Nonthermal viscosity in magnets: Quantum tunneling of the magnetization (invited)

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In this article we present experimental results on the magnetic relaxation in different systems (single domain particles, magnetic grains, and random magnets). The existence of two relaxation regimes is demonstrated. At high temperatures, the magnetic viscosity $S \equiv 1/M_0 \partial M/\partial \ln(t)$ is proportional to temperature in accordance with theoretical expectation for thermally activated processes. At low temperatures, the viscosity is independent of temperature, providing evidence to quantum tunneling of magnetization. Qualitative agreement between theory and experiment is found.

I. INTRODUCTION

One of the most fascinating recent discoveries in condensed matter physics is the observation of quantum tunneling of magnetic flux in superconductors.[1] This observation has been based upon a theoretical prediction,[2] that in the limit of weak dissipation a macroscopic object can tunnel through a small energy barrier with a rate significant enough to be detected experimentally.

In a more recent time, it has been predicted that the macroscopic quantum tunneling can also be observed in magnetic systems. The tunneling variable is the magnetization $M(r)$. It has been shown theoretically that $M(r)$ can tunnel quantum mechanically between different configurations corresponding to metastable states.[3–7] Let such a process occur with a transition rate $\Gamma_Q = \nu \exp(-B)$, B being the WKB exponent at zero temperature and ν is the attempt frequency. As the temperature is growing, one should also be concerned with thermal transitions through energy barriers. The rate of thermal transitions is determined by the Boltzmann factor $\Gamma_T \sim \nu \exp(-U/k_B T)$, where U is the barrier height. The prefactor ν is usually of the order of the FMR frequency $\nu \sim 10^{10} - 10^{11}\ \mathrm{s}^{-1}$. One can define a crossover temperature $k_B T_c \sim U/B$ below which quantum underbarrier transitions dominate. In fact, quantum transitions will reveal themselves on the time scale of the experiment if the following condition is satisfied: $k_B T < U/30 \leqslant k_B T_c$.

It has been demonstrated that up to a numerical factor of the order of one, $k_B T_c$ for different tunneling mechanisms (uniform rotation of M in small particles,[3] quantum nucleation of magnetic bubbles,[4] tunneling of domain walls[6,7]) is always of the order of $\mu_B H_k$. It corresponds to the temperature range $T \sim 0.1$–10 K for typical values of the anisotropy field H_k.

There are two kinds of barriers in magnetic systems: intrinsic barriers due to magnetic anisotropy and barriers due to the pinning of domain walls by defects. Both are responsible for the pronounced metastability of magnetic materials. Anisotropy barriers are of the order of $H_k M_0 V$, while pinning barriers are of the order of $H_c M_0 V$, where H_k is the anisotropy field, M_0 is the magnetic moment of the unit volume, H_c is the coercive field, and V is the volume involved in the tunneling process. These will produce the WKB exponent $U/k_B T_c$,[3] of the order of $(H_\parallel / H_\perp)^{1/2} J$ for anisotropy barriers; $H_\parallel$ and $H_\perp$ being the anisotropy fields responsible for the equilibrium orientation of M and for the noncommutation of $M_\parallel$ with the Hamiltonian and $J \sim M_0 V/\mu_B$ being the total tunneling spin. In the case of pinning barriers, the WKB exponent is of the order of $(H_c/\sqrt{H_\parallel H_\perp})J$. To increase the total tunneling magnetic moment, $J \gg 30$, one may think to work with materials for which $H_\parallel \ll H_\perp$ (particles with strong easy plane anisotropy and relatively weak uniaxial anisotropy in the plane) or with materials with low coercitivity compared to the anisotropy fields (random magnets). The decrease of energy barriers due to the external applied field can also help to increase the total tunneling magnetic moment.

Consider a magnetic system with high metastability and a broad distribution of energy barriers. It may be a system of interacting single-domain particles, highly defected ferromagnetic crystal, random magnets, etc. As one applies the magnetic field, the magnetic moment of such a system typically has a two-step evolution. In the first, rapid stage, it changes due to the rotation of the local magnetization in areas where the barriers are removed by the field. As the magnetization of the system changes, so does the internal field inside the magnet. Eventually, the evolution of the system gets stuck in the state where the barriers just begin to develop. The later, slow phase of the evolution, is due to thermal or quantum transitions in the presence of the barriers. Thus starting from zero barriers, the relaxing system automatically arrives to the barrier heights for which the lifetime of the metastable states corresponds to the actual time scale of the experiment. For thermally activated processes, the rate of the slow relaxation discussed above must decrease as the temperature goes down. Independence of this rate on temperature, below some temperature, is the vestige of quantum tunneling.

As is well known in magnetism, that in the presence of a wide distribution of energy barriers, the relaxation of the magnetization has $\log(t)$ dependence on time. Each individual metastable state decays as $\exp(-\Gamma t)$, where Γ is the rate of the decay $\Gamma = 1/\tau$ being τ the lifetime. The escape rate Γ at any temperature can be written as

$$\Gamma = \nu(T) \exp[-U/k_B T^*(T)], \tag{1}$$

with $T^*(T) \simeq T$ at $T > T_c$ and $T^*(T) > T$ at $T < T_c$. A

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more detailed analysis shows that the dependence of T^* on T bring information on the relative contribution of the dissipation to the tunneling rate. In the absence of dissipation, the transition from thermal to quantum decay is rather sharp with a well-established plateau in the $T^*(T)$ dependence.[8,9] It can be shown, however,[10–12] that in the presence of the distribution of energy barriers their commutative effect transforms the exponential decay of the magnetization into $\log(t)$ dependence. This occurs if at any moment of observation t, there are barriers for which t is a typical lifetime. The coefficient in front of $\log(t)$ is the so-called magnetic viscosity S:

$$M(t)=M(t_0)[1-S(T)\ln(t/t_0)], \tag{2}$$

where the magnetic viscosity is given by[12]

$$S(T)=[k_B\langle T^*(T)\rangle]/\langle U\rangle. \tag{3}$$

$\langle T^*(T)\rangle$ means averaging over the barriers of a different shape and $\langle U\rangle$ is the average energy barrier, which determines the blocking temperature. $k_BT_B\sim\langle U\rangle/30$ in static $M(T,H)$ measurements. Equation (2) is valid if we assume that the relaxing part of the magnetic moment is small compared to the total moment. One should expect this condition to become invalid at large t, $t\sim t_0^{1/S(T)}$. Another important consequence of the theory is that T^* does not depend on the extensive parameters like, e.g., the tunneling volume. Even for a broad distribution of tunneling volumes, one should expect a relatively narrow distribution in $T^*(T)$ at a low temperature. This suggests that the magnetic viscosity $S(T)$ must have a universal behavior similar to that of the escape rate Γ. In other words, for thermally activated processes we define a thermal viscosity S_T, which is linear on T and is zero at $T=0$; below T_c we may define a quantum viscosity S_Q, which does not vanish at $T=0$ and has a well-defined plateau. Moreover, the constancy of S_Q does not depend on the quantum mechanism involved in the relaxation of the magnetization.

In this paper we report observation of such behavior in single domain particles, magnetic grains, and random magnets and we will attempt to make a link between theory and experiment.

Recently Awschalom *et al.*[13,14] reported some evidence of tunneling of magnetic moment in small ferromagnetic particles and evidence of tunneling of the Néel vector in small antiferromagnetic particles. Paulsen *et al.*[15] have also published relaxation results which suggest the occurrence of the tunneling of the domain wall in small ferromagnetic particles.[11]

II. EXPERIMENT

A. Samples

The materials studied are ferrofluids, magnetic grains, and amorphous rare-earth (RE)-transition metal (TM) alloys.

The three ferrofluid samples[16,17] used in these experiments contain very small particles of FeC, Fe_3O_4, and $CoFe_2O_4$, respectively. In order to avoid clustering, the particles were coated with a nonmagnetic surfactant layer.

TABLE I. Composition of samples, thickness, and other parameters.

Sample	Substrate	Buffer fundamental sequence (F,S)	Repetition of F,S	RE/TM
Fe/Sm	Kapton foil	100 Å Cu[Fe(3 Å)/Sm(2 Å)]$_{x6}$	15	Fe_4Sm
Fe/Tb	Kapton foil	100 Å Ag Fe(3 Å)/Tb(4 Å)]	50	Fe_3Tb
Co/Sm	Kapton foil	100 Å Cu[Co(4 Å)/Sm(3 Å)]$_{x5}$	11	Co_4Sm

The size distribution of the particles were studied by using electron microscopy. In the case of the FeC ferrofluid the mean particle diameter is of 36 Å and the average distance between the particles is of 65 Å. For the Fe_3O_4 and $CoFe_2O_4$ ferrofluids the mean particle diameter is of 60 Å.

The magnetic grains of Fe and Dy were obtained by depositing a very thin film of few angstroms thickness of the corresponding metal onto crystalline Cu(111).[18,19] In order to increase the magnetic signal, we formed a multilayer system by evaporating very thin metallic layers between Cu layers of 100 Å thickness. No peaks corresponding to crystalline Fe and Dy in the x-ray diffraction patterns have been observed.

Our amorphous TM-RE alloys[20,21] were prepared by using two electron beam evaporators in a high vacuum chamber. The pressure during the evaporation of metals was maintained at 5×10^{-7} Torr. The evaporation rate was 0.5 Å/s and a very thin kapton film was used as a substrate. The substrate was kept at 260 °C during the evaporation. The composition of the samples, the thickness and other parameters of the layers are summarized in Table I. For the two samples containing iron, we also recorded Mössbauer spectra between 4 K and room temperature. The Mössbauer data confirm the random magnetic properties of the films. In order to increase the magnetic signal, we used samples containing 40 films of each composition.

B. Equipment

Magnetic measurements were performed by using a SHE SQUID magnetometer. During the relaxation experiments, the data were corrected to take into account the spurious drift of the SQUID base by doing the following procedure: after resetting the SQUID sensor, we measure the magnetization as the induced current in the second derivative coil. If v_1, v_2, and v_3 are, respectively, the two minima and the maximum of the SQUID signal, the magnetization value is proportional to $(v_2-v_1)-(v_3-v_1)/2$. Long-time shifts are also avoided because the total magnetization is determined at each point. As the elapsed time from the beginning of the measurements increases, eight measurements are averaged to improve accuracy of the values. The applied magnetic field in the relaxation experiments were produced by using a power supply which has a record constancy in the generated current. The accuracy in the field measurement was better than 0.1 Oe. The applied field was calibrated by using a pure Pb sample in the

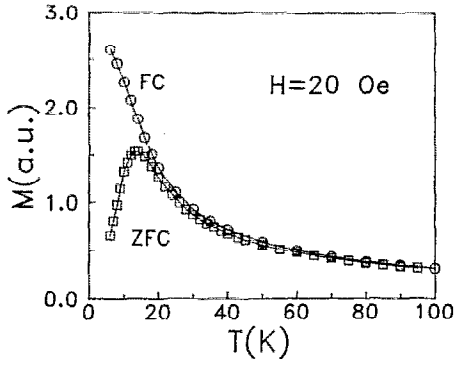


FIG. 1. ZFC and FC magnetization curves for the FeC ferrofluid sample with an applied field $H=20$ Oe.

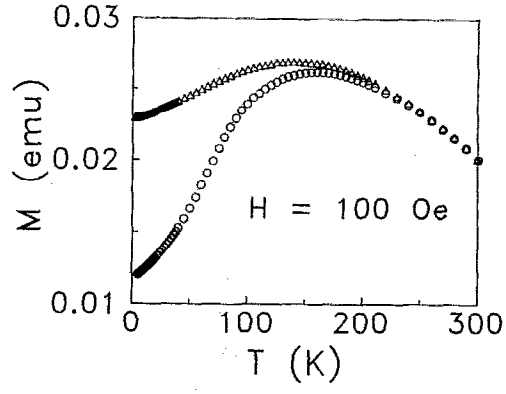


FIG. 2. ZFC and FC magnetization curves for the FeSm random magnet with an applied field $H=100$ Oe parallel to the film plane.

superconducting state. The constancy of the applied field was checked by measuring the signal from a pure paramagnetic sample during two hours at different temperatures.

We have studied the time dependence of the magnetization using two different criteria:

(i) The sample was cooled from room temperature to a well-defined final temperature in the presence of a magnetic field H_1. Then the field was rapidly changed from H_1 through zero to H_2. The first measurement of the magnetization was taken four seconds after the change of the field. The variation of the magnetization with time was then followed during the time period up to a few hours. The time interval between two consecutive $M(t)$ points was 5 s.

(ii) After saturation of the hysteresis loop, the field is reversed and stabilized at a given value H close to the coercive field H_c and relaxation measurements as a function of time are collected until the magnetization goes to zero.

III. RESULTS

A. Static measurements

1. Low field measurements

Zero field cooled (ZFC) and field cooled (FC) measurements of $M(T)$ at low field were performed to obtain information about the irreversibility and freezing of magnetic moments.

The remarkable feature of the ZFC curve for the ferrofluid and magnetic grain samples is the existence of a narrow peak in the $M(T)$ dependence. The two curves of $M_{FC}(T)$ and $M_{ZFC}(T)$ are the same for temperatures above the blocking temperature T_B, with the magnetization following a Curie–Weiss law with a very small Curie temperature being indicative of the weak magnetic coupling between the particles. We interpret the peak in the M_{ZFC} data as due to random orientations of the anisotropy directions below T_B. In Fig. 1 we show the ZFC and FC curves for the FeC ferrofluid sample with an applied field of 20 Oe.

In the case of random magnets, the most remarkable feature of the ZFC curve is the existence of a broad peak. The onset of the irreversibility starts before the cusp of the peak, indicating the existence of clusters of spins that are involved in the freezing phenomenon. This also indicates the presence of a broad distribution of relaxation times for the metastable states related to the orientation of the ferromagnetic clusters with respect to the applied field and the effective anisotropy axes. One cannot exclude also that defects and irregularities of the geometry of the film are responsible for some of the metastable states. Figure 2 shows the low field data for the Fe_4Sm sample.

2. High field measurements

Hysteresis measurements at different temperatures were performed for all the samples in order to get the variation with temperature of the coercive field H_c, the remanence magnetization M_r, the saturation magnetization M_s, and the anisotropy field H_k. In the case of the random magnets, these measurements were done for the applied field parallel and perpendicular to the film plane. The total magnetization of these samples has been found to lie in the plane.

The $M(H)$ curves for both magnetic grains and ferrofluid systems at $T > T_B$ are well fitted to Langevin functions and scale in H/T confirming the superparamagnetic behavior of these systems. For $T < T_B$, both the coercitivity and remanence increase as the temperature decreases, indicating that corresponding to our small magnetic particles and grains, there is a broad energy barrier distribution and magnetic relaxation is expected at low temperatures. The hysteresis loops on our FeC ferrofluid sample at temperatures down to 150 mK are shown in Fig. 3. Below 0.5 T the coercive field H_c is independent of temperature, suggesting the existence of irreversible changes without thermal activation.

For all three random magnet samples, the best fit to the $M(H)$ data at $T < T_B$ is provided by the $1/H$ law at low fields and by $1/H^2$ at high fields. Then according to the correlated spin-glass model[22,23] our RE-TM systems behave as two-dimensional amorphous ferromagnets: the atomic magnetic moments are ferromagnetically correlated on a small scale, while on a large scale the magnetization rotates stochastically over the sample. In Fig. 4 we show the $M(H)$ data for the SmCo samples and in Figs. 5(a)

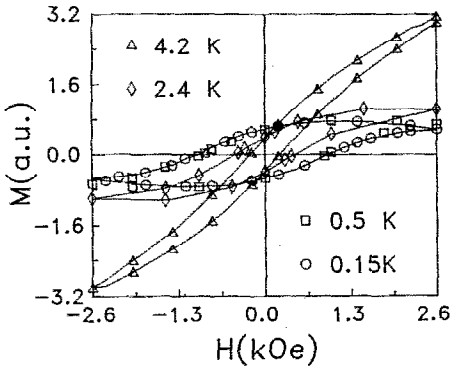


FIG. 3. Hysteresis loops on the FeC ferrofluid at temperatures down to 150 mK.

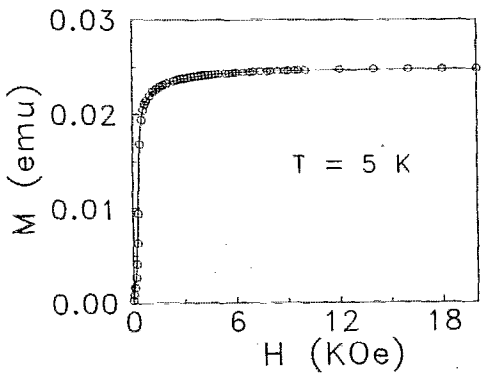
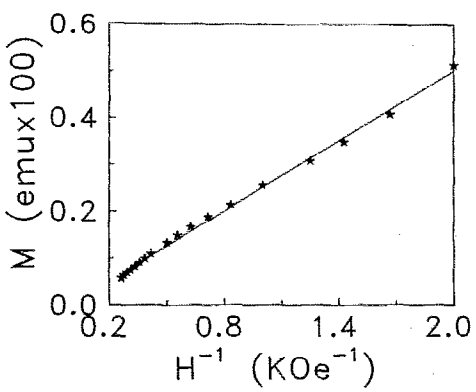


FIG. 4. $M(H)$ data for the Co_4Sm random magnet sample at $T=5$ K.



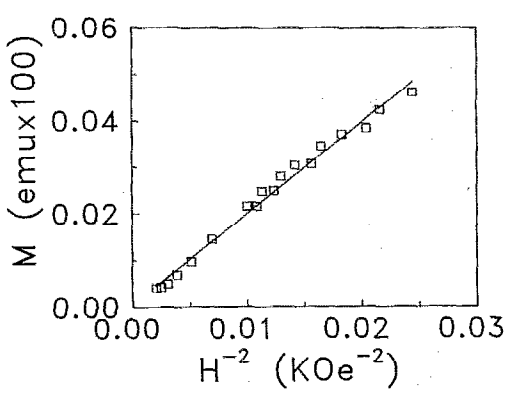


FIG. 5. Two regimes in approaching to saturation for the Co_4Sm sample: (a) $\Delta M \alpha 1/H$; (b) $\Delta M \alpha 1/H^2$.

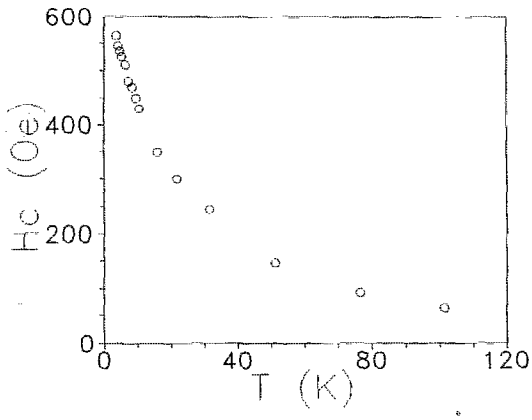


FIG. 6. Varitation with temperature of the coercive field for the Fe_4Sm sample.

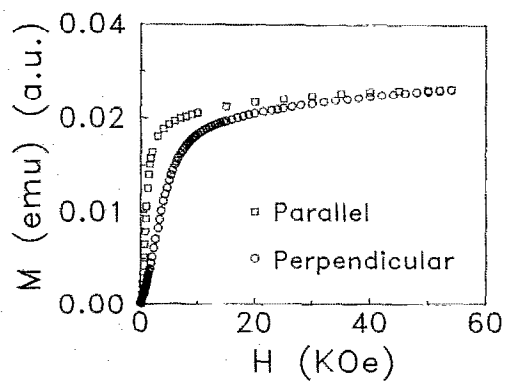


FIG. 7. $M(H)$ dependence at $T=5$ K for the Fe_3Tb sample with H applied parallel and perpendicular to the film plane.

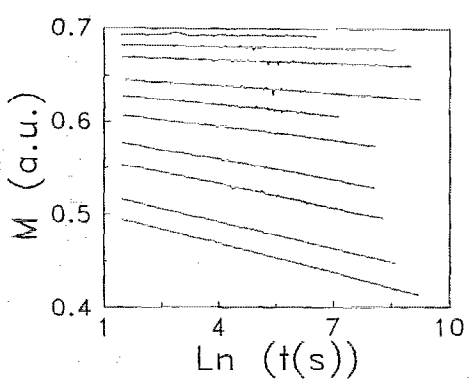


FIG. 8. Time dependence for the thermoremanence magnetization of the $CoFe_2O_4$ ferrofluid sample ($H_1=100$ Oe and $H_2=0$ Oe).

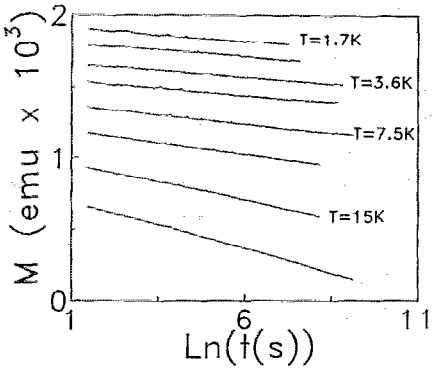


FIG. 9. Time dependence for the thermoremanence magnetization of the FeTb sample ($H_1 = 100$ Oe, $H_2 = -100$ Oe, both H_1 and H_2 are parallel to the film plane).

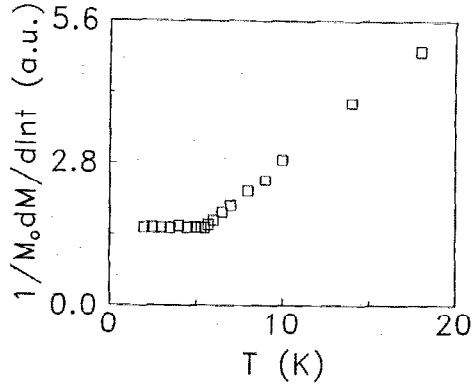


FIG. 11. $S(T)$ values for the FeTb sample. These data are partially given in Fig. 9.

and 5(b) we show the two regimes approaching saturation for the data of Fig. 4. Figure 6 shows the variation with temperature of the coercive field for the FeSm sample and Fig. 7 shows the $M(H)$ curves for the FeTb sample when the external field is applied parallel and perpendicular to the film plane.

B. Magnetic relaxation

Let us list the major features of the relaxation process observed experimentally:

(1) The relaxation of the thermoremanent magnetization follows the $\log(t)$ law (Figs. 8 and 9).
(2) A small part of the total magnetization is relaxing.
(3) The magnetic viscosity $S(T)$ depends linearly on T at high temperatures. This dependence extrapolates to zero at $T=0$ (Figs. 10 and 11).
(4) $S(T)$ is independent of T at low temperatures (Figs. 10 and 11).
(5) The transition from the thermal to the nonthermal relaxation regime is sharp (Fig. 11).
(6) The crossover temperature occurs at a few K and depends on the value of the applied magnetic field (Fig. 12).

IV. DISCUSSION

The observation of the logarithmic in time dependence for the thermoremanence magnetization is what one should expect by considering a broad distribution of energy barriers or the variation with time of the demagnetizing field during the relaxation process.

The fact that only a small part of the total magnetization is relaxing agrees well with the broad maximum observed in the ZFC curve suggesting the existence of metastable states separated from the absolute energy by barriers much lower than average. As follows from Eqs. (2) and (3), the relaxing fraction of the total magnetization, on the time scale of the experiment must be of the order T^*/T_B in agreement with the experimental data.

The proportionality of $S(T)$ to T at $T_c \ll T \ll T_B$ indicates a weak correlation between the particles or clusters. For the ferrofluid and magnetic grain systems it is in agreement with the low value for the Curie temperature deduced from the Curie–Weiss law fitting of the FC magnetization data above T_B. In the case of random magnets, this indicates that the interactions between clusters do not affect the relaxation inside each cluster. This is a reasonable consequence of the large magnetic anisotropy values for these systems. Since a small fraction of the total moment is relaxing, the clusters involved simultaneously in the relax-

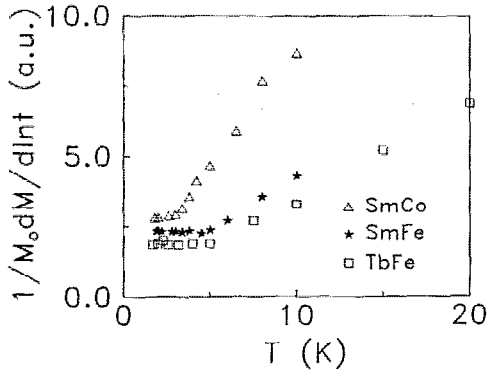


FIG. 10. Temperature dependence of the magnetic viscosity $S(T)$ for the three random magnet samples.

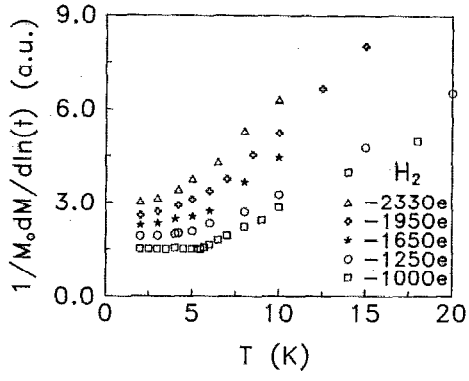


FIG. 12. Variation of the $S(T)$ and T_c with the value of the applied magnetic field H_2. In all cases $H_1 = 100$ Oe.

TABLE II. Experimental values of the anisotropy constants and magnetization.

Sample	$T(k)$	H_k (Oe)	K (erg/cm^3)	H_c (Oe)	M_s (emu/cm^3)
Fe_4Sm	5	2.6×10^4	9×10^6	650	6.9×10^2
Fe_3Tb	5	2.2×10^4	6×10^6	400	5.5×10^2
Co_4Sm	5	1.4×10^4	4×10^6	300	5.7×10^2
FeC (ferrofluid)	5	1.5×10^4	3×10^6	10^3	400
$CoFe_2O_4$ (ferrofluid)	5	3.0×10^4	6×10^6	10^4	425
Fe_3O_4 (ferrofluid)	5	5.0×10^3	10^6	130	480

ation process must be separated by a large distance which should additionally decrease their interactions.

With decreasing temperature, thermal effects become less important and we observe a plateau for the $S(T)$ values. This is exactly what one would expect in the presence of quantum tunneling of magnetization. Moreover, the constancy of the quantum viscosity S_Q does not depend on the quantum mechanism involved in the relaxation of the magnetization.

Another important observation concerns the sharpness of the transition from the thermal to the quantum regime. According to the theory it should be rather sharp in the case of weak dissipation.[24–27]

The theoretical prediction for the crossover temperature T_c for single domain particles with uniaxial anisotropy can be estimated as $k_B T_c \approx \mu_B H_k$. The values of H_k for the three ferrofluid samples are given in Table II. We have observed that the T_c(FeC ferrofluid) ≈ 1 K and T_c($CoFe_2O_4$ ferrofluid) ~ 2 K in agreement with the theoretical predictions. For random magnets $H_{\parallel} \sim H_{\perp} \sim H_k$, see Fig. 7, and the crossover temperature can be estimated using the quantum nucleation theory of Chudnovsky and Gunther[4] by substituting the experimental values of the anisotropy constants and magnetization (see Table II) in the theoretical expression for T_c. This gives T_c of about a few K for our three random magnets in agreement with the experimental observations.

As the theory suggest we have observed that the T_c values decrease when increases the magnetic field in the relaxation experiments as a consequence of the reduction of the barrier heights. We have also observed that the increasing values of the applied fields tend to smear the transition (Fig. 12). This may be due to have higher dissipation effects when barrier heights decrease. However, a convincing comparison between theory and experiments on dissipation cannot be made at this point.

V. CONCLUSIONS

As a conclusion in this paper we have shown experimental results on the relaxation of single domain particles and magnetic clusters which strongly support the existence of quantum, in nature, relaxation processes. Relaxation experiments provide an ideal situation for the observation of this effect. They automatically drive the system into a state with extremly low energy barriers for which nonthermal sub-barrier transitions of the magnetization from metastable states occurs.

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