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LETTER TO THE EDITOR

Quantum tunnelling effects in Fe/Sm multilayers

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Abstract. Magnetic relaxation investigations of Cu/Fe_4 Sm/Cu multilayers from 20 K to 1.6 K at low and high fields have been performed. The constancy of both the magnetic viscosity and the coercive field below 2.5 K suggests that this temperature corresponds to the crossover between thermal and quantum regimes.

In the classical model of both thermal activated magnetization reversal in singledomain particles and nucleation of ferromagnets, it is clear that the time dependence of magnetization or 'magnetic viscosity' disappears at the absolute zero of temperature. Recently [1-3] it has been found that there is a non-zero probability for the quantum tunnelling of both the magnetization in small particles and nucleation in bulk ferromagnets. The quantum switching process is independent of temperature and is acting below a certain temperature T^* which corresponds to the crossover from the classical to the quantum regime.

Quantum theories [4] of tunnelling can be applied in magnetism in a wide variety of situations including isolated particles as well as strongly interacting particle systems with different forms of magnetic anisotropy and nucleation around defects and surfaces etc. However, attention has only been focused on the tunnelling of the magnetization in single-domain particles through an energy barrier between easy directions [1,2], quantum nucleation within a homogeneous region of a sample described by the simplest form of the magnetic anisotropy energy density [3] and in the motion of a single-domain wall pinning by a defect [5].

In general, magnetization processes are hindered by energy barriers arising from anisotropy in single-domain particles and pinning in materials in which domain wall motion is the dominant magnetization process. Therefore, time dependence of magnetization at sufficiently high temperatures is the natural classical consequence of the effects of thermal agitation in all materials. The predicted relaxation law for the intensity of the magnetization M(t) is [6]

$$M(t) = \text{constant} - S \ln t \tag{1}$$

where S is the coefficient of magnetic viscosity and the constant is related to the magnetization at the beginning of the measurement. This law has been observed for most materials [7] and also obtained in Monte Carlo simulations [8].

The contribution to the magnetic viscosity due to thermal processes, in domain wall pinning and single-domain particles, is given by [6-9]

$$S \sim T/V$$
 (2)

where V is the volume of the switching unit.

Below the crossover temperature, \overline{T}^* , from the classical to the quantum regime, the viscosity is mainly associated with tunnelling through the energy barrier and the resultant viscosity has a major contribution from a term that is independent of the temperature. Hence the tendency of the viscosity to have a non-zero value in the limit $T \to 0$ and the plateau in the S(T) values must be evidence of the quantum, rather than thermal, nature of the effect [10].

The purpose of this letter is to show the occurrence of this new and fascinating effect—quantum tunnelling—occurring at temperatures below 3 K in SmFe multilayers. Our conclusions are based on the experimental finding of the constancy of both magnetic viscosity and coercive field as temperature is varied.

The materials under study are compositionally modulated FeSm multilayers with the composition

The samples were prepared by electron beam evaporation in a high-vacuum chamber. The evaporation rate for Fe and Sm was 0.3 Å s⁻¹. The pressure during the evaporation was always lower than 10^{-7} Torr. Kapton foil was used as the substrate. The temperature of the substrate was 260 °C. The atomic composition of the FeSm layers was obtained from electron probe investigation: Fe₄Sm. The high-angle x-ray data show only peaks corresponding to Cu(111). Hence we believe that there are Fe₄Sm alloys of thickness 30 Å separated by Cu(111) of thickness 100 Å.



Figure 1. The in-plane magnetization dependence on the magnetic field at 5 K.

Hysteresis curves obtained between 5 K and 100 K with the field applied parallel and perpendicular to the film plane show that the total magnetization lies in the film plane. In figures 1 and 2 we show the hysteresis loop at 5 K and the variation of the coercive field with temperature between 5 K and 100 K. The high value of the anisotropy field of these samples, $H_{\rm K} = 2.5$ T, will favour a high crossover temperature from the classical to the quantum regime. In figure 3 we show the results for the



Figure 2. The variation of the coercive field with temperature.

zero-field-cooled (ZFC) and field-cooled (FC) magnetization measurements. The first remarkable feature of the low-field ZFC-FC curves 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. Therefore the broad maximum should reflect a large distribution of freezing temperatures due to the existence of a broad distribution of ferromagnetic clusters.



Figure 3. ZFC-FC magnetization processes for an applied field of 100 Oe parallel to the film plane.

Relaxation measurements were also performed with a SHE SQUID magnetometer. The experimental procedure is the same as that described in [10]. We have measured the time dependence of the sample magnetization using two different criteria: (i) first the sample is cooled at constant field, H_1 , from high temperature to a well established final temperature (FC process), then the field is switched to a new value H_2 and relaxation measurements as a function of time are collected—in this case both H_1 and H_2 are lower than the coercive field at the final temperature; (ii) after saturation of the hysteresis loop in a field of 5 T, 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 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.

Typical relaxation curves for the thermoremanent magnetization after switching the field from 100 Oe to -100 Oe are shown in figure 4. The magnetic viscosity, S, is plotted in figure 5 as a function of temperature. As can be observed, S shows two linear dependences on T: above 3 K, S shows a linear dependence on the temperature indicating that the heights of the energy barriers, U, do not depend on



Figure 4. The time dependence for the thermoremanent magnetization at different temperatures, after switching the magnetic field from 100 Oe to -100 Oe.



Figure 5. The temperature dependence of the magnetic viscosity $S \equiv dM/d \ln t$.

temperature; below 3 K it can be assumed that S does not change with temperature, indicating the existence of a non-thermal contribution to the relaxation process. The most reasonable explanation for this experimental fact, in agreement with theory [1,3], is the occurrence of quantum tunnelling of magnetization. This suggests that the crossing temperature from classical, thermally activated, to quantum processes is about 2 K in this system, in agreement with theory [1-3].

As we said before, we have also measured the time dependence of the magnetization near the coercive field H_c . After saturating the sample in a field of 6 T, we changed the field to a new value and waited until the magnetization reached the value M = 0. The time necessary for that $\tau \sim M_s (dt/dM)_{M=0}$, constitutes the timescale of the experiment for given H and T. We have plotted this time, τ , versus 1/H for different temperatures (figure 6).

Above 2.5 K all the data points are distributed over a convergent set of segments extrapolating to the point with coordinates $(1/H = 0.0014, \tau_0 = 1.4 \times 10^{-11} \text{ s})$. The mean relaxation time τ can be related to an effective temperature T_e via

$$\tau = \tau_0 \mathrm{e}^{U(H)/T_{\mathrm{e}}} \tag{3}$$

where $T_e \equiv T$ for $T \gg T^*$ and $T_e \simeq T^*(H)$ for $T \ll T^*$, and U(H) represents the



Figure 6. The mean relaxation time τ versus 1/H for different temperatures.



Figure 7. The temperature dependence of the effective temperature, T_{e} , defined in equation (3).

dependence of the barriers on the applied field.

From the slopes of the segments of figure 6 we get the values of T_e for each temperature. Figure 7 shows the variation $T_e(T)$. Above 2.5 K we find, as expected, that T_e is equal to T (for a mean energy barrier E = 0.003 eV). Below this temperature T^* shows an upturn (25%), while the magnetic viscosity (figure 5) shows a plateau below 2.5 K. The simplest explanation for this apparent discrepancy is the following: in the magnetic viscosity experiment the magnetic field was the same at each temperature, while this was not the case for the experiments near the coercive field because $H_e(T)$ is a decreasing function. As a result, one might expect an increase of the crossover temperature, which suggests that our multilayers are more or less continuous over rather large length scales, leading to the existence of domain walls.

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