Measuring dc susceptibility of particulate media

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Abstract

The initial dc susceptibility χ of an array of single domain uniaxial particles is calculated as a function of temperature, sample texture, distribution of activation volumes and measurement duration. Interactions are modeled by the mean field approximation within which a self-consistent expression for χ is derived.

The response of a thermally relaxing system to outside stimuli is delayed since thermal relaxation takes place on a finite time scale. We consider here a uniaxial, single domain ferromagnetic particle with (activation) volume V , anisotropy constant K and saturation magnetization M_s , an external field is applied in the z -direction with which the particle's easy axis spans an angle β . The energy of the system is then $E/KV = \sin^2(\theta - \beta) - 2h \cos \theta$, where $h = H/H_n$, $H_n = 2K/M_s$, is the reduced applied field and θ is the angle spanned by the magnetization vector $\mathbf{M}$ and the z -axis. At sufficiently small fields the energy function has two minima at θ_1 and θ_2 . We shall treat an array of identical noninteracting particles as having two levels [1] (low temperature assumption, $q \gg 1$, see below) so that its reduced magnetization $m = M_z/M_s = n_1 \cos \theta_1 + n_2 \cos \theta_2$ where n_i is the probability that the i -th local minimum is occupied. The probabilities n_i satisfy the obvious relation $n_1 + n_2 = 1$ and the master equation [1]

$$\dot{n}_1 = -\kappa_{12}n_1 + \kappa_{21}n_2 = -\dot{n}_2, \quad (1)$$

$\dot{n}_i = dn_i/dt$, where $\kappa_{ij}(h) = \kappa_{i \rightarrow j} = f_0 \exp[-Q_i(h)/k_B T]$ is the transition rate of the process $i \rightarrow j$, the prefactor $f_0 \approx e^{25}$ Hz [1–3], Q_i is the barrier height to be overcome on leaving the i -th well and T is temperature.

Let us now assume that the system initially was in a demagnetized state in zero field with $n_1 = n_2 = \frac{1}{2}$ (thermal equilibrium) and that an infinitesimal field $h > 0$ is applied in the z -direction at time $t = 0$ and maintained. Substituting the expansion $\theta_i \rightarrow \theta_i + h\delta\theta_i$ into the extremal equation $dE/d\theta = 0$ we find the perturbed positions of the barrier and local minima. The expansions $n_i \rightarrow n_i + h\delta n_i$

and $\kappa_{ij}(h) \approx \kappa_{ij}(0) + h\kappa'_{ij}(0)$ in Eq. (1) yield, in the first order of h , a differential equation for δn_i with the initial condition $\delta n_i(0) = 0$. The perturbed reduced magnetization becomes $m \rightarrow m + h\delta m$ and, by definition, δm is the reduced initial dc susceptibility $\chi_\beta(T, t)$ which finally takes on the form

$$\chi_\beta(T, t) = \sin^2\beta + 2q(1 - e^{-2\kappa'}) \cos^2\beta, \quad (2)$$

where $q = KV/k_B T$ is a reduced barrier height and $\kappa = f_0 e^{-q}$ is the relaxation rate in zero field. The time independent term corresponds to a shift in the positions of the local minima and the remaining term describes the gradual relaxation towards the new equilibrium state at field h . If $\kappa t \gg 1$, Eq. (2) yields the usual equilibrium susceptibility whereas at $\kappa t \ll 1$ the system cannot relax during the measurement period and Fig. 1 shows the function $\chi_\beta(T)|_{t=\text{const}}$ with a sharp peak at the blocking temperature [2] where $\kappa(T)t \approx 1$. However, when averaged [3] over V Eq. (2) yields a very broad peak for which the concept of blocking temperature is no longer well defined. The activation volume is usually [2] assumed to have a log-normal distribution $P(V) \sim V^{-1} \exp[-\ln^2(V/V_0)/(2\rho^2)]$, the parameter ρ expresses the distribution width. The susceptibility of a textured [4] array is obtained by averaging Eq. (2) with respect to the inclination angle β .

Interparticle interactions may be included within the mean field approximation [5] in which the applied field h is replaced by the effective field $h_{\text{eff}} = h + \alpha \langle m \rangle$ where $\langle m \rangle$ is the instantaneous net magnetization of the array and α is a small interaction constant. For simplicity we shall assume that the texture of the array has axial symmetry so that the vector $\langle m \rangle$ is in the z -direction. Eq. (1) yields

$$\delta \dot{n}_{1\beta} = -2\kappa \delta n_{1\beta} + 2q\kappa(1 + \langle \delta m \rangle) \cos \beta, \quad (3)$$

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Fig. 1. The reduced susceptibility (2) plotted versus the reduced inverse temperature $q = KV/k_B T$ (solid unlabeled line). Also shown is the mean field susceptibility (6) with $\alpha = 4 \times 10^{-3}$ (short dash) and $\alpha = -5 \times 10^{-2}$ (long dash) and the susceptibility (2) averaged over activation volumes with log-normal distribution and $\rho = 0.24$ (solid labeled line). For this curve $q = KV_0/k_B T$. In all cases the inclination angle $\beta = 0$ and the waiting time $t = 10$ s. At larger waiting times the peaks shift towards lower temperatures.

for every β and the unknown function $\langle \delta m(t) \rangle$ satisfies the relation

$$\bar{s} + 2\langle \delta n_{1\beta} \cos \beta \rangle = \langle \delta m \rangle (1 - \alpha \bar{s}), \quad (4)$$

obtained by perturbing the mean magnetization $\langle m \rangle$, $\bar{s} = \langle \sin^2 \beta \rangle$ and $\bar{c} = \langle \cos^2 \beta \rangle$ for brevity. Substituting the solution of Eq. (3), with $\delta_{1\beta}(0) = 0$, into Eq. (4) results in a Volterra integral equation

$$\begin{aligned} \bar{s} + 4q\kappa\bar{c} \int_0^t dt' e^{-2\kappa(t-t')} [1 + \alpha \langle \delta m(t') \rangle] \\ = \langle \delta m(t) \rangle (1 - \alpha \bar{s}), \end{aligned} \quad (5)$$

whose solution $\chi(T, t) = \langle \delta m(t) \rangle$ is

$$\chi(T, t) = \sigma^{-1} \bar{s} e^{-2\Gamma t} + \xi (1 - \alpha \xi)^{-1} (1 - e^{-2\Gamma t}), \quad (6)$$

$\sigma = 1 - \alpha \bar{s}$, $\xi = \bar{s} + 2q\bar{c}$ and the new relaxation rate $\Gamma = \kappa \sigma^{-1} (1 - \alpha \xi)$. Interactions alter the minima positions, the relaxation rate and the asymptotic ($t \rightarrow \infty$) susceptibility. Ferromagnetic coupling, $\alpha > 0$, slows down the relaxation rate and increases the slope of the initial dc susceptibility while antiferromagnetic coupling has the opposite effect (see Fig. 1). Textured arrays yield a variety of similar curves.

So far the perturbing field was turned on instantaneously, a more detailed analysis follows: Let $h(t) = ht/t_1$ on the time interval $0 \leq t \leq t_1$. Eq. (1) has, in this case, the solution

$$\delta n_{1\beta}(t_1) = q[1 - f(t_1)] \cos \beta$$

with $f(t) = (1 - e^{-2\kappa t})/(2\kappa t)$. At time t_1 the field reaches its final value at which it is maintained for time t_2 . The net response of the noninteracting array is then

$$\chi_\beta(T, t_1 + t_2) = \sin^2 \beta + 2q[1 - e^{-2\kappa t_2} f(t_1)] \cos^2 \beta. \quad (7)$$

The formulae (2), (6) and (7) show that the initial susceptibility of a particulate system depends on the distribution of activation volumes, on the interaction strength, on the texture of the system and on measurement duration.

References

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