Dynamic susceptibility of concentrated magnetic liquids

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A possible reason for the logarithmic dispersion law of the susceptibility which has been observed in magnetic colloids is the appearance of a dipole-spin-glass state in these systems.

Experiments indicating a similarity between magnetic spin glasses and magnetic liquids—concentrated suspensions of one-domain particles of ferro- or ferrimagnets—were first reported[1] in 1980. Tari *et al.*[1] observed a characteristic peak in the temperature dependence of the susceptibility, $\chi(T)$, of suspensions of ultrasmall ($\sim$100-Å) particles of magnetite—the most common material for magnetic liquids. In the present letter we report measurements of the dynamic properties of magnetite magnetic liquids which yield further evidence for the spin-glass model. We discuss the content of this model for these particular systems.

At first glance, the fundamental difference between ordinary spin glasses and magnetic liquids would seem to be that the spatial structure of the latter is not fixed: The magnetic grains of magnetic liquids have mechanical (translational and rotational) degrees of freedom in the liquid matrix. The existence of this microscopic mobility must, of course, be taken into account in a determination of the response of a system to an alternating external field. In the approximation of independent grains (low concentrations), calculations[2] yield a Debye length $\chi' \sim (1+\omega^2\tau_B^2)^{-1}$, for magnetic liquids, where $\tau_B = 3V\eta/k_BT$ is the Brownian time for the rotational diffusion of a spherical particle of volume V in a liquid with a dynamic viscosity η. Typical values of τ_B for $V \sim 10^{-18}$ cm^3 would be about 10^{-6} s. Martsenyuk *et al.*[2] have suggested that a one-domain particle has a magnetic moment $\mu = M_sV$, where M_s is the saturation magnetization of the material. For highly dilute magnetic liquids, their model,[2] corrected for

the Néel superparamagnetism, predicts results in satisfactory agreement with experiment.[3]

The situation changes fundamentally when we look at concentrated systems, where the fraction of the solid phase reaches 10–20% by volume. In such magnetic liquids, as many observations have shown (see, e.g., the review by Krueger[4]), the magnetic dipole-dipole interaction forces the particles to combine into aggregates, which are quasispherical in the absence of an external field. In an aggregate containing a sufficiently large number of particles (ten or more), it would be difficult to suggest the appearance of an uncompensated magnetic moment: Ferromagnetic clusters are unfavorable from the energy standpoint. If, on the other hand, we take into account both the finite size of the aggregate and the polydisperse nature of the grains of a real magnetic liquid, then we should acknowledge that the formation of a regular structure with an antiferromagnetic order would be improbable. The most likely situation appears to be a packing of the aggregate in a spin-glass fashion, with the directions of the magnetic moments of the individual regions uncorrelated, and with the sum of these moments over an aggregate vanishing. In this case an alternating magnetic field cannot cause a rotation of a cluster of grains (an aggregate), since the magnetization of the cluster is always parallel to the external field, and the torque is strictly zero. In other words, the formation of clusters "turns off" the mechanical degrees of freedom of a magnetic liquid in an alternating field, explaining the close relationship between such a system and a solid spin glass.

To analyze the dynamics of the magnetization of a concentrated magnetic liquid, we use the simple cluster model of a spin glass,[5,6] which treats the spin glass as a set of noninteracting regions characterized by a superparamagnetic behavior. According to this model, the magnetization of the system occurs through a surmounting of energy barriers $\sim Kv$, where v is the volume of the cluster whose magnetization is undergoing reversal, and K is an effective anisotropy constant. In our case, K is of a magnetostatic nature, so that we have $K \sim M_S^2$. The freezing temperature is thus given in order of magnitude by $T_f \sim M_s^2 V/k_B \lesssim 10^3$ K and is determined by the average of the blocking temperatures for the orientational fluctuations of the magnetic moments of the individual clusters. The formation of a spin-glass phase always leads to a broad distribution of v. The nature of the distribution, $f(v)$, is determined[6] by comparison with experiment.

For the present experiments we used samples of magnetic liquids (magnetite colloids in kerosene) with an average grain size ~ 100 Å and a saturation magnetization $M_0 = 54$ or 79.5 G. The dynamic susceptibility was measured at room temperature over the range from 20 Hz to 30 kHz by a mutual-inductance bridge method, and it was measured over the range from 20 kHz to 1 MHz by a resonance method. We found that these two liquids exhibit a similar behavior in terms of the frequency dependence of the real (χ') and imaginary (χ'') components of the susceptibility. The data from the measurements of sample No. 2 are shown in Fig. 1. Here we can clearly see the quasilinear decrease in χ' with the logarithm of the frequency at $T \lesssim T_f$, which is the typical behavior of the spin glasses CuMn, etc.[7]

Can we interpret these results in the cluster model? To estimate the time (τ)

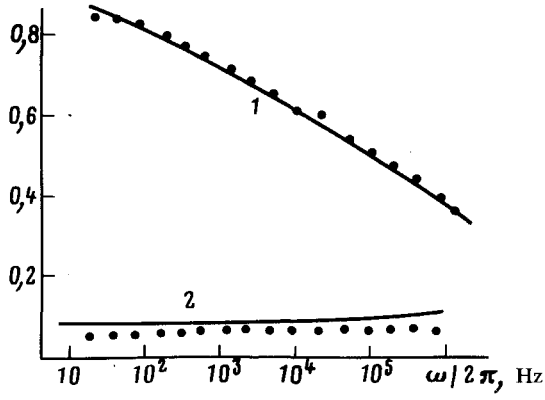

FIG. 1. Frequency dependence of the magnetic-susceptibility components of a magnetic liquid. The points are experimental, and the lines are calculated from Eqs. (4). 1—χ'; 2—χ''.

required for the magnetic moment to cross the potential barrier Kv, we use the Néel formula

$$\tau = \tau_0 \exp(Kv/k_B T), \tag{1}$$

where τ_0 is a nucleation constant, $\sim 10^{-9}$ s. The static (equilibrium) susceptibility of a cluster of volume v is $\chi_0 = M_s^2 v/3k_B T$, so that in a system of clusters with a distribution of v values we find the susceptibility components to be

$$\chi' = \frac{NM_s^2}{3k_B T} \int_0^\infty \frac{v^2 f(v)dv}{1+\omega^2\tau^2}, \qquad \chi'' = \frac{NM_s^2}{3k_B T} \int_0^\infty \frac{\omega\tau v^2 f(v)dv}{1+\omega^2\tau^2}; \tag{2}$$

here N is the total number of clusters. As $f(v)$ we use an inverted gamma distribution,

$$f(v) = \frac{V_0^{1+\gamma}}{\Gamma(1+\gamma)v^{2+\gamma}} \exp(-V_0/v), \tag{3}$$

where V_0 and γ are parameters, $\Gamma(x)$ is the gamma function, and $f(v)$ has been normalized to one. The function $f(v)$ has a power-law asymptotic behavior at $v > V_0$ and is thus convenient for describing a broad distribution of v.

Over the frequency range studied ($\omega/2\pi \lesssim 1$ MHz), we can use the approximation $|\ln\omega\tau_0| \gg 1$. Substituting (1) and (3) into (2), and integrating, we thus find the following frequency dependence for the susceptibility components, with a logarithmic accuracy in $\omega\tau_0$:

$$\chi' = C(-\ln\omega\tau_0)^{1-\gamma}, \qquad \chi'' = 2C(1-\gamma)(-\ln\omega\tau_0)^{-\gamma}. \tag{4}$$

Comparison of (4) with the experimental curves yields $\gamma = 0.32$ and $\tau_0 = 2\times 10^{-9}$ s. The solid lines in Fig. 1 show results calculated for these parameter values. The coefficient C in (4) is a function of M_0, K, and V_0. Comparing its theoretical value with the value $C = 0.13$ found experimentally, we find, with $K \approx M_s^2$, the estimate $V_0 \approx 10V$. Consequently, the scale size of the region that undergoes magnetization reversal (the cluster) turns out to be an order of magnitude greater than the volume of an individual

grain in a magnetic liquid, in agreement with the suggestion of a cooperative nature of these phenomena.

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[1]A. Tari, J. Popplewell, and S. W. Charles, J. Magn. Magn. Mater. 15-18, 1125 (1980).
[2]M. A. Martsenyuk, Yu. L. Raĭkher, and M. I. Shliomis, Trudy Mezhdunarodnoĭ konferentsii po magnetizmu ICM-73 (Proceedings of the International Conference on Magnetism ICM-73), Vol. 3, Nauka, Moscow, 1984, p. 540.
[3]M. M. Maĭrov, Magnitnaya gidrodinamika (Magnetic Hydrodynamics), No. 2, 21 (1979).
[4]D. A. Krueger, IEEE Trans. Magn. **MAG-16**, 251 (1980).
[5]E. P. Wohlfarth, Phys. Lett. **70A**, 489 (1979).
[6]A. Aharoni, Phys. Lett. **99A**, 458 (1983).
[7]A. J. Van Duyneveldt and C. A. M. Mulder, Physica (Utrecht) **114B**, 82 (1982).

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