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Magnetic relaxation in single-crystal Co/Cu(100) superlattices

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By measuring the relaxation of the magnetization in a fixed applied field for antiferromagnetic-coupled, single-crystal Co/Cu(100) superlattices, we show that the temperature dependence of the magnetic viscosity is markedly different for samples almost identical from the structural point of view. If the structure of the samples is properly characterized, deviations of the expected behavior for the temperature dependence of the magnetic viscosity can be uniquely interpreted. The results further indicate that care should be taken when assigning a temperature-independent behavior of the viscosity to quantum tunneling of the magnetization without additional experimental support. [S0163-1829(97)03517-0]

Magnetic relaxation is a time-dependent magnetization change following a change in an applied magnetic field that takes place as the system overcomes certain energy barriers.[1] Magnetic relaxation is very important in magnetization-reversal processes such as those employed in magnetic recording. As the size of the recorded domains in storage media decreases continuously and new media (e.g., an array of single-domain magnetic structures with only two quantized states and a density of 65 Gbits/in.^{2}) are being developed,[2] there is an increasing need to understand the details of the switching mechanisms.

On the other hand, there has been an important upsurge of interest on low-temperature magnetic relaxation driven by the possibility to observe quantum tunneling of the magnetization (QTM),[3–6] a manifestation of macroscopic quantum tunneling, i.e., tunneling between macroscopically distinguishible states.[7] There is now strong evidence for tunneling between different flux states in superconducting Josephson junctions[8] and the evidence is growing for QTM. Resonant tunneling could be detected by measuring the resonance between degenerate states of certain molecular antiferromagnets as they are *coherently* driven by tunneling between two potential wells[9] or by field-tuning different spin states in molecular magnets.[10] A much more popular[3–6] approach to QTM studies the decay of a metastable state by tunneling by measuring the temperature dependence of the magnetic relaxation after a sudden change of applied field. The observation of a relaxation rate *independent* of the temperature below a certain transition temperature T_t is sometimes interpreted as evidence of quantum tunneling of the magnetization, even if T_t is fairly high (20 K).[6]

The main difficulty with the interpretation of isothermal relaxation measurements comes from the characterization of the samples, which in most cases consist in collections of objects with a poorly characterized distribution of sizes, shapes, and interactions. Even for ''nanostructured'' samples such as multilayers prepared by sputtering, there is a large number of defects of uncontrolled nature. In this work we aimed at studying the temperature dependence of relaxation effects of the magnetization in two selected metallic single-crystal superlattices whose structural and magnetic properties have been thoroughly characterized by scanning tunneling microscopy (STM), thermal energy atom scattering (TEAS), polarized neutron diffraction, and magnetometry. We will show that if the samples are carefully characterized, magnetic relaxation is a powerful technique, yielding detailed information not easily available by standard structural techniques. Furthermore, deviations from the expected behavior detected at low temperatures can be interpreted as due to subtle structural differences, without implying quantum tunneling of the magnetization.

For most magnetic materials the time evolution of the magnetization M at a given temperature T and applied field can be described by a logarithmic law

$$M(t)=M(t_0)\left[1-S(T)\ln\left(\frac{t}{\tau_0}\right)\right], \tag{1}$$

where $S(T)$ is the magnetic viscosity caused by the thermal activation of magnetization reversals over activation energy barriers.

In fact, this logarithmic dynamic response during isothermal relaxation of metastable systems is fairly universal. Examples of this behavior include structural relaxation of metallic glasses,[11] the decay of charge carriers in amorphous semiconductors,[12] the trapped flux[13] in superconductors, and magnetization reversal in systems composed of small magnetic particles.[14] The ln t kinetics can have a variety of origins, including a non-Arrhenius type of kinetics, interacting and noninteracting particles with a distribution of energy barriers, traditionally ascribed to disorder.[1]

Assuming that a distribution of energy barriers is the origin of the ln t kinetics, let $q(E)$ be the number of barriers per unit energy with activation energy E and $c(E)$ the average change of the measured property, magnetization in our case, when only one of the barriers with activation energy E is thermally activated per unit volume. It can easily be shown that the ln t kinetics can be obtained *if* the reactions are

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Arrhenius type *and* the product $c(E)q(E)$ is roughly constant in the activation energy range swept by the isothermal energy $k_BT\ \ln(t/\tau_0)$ between the initial t_i, and final t_f time of observation. In that case, the total number density of barriers activated after time t at a temperature T is

$$q(E_t)=q(E)\Theta(E,T,t), \tag{2}$$

where $\Theta(E,T,t)=1-\exp[-\nu_0 t\ \exp(-E/k_BT)]$ is a function that, for a given T and t, changes abruptly from 0.01 to 0.99 over a narrow range of energy barriers centered at $E_t=k_BT\ln(\nu_0 t)$. Hence the magnetization would relax in an isothermal experiment as

$$M(t)=M_0[1-c(\langle E\rangle)q(\langle E\rangle)]k_BT\ \ln(\nu_0 t), \tag{3}$$

where $\langle E\rangle$ is the average value of the barriers explored in the interval $k_BT(\ln t_f-\ln t_i)$. The slope of the M vs $\ln t$ straight line is defined as the magnetic viscosity

$$S=\frac{dM}{d\ \ln t}=c(\langle E\rangle)q(\langle E\rangle)k_BT. \tag{4}$$

For thermal processes S is linear with T *if and only if* $c(\langle E\rangle)q(\langle E\rangle)$ is constant in the whole energy interval interrogated by the experiment. For each measuring temperature there is an activation energy range swept in the observation time, $k_BT(\ln t_f-\ln t_i)$, which is obviously much smaller than the total interval explored by the set of isotherms. As a result, $c(\langle E\rangle)q(\langle E\rangle)$ may behave as roughly constant in each small energy interval, but change from one interval to the other. It is obvious that a departure from linearity in the temperature dependence of S, in particular a constant value below some temperature, can be as well ascribed to quantum tunneling or to an appropriate change of $c(E)q(E)$.

The samples studied are two selected single-crystal Co/Cu(100) superlattices, namely, $A=[6$ monolayers (ML) Co/8 ML Cu$]_{62}$ and $B=(9$ ML Co/5 ML Cu$)_{103}$, grown in a molecular beam epitaxy system by alternate deposition of Co and Cu at room temperature on single-crystal Cu(100) substrates. In these conditions, both Co and Cu grow layer by layer on top of the other[16] with the fcc structure, with identical lateral parameters ($a=3.61$ Å) and slightly different vertical lattice parameters ($a_\perp^{\mathrm{Co}}=3.52$ Å, $a_\perp^{\mathrm{Cu}}=3.61$ Å).[17] The number of Co or Cu monolayers in each period was controlled with high precision by counting oscillations in the intensity of the specularly reflected beam of thermal He atoms.[16] The oscillations have monolayer periodicity. The samples were single-crystal superlattices as judged by x-ray and neutron diffraction.[18] They both display partial antiferromagnetic coupling of the magnetization of adjacent Co layers across the Cu spacers as demonstrated by polarized neutron diffraction.[18] On the other hand, there is a fraction of the layers that is ferromagnetically (FM) coupled as deduced from the presence of remanence in magnetization measurements,[19] either by direct contact of Co layers through pinholes in the Cu spacer or simply by thickness fluctuations of the latter (since the coupling changes sign with the spacer thickness[16]). The FM fraction amounts to 56% for sample A and 50% for the sample B.

The only structural difference between samples A and B detected by diffraction techniques was due to the existence of a mosaic in substrate B, giving rise to two superlattices inclined with respect to each other by 1.1°. Detailed studies by scanning tunneling microscopy[20,21] (STM) have revealed the microstructure of Co/Cu interfaces at room temperature. On the atomically flat terraces, there are large Co islands with well-defined thickness and isolated atoms of Co intermixed with Cu, while at the monoatomic steps separating the terraces there are smaller Co clusters surrounded by Cu removed from the substrate. Thus a bimodal distribution of Co sizes (small clusters and well-defined layers) is expected for sample A. The smaller terraces and higher density of steps and dislocations of the Cu substrate with mosaic (B) give rise to enhanced reaction between Co and the Cu substrate[22] with a wider distribution of Co cluster sizes at the interface.

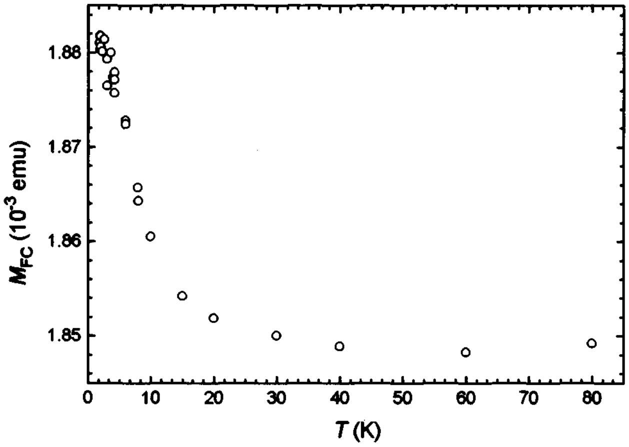

FIG. 1. Temperature dependence of the field-cooled magnetization for the superlattice $A=(6$ ML Co/8 ML Cu$)_{62}$/Cu(100). It was measured while cooling with a field of 200 Oe applied parallel to the layers.

Figure 1 shows the magnetization of sample A during cooling with an applied field of 200 Oe. The magnetization increases by 2% below 10 K following a $1/T$ behavior. This is due to the presence of a small fraction of superparamagnetic or paramagnetic Co clusters that follow the Curie law and coexist with ferromagnetically coupled entities. Note that the AF-coupled layers do not contribute to this process. We ascribe the paramagnetic behavior to small Co clusters diluted in the Cu layers close to each Co/Cu interface. While dilute Co atoms in the bulk of Cu do not show a magnetic moment, isolated Co atoms at a Cu surface were shown to constitute a surface Kondo system with a maximum in the rate of magnetic scattering at 23 K.[23] The isolated Co clusters behave as ‘‘loose’’ spins, i.e., magnetic moments that do not interact with each other.[24]

Figure 2 reproduces some representative $M(t)$ curves taken during relaxation of sample A at different temperatures. The magnetization decays with a logarithmic time dependence in all cases. Notice that the total relaxed fraction of magnetization in these kinetic experiments is of the order of 2%; i.e., it corresponds to the paramagnetic fraction detected in Fig. 1. The slope of the relaxation curves is the magnetic viscosity S, whose behavior (corrected by M_0) as a function of T is plotted in Fig. 3. There is a dramatic *increase* of S as T decreases below 5 K. This behavior is obviously inconsistent with an interpretation in terms of quantum tunneling of the magnetization, which can only justify a behavior of S

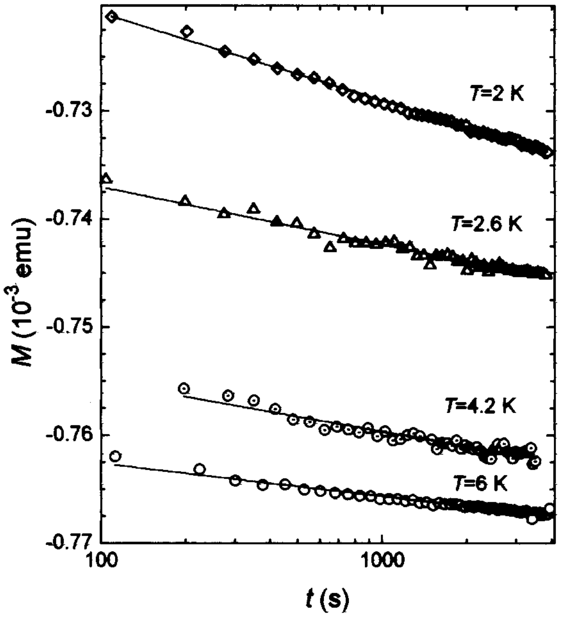

FIG. 2. Representative isothermal relaxations of the magnetization for sample A. The sample was cooled to the temperature of measurement with a field of 200 Oe applied; then, the field was reversed to -120 Oe and the time dependence of M recorded.

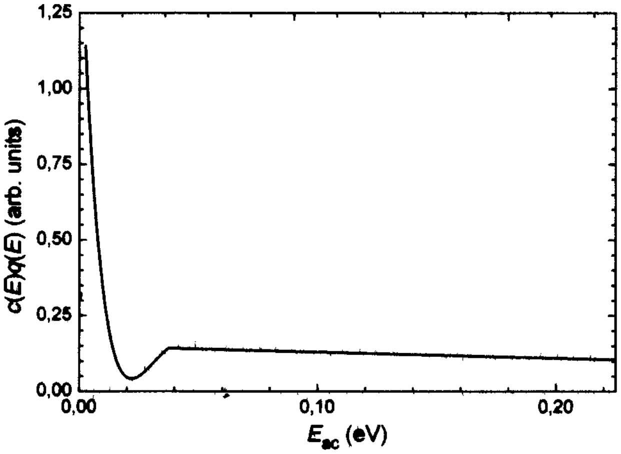

FIG. 4. Distribution function of the energy barriers for a (6 ML Co/8 ML Cu)$_{62}$/Cu(100) sample as a function of the barrier energy for an applied field of 200 Oe.

independent of T. An increase of the kinetics at lower temperature seems difficult to accept. The anomalous behavior of $S(T)$ can be better associated to an increase of $c(E)q(E)$, i.e., to a larger number of barriers being activated at low T. In Fig. 4 we plotted the $c(E)q(E)$ profile deduced from Fig. 3. The abrupt increase in the low-energy range comes from the paramagnetic clusters detected in Fig. 1. The distribution of barriers has a peak at low energy that can be estimated considering that, for $T=2.5$ K, E_t is located at 6 meV after a measuring time of 500 s. Since the corresponding height of the barrier is of the order of 3×10^{-6} eV per atom (as estimated from the anisotropy constant of Co, 5×10^4 J/m^3), the clusters with activation energy below 6 meV have less than 2000 atoms. This estimation corresponds to square islands of Co six layers high with a lateral dimension of 5–6 nm, in excellent agreement with the island size and shape visualized in STM images.[21]

The temperature dependence of the viscosity of sample B in the low-temperature range is shown in Fig. 5. It also deviates from a linear behavior but in a different way from Fig. 3. There are *two* temperature regions where S could be considered ''independent'' of the temperature (within the noise), the first plateau starting at the surprisingly high temperature of 16 K, very near the one reported for Dy/Mo and Tb/Mo sputtered multilayers.[6] It has to be kept in mind that the multilayers prepared by sputtering are polycrystalline with mosaic spreads of the order of 4°–7°. Taking into account the data presented for sample A, there is no reason to interpret those of sample B as evidence of QTM. It rather seems that the apparent independence of S with respect to T for sample B is the result of a broader distribution of barriers due to structural disorder within the layers. It is easy to demonstrate that a density of barriers depending with the energy as $1/E$ would rigorously yield a magnetic viscosity indepen-

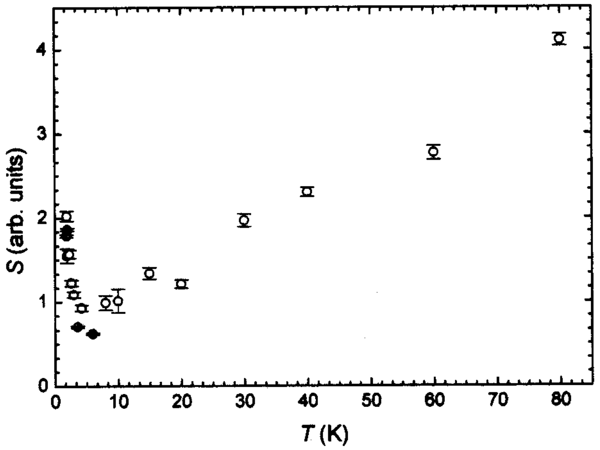

FIG. 3. Magnetic viscosity S as a function of temperature for $A=(6$ ML Co/8 ML Cu$)_{62}$/Cu(100). The magnetic viscosity is defined as the slope of the relaxation curves of Fig. 2 normalized by the corresponding M_0^{FG} value of Fig. 1.

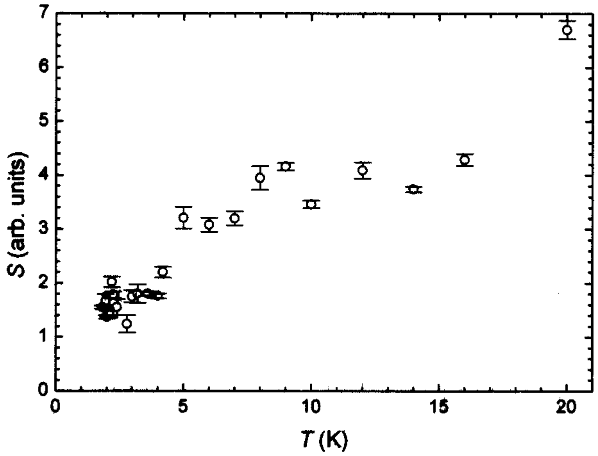

FIG. 5. Temperature dependence of the magnetic viscosity for sample $B=(9$ ML Co/5 ML Cu$)_{103}$/Cu(100) deduced from relaxation experiments such as displayed in Fig. 2.

dent of T.[15] The wider size distribution of Co clusters at the interface induced by the mosaic spread of the substrate is enough to obscure the behavior observed in sample A.

In summary, we have shown using carefully characterized magnetic superlattices that the magnetic-viscosity can be substantially different for samples almost identical from the point of view of standard structural techniques. This reveals the power of magnetic relaxation measurements if the samples are adequately characterized. On the contrary, one has to caution against simplistic interpretations of deviations from the expected linear temperature dependence of the viscosity. In fact, while $S(T)$ in sample A apparently *increases* as the temperature decreases, in sample B a small departure from perfection results in a viscosity ‘‘independent’’ of T that could easily be misinterpreted as evidence of quantum tunneling of the magnetization.[6] Without additional experimental support, any claim in this direction should be taken with caution, since S is weighted by the energy barrier distribution, which may differ from the usually assumed log-normal size distribution of magnetic entities giving rise to a magnetic viscosity ‘‘independent’’ of the temperature. The recently detected complex dynamics of magnetization reversal even in single-domain particles[25] also points in the same direction.

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