Neutron-depolarization theory in particulate media

R. Rosman and M. Th. Rekveldt
Interfacultair Reactor Instituut, Delft University of Technology, 2629 JB Delft, The Netherlands
(Received 15 May 1990)

Neutron-depolarization (ND) theory in particulate media is discussed. The relations between the correlation matrix $\underline{\omega}$, derived from a neutron-depolarization experiment, and parameters describing the micromagnetic state of a particulate medium are derived and discussed extensively. The latter involves the individual particles, magnetic interparticle correlations as a result of particle interactions or particle orientational correlations, density variations within the medium, and variations in the neutron transmission length over the cross section of the medium. These theoretical relations have been used to interpret some of the results of ND measurements on a CrO_2 pigment. It is shown that the neutron-depolarization technique yields relevant quantitative information about the micromagnetic state of particulate media.

I. INTRODUCTION

The three-dimensional neutron-depolarization (ND) technique is a powerful method to study static and dynamic properties of magnetic structures in the micrometer and submicrometer regions (e.g., Refs. 1–7). In this technique the polarization vector of a polarized neutron beam is analyzed after transmission through a magnetic medium. The polarization change during transmission is related to the micromagnetic state of the medium. The mean magnetic induction results in a net precession of the polarization vector around the mean magnetic induction, while variations in the local magnetic induction result in an effective shortening of the polarization vector, called depolarization henceforth. The ND technique in general yields the correlation length along the neutron path of the variations in the local magnetic induction, the mean local orientation of these variations and the mean magnetic induction. The range of magnetic correlation lengths that can be measured covers 10 nm up to mm's.

Although the ND technique has been known for quite some time, it has only recently been applied to study magnetic correlations in particulate media,[8] i.e., media containing magnetic single-domain particles. In general these media have been object of thorough studies because of their practical applications (e.g., as magnetic recording tapes, ferrofluids). Besides they are useful in a more fundamental study of magnetic particle interactions. The properties of these media are highly affected by magnetic particle interactions,[9,10] resulting in correlated magnetization orientations of neighboring particles, by possible orientational correlations between particles and by density variations within the medium. As a result, it is important to have access to measuring techniques which can characterize the micromagnetic state of particulate media. The measuring techniques commonly used, i.e., magnetization, susceptibility, and noise measurements, generally yield qualitative information about the micromagnetic state. The ND technique is one of the very few techniques which may yield detailed quantitative information.

In order to interpret the results of ND measurements on particulate media, a ND theory is necessary which takes the influence on the depolarization of correlations between the particle shape and the orientation of the particle magnetization $\mathbf{M}_s$ and the influence of the demagnetizing fields of single-domain particles into account. Both influence highly affect the depolarization in particular media. The ND theory of Maleev and Ruban[11,12] accounts for the influence of the demagnetizing fields on the depolarization. However, it neglects the influence of the correlations between the particle shape and $\mathbf{M}_s$. The ND theory formulated by Rekveldt,[3] being developed for ND in continuous ferromagnets in particular, neglects demagnetizing fields as well as these correlations. The effect of interdomain correlations and the domain shape on the depolarization was discussed in a subsequent paper.[14] It was only recently that a more general ND theory, which takes both demagnetizing fields and correlations between the particle shape and the orientation of the particle magnetizations into account, has been formulated (Rosman *et al.*[15,16]).

The paper deals with an extension of the latter theory in view of its application to particulate media. A correlation matrix $\underline{\omega}$ is introduced describing the micromagnetic state of the medium. The relations between $\underline{\omega}$ and various parameters are derived and discussed extensively. These parameters describe the intrinsic particle properties, magnetic interparticle correlations as a result of particle interactions or particle orientational correlations, density variations within the medium and variations in the neutron transmission length over the cross section of the medium. The extended ND formulas are derived and discussed in Sec. II. A brief discussion covering the various contributions and the application of the theory to interpret results of ND measurements on a CrO_2 pigment are given in Sec. III. Section IV presents the final conclusions.

II. NEUTRON DEPOLARIZATION IN PARTICULATE MEDIA

The polarization vector of a polarized neutron beam ($\mathbf{P}$) generally shortens during transmission through a magnetic medium. This shortening is related to the mean correlation length along the neutron path of variations in the local magnetic induction $\mathbf{B}(\mathbf{r})$ around the mean magnetic induction $\langle \mathbf{B} \rangle$, denoted $\Delta\mathbf{B}(\mathbf{r})=\mathbf{B}(\mathbf{r})-\langle \mathbf{B} \rangle$, and to the mutual orientation between $\mathbf{P}$ and $\Delta\mathbf{B}(\mathbf{r})$. These variations always exist in particulate media, due to a particle volume fraction ϵ less than 1.

Apart from the individual particles magnetic interparticle correlations also affect $\Delta\mathbf{B}(\mathbf{r})$. These correlations may be the result of particle interactions, orientational correlations between neighboring particles, and the presence of density variations within the medium. Furthermore, a varying neutron transmission length over the cross section of the medium will affect the determination of $\Delta\mathbf{B}(\mathbf{r})$. In the paper, magnetic correlations refer to only those correlations which differ from $\langle \mathbf{B} \rangle$.

This section deals with the relations between $\underline{\omega}$ on the one hand and parameters describing the intrinsic particle properties, the various types of correlations and variations in the neutron transmission length on the other. In Sec. II A a short review of the ND theory based on Refs. 15 and 16 is given and the general expression for the correlation matrix $\underline{\omega}$ in particulate media is derived. The correlation matrix $\underline{\omega}$ for magnetically uncorrelated single-domain particles is discussed in Sec. II B. The effect of magnetic interparticle correlations on $\underline{\omega}$ is dealt with in Sec. II C. Section II D discusses the contribution to $\underline{\omega}$ of variations in the neutron transmission length.

A. General neutron depolarization theory

In Refs. 15 and 16, the general neutron depolarization theory has been discussed using two approaches, i.e., the Larmor and the scattering approach. The former approach is based on the Larmor precession of a polarized neutron beam around variations in $\mathbf{B}(\mathbf{r})$, the latter on small-angle neutron scattering by these variations. It was shown for the first time that both approaches are fully equivalent.

The depolarization matrix in a ND experiment, $\underline{\mathbf{D}}$, expresses the relation between the polarization vector before ($\mathbf{P}^0$) and after ($\mathbf{P}^1$) transmission through the medium ($\mathbf{P}^1=\underline{\mathbf{D}}\mathbf{P}^0$). According to Ref. 15, the depolarization matrix yields the mean magnetic induction $\langle \mathbf{B} \rangle$ as well as the correlation matrix $\underline{\alpha}$. The components α_{ij} $(i,j=x,y,z)$ of $\underline{\alpha}$ are given by

$$\alpha_{ij}=\frac{1}{W}\int_W d^3\mathbf{r}\int_{z_0}^{z} dz'\,\Delta B_i(x,y,z)\Delta B_j(x,y,z') , \tag{1}$$

where W is a representative subvolume of the medium with size L_W along the propagation direction of the beam. It is assumed that L_W is large compared to the mean magnetic correlation length in the medium but still small, so that the polarization change in W is much smaller than 1. The propagation direction of the neutron beam ($\mathbf{e}_0$) is along the z direction. In the paper media for which $\underline{\alpha}$ is diagonal are considered only, implying that no correlations between $\Delta B_i(\mathbf{r})$ and $\Delta B_j(\mathbf{r})$ $(i\neq j)$ along the neutron path exist. This assumption is generally valid in particulate media provided $\mathbf{e}_0$ is along a main direction of the magnetization orientation distribution. In that case, the quantities ξ and γ_i, defined

$$\xi=\sum_i \alpha_{ii} , \tag{2}$$

$$\gamma_i=\alpha_{ii}/\xi , \tag{3}$$

are used to characterize the micromagnetic state of the medium. The quantity ξ is proportional to the correlation length of $[\Delta B(\mathbf{r})]^2$ along $\mathbf{e}_0$. The quantities γ_i yield the mean local orientation of $\Delta\mathbf{B}(\mathbf{r})$ (magnetic texture).

No general relation between $\underline{\mathbf{D}}$ on the one hand and $\underline{\alpha}$ or ξ and γ_i on the other can be given. If $\langle B \rangle=0$, the components D_{ij} of $\underline{\mathbf{D}}$ are given by[15]

$$D_{ij}=\delta_{ij}e^{-c_1 L\xi(1-\gamma_i)} . \tag{4}$$

Here, δ_{ij} is the Kronecker delta, $c_1=\gamma^2/v^2=2.18\times10^{29}\lambda^2\,\mathrm{m}^{-4}\,\mathrm{T}^{-2}$, γ the gyromagnetic ratio, v the neutron velocity, λ the neutron wavelength, and L the neutron transmission length through the medium. If $\langle \mathbf{B} \rangle$ is oriented along the y direction and provided $\alpha_{xx}=\alpha_{zz}$ the elements D_{ij} are given by

$$\begin{aligned}
&D_{xx}=D_{zz}=\cos\phi\, e^{-c_1 L\xi[(1+\gamma_y)/2]} ,\\
&D_{xz}=-D_{zx}=\sin\phi\, e^{-c_1 L\xi[(1+\gamma_y)/2]} ,\\
&D_{yy}=e^{-c_1 L\xi(1-\gamma_y)} ,\\
&D_{xy}=D_{yx}=D_{yz}=D_{zy}=0 ,
\end{aligned} \tag{5}$$

with the rotation angle $\phi=\mu_0\langle M \rangle L\sqrt{c_1}$. If the precessional motion of the polarization vector around $\langle \mathbf{B} \rangle$ is in the order of or larger than $\pi/2$ no distinction can be made between the two directions perpendicular to $\langle \mathbf{B} \rangle$. The high value of c_1 ($c_1=2.7\times10^{10}\,\mathrm{m}^{-2}\,\mathrm{T}^{-2}$ at $\lambda=0.35$ nm) shows the sensitivity of the polarization vector for the mean magnetic induction. It should be noted that generally $\alpha_{xx}\neq\alpha_{zz}$. Then a modulation in D_{xx}, D_{xz}, D_{zx}, and D_{zz} as a function of $\langle B \rangle$ is expected to be seen, the amplitude of which is independent of $\langle B \rangle$.[15]

Equation (1) gives the relation between $\underline{\alpha}$ and $\mathbf{B}(\mathbf{r})$. However, information about the local magnetization $\mathbf{M}(\mathbf{r})$ may be wanted instead of information about $\mathbf{B}(\mathbf{r})$. A relation between $\underline{\alpha}$ and $\mathbf{M}(\mathbf{r})$ can be derived from Eq. (1) by working in Fourier space, resulting in[15]

$$\alpha_{ij}=\alpha_{ij}^1-L_W\langle B_i \rangle\langle B_j \rangle/2 \tag{6}$$

with

$$\alpha_{ij}^1=\frac{8\pi^4}{W}\int_S d^2\mathbf{s}\; B_i(\mathbf{s})B_j(-\mathbf{s}) , \tag{7}$$

$$\mathbf{B}(\mathbf{s})=\frac{\mu_0}{(2\pi)^3}\int_W d^3\mathbf{r}\,\mathbf{M}^*(\mathbf{r},\hat{\mathbf{s}})e^{i\mathbf{s}\cdot\mathbf{r}} , \tag{8}$$

$$\mathbf{M}^*(\mathbf{r},\hat{\mathbf{s}})=\hat{\mathbf{s}}\times(\mathbf{M}(\mathbf{r})\times\hat{\mathbf{s}}) . \tag{9}$$

Here, $\mathbf{s}$ is the reciprocal lattice vector, S is the reciprocal xy plane, and $\hat{\mathbf{s}}=\mathbf{s}/|\mathbf{s}|$. The second term on the right hand side of Eq. (6) corrects for the polarization change within W due to $\langle \mathbf{B} \rangle$. The latter, which is included in $\underline{\alpha}^1$ does not result in depolarization. The fact that the right-hand side of Eq. (8) contains the quantity $\mathbf{M}^*(\mathbf{r},\hat{\mathbf{s}})$

instead of $\mathbf{M}(\mathbf{r})$ accounts for the fact that $\mathbf{B}(\mathbf{r})$ obeys the Maxwell law $\nabla \cdot \mathbf{B}(\mathbf{r})=0$.

In the derivation of Eq. (6) an approximation has been made, which is not explicitly mentioned in Ref. 15. The approximation is that demagnetizing fields present outside W arising from W are included in $\underline{\alpha}$ and $\underline{\alpha}^1$ (see Ref. 17). In this way it is implicitly assumed that no overlap exists between demagnetizing fields arising from different subvolumes W. As this overlap is small, in particular in particulate media, the approximation is valid here. Note that Eq. (6) is exact for infinitely diluted media. If the demagnetizing fields outside W arising from W are also taken into account in the calculation of $\langle \mathbf{B} \rangle$, $\langle \mathbf{B} \rangle = \int_\infty \mathbf{B}(\mathbf{r}) dr / W = \frac{2}{3}\mu_0 \langle \mathbf{M} \rangle$ independent of the shape of W. Here, $\langle \mathbf{M} \rangle$ is the mean magnetization within W. However, in a ND experiment on a magnetized medium, the latter generally is positioned in a magnetic yoke, which is used to short-circuit any flux from the medium (see Sec. III). This short-circuiting results in $\langle \mathbf{B} \rangle = \mu_0 \langle \mathbf{M} \rangle$, while the quantity $\Delta\mathbf{B}(\mathbf{r})$ is hardly affected. In the paper the influence of a yoke on $\langle \mathbf{B} \rangle$ will be taken into account, as a result of which $\langle \mathbf{B} \rangle$ is assumed to equal $\mu_0 \langle \mathbf{M} \rangle$ when analyzing the rotation angle ϕ. Any influence of the yoke on $\Delta\mathbf{B}(\mathbf{r})$ and therewith on the depolarization will be neglected. As a consequence of the latter $\langle \mathbf{B} \rangle$ in Eq. (6) is assumed to equal $\mu_0 \langle \mathbf{M} \rangle$. As no significant approximations have been used in the derivation of Eqs. (4) and (5) it appears that a ND experiment directly yields the length over which the deviations in the local magnetization (magnetic induction) from the mean magnetization (magnetic induction) are correlated. This can deliver useful information on particulate media.

In particulate media the use of the correlation matrix $\underline{\omega}$, defined

$$\underline{\omega} = \frac{2\underline{\alpha}}{\epsilon(\mu_0 \langle M_s \rangle)^2} , \tag{10}$$

is more suitable than the use of $\underline{\alpha}$, as $\underline{\omega}$ has the dimension of length and does not trivially depend on the particle volume fraction ϵ. Here, $\langle\ \rangle$ means an average over all particles. Therefore, $\underline{\omega}$, its trace ζ and the quantities γ_i will be used in the following. The quantity ζ is the correlation length of $[\Delta\mathbf{B}(\mathbf{r})]^2$ along $\mathbf{e}_0$ normalized on $\Delta\mathbf{B}(\mathbf{r}) = \mu_0 M_s$ and $\epsilon = 1$. A decrease in $\Delta\mathbf{B}(\mathbf{r})$ with a factor f results in a decrease of ζ with a factor f^2. Note that $\zeta = 2\xi / [\epsilon(\mu_0 \langle M_s \rangle)^2]$ and that $\gamma_i = \omega_{ii}/\zeta$. A factor 2 is added in the definition of $\underline{\omega}$ in order that ζ approximates to the average particle size along $\mathbf{e}_0$ in case of magnetically uncorrelated particles (see Sec. II B). Furthermore, if a quantity or matrix refers to a medium with $\langle B \rangle = 0$, a superscript zero will be added (e.g., $\underline{\omega}^0, \zeta^0, \gamma_i^0$).

If Eqs. (6) and (10) are applied to an ensemble of identical single-domain particles with volume V and spontaneous magnetization $\mathbf{M}_s$, it follows that

$$\omega_{ij} = \frac{1}{4\pi^2 N \langle V \rangle} \sum_{k=1}^{N} \sum_{l=1}^{N} \left[\int_S d^2\mathbf{s}\, n_i^{*k}(\hat{\mathbf{s}}) n_j^{*l}(\hat{\mathbf{s}}) F^k(\mathbf{s}) F^l(\mathbf{s}) e^{i\mathbf{s}\cdot(\mathbf{E}^k - \mathbf{E}^l)} \right] - \tfrac{4}{9} L_W \epsilon m_i m_j . \tag{11}$$

Here the superscripts k and l refer to the particles in W (total number N), $F(\mathbf{s}) = 1/V \int_V e^{i\mathbf{s}\cdot\mathbf{r}} d^3\mathbf{r}$ is the particle form factor, $\langle V \rangle$ is the average particle volume, $\mathbf{n}(\hat{\mathbf{s}}) = \hat{\mathbf{s}} \times (\mathbf{n} \times \hat{\mathbf{s}})$ with $\mathbf{n}$ the unit vector along $\mathbf{M}_s$, and m_i is the i component of the reduced magnetization $\mathbf{m}(\langle \mathbf{M} \rangle = \epsilon M_s \mathbf{m}, |\mathbf{m}| \le 1)$. The vector $\mathbf{E}$ gives the position of the center of mass of a particle. The $k=l$ terms of Eq. (11) represent the contribution to $\underline{\omega}$ of the individual particles. The terms for which $k \neq l$ represent the contribution to $\underline{\omega}$ of interparticle correlations and of overlap of demagnetizing fields of different particles within W.

B. The correlation matrix $\underline{\omega}$ for magnetically uncorrelated single-domain particles

This section discusses the matrix $\underline{\omega}$ for an ensemble of magnetically uncorrelated single-domain particles. In order to simplify the calculations involved, the particles are assumed to be ellipsoids with axial dimension $2b$ and radial dimension $2a$. The form factor of such an ellipsoid is given by

$$F(\mathbf{s}) = \frac{3(\sin u - u \cos u)}{u^3} \tag{12}$$

with

$$u = |\mathbf{s}| \sqrt{[(\mathbf{n}\cdot\mathbf{s})^2 (b^2 - a^2) + a^2]} .$$

The spontaneous particle magnetization $M_s \mathbf{n}$ is oriented along the b axis.

The correlation matrix $\underline{\omega}$ is derived from Eq. (11) by taking W as the subvolume containing a single particle ($W = V/\epsilon$) and by averaging the result obtained over the particle orientation

$$\omega_{ij} = \frac{\delta_{ij}}{4\pi^2 \langle V \rangle} \int_S d^2\mathbf{s} \langle V^2 [n_i^*(\hat{\mathbf{s}})]^2 F^2(\mathbf{s}) \rangle - c_2 \bar{z} m_i m_j . \tag{13}$$

Here, $\bar{z}$ is the mean particle size along $\mathbf{e}_0$, $c_2 = (4\pi\epsilon^2/81)^{1/3}$, and $\langle \cdots \rangle$ is an average over all particles. In order to derive Eq. (13), the quantity L_W has been approximated by

$$L_W = \bar{z} \left[\frac{9\pi}{16\epsilon} \right]^{1/3} , \tag{14}$$

a value which is exact when the axes of the ellipsoids coincide with the system directions. For spheres $L_W = (V/\epsilon)^{1/3}$. Equation (13) can also be obtained by taking W large, so that W is representative for the total medium, and by neglecting the $k \neq l$ terms of Eq. (11). In this case no demagnetization fields outside W arising from W exist. However, the overlap of demagnetization fields of particles within W has been neglected now (due to the negligence of the $k \neq l$ terms).

Substitution of Eq. (12) in Eq. (13) and integrating over $|\mathbf{s}|$ results in

$$\omega_{ij}=\delta_{ij}\frac{3}{2\langle a^2 b\rangle}\left[\frac{1}{2\pi}\int_S d\hat{\mathbf{s}}\left\langle\frac{a^4 b^2(n_i^*(\hat{\mathbf{s}}))^2}{(\mathbf{n}\cdot\hat{\mathbf{s}})^2(b^2-a^2)+a^2}\right\rangle\right] - c_2\bar{z}m_i m_j \ . \quad (15)$$

Equation (15) can be solved analytically for spheres only. In case $\langle \mathbf{M}\rangle$ is along x, y, or z, the application of Eq. (15) to identical spherical particles with radius R results in

$$\gamma_x=\frac{\frac{3}{4}\langle n_x^2\rangle+\frac{1}{4}\langle n_y^2\rangle-\tau_x}{1+\langle n_z^2\rangle-\tau}\ ,$$
$$\gamma_y=\frac{\frac{1}{4}\langle n_x^2\rangle+\frac{3}{4}\langle n_y^2\rangle-\tau_y}{1+\langle n_z^2\rangle-\tau}\ , \quad (16)$$
$$\gamma_z=\frac{2\langle n_z^2\rangle-\tau_z}{1+\langle n_z^2\rangle-\tau},\quad \zeta=\tfrac{3}{4}R\,(1+\langle n_z^2\rangle-\tau)\ ,$$

or inversely,

$$\langle n_x^2\rangle=\frac{(\frac{3}{2}\gamma_x+\frac{1}{2}\gamma_y)[2(1-\tau)+\tau_z]}{2-\gamma_z}+(\tfrac{3}{2}\tau_x-\tfrac{1}{2}\tau_y)\ ,$$
$$\langle n_y^2\rangle=\frac{(\frac{3}{2}\gamma_y-\frac{1}{2}\gamma_x)[2(1-\tau)+\tau_z]}{2-\gamma_z}+(\tfrac{3}{2}\tau_y-\tfrac{1}{2}\tau_x)\ , \quad (17)$$
$$\langle n_z^2\rangle=\frac{\gamma_z(1-\tau)+\tau_z}{2-\gamma_z},\quad R=\frac{\frac{4}{3}\zeta(2-\gamma_z)}{2(1-\tau)+\tau_z}\ ,$$

with $\tau_i=16m_i^2c_2/9$ and $\tau=16m^2c_2/9$.

At $m=0$, a single particle can be considered as a magnetic inhomogeneity with magnetic amplitude $\Delta B=\mu_0 M_s$. As a result, the quantity ζ^0 approximates to the mean particle size along the neutron path. The latter is illustrated by Fig. 1, which gives the quantities ζ^0/a and ζ^0/b, numerically calculated from Eq. (15), versus b/a in case of an isotropic particle orientation distribution. For such a distribution ζ^0 is roughly proportional to b for $b/a<0.1$ and to a for $b/a>10$. If b/a approaches infinity, ζ^0/a approaches $3\pi/4$. The correlation length ζ^0 equals $3b/2$ independent of the particle orientation, if b/a approaches zero.

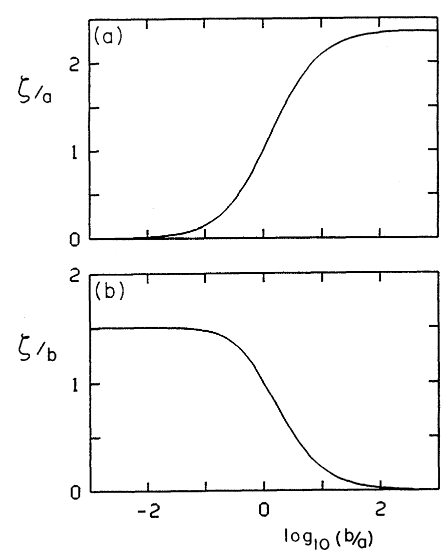

FIG. 1. The quantities ζ^0/a (a) and ζ^0/b (b) vs $\log_{10}(b/a)$ in case of an isotropic particle orientation distribution.

For magnetically uncorrelated particles, γ_i^0 is dependent on the particle shape and orientation. The former dependence is due to the correlation between the particle shape and the orientation of $\mathbf{M}_s$ and to the demagnetization fields of the particles. In general, the more the particles are oriented along the i direction the larger the value of γ_i^0 (in the theory of Rekveldt $\gamma_i^0=\langle n_i^2\rangle$ (Ref. 13)).

Figures 2 and 3 give γ_i^0 versus θ_0 for several values of b/a in case identical ellipsoidal particles are oriented within a cone with apex θ_0 along the y and z directions, respectively. The dependence of γ_i^0 on the particle shape appears to be much stronger when the cone is oriented along the y direction than when it is oriented along the z direction. In case of an isotropic particle magnetization orientation distribution, $\gamma_x^0=\gamma_y^0=\frac{1}{4}$ and $\gamma_z=\frac{1}{2}$, independent of the particle shape. This phenomenon, called *the*

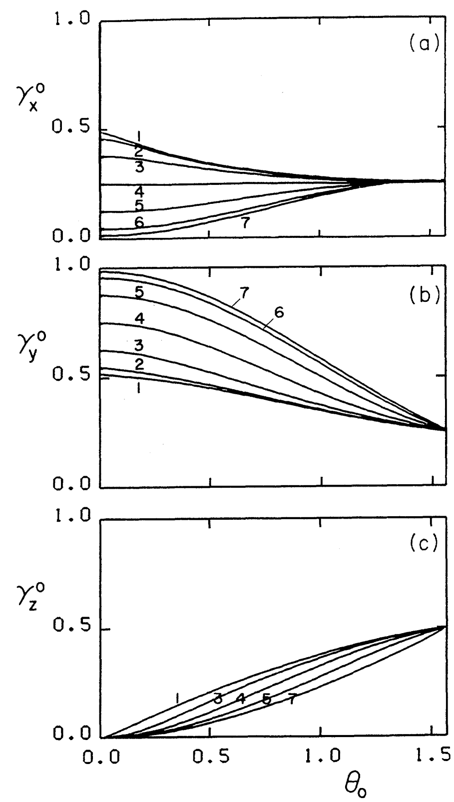

FIG. 2. The quantities γ_x^0 (a), γ_y^0 (b), and γ_z^0 (c) vs the apex θ_0 of the cone the particle magnetizations are oriented in. The cone is along the y direction [$b/a=\frac{1}{30}$ (1), $\frac{1}{10}$ (2), $\frac{1}{3}$ (3), 1 (4), 3 (5), 10 (6), 30 (7)].

intrinsic anisotropy, is extensively discussed in Ref. 15. If all particles are oriented along the $\pm z$ direction, γ_z^0 equals 1 while in case all particles are oriented along the $\pm y$ direction ($\pm x$ direction) the quantity $\gamma_y^0(\gamma_x^0)$ is smaller than 1, due to the influence of the local demagnetization fields on $\underline{\omega}$. As these fields are related to the particle shape, this effect is dependent on the particle shape. Figure 4 gives γ_x^0 and γ_y^0 ($\gamma_z^0=0$) versus b/a for magnetically uncorrelated identical ellipsoidal particles oriented along the $\pm y$ direction. When b/a approaches 0, γ_x^0 and γ_y^0 approach $\frac{1}{2}$, while when b/a approaches infinity γ_x^0 and γ_y^0 approach 0 and 1, respectively. In the latter case the demagnetization fields of the particles equal zero, as a result of which $\gamma_i^0=\langle n_i^2\rangle$. The examples given here illustrate the strong dependence of γ_i on the particle shape. This dependence, which has been fully taken into account in the ND formulas for the first time in Ref. 15, is important when deriving the particle orientation distribution from a ND experiment.

In case $m\neq 0$ an increase in ϵ or m trivially results in a decrease in $\Delta\mathbf{B}(\mathbf{r})$ and therefore in ζ and $\gamma_\parallel$, with $\gamma_\parallel$ the value of γ_i in the direction parallel to $\langle\mathbf{M}\rangle$. This is important when analyzing the dependence of ζ and γ_i on the magnetic state. Whenever $\langle\mathbf{M}\rangle$ is oriented perpendicular to $\mathbf{e}_0$ (e.g., along the y direction), as is the case in most ND experiments,

$$\zeta=\zeta^0(1-c_2c_3m^2)\;, \tag{18}$$

$$\gamma_y=\frac{\gamma_y^0-c_2c_3m_y^2}{1-c_2c_3m^2}\;, \tag{19}$$

or inversely,

$$\zeta^0=\frac{\zeta}{1-c_2c_3m^2}\;, \tag{20}$$

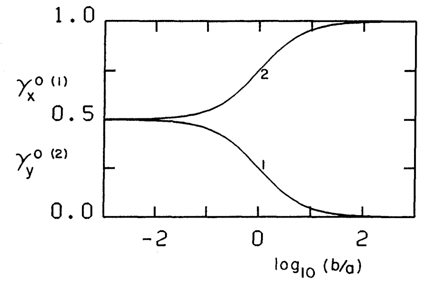

FIG. 4. The quantities γ_x^0 (1) and γ_y^0 (2) vs $\log_{10}(b/a)$ when the particles are oriented along the $\pm y$ direction ($\gamma_z^0=0$).

$$\gamma_y^0=\gamma_y(1-c_2c_3m^2)+c_2c_3m_y^2\;, \tag{21}$$

with $c_3=\overline{z}/\zeta^0$ and $m=\langle M\rangle/(\epsilon M_s)$. The quantity c_3 is of the order of 1, the exact value being dependent on the average particle shape and orientation and spread in particle volume. Note that c_3 does not depend on the volume itself. The larger the spread in particle volume, the smaller is c_3. For identical spheres $c_3=\frac{16}{9}/(1+\langle n_z^2\rangle)$. In most particulate media ϵ is smaller than 0.4, resulting in c_2 less than 0.3. Then, assuming c_3 equal to 1, ζ/ζ^0 has a value between 0.9 and 1 for isotropically distributed media ($m\leq\frac{1}{2}$) and a value between 0.7 and 1 for fully aligned media ($m\leq 1$). The fact that ζ/ζ^0 in particulate media is of the order of 1 for all m values is due to a varying neutron transmission length over the cross section of a particle, to local demagnetization fields around the particles and to a value of ϵ substantially smaller than 1. In homogeneous ferromagnets ($\epsilon=1$), ζ/ζ^0 equals 0 at $m=1$. In calculating ζ^0 and γ_y^0 from ζ

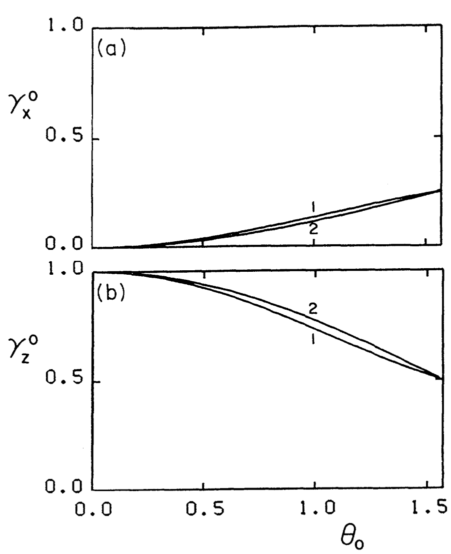

FIG. 3. The quantities γ_x^0 (a), and γ_z^0 (b) vs the apex θ_0 of the cone the particle magnetizations are oriented in ($\gamma_y^0=\gamma_x^0$). The cone is along the z direction [$b/a=\frac{1}{30}$ (1), 30 (2)].

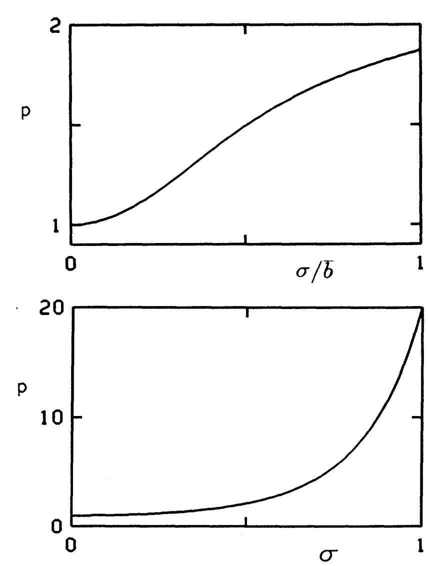

FIG. 5. The quantity p vs $\sigma/\bar{b}$ (a) and σ (b) in case b/a is constant and b follows a Gaussian (a) or a log-normal (b) distribution.

and γ_y, it should be realized that c_3 generally is not known exactly and furthermore that demagnetization fields of neighboring particles may overlap. Errors in ζ^0 and γ_y^0 due to the latter and due to an uncertainty in c_3 increase with increasing $c_2c_3m^2$.

In case of polydisperse particles, the depolarization will be dominated by the depolarization arising from the largest particles. Following from Eq. (15) the contribution to ω_{ij} of a single particle is proportional to a^2b^2. As a result, a spread in particle size which is independent on the particle orientation results in an increase in ζ with a factor

$$p=\frac{\langle a^2b^2\rangle}{\langle a^2b\rangle\langle b\rangle} . \tag{22}$$

Figure 5(a) gives p versus the quantity $\sigma/\bar{b}$ in case b/a is constant and b follows a Gaussian distribution

$$f(b)=\frac{1}{\sigma\sqrt{(2\pi)}}e^{-(b-\bar{b})^2/(2\sigma^2)} . \tag{23}$$

Figure 5(b) gives p versus σ when b/a is constant and b follows a log-normal distribution

$$f(b)=\frac{1}{b\sigma\sqrt{(2\pi)}}e^{-\ln^2(b/\bar{b})/(2\sigma^2)} . \tag{24}$$

In Eq. (23) $\bar{b}$ is the average value of b, while $\ln(\bar{b})$ is the average value of $\ln(b)$ in Eq. (24).

C. The effect of interparticle correlations on $\underline{\omega}$

When the particles are magnetically uncorrelated, $\underline{\omega}$ yields information about the particle properties. As already mentioned, interparticle properties may also highly contribute to $\underline{\omega}$. Generally speaking, positive correlations along $\mathbf{e}_0$ in the i component ($i=x,y,z$) of $\mathbf{n}$ of neighboring particles result in an increase in ω_{ii} and therewith in ζ and γ_i. Negative correlations along $\mathbf{e}_0$ result in a decrease of ω_{ii}. Due to the demagnetization fields of the particles, correlations along $\mathbf{e}_0$ in n_i may not only affect ω_{ii} but also ω_{vv} ($i\neq v$). Correlations between n_i and n_j ($i\neq j$) of different particles are likely to occur only in case of a nonisotropic orientation distribution of particle magnetizations of which the main directions differ from the system directions. Then the nondiagonal elements ω_{ij} ($i\neq j$) may yield the main directions of the distribution (e.g., Ref. 18).

Magnetic interparticle correlations can be the consequence of (magnetic) particle interactions, orientational correlations between neighboring particles and the presence of density variations within the medium. In the next three paragraphs these different causes of correlations are discussed for particulate media in the remanent state. It is assumed that the particle anisotropy is large, such that the particle magnetization is along an easy axis. This assumption is generally valid in particulate media.

Particle interactions may result in positive as well as negative correlations as they favor a parallel magnetization orientation in particles located in the direction along $\mathbf{M}_s$ and an antiparallel magnetization orientation in particles located in the direction perpendicular to $\mathbf{M}_s$ [Fig. 6(a)]. As a result, particle interactions may cause that magnetization reversal takes place along lines parallel to $\langle\mathbf{M}\rangle$. Then the superdomains during the magnetization reversal process exceed the particle volume and are elongated along $\langle\mathbf{M}\rangle$. A superdomain should be taken as a subvolume of the medium the mean magnetization of which differs from $\langle\mathbf{M}\rangle$. If the particle magnetization is along an easy axis, the effect of particle interactions on the local magnetization distribution is zero in the state of maximum remanence. It is generally maximum around $m=0$. The precise influence of particle interactions on the micromagnetic state is complicated as the dipole interactions have a long range and as the interactions depend on many aspects of the microstructure, such as the exact particle location and the intrinsic particle properties. It is discussed extensively in the literature (e.g., Refs. 19 and 20).

Particle orientational correlations may result in positive magnetic particle correlations along $\mathbf{e}_0$ in two ways, i.e., *directly* and *indirectly*. *Directly* they result in correlations in magnetized media, as particle orientational correlations are spatial particle correlations and these trivially form magnetic correlations at $m\neq0$. As an example, subvolumes in which the mean particle orientation differs from that in the medium *directly* act as large superdomains at $m\neq0$. This effect is due to the correlation between $\mathbf{M}_s$ and the particle shape [Fig. 6(b)] and increases with increasing m. Particle orientational correlations may *indirectly* result in correlations by the fact that the particle coercive field depends on the particle orientation. Therefore, particle orientational correlations result in correlations of the particle coercive field and subvolumes in which the particle orientations are highly correlated may form superdomains during the magnetization reversal process which exceed the particle volume.

Similar to particle orientational correlations, density variations within a particulate medium may result in pos-

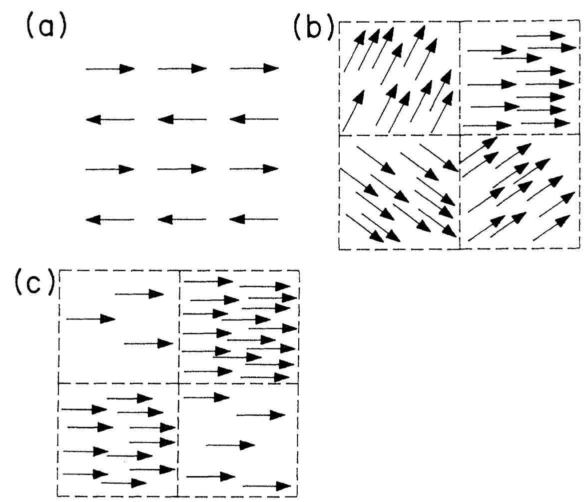

FIG. 6. Magnetic interparticle correlations as a result of magnetic interactions (a), particle orientational correlations (b), and density variations (c).

itive correlations *directly* and *indirectly*. They may *directly* contribute to $\underline{\omega}$ as density variations within a particulate medium result in additional variations in $\mathbf{B}(\mathbf{r})$ and therefore in an increase in $\underline{\omega}$ (corresponding to positive correlations). If this contribution is considered, subvolumes with a particle volume fraction larger (smaller) than the average value act as superdomains with a mean magnetization parallel (antiparallel) to $\langle \mathbf{M} \rangle$ [Fig. 6(c)]. Density variations are likely to affect $\underline{\omega}$ substantially in this way only when they exist over a length much larger than the particle size. Similar to particle orientational correlations, density variations may also *indirectly* affect $\underline{\omega}$ as the local coercive field depends on the local particle density. Consequently, also density variations may result in superdomains during the magnetization reversal process which exceed the particle volume. Note that the *direct* effects of particle orientational correlations and density variations are related to the degree of the particle dispersion. Often, the contributions of the different types of correlations to $\underline{\omega}$ can be (partially) disentangled by studying the dependence of $\underline{\omega}$ on m (see Sec. III).

In the above, several causes of correlations have been discussed. The exact form of the correlations has not yet been dealt with. Assumptions on this form have to be made in order to obtain expressions for their contributions to $\underline{\omega}$, which are more specific than Eq. (11). In the following sections two forms of correlations will be discussed. At first, the contribution to $\underline{\omega}$ of long-range positive correlations existing within large 3D subvolumes is discussed (Sed. II C 1). These subvolumes can be taken as superdomains with a magnetization depending on the mean particle density and orientation within the subvolume. Secondly, short-range positive as well as negative correlations, which require a different description method, are dealt with (Secs. II C 2 and II C 3, respectively).

1. The effect of long-range positive correlations on $\underline{\omega}$

Positive magnetic correlations existing in large 3D superdomains V_d are considered. The superdomain magnetization is $\mathbf{M}_d = \epsilon m_d M_s \mathbf{n}_d$ ($|\mathbf{n}_d| = 1$). If the contribution of the individual particles to $\underline{\omega}$ is neglected, the correlation matrix of a medium consisting of these superdomains is equivalent to $\underline{\omega}$ of a medium of magnetically uncorrelated single-domain particles with a particle volume fraction of 1 (in principal not possible), a particle size and shape equal to those of the superdomains and a particle spontaneous magnetization equal to $\mathbf{M}_d$ [see Eq. (13)]. Note that the first term on the right-hand side of the obtained result has to be divided by ϵ due to the definition of $\underline{\omega}$ [see Eq. (10)]:

$$\omega_{ij} = \delta_{ij} \frac{\epsilon m_d^2}{4\pi^2 \langle V_d \rangle} \int_S d^2\mathbf{s} \langle V_d^2 [n_d^*(\hat{\mathbf{s}})]_i^2 F_d^2(\mathbf{s}) \rangle - \epsilon c_4 \bar{z}_d m_i m_j \; . \tag{25}$$

Here, $F_d(\mathbf{s}) = 1/V_d \int_{V_d} e^{i\mathbf{s}\cdot\mathbf{r}} d^3\mathbf{r}$ is the superdomain form factor, $\bar{z}_d$ is the mean superdomain size along $\mathbf{e}_0$, $c_4 = (4\pi/81)^{1/3} (=0.54)$ and $\mathbf{n}_d^*(\hat{\mathbf{s}}) = \hat{\mathbf{s}} \times (\mathbf{n} \times \hat{\mathbf{s}})$. Following from Eq. (25) the contribution to ζ of a superdomain with size L_d along $\mathbf{e}_0$ is of the order of $L_d \epsilon (m_d^2 - m^2)$. This is consistent with the picture that a superdomain approximates to a magnetic inhomogeneity with magnetic amplitude $\Delta B = \epsilon (m_d - m) \mu_0 M$, which exists along $\mathbf{e}_0$ over a length L_d.

The reduced superdomain magnetization m_d is directly related to the particle orientation distribution within a superdomain. The narrower this distribution is, the larger m_d and the larger the contribution of the superdomains to $\underline{\omega}$. In case the superdomains are the result of particle interactions only, the average orientation distribution within a superdomain equals the distribution within the medium. Then the maximum value of m, denoted $m_{\max}$, equals the maximum value of m_d. At $m = m_{\max}$, the superdomains do not exist anymore as observable magnetic inhomogeneities and hence they do not contribute to $\underline{\omega}$. The values of γ_i corresponding to superdomains as a result of particle interactions depend on the local orientation and shape of these superdomains. During the demagnetization process $\mathbf{M}_d$ is expected to be oriented closely around $\langle \mathbf{M} \rangle$. Then $\gamma_\parallel$ approaches a value close to 1. The exact value of γ_ω depends on the superdomain shape (see Fig. 4, curve 2).

If the superdomains are due to particle orientational correlations, the average spread in particle orientation within a superdomain is smaller than that within the medium. Then the maximum value of m_d always exceeds m and the superdomains may contribute to $\underline{\omega}$ over the entire m range. The values of γ_i corresponding to superdomains as a result of orientational correlations are related to the spread in $\mathbf{n}_d$.

The direct effect (see above) of density variations on $\underline{\omega}$ is substantial only when V_d is much larger than the particle volume. Then $\mathbf{M}_d$ is along $\langle \mathbf{M} \rangle$ and $m_d = m \Delta\epsilon / \bar{\epsilon}$, with $\bar{\epsilon}$ the average particle volume fraction and $\epsilon + \Delta\epsilon$ the particle volume fraction of V_d ($\langle \Delta\epsilon = 0 \rangle$). The direct contribution of the density variations to $\overline{\omega}$ is now given by Eq. (25), provided the second term on its right-hand side is omitted (due to $\langle \Delta\epsilon \rangle = 0$). The contribution of a subvolume V_d approximates to $L_d m^2 (\Delta\epsilon)^2 / \bar{\epsilon} \ll L_d$. If $\underline{\omega}$ is dominated by depolarization arising from the direct effect of density variations, ζ is expected to be proportional to m^2 and $\gamma_\parallel$ will approach a value close to 1. If the superdomains are due to H_c correlations arising from the density variations, Eq. (25) is valid with $0 \le m_d \le 1$.

Only the depolarization arising from the mean magnetization of the subvolumes V_d has been considered so far. The contributions to $\underline{\omega}$ of the individual particles and short-range correlations within V_d, being much smaller, have been neglected. The contribution to $\underline{\omega}$ of the individual particles within a superdomain is given by Eq. (13), in which $m_i m_j$ should be replaced by $\delta_{ij} [(n_d)_i m_d]^2$. The contribution to $\underline{\omega}$ of short-range positive and negative correlations is discussed in Secs. II C 2 and II C 3, respectively.

2. The effect of short-range positive correlations on $\underline{\omega}$

The particle shape and precise particle location hardly interfere with the contribution to $\underline{\omega}$ of long-range positive correlations. As a result, this contribution can be described well using the rather simple model of superdomains. However, the particle shape and precise particle location do effect the contribution of short-range correlations to $\underline{\omega}$. Therefore, a more profound model is necessary to take the contribution of short-range correlations to $\underline{\omega}$ into account. In this section the latter contribution is discussed by considering the effect on $\underline{\omega}$ of positive correlations in n_i of N identical particles located in a linear row. Such a row, in which the components of $\mathbf{M}_s$ along $\langle \mathbf{M} \rangle$ have the same sign, will be called a *chain*. The contribution to $\underline{\omega}$ of these chains is dependent on their orientation with respect to $\mathbf{e}_0$ and the spread in orientation and size of the particles within them, as will be discussed below.

The contribution to $\underline{\omega}$ of chains oriented along $\mathbf{e}_0$ is given by [see Eqs. (11) and (13)]

$$\omega_{ij} = \frac{\delta_{ij}}{4\pi^2 \langle V \rangle} \int_S d^2\mathbf{s} \{ \langle V^2 [n_i^*(\hat{\mathbf{s}})]^2 F^2(\hat{\mathbf{s}}) \rangle + (N-1) \langle V n_i^*(\hat{\mathbf{s}}) F(\hat{\mathbf{s}}) \rangle^2 \} - N c_2 \bar{z} m_i m_j \,, \tag{26}$$

where $\langle\ \rangle$ is an average over the particles within the chains. The terms on the right-hand side of Eq. (26) describe the contribution to $\underline{\omega}$ of the individual particles, the contribution to $\underline{\omega}$ of the correlations within the chain and the "correction" for the mean magnetization (see Sec. II B), respectively. When n_y or n_z is positively correlated within chains consisting of identical spheres with radius R, Eq. (26) corresponds to

$$\begin{aligned} \gamma_x^0 &= (3\langle n_x^2 \rangle + \langle n_y^2 \rangle + (N-1)\langle n_y \rangle^2)/(4f) \,, \\ \gamma_y^0 &= (\langle n_x^2 \rangle + 3\langle n_y^2 \rangle + 3(N-1)\langle n_y \rangle^2)/(4f) \,, \\ \gamma_z^0 &= 2\langle n_z^2 \rangle / f, \quad \zeta^0 = \tfrac{3}{4} R f \,, \end{aligned} \tag{27}$$

and

$$\begin{aligned} \gamma_x^0 &= (3\langle n_x^2 \rangle + \langle n_y^2 \rangle)/(4g) \,, \\ \gamma_y^0 &= (\langle n_x^2 \rangle + 3\langle n_y^2 \rangle)/(4g) \,, \\ \gamma_z^0 &= 2(\langle n_z^2 \rangle + (N-1)\langle n_z \rangle^2)/g \,, \\ \zeta^0 &= \tfrac{3}{4} R g \,, \end{aligned} \tag{28}$$

respectively. Here, $f = 1 + \langle n_z^2 \rangle + (N-1)\langle n_y \rangle^2$ and $g = 1 + \langle n_z^2 \rangle + 2(N-1)\langle n_z \rangle^2$. The contributions of the correlations to ζ^0 and γ_i^0 in Eqs. (27) and (28) equal the values of magnetically uncorrelated spherical particles with radius $(N-1)R\langle n_y \rangle^2$ and $(N-1)R\langle n_z \rangle^2$ oriented along the $\pm y$ direction and $\pm z$ direction, respectively. These contributions are dependent on $\langle n_y \rangle^2$ and $\langle n_z \rangle^2$ and therewith on the spread in the particle orientation within a chain.

This dependence on the particle orientation is even stronger with increasing b/a ratio, as shown in Fig. 7. In this figure, the quantity $\nu' = \nu/(N-1)$ with $\nu = \zeta_1^0/\zeta_2^0$ is plotted versus the apex angle θ_0 of cones within which the particles are assumed to be oriented (cone either along the y direction [Fig. 7(a)] or along the z direction [Fig. 7(b)]). Here, ζ_1 is the correlation length corresponding to the contribution to $\underline{\omega}$ of the correlations within the chains and ζ_2 the correlation length of the individual particles, numerically calculated from Eq. (26). Consequently, ν gives the ratio of the contributions to ζ of the correlations and the individual particles within the chains. Its value rapidly decreases with increasing θ_0 and with increasing b/a for $b/a \geq 1$. The decrease of ν with increasing θ_0 is due to the fact that only the component of $\mathbf{M}_s$ which is correlated along $\mathbf{e}_0$ contributes to $\underline{\omega}$. The average value of this component decreases with increasing θ_0. The decrease of ν with increasing $b/a (b/a > 1)$ is due to the fact that only correlations along $\mathbf{e}_0$ contribute to $\underline{\omega}$. The larger b/a is the smaller the projection of a particle on its neighboring particle along $\mathbf{e}_0$ (see Fig. 8).

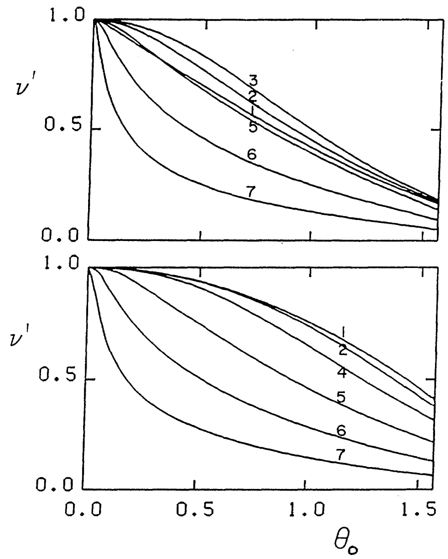

FIG. 7. The quantity ν' vs the apex angle θ_0 of the cone the particle magnetizations are oriented in. The cone is either along the y (a) or along the z direction (b) [$b/a = \frac{1}{30}$ (1), $\frac{1}{3}$ (2), 1 (3), 3 (4), 10 (5), 30 (6), 100 (7)].

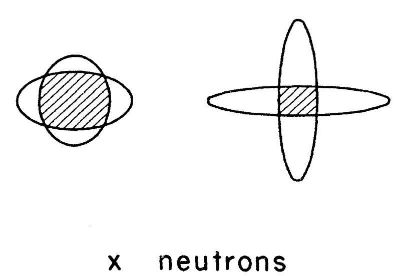

FIG. 8. The contribution to $\underline{\omega}$ of correlations between two particles is roughly proportional to the projection of a particle on its neighboring particle along $\mathbf{e}_0$. This projection (shadowed region) decreases with increasing b/a when the particle orientations differ.

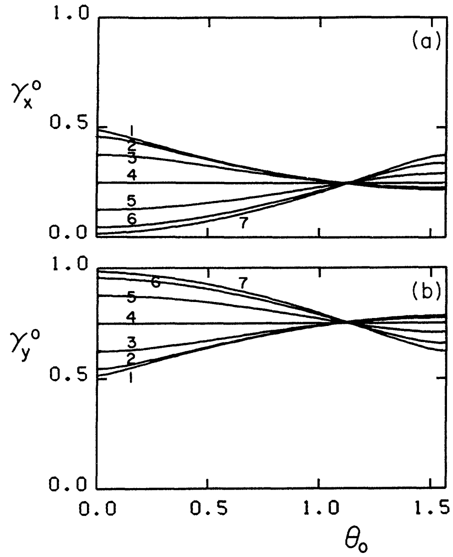

FIG. 9. The quantities γ_x^0 (a) and γ_y^0 (b) corresponding to chains along $\mathbf{e}_0$ vs the apex angle θ_0 of the cone the particle magnetizations are oriented in ($\gamma_z^0=0$). The quantity n_y is positively correlated within the chains and the cone is along the y direction [$b/a=\frac{1}{30}$ (1), $\frac{1}{10}$ (2), $\frac{1}{3}$ (3), 1 (4), 3 (5), 10 (6), 30 (7)].

The contribution of the correlations to $\underline{\omega}$ is roughly proportional to the area of this projection.

Figure 9 gives the values of γ_x^0 and γ_y^0 corresponding to the correlations within chains along $\mathbf{e}_0$ versus θ_0 ($\gamma_z^0=0$), in case n_y is positively correlated within the chains and in case the particle orientations are within a cone along the y direction with apex θ_0. Dependent on the particle shape, the quantity γ_x^0 has a value between 0 and 0.5 and the quantity γ_y^0 between 0.5 and 1. From a comparison between Figs. 2 and 9 it follows that the existence of chains along $\mathbf{e}_0$ in which n_y is positively correlated results in a small increase in γ_y^0 and a decrease in γ_z^0, in comparison with magnetically uncorrelated particles. The quantity γ_x^0 is only slightly affected by the correlations. If n_z is positively correlated in chains along

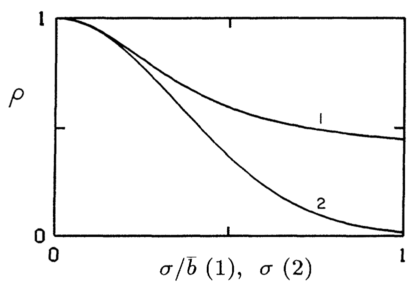

FIG. 10. The quantity ρ vs $\sigma/\bar{b}$ (1) and σ (2) when b/a is constant and b follows a Gaussian (1) or a log-normal (2) distribution.

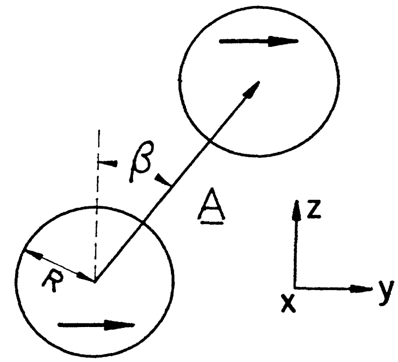

FIG. 11. A chain consisting of two identical spherical particles with radius R. The particle magnetization (small arrows) is along y.

$\mathbf{e}_0$, the contributions to $\underline{\omega}$ of the correlations correspond to $\gamma_x^0=\gamma_y^0=0$ and $\gamma_z^0=1$. Consequently, correlations in n_z along $\mathbf{e}_0$ result in an increase in γ_z^0 and a decrease in γ_x^0 and γ_y^0. However, it should be realized that the contribution of the correlations within the chains to γ_i^0 of the medium, being proportional to the parameter ν, decreases with increasing θ_0 and increasing $b/a\,(b/a\geq 1)$ (see above).

A spread in particle size within a chain results in a decrease in ν. When this spread is independent of the particle orientation and when b/a is constant, a spread in particle size results in a decrease in ν with a factor

$$\rho=\langle ab\rangle^2/\langle a^2b^2\rangle \tag{29}$$

[see Eqs. (11) and (15)]. Figure 10 gives the latter factor versus the quantity $\sigma/\bar{b}$, in case the size distribution is Gaussian with standard deviation σ, and versus σ when it is a log-normal distribution with log-normal standard deviation σ [see Eqs. (11) and (15)].

So far, it has been assumed that the chains are oriented along $\mathbf{e}_0$. Deviations from this alignment result in a decrease of the contribution to $\underline{\omega}$ of the correlations. In order to illustrate the latter, a chain consisting of two identical spherical particles with radius R is considered (Fig. 11). The particle spontaneous magnetization is along y. The vector between the centers of the spheres, $\mathbf{A}$, is in the yz plane and makes an angle β with the z direction. At $m=0$, the ratio μ of the contributions to $\underline{\omega}$ of such a chain and of two uncorrelated particles is given by [see Eq. (11)]

$$\mu=\frac{\int_S d^2\mathbf{s}\,F^2(\mathbf{s})[1+\cos(\mathbf{s}\cdot\mathbf{A})]}{\int_S d^2\mathbf{s}\,F^2(\mathbf{s})}\ . \tag{30}$$

Figure 12 gives μ versus β for $A=2R$, $A=4R$, and $A=6R$. Obviously, the quantity μ decreases with increasing β. An increase in A results in a decrease in μ. At $\beta=\pi/2$, μ exceeds 1, which is due to the overlap of demagnetization fields. The value of μ averaged over β is 1.54 for $A=2R$, 1.32 for $A=4R$, and 1.21 for $A=6R$.

It follows from the above that one-dimensional correla-

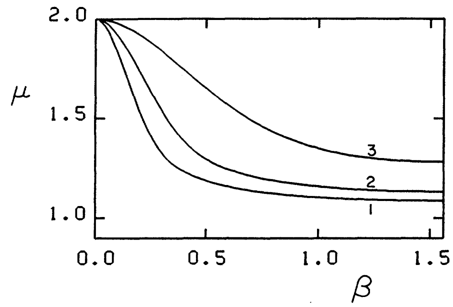

FIG. 12. The quantity μ vs β for $A/R=6$ (1), 4 (2), and 2 (3).

tions can only substantially affect ω if they occur along directions closely oriented along $\mathbf{e}_0$ (see Fig. 12) and provided that the spread in particle size is relatively small. Chains closely oriented along $\mathbf{e}_0$ may be found in dc-demagnetized media, in highly oriented media such as recording tapes or in media containing strong particle-orientation correlations.[8,21] Unless elongated particles are highly oriented, they generally have several neighbors along $\mathbf{e}_0$ and the model of chains is not valid anymore. In that case, only positive correlations existing in 3D superdomains as discussed in Sec. II C 1 may contribute to ω.

3. *The effect of short-range negative correlations on ω*

Negative correlations between particle magnetizations, which may arise from magnetic interactions, generally occur on a small scale. This section deals with chains of two particles with an antiparallel orientation of n_y or n_z.

If such chains are oriented along $\mathbf{e}_0$ the contribution to ω is given by Eq. (26), with $(N-1)$ replaced by -1. Consequently, the ratio of the contributions to ω of the negative correlations within chains along $\mathbf{e}_0$ and the individual particles is given by $-\nu'$ (see Fig. 7). In principle, negative correlations along $\mathbf{e}_0$ may result in a ζ equal to 0. However, the influence of the correlations on ω is generally smaller, due to a spread in the size or the orientation of the particles within the chains or to chain orientations different from $\mathbf{e}_0$.

Figure 13 gives the values of γ_i of chains along $\mathbf{e}_0$ in which n_y is negatively correlated. Negative correlations along $\mathbf{e}_0$ in n_y result in a decrease of ζ and γ_y^0 and an increase in γ_x^0 and γ_z^0, following from a comparison between Figs. 2 and 13. Negative correlations along $\mathbf{e}_0$ in n_z are not likely to occur for physical reasons [see Fig. 6(a)].

D. The influence of variations in the total neutron transmission length on ω

A neutron transmission length varying over the cross section of the medium may contribute to ω. Such a spread always exists in particulate media due to the fact that the neutron transmission length varies over the cross section of a single-domain particle. The latter contribution is included in ω of magnetically uncorrelated parti-

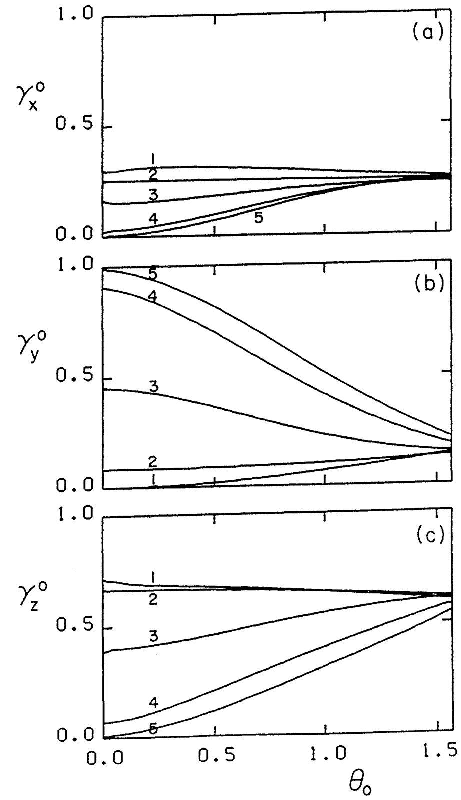

FIG. 13. The quantities γ_x^0 (a), γ_y^0 (b), and γ_z^0 (c) of chains along $\mathbf{e}_0$ vs the apex angle θ_0 of the cone ($\| y$) in which the particle magnetization orientation is. The chain consists of two identical particles with an antiparallel orientation of n_y [$b/a=\frac{1}{30}$ (1), 1 (2), 3 (3), 10 (4), 30 (5)].

cles (see Sec. II B). A varying number of particles along the neutron path, however, may give rise to an additional spread in neutron transmission length. The latter does almost not affect the depolarization when the mean magnetic induction equals zero. If $\langle B\rangle\neq 0$, however, this spread results in an additional variation in the rotation angle along a neutron path and therefore in an additional depolarization. This additional depolarization cannot always be described in terms of ω. If so, the total depolarization matrix $\underline{D}$ has to be considered.

If $\langle \mathbf{B}\rangle$ is along y and $f(N)$ is the normalized distribution function of the number of particles (N) along a neutron path, then due to a nonconstant $f(N)$ the depolarization elements D_{xx}, D_{zx}, D_{zx}, and D_{zz} will be decreased by a factor

$$D_a=\int_0^\infty f(N)\cos[(N-\bar{N})\vartheta]dN\ . \tag{31}$$

Here $\bar{N}$ is the mean value of N and $\vartheta=\mu_0\langle M\rangle \bar{z}\sqrt{c_1}$ the average rotation angle per particle. The additional depolarization described by Eq. (31) can be substantial for magnetized media with a high degree of alignment.

As an example, D_a is considered for a recording tape with all particles aligned along the $\pm y$ direction and with

$m = 1$. The particles are assumed to be ellipsoids with b/a approaching infinity. The distribution function $f(N)$ is assumed Gaussian [see Eq. (23)] with a squared standard deviation $\sigma_N^2 = q\bar{N}$. Substitution of Eq. (23) in Eq. (31) and using $\langle B \rangle = \mu_0 \langle M \rangle$ (rotation is considered!) and $L = \bar{N}\bar{z}/\epsilon$ yields

$$D_a = e^{-(2/3)c_1 \epsilon L (\mu_0 M_s)^2 a m^2 q} . \quad (32)$$

In this specific example, D_a can also be described in terms of $\underline{\omega}$ as σ_N^2 is proportional to $\bar{N}$. The decrease of D_{xx}, D_{xz}, D_{zx}, and D_{zz} with a factor D_a corresponds to an increase of ω_{yy} with

$$\omega_a = \tfrac{4}{3} a q m^2 . \quad (33)$$

Even for $q = 1$ this increase can be in the order of ω_{yy}^0, resulting in a strong increase in ζ and γ_y.

If $\underline{\omega}$ is dominated by a spread in neutron transmission length, ζ will be proportional to m^2 and $\gamma_\parallel$ should approach a value close to 1. It should be noted, however, that Eqs. (31) and (32) give an upperbound of D_a. As a consequence of demagnetization fields in those parts of the medium where no particles are located, D_a can be (much) smaller than these equations indicate.

Variations in the neutron transmission length that substantially contribute to $\underline{\omega}$ are likely to exist mainly in particulate media with a small neutron transmission length, e.g., in recording tapes.

III. DISCUSSION

The ND technique can in principle be applied to all particulate media, provided the magnetic correlation length ζ is larger than about 10 nm. The precise minimum ζ value which can be determined depends on M_s, ϵL, and λ.

It has been shown in Sec. II that the correlation matrix $\underline{\omega}$ is affected by the intrinsic particle properties, magnetic interparticle correlations as a result of (magnetic) particle interactions, or particle orientational correlations, particle density variations within the medium, and a varying neutron transmission length. The separation of these different contributions to the correlation matrix may be problematic in the analysis of ND results. However, generally the mean particle size, shape, and orientation are known from other techniques, so that $\underline{\omega}$ can be corrected for the contribution of the individual particles. Furthermore, the determination of the dependence of $\underline{\omega}$ on m as part of the analysis allows to (partially) disentangle the other contributions.

In the state of maximum remanence $\underline{\omega}$ is affected by the individual particles, a varying neutron transmission length, and the direct effects of particle orientational correlations and (long-range) density variations. The effect of a varying neutron transmission length is important mainly in recording tapes. The contributions to $\underline{\omega}$ of a varying neutron transmission length and the (direct effect of) density variations can be distinguished from those of the individual particles and the (direct effect of) particle orientational correlations by the fact that the former two contributions yield a value of $\gamma_\parallel$ close to 1, while the latter two in principle may yield all values of $\gamma_\parallel$ between 0 and 1. Generally, the contributions to $\underline{\omega}$ in the state of maximum remanence of particle orientational correlations yield a $\gamma_\parallel$ which is higher than the value of $\gamma_\parallel$ corresponding to the individual particles.

Around $m = 0$, $\underline{\omega}$ may consist of contributions from the individual particles, particle interactions and the indirect effects (H_c correlations) of particle orientational correlations and density variations. The value of $\gamma_\parallel$ corresponding to the contributions of interactions resulting in positive correlations and of (indirect effect of) density variations will be close to 1, while $\gamma_\parallel$ of the other contributions may have all values between 0 and 1.

A correlation length smaller than that expected for individual particles can only be due to particle interactions resulting in negative correlations along $\mathbf{e}_0$. Furthermore, the analysis of $\underline{\omega}$ along different $\mathbf{e}_0$ directions helps to separate some of the main contributions. Such an analysis in general also yields the average particle or superdomain shape (e.g., Refs. 8, 17, and 21).

The techniques commonly used to study the micromagnetic state of particulate media are magnetization, susceptibility, and noise measurements. The bulk methods as magnetization and susceptibility measurements obviously yield rather restricted information about correlations in the micromagnetic state. On the other hand, the noise technique, which is mainly applied to recording materials, is also sensitive to the individual particles, particle correlations, and density variations. However, when comparing the results of noise and ND measurements the following aspects should be taken into account. In the first place, not only the micromagnetic state but also the surface roughness contributes to the noise. Secondly, the noise is sensitive to the volume of magnetic inhomogeneities whereas the ND is sensitive to the size along $\mathbf{e}_0$ of the inhomogeneities. Thirdly, the contribution of a magnetic inhomogeneity to the noise depends on its distance to the recording head and therefore on its location in the coating. The contribution of a magnetic inhomogeneity to the correlation length is independent on its exact location in ND. Often, it appears to be difficult to separate the various contributions to the noise. Besides, the information obtained about magnetic correlations is general qualitative instead of quantitative.

In summary, in comparison with the more conventional techniques mentioned above, the ND technique can yield more detailed information about the micromagnetic state of particulate media. Moreover, the information obtained about magnetic correlations is quantitative. However, magnetization, susceptibility, and noise measurements are cheaper and faster than ND measurements, and the noise level of recording materials in itself is an important quantity to study. Besides, as the different techniques are partially complementary, the most detailed information is expected to be obtained by applying a combination of the techniques (e.g., Ref. 21).

The small-angle neutron scattering (SANS) technique is the only technique which may yield information comparable to that obtained with the ND technique. In contrast with the ND technique, SANS is sensitive to both nuclear and magnetic inhomogeneities. However, often nuclear and magnetic scattering in particulate media cannot be separated. Furthermore, the total range of ζ in particulate media, typically from 10 mn up to mm's, cannot be covered with a SANS experiment. A SANS experiment in which a polarized neutron beam is used would sometimes yield more information than a ND experiment. However, SANS instruments using a polarized neutron beam are not easily available thus far.

In a ND experiment, magnetic fields larger than the coercive field of the medium (H_c) have to be used in order to cover $\underline{\omega}$ during the total magnetization reversal process. As the ND technique is very sensitive to stray fields which arise from equipment that produces these fields, the ND technique can generally be performed on particulate media only if they are in the remanent state (H_c on the order of 5–80 kA/m). During such an experiment the medium is positioned in a magnetic yoke in order to short-circuit any flux from the medium. As the neutron beam may not pass through the yoke, ND measurements on media with a remanent magnetization parallel to $\mathbf{e}_0$ are difficult to perform. Such experiments would be interesting in particular to study magnetic correlations along the field direction.

Up to now, ND measurements have been performed on various magnetic pigments,[22] recording audio and video tapes[8,21,23] and ferrofluids in order to study their micromagnetic state. The theory formulated in this paper has successfully been used to interpret these measurements. The dynamics of the micromagnetic state is under study at present.

As an example some of the results of ND measurements on a commerically available CrO_2 pigment used for video tapes are presented and discussed. The particles studied are acicular with a mean diameter of 20 nm, an aspect ratio of 10, and a spontaneous magnetization along the longitudinal axis.[24] The mean magnetization $\langle \mathbf{M} \rangle$ of the pigment is along y. Figure 14 gives the correlation length ζ versus the reduced remanent magnetization m during the magnetization (reversal) process of pigment (starting from the virgin state [Fig. 14(a)] and starting from the state of maximum remanence [Fig. 14(b)]). The correlation length $\zeta^0 = 36 \pm 2$ nm in the virgin state. It increases about tenfold with increasing m during the magnetization process. During the magnetization reversal process ζ decreases and increases again, with a minimum ($\zeta^0 = 100 \pm 10$ nm) which is three times the value in the virgin state. Figure 15 gives the quantity γ_y versus m. The oscillations in γ_y are artifacts as a result of experimental circumstances.[22] The latter, which affect $\underline{\omega}$ at $m \neq 0$, also account for the small oscillations observed in ζ. It follows that γ_y has a value between 0.2 and 0.5. At $m=0$, $\gamma_x^0 = 0.27$, $\gamma_y = 0.24$, $\gamma_z^0 = 0.49$ (± 0.02, virgin state) and $\gamma_x^0 = 0.30$, $\gamma_y^0 = 0.47$, $\gamma_z^0 = 0.23$ (± 0.05, demagnetized state).

Assuming the virgin 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^0 in the virgin state corresponds to $a = 17 \pm 2$ nm. This value exceeds the value given by supplier (10 nm), which may be due to possible correlations already present in the virgin pigment. However, it is more likely due to a large spread in the particle size, resulting in an increased value of ζ (see Fig. 5). The increase of ζ with increasing m, together with a value of γ_y smaller than 0.5, indicates the presence of subvolumes in which the particle orientations are highly correlated. With increasing m these subvolumes become magnetized and act as large 3D superdomains. The low value of γ_y

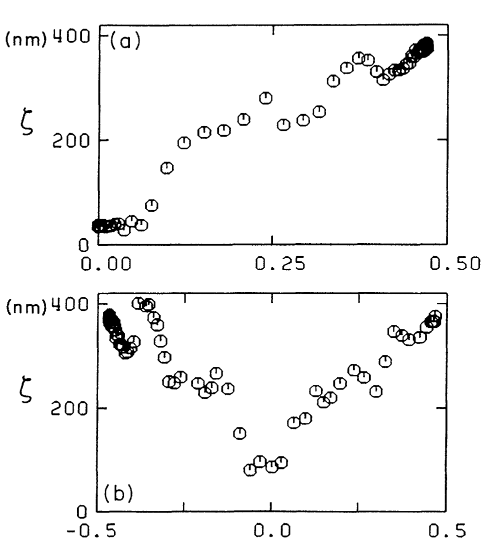

FIG. 14. The correlation length ζ vs the reduced remanent magnetization m during the magnetization (reversal) process of CrO_2 pigment [starting from the virgin state (a) and from the state of maximum remanence (b)].

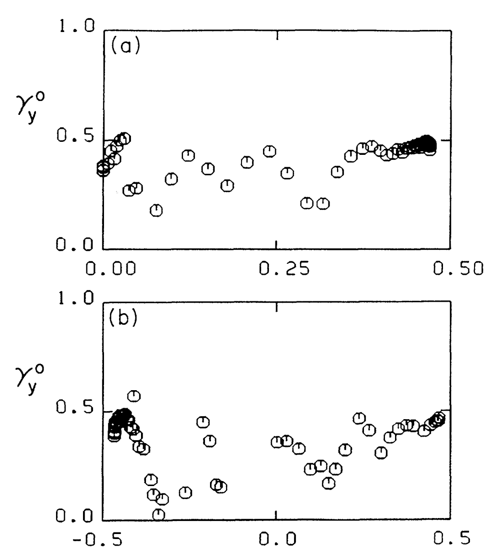

FIG. 15. The quantity γ_y vs the reduced remanent magnetization m during the magnetization (reversal) process of CrO_2 pigment [starting from the virgin state (a) and from the state of maximum remanence (b)].

over the whole m range makes large effects of density variations unlikely. The switching volumes in the magnetization reversal process are much smaller than these superdomains. Their size along $\mathbf{e}_0$ still exceeds the particle size, however, following from a comparison of the value of ζ^0 in the virgin and the demagnetized states. The latter is consistent with the value of γ_y^0 in the demagnetized state, which exceeds the value in the virgin state. A more extensive discussion of these measurements together with ND results on other pigments will be given in Ref. 22.

IV. SUMMARY AND CONCLUSIONS

Neutron depolarization theory in particulate media has been discussed. The relation between the correlation matrix $\underline{\omega}$, derived from a neutron depolarization experiment, and various parameters describing the micromagnetic state of a medium have been derived. The latter involve the individual particles, magnetic particle correlations as a consequence of particle interactions, particle orientational correlations, and density variations within the medium, and a varying neutron transmission length. The several contributions to $\underline{\omega}$ may (partially) be disentangled by the dependence of $\underline{\omega}$ on the reduced remanent magnetization m and on the transmission angle through the medium. The contributions of the individual particles to $\underline{\omega}$ is hardly dependent on m. Particle interactions do not affect $\underline{\omega}$ in the state of maximum remanence, while the contribution to $\underline{\omega}$ of a varying neutron transmission length equals zero at $m=0$. Using the relations derived, the neutron depolarization technique can successfully be used to yield detailed quantitative information about the micromagnetic state of particulate media. One-dimensional correlations can only substantially affect $\underline{\omega}$ if they occur along directions closely oriented along $\mathbf{e}_0$ and provided that the spread in particle size and orientation is relatively small.

[1]G. M. Drabkin, E. I. Zabidarov, Ya. A. Kasman, and A. I. Okorokov, [Sov. Phys.—JETP **29**, 261 (1969)].

[2]M. Th. Rekveldt, J. Phys. C1 **32**, C1–579 (1971).

[3]A. I. Okorokov, V. V. Runov, V. I. Volkov, and A. G. Gukasov, Zh. Eksp. Teor. Fiz. **69**, 590 (1975) [Sov. Phys.—JETP **42**, 300 (1975)].

[4]G. M. Drabkin, E. I. Zabidarov, and A. V. Kovalev, Zh. Eskp. Teor. Fiz. **69**, 1804 (1975) [Sov. Phys.—JETP **42**, 916 (1975)].

[5]M. Th. Rekveldt, J. Phys. C1 **38**, C1–23 (1977).

[6]M. Th. Rekveldt, F. J. van Schaik, and W. H. Kraan, Nukleonika **24**, 809 (1979).

[7]N. Stuesser, M. Th. Rekveldt, and T. Spruyt, Phys. Rev. B **33**, 6423 (1980).

[8]R. Rosman, M. Th. Rekveldt, and H. A. J. Cramer, J. Phys. **C8**, 1983 (1988).

[9]L. F. G. Thurlings, IEEE Trans. Magn. **MAG-19**, 84 (1983).

[10]M. Fearon, R. W. Chantrell, A. Lyberatos, and E. P. Wohlfarth, IEEE Trans. Magn. **MAG-23**, 174 (1987).

[11]S. V. Maleev and V. A. Ruban, Zh. Eksp. Teor. Fiz. **62**, 416 (1972) [Sov. Phys.—JETP **35**, 222 (1972)].

[12]S. V. Maleev and V. A. Ruban, Fiz. Tver. Tela (Leningrad) **18**, 2283 (1976) [Sov. Phys. Solid State **18**, 1331 (1976)].

[13]M. Th. Rekveldt, Z. Phys. **259**, 391 (1973).

[14]M. Th. Rekveldt, J. Magn. Magn. Mater. **1**, 342 (1976).

[15]R. Rosman and M. Th. Rekveldt, Z. Phys. B **79**, 61 (1990).

[16]R. Rosman, H. Frederikze, and M. Th. Rekveldt, Z. Phys. B **81**, 149 (1990).

[17]R. Rosman and M. Th. Rekveldt, J. Magn. Magn. Mater. (to be published).

[18]M. Th. Rekveldt, Textures Microstructures **11**, 127 (1989).

[19]H. N. Bertram and A. K. Bhatia, IEEE Trans. Magn. **MAG-9**, 127 (1973).

[20]A. Lyberatos, E. P. Wohlfarth, and R. W. Chantrell, IEEE Trans. Magn. **MAG-21**, 1277 (1985).

[21]R. Rosman, M. D. Clarke, M. Th. Rekveldt, R. P. Bissel, and R. W. Chantrell, IEEE Trans Magn. **MAG-26** (to be published).

[22]R. Rosman and M. Th. Rekveldt, J. Appl. Phys. (to be published).

[23]R. Rosman and M. Th. Rekveldt, J. Appl. Phys. (to be published).

[24]H. A. J. Cramer (private communication).