.5. The values of γ_i^o at m=0 have been measured separately, yielding γ^o=0.27, γ^o=0.24, γ^o=0.49 (±0.02, virginal state) and γ_x^α=0.30, γ_y^β=0.47, γ_z^α=0.23 (±0.04, demagnetized state).

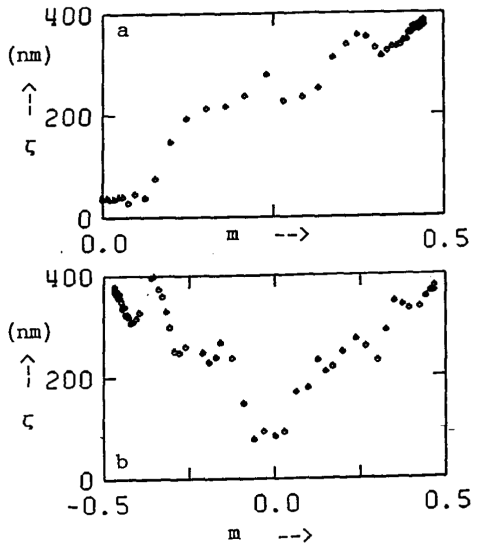

Fig.3 The correlation length ζ versus m during the magnetization (a) and magnetization reversal (b) process.

3.3 Discussion.

Assuming the virginal state of the CrO_2 particles to consist of uncorrelated ellipsoidal particles with a longitudinal axis of 2b and a radial dimension of 2a with b/a>10, the value of ζ in the virginal state corresponds to a=17±2 nm. This value is somewhat higher than the value given by the supplier (a≈10 nm). This may be due to possible correlations already present in the virginal powder. However, it is more likely due to the large spread in the particle size, resulting in an increased value of ζ.

The increase of ζ with increasing m, together with a value of γ_y much smaller than 1, indicates the presence of subvolumes in which the particle orientations are highly correlated. With increasing m these subvolumes become magnetized and act as superdomains. The low value of γ_y over the whole m-range excludes large effects of density variations. The switching volumes in the magnetization reversal process are much smaller than these superdomains. However, the size of the switching volumes along z exceeds the particle size, following from a comparison of the value of ζ in the virginal and the demagnetized state.

The Henkel curve derived from fig.3 (not shown) shows a deviation from a straigth line towards lower m values, indicating the presence of interactions with a demagnetizing effect.

Similar measurements on CoZnferrite particles yielded basically the same results. A more extensive discussion both on the theory and the measurements is given in [7].

3 CONCLUSIONS

The relation between the correlation matrix ω, derived from a ND experiment, and the micro-magnetic state of a particulate medium has been derived. The matrix ω may yield quantitative information about the individual particles and several types of superdomains. The contribution of the several types of superdomains to ω can be separated by analyzing the ratio of the diagonal elements of ω and the dependence of ω on the mean magnetization.

References.

[1] G.M.Drabkin, E.I.Zabidarov, Ya.A.Kasman and A.I.Okorokov, JETP 29 (2), 261 (1969).
[2] M.Th.Rekveldt, J.de Physique 38, C1-23 (1977).
[3] N.Stuesser, M.Th.Rekveldt and T.Spruyt, Phys.Rev.B 33 (9), 6423 (1980).
[4] R.Rosman, M.Th.Rekveldt and H.A.J.Cramer, J.de Phys., C8 (12), 1983 (1988).
[5] L.F.G.Thurlings, IEEE Trans.Magn., vol MAG-19, 84 (1983).
[6] R.Rosman and M.Th.Rekveldt, Z.Phys.B., Jan 1990.
[7] R.Rosman and M.Th.Rekveldt, to be published
[8] Philips Research Laboratories, The Netherlands.