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Nonmonotonic dependence of magnetic viscosity on thermal relaxation rate

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Magnetic viscosity is studied as a function of the conditions under which the measured sample is prepared and it is shown that rapidly relaxing components cannot contribute to the observed relaxation process. The resultant magnetic viscosity then does not monotonically increase with increasing thermal relaxation rate but exhibits a local maximum associated with the transition between the fast and slow relaxation regimes. Related transitional phenomena and experimental results are discussed.

In a recent paper Ibrahim *et al.*[1] reported on observations of magnetic viscosity in nanoscale FeOOH particles. The authors measured the magnetic viscosity $S(T)$ as function of temperature and found it to exhibit a prominent local maximum in the vicinity of the blocking temperature of the sample. By contrast, theoretical studies predict monotonous temperature dependence,

$$S(T) \propto T^{\gamma}, \tag{1}$$

where for the exponent one usually[2] takes the value $\gamma = 1$ though also the value $\gamma \gtrsim 1/2$ has been reported.[3] Ibrahim *et al.*[1] thus state that their data cannot be interpreted within the present theory of magnetic viscosity.

We show here that relation (1) is associated with a special and, in general, rather unrealistic set of initial conditions imposed on the measured sample and that deviations from these initial conditions lead quite naturally to nonmonotonic temperature dependence of $S(T)$. As an example we trace the experimental procedure of Ibrahim *et al.*[1] and in a simple model calculation we reproduce (qualitatively) their experimental results. We find that the occurrence of a local maximum in the function $S(T)$ heralds a transition between two relaxation regimes and compare this effect with previously studied transitional behavior of magnetic switching field distribution[4,5] and initial dc susceptibility.[6] The work is concluded by a brief discussion of the dependence of magnetic viscosity on applied external field where analogous behavior is well known.[7]

For simplicity we assume an ensemble of identical, noninteracting uniaxial particles with energies $E = KV(1 - \alpha_3^2) - HM_sV\alpha_3$. Here K is the anisotropy constant, V the activation volume, M_s the saturation magnetization, H is an external field applied in the z direction parallel to the particle's easy axis, and α_3 is the direction cosine of the magnetization vector. The nucleation field of the particle is $H_n = 2K/M_s$ and we introduce the reduced field $h = H/H_n$. At sufficiently low temperatures, in fields $|h| < 1$, the particle has essentially only two states (levels) with magnetization parallel, respectively, antiparallel, to the applied field. The probabilities n_1 and $n_2 = 1 - n_1$ of finding the particle in one of the two available states is given by the master equation[8]

$$dn_1/dt = -\kappa_{12}n_1 + \kappa_{21}n_2 = -\Gamma n_1 + \kappa_{21}, \tag{2}$$

where $\Gamma = \kappa_{12} + \kappa_{21}$, the rates of thermally activated transitions between the two levels are $\kappa_{ij} = f_0 \exp[-Q(1 \pm h)^2]$ and $Q = KV/T$ ($k_B = 1$). For the prefactor we choose the value[3,9] $f_0 = e^{25}$ Hz. The mean magnetization of the ensemble is $M(t) = M_s[n_1(t) - n_2(t)]$.

In order to model[2,3] a real system we shall now assume that the activation volume V has a log-normal distribution within the ensemble:

$$P(V) = \mathcal{N} V^{-1} \exp[-(2\sigma^2)^{-1} \ln^2(V/V_0)], \tag{3}$$

where $\mathcal{N}$ is a normalization constant. The most probable activation volume $V_p = e^{-\sigma^2}V_0$ while the average activation volume $\langle V \rangle = e^{\sigma^2/2}V_0$. For simplicity we neglect all variations of the nucleation field[3] and assume that all particles contribute the same magnetic moment, regardless of their activation volume.[10] At constant T and h the mean reduced magnetization of the ensemble is obtained by averaging the solution of Eq. (2) over the distribution (3):

$$\langle M(t) \rangle = \left\langle \Delta_M(V) e^{-\Gamma(V)t} \right\rangle + \langle M_{\rm eq}(V) \rangle , \tag{4}$$

where $\Delta_M(V) = M_{\rm in}(V) - M_{\rm eq}(V)$, the equilibrium magnetization $M_{\rm eq}(V) \approx M_s \tanh 2Qh$ and $M_{\rm in}(V)$ is the initial mean magnetization of a particle with activation volume V. The magnetic viscosity S is then defined[3] as the peak value of the function

$$\tilde{S}(t) = -\frac{\partial \langle M(t) \rangle}{\partial \ln(t/t_0)} = \left\langle \Delta_M(V)\Gamma(V)te^{-\Gamma(V)t} \right\rangle \tag{5}$$

(t_0 is an arbitrary time scale), i.e., by the relation $S = \max_t \tilde{S}(t)$.

The integral (5) is usually studied[2,3,10] for $\Delta_M \equiv M_s$ (a saturated array in zero field) or for $\Delta_M \equiv 2M_s$ (a

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saturated array in a reversing field) and in this case $S = \Delta_M \max_t \langle \Gamma t e^{-\Gamma t} \rangle \propto T^{\gamma}$ in accordance with Eq. (1). In general, however, it is certainly not true that all particles are in their saturated state at the initial instant of measurement. The variation of Δ_M with V then depends on the sample preparation (history) and this dependence modifies the temperature properties of the magnetic viscosity $S(T)$. In particular, rapidly switching components of the ensemble may reach thermal equilibrium *before* the magnetic viscosity measurement is undertaken and these components therefore do not contribute to the observed relaxation process, introducing, in effect, a cutoff into the function Δ_M.

As an example we review here the experimental procedure of Ibrahim *et al.*:[1] The sample is cooled in applied field $h_a < 1$ until the measurement temperature is reached, then the field is reversed ($h_a \to -h_a$) and a measurement of magnetic viscosity is carried out under the constant reversing field $-h_a$. We shall assume that the applied field reverses as $h(t) = h_a \cos 2\pi f t$ so that the finite reversal duration is $t_d = 1/(2f)$. The nonequilibrium magnetization corresponding to the minor hysteresis loop driven by this field is obtained by numerical integration[11] of Eq. (2). For definiteness we also assume that the cooling sample remains in thermal equilibrium at the constant applied field h_a and this assumption yields the initial condition for the hysteresis loop calculation. The resultant final magnetization at time $t = t_d$ (in reversing field $-h_a$) represents the initial magnetization $M_{\rm in}(V)$ for the subsequent magnetic viscosity measurement. Obviously, a slowly relaxing particle with large V cannot relax appreciably during the field reversal and $\Delta_M \lesssim 2M_s$, while for a rapidly relaxing particle one has $\Delta_M \approx 0$ which is the above-mentioned cutoff. Sample plots of the function $\Delta_M(T/KV, f)$ [obtained by integrating Eq. (2)] are shown in Fig. 1 for three values of the sweep rate f. Note that only very small particles can reverse during a fast sweep and $\Delta_M \approx 2M_s$ over a larger temperature interval. The average in Eq. (5) is conveniently carried out using 16-point Gaussian quadratures[12] whence the magnetic viscosity $S(T) = \max_t \tilde{S}(t,T)$ follows (see Fig. 1). The small V (large T) cutoff in $\Delta_M(T/KV)$ now gives rise to the prominent peak associated with a transition from the low temperature region of slow reversals to the high temperature region of fast reversals, i.e., with the blocking temperature (at given sweep rate) as reported by Ibrahim *et al.*[1]

Analogous nonmonotonic dependence of magnetic viscosity on temperature is to be expected whenever the function $\Delta_M(T/KV)$ appreciably decreases over the

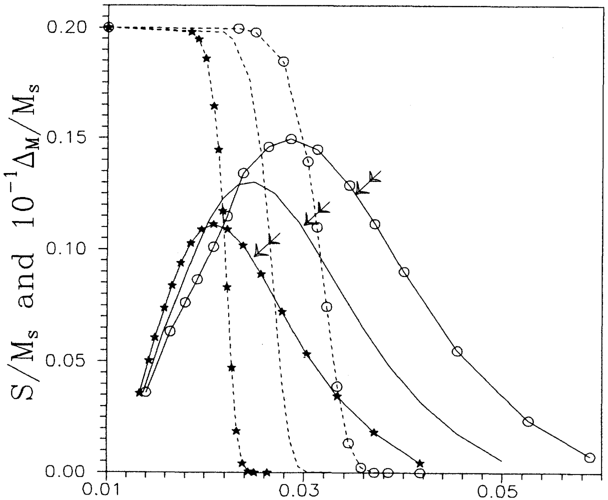

FIG. 1. The single-particle function $\Delta_M(T/KV)$ (dashed line, $V \equiv V_p$) and the magnetic viscosity $S(t)$ (solid line) vs scaled temperature. Sweep rate $f = 1$ Hz ($\star$), 50 Hz (no marks), and 10^3 Hz (○). The amplitude $h_a = 0.3$ and $\sigma = 0.28$. The arrows mark values at the (f-dependent) temperature for which $\langle M_{\rm in}(V) \rangle = 0$.

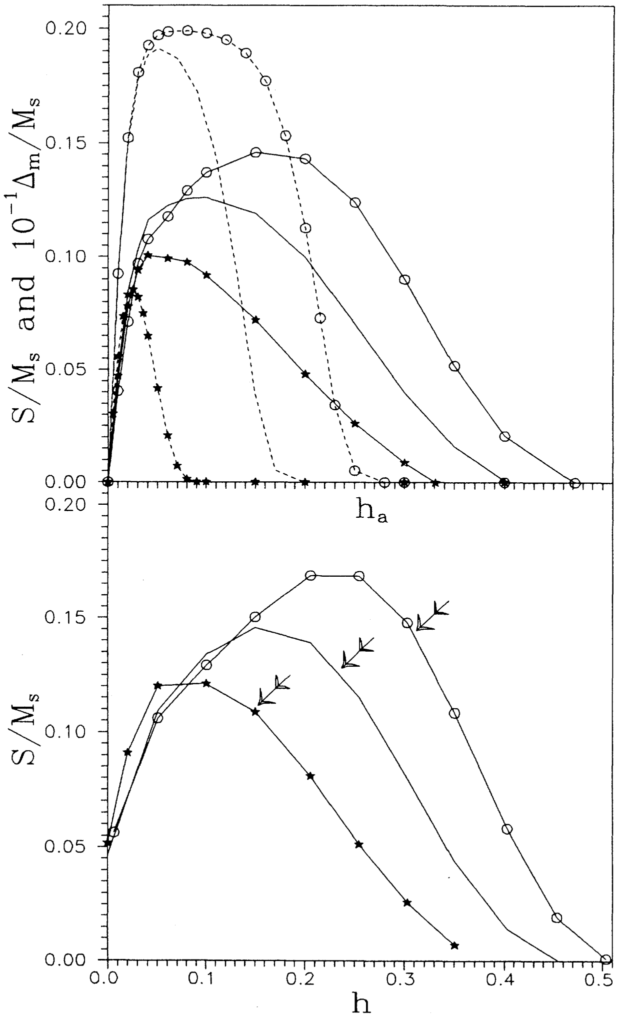

FIG. 2. Top: The single-particle function $\Delta_M(h_a)$ (dashed line, $KV/T = 25$) and magnetic viscosity $S(T)$ (solid lines, $KV_p/T = 25$) vs field amplitude. The model experimental conditions are described in text. Magnetic viscosity $S(T)$ along a major hysteresis loop driven by field $h(t) = -\cos 2\pi f t$. The arrows mark values at the (f-dependent) coercive fields. Markings as in Fig. 1.

measured temperature interval and our results remain essentially unchanged if we assume[10] that the magnetic moment of the particle is proportional to the activation volume V, i.e., if in place of Eq. (5) we write

$$\tilde{S}(t) = \langle V\rangle^{-1} \left\langle V\Delta_M(V)\Gamma(V)te^{-\Gamma(V)t}\right\rangle , \qquad (6)$$

though the local peak of $S(T)$ becomes in this case somewhat sharper. In either case, however, the peak in the transition region is smooth and we find no reason to introduce a singular critical point into the function $S(T)$ as proposed by Ibrahim *et al.*[1]

The presence of a local maximum in the function $S(T)$ is by no means the only exceptional phenomenon accompanying a transition between two relaxation regimes; indeed, such effects seem to be the rule rather than an exception: The peak value of the switching field distribution (SFD) measured along a major hysteresis loop varies[4] at low temperatures as $T^{-1/2}$ and at high temperatures as T^{-1}. In the transition region (around the f-dependent blocking temperature) the SFD peak value exhibits a local minimum and a local maximum.[4]

Another transition effect was recently predicted by Klik and Chang[5] who studied thermal relaxation in weakly coupled uniaxial particles using a multidimensional master equation formalism. The particles were deposited on a regular planar lattice and Klik and Chang found that stretching of the lattice may change the effective number of levels (states) contributing to the relaxation process and that this change is again accompanied by a local maximum and minimum in the SFD peak value. The transition between the two relaxation regimes distinguished by the number of contributing levels thus has a similar effect on the SFD function as a transition from low to high temperature magnetization reversals; however, the former transition was also found[6] to be accompanied by significant enhancement of the initial dc susceptibility.

We wish to conclude this paper by citing a transition phenomenon associated with variations of the applied field. To this end we consider the following (model) experimental conditions: Let a sample be initially in thermal equilibrium in applied field $h_a < 1$. The field is reversed and magnetic viscosity is measured in the field $-h_a$ as before, this time, however, at constant temperature as a function of h_a. In our model calculation we assumed that the sweep rate is constant, independent of the amplitude h_a, and in Fig. 2 we plot the function $\Delta_M(h_a)$ which vanishes at $h_a = 0$ and also at sufficiently large reversing fields where the particle certainly switches during the field sweep. The magnetic viscosity $S(h_a)$, computed from the function $\tilde{S}(t)$ of Eq. (5), has then a local maximum in the transition region between slow (small reversing field but large Δ_M) and fast (large reversing field but $\Delta_M \to 0$) relaxation regimes. The function $S(h_a)$ shown in Fig. 2 is, not surprisingly, similar to magnetic viscosity measured along the major hysteresis loop[7] which has a local maximum near coercivity (see Fig. 2) though in this case $\Delta_M(h)$ does not vanish at zero field.

In summary one may say that the nonmonotonic behavior of magnetic viscosity is due to the presence of two competing processes: The function $\Gamma te^{-\Gamma t}$ yields a contribution which monotonically increases with increasing decay rate Γ. On the contrary, the function Δ_M simultaneously decreases since all rapidly relaxing components reached thermal equilibrium during the preparation of the sample, i.e., before the actual measurement commenced.

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[12] This corresponds to 32 sampling values of the activation volume V, see, e.g., J. Pachner, *Handbook of Numerical Analysis Applications* (McGraw-Hill, New York, 1984), Sec. 3.3. In Eq. (3) we exclude extremes of volume by constraining V/V_0 to the interval $(0.6, 2)$.