Mesoscopic spin tunneling in the hard-random-axis-magnet amorphous alloy Tb_2Fe

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Measurements of the time dependence of the remanent magnetization for the strong random magnetic anisotropy amorphous alloy Tb_2Fe show a temperature-independent time-logarithmic relaxation rate below the crossover temperature $T_Q = 8$ K. Our results are consistent with the occurrence of quantum tunneling of magnetization, T_Q being one of the *highest* crossover temperatures found to date. The estimated mesoscopic tunneling volumes, including around 100 spins, compare with the short-range ferromagnetically correlated volumes existing in the sample.

During the past few years the study of quantum tunneling of magnetization (QTM) has become an active field of research.[1] Many experiments concerning the temperature dependence of the magnetic relaxation showed that, at low enough temperatures, metastable spin configurations decay through underbarrier quantum transitions, and not via the expected pure thermally activated (TA) processes, which would lead to a vanishing zero-temperature relaxation rate. In the former experiments the observed quantum event was the nucleation of reversed domains in disordered ferromagnetic crystals.[2] The tunneling of the magnetic moment in ferro- and antiferromagnetic (AF) nanoparticles and microparticles was reported by several authors[3-5] (a different approach of studying the QTM based on resonance experiments was also undertaken in nanometer-scale AF particles[6]). Recent experiments[7] also showed QTM events of single portions of planar magnetic domain walls. The QTM was even observed in systems possessing weak[8] and strong[9] random magnetic anisotropy (RMA). Theoretical predictions[10,11] concerning the estimates of the probability of quantum decay of metastable spin configurations and the crossover temperature from thermal to quantum regime agree reasonably well for ferro- and AF materials.[3,4,12] For the strong RMA amorphous system Tb_2Cu, our experimental results[9] suggest a crossover temperature $T_Q \approx 4$ K, which compares well with the calculated value $T_Q \approx 3.7$ K, obtained by using a theoretical expression of T_Q deduced for hard-random magnets.[13]

We here report on an experiment suggesting the existence of QTM in the amorphous magnet a-Tb_2Fe, which seems to have the highest T_Q (≈ 8K) found to date in strong RMA magnets and, to our knowledge, in any magnetic system. Low-field magnetization[14] and small-angle neutron scattering[15] (SANS) experiments were performed on a-$Tb_2(Fe_{1-x}Ni_x)$ alloys. Zero-field-cooled (ZFC) magnetization values at the ordering transition temperatures, $M(T_c)$, kept nearly constant on varying Ni content. Thus, accordingly with the theoretical prediction[16] that $M(T_c) \propto (J/D)^4$, where D stands for the RMA crystal-field strength and J is the mean value of the exchange interaction between the Tb^{3+} ions, a constant ratio of J/D was suggested for this series.[14] The analysis of the SANS data showed a magnetic correlation length $\xi(56 \pm 5\text{Å})$ almost temperature independent below the transition temperatures and with differences smaller than 20% in ξ for different Ni contents. Thus, since at low temperatures $\xi \propto (J/D)^2$, [17] a constant J/D ratio is again deduced for these alloys.[15] On the other hand, the short ferromagnetic correlation length found is consistent with a strong RMA. However, there is a significant difference between the Fe and Ni atoms which affects J value along the series. Such atoms do not carry magnetic moment in the isomorphous a-Y_2Fe and a-Y_2Ni alloys, but an applied magnetic field induces a moment on the iron because of the exchange-enhanced susceptibility of the d band (as opposed to nickel).[18] Although the magnetic moment on Fe in a-Tb_2Fe is insignificant, there is a strong antiferromagnetic coupling between the Fe and Tb ions (a-Tb_2Fe is a sperimagnet), which in turn increases the value of the ferromagnetic coupling constant J between the Tb^{3+} ions. Thus, substitution by Ni will decrease J, in agreement with the decrease of T_c (Ref. 15). This result, combined with the above-mentioned constancy of J/D, suggests that D decreases with the Ni concentration.

In the isomorphous compound a-Tb_2Cu, the Cu ions do not carry a magnetic moment, the magnetic properties of this alloy being similar to those of a-Tb_2Ni.[14] Therefore, from the preceding discussion, we can expect for D^2/J a value higher in a-Tb_2Fe than in a-Tb_2Cu. The same happens for the volume anisotropy constant K, since $K \propto D^2/J$ in these RMA systems.[9,19,20] The higher magnetic anisotropy in a-Tb_2Fe compared with the anisotropy in the Cu alloy motivated us to study the QTM in a-Tb_2Fe, due to the predicted increase of the quantum relaxation regime crossover temperature T_Q, when K increases [see Eq. (1) below].[13]

Amorphous ribbons of a-Tb_2Fe were prepared by melt spinning and the amorphous structure of the samples

checked by x-ray diffraction. Low-field magnetization measurements were carried out with a superconducting quantum interference device magnetometer. The large difference observed below $T_s = 160$ K, between the low-field magnetization values for field-cooled (FC) and zero-field-cooled (ZFC) processes, indicates that the system passes from the paramagnetic phase to a low-temperature sperimagnetic phase, due to the strong RMA of the Tb^{3+} ions. We measured the magnetization at high fields by using a vibrating sample magnetometer fitted with a 12-T superconducting solenoid, from 1.9 up to 150 K. First-magnetization isotherms and hysteresis loops were performed using a maximum applied field of 9.5 T, at a constant sweep rate of 250 Oe/s. The 9.5-T field was high enough to fully close the hysteresis loops and to produce an isothermal remanent magnetization, M_r, without memory effects. In Fig. 1 we present the hysteresis loop at 3.2 K, showing the large magnetization jumps, at the coercive fields H_c, typical of strong RMA systems. The M_r values obtained for zero applied magnetic field are shown in the inset of Fig. 1, as well as the irreversible susceptibility at remanence, χ_{irr}, obtained from the hysteresis loops and the low-field magnetization data. The H_c field behaves similarly to M_r, having large values at low temperatures, e.g., $H_c \approx 3$ T at 1.8 K and almost disappearing at around T_s.

We measured the time dependence of the magnetization of the metastable remanent state obtained after application of a 9.5-T field. Figure 2 shows the $M_r(t)$ values taken at time intervals of 2 s, between the instant in which the applied magnetic field reached zero and ~ 1800 s later. The semilog plots of Fig. 2 are quite linear, thus, a logarithmic law $M_r(t) = A - S \ln T$, where S is the magnetic viscosity, is well followed within the time window of our experiments, especially at low temperatures. We obtained fits of similar quality to the logarithmic ones, or even better at high temperatures, by using an algebraic law, i.e., $M_r(t) = Bt^{-\beta}$. To compare both kind of fits, we show in Fig. 3 the thermal dependencies of the characteristic parameters S and $\beta' = \beta M_r(0)$. The logarithmic and algebraic decay laws arise from quite different probability distributions of energy barriers, $P(U)$, i.e., flat or uniform[21] and exponential[22] respectively. Nevertheless, both should behave similarly at low temperatures,[9] and this is what we observe in Fig. 3 at temperatures below ≈ 15 K, where $S(T) \approx \beta'(T)$. Let us consider now the occurrence of TA relaxation processes and the existence of any of the above $P(U)$. In both cases, a linear decrease with temperature for the magnetic viscosity, S, and for the algebraic law exponent at low temperatures, is predicted.[9,21] This linear behavior is in fact observed in Fig. 3 on approaching the low-temperature regime, for $T \lesssim 20$ K, but it is not at all followed below ≈ 8 K, where S and β' are nearly temperature *independent*. We ascribe this low-temperature behavior to the occurrence of quantum tunneling of the magnetization below a crossover temperature $T_Q \approx 8$ K. This temperature T_Q is defined[10] in terms of the rate Γ of quantum decay of a metastable spin configuration, $\Gamma \propto \exp(-U/k_B T_Q)$, compared to the rate of thermal transitions over an energy barrier of height U, $\Gamma \propto \exp(-U/k_B T)$; thus, for $T < T_Q$ quantum transi-

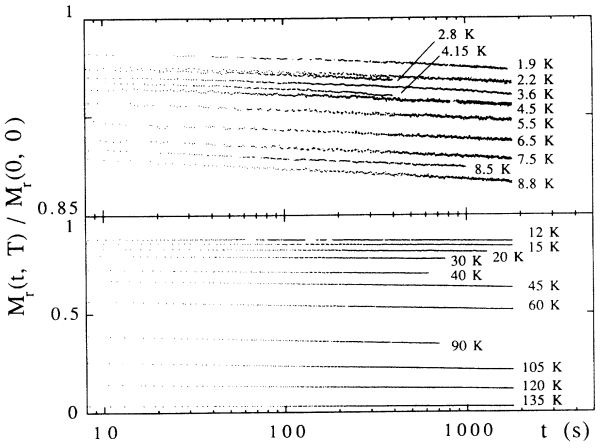

FIG. 2. Time decay of the normalized remanent magnetization for a-Tb_2Fe.

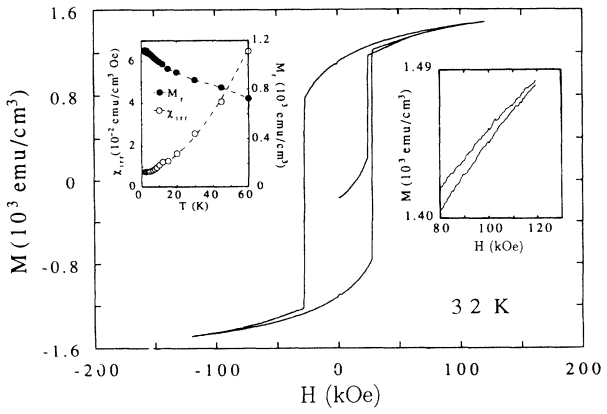

FIG. 1. Hysteresis loop at 3.2 K for a-Tb_2Fe. Right inset: detail of the high-field region. Left inset: irreversible susceptibility at remanence and remanent magnetization.

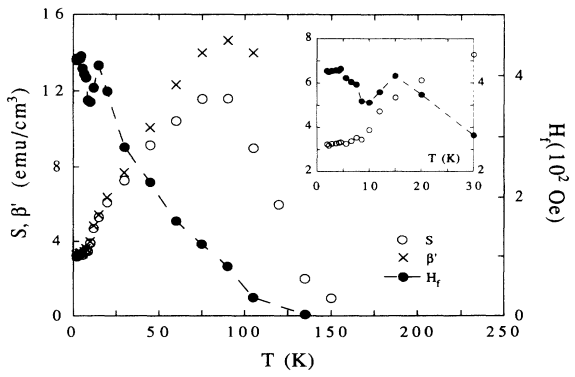

FIG. 3. Temperature dependences of the magnetic viscosity S, exponent of the algebraic magnetization decay law β', and fluctuation field H_f for a-Tb_2Fe. Inset shows clearer the non-thermal behavior of S and H_f below the crossover temperature $T_Q \approx 8$ K.

tions prevail. The expression giving T_Q in terms of macroscopic parameters has been obtained by Chudnovsky for hard-random magnets and spin glasses,[13]

$$T_Q=(\mu_B/k_B)(K/\chi)^{1/2}\ , \tag{1}$$

where K is the volume anisotropy constant and χ the zero-field susceptibility; Eq. (1) is valid only if $\chi \ll 1$. We can estimate K from the anisotropy field H_h, defined as the field necessary to close the up and down branches of the hysteresis loop, and using the expression $K=(1/2)H_hM_s$, where M_s is the saturation magnetization. For our a-Tb_2Fe sample, at 3.2 K, we obtain, from Fig. 1, a closing field $H_h \gtrsim 9.5$ T, in agreement with the assumption of no irreversibility above that field, used to perform the relaxation measurements. However, a detailed analysis of the high-field regime (see inset in Fig. 1) gives us a value of $H_s \gtrsim 12$T, above which the hysteresis loop fully closes. Thus, by using the above values of H_h and $M_s=1500$ emu/cu^3, we obtain a minimum K value of 7.1×10^7 erg/cm^3 and an estimated value of $K \gtrsim 9\times10^7$erg/cm^3, at 3.2 K. Note that, due to the little thermal variation of H_h and M_s at low temperatures, K is nearly temperature independent below $\sim$15 K. Substituting into Eq. (1) the values obtained for K and the low-temperature differential susceptibility $\chi \approx 7.9\times10^{-3}$ emu/Oe cm^3, we get a lower limit for T_Q of 6.4 K, and an estimated value $T_Q \gtrsim 7.2$ K. This later T_Q value is in fair agreement with the experimentally measured $T_Q \approx 8$ K. On the other hand, the high value of T_Q obtained for a-Tb_2Fe is due to the high magnetocrystalline anisotropy introduced by the Tb^{3+} ions, as anticipated, and to the low value of χ in our strong RMA system a-Tb_2Fe.

Let us consider now the average volume v_Q of the entity, or "particle," which tunnels. Before determining v_Q we will obtain the average TA volume v making use of the concept of thermal fluctuation field, $H_f=S/\chi_{\rm irr}$.[23] This field, expressed in terms of the field derivative of the energy barrier, is $H_f=k_BT/(\partial U/\partial H)_T$, and describes more clearly than through S the effects of thermal activation. This is so because of the independence H_f nor on the details of $P(U)$, or on the sample and experimental conditions. Thus, it can be shown[9,21,24] that

$$v(T)\cong 1.2\frac{k_BT}{H_f(T)M_S(T)}\ , \tag{2}$$

where the prefactor 1.2 is valid for an RMA system under zero applied magnetic field,[9] our experimental situation at remanence. In Fig. 3 we plot the temperature dependence of H_f. Above $\sim$8 K, H_f displays the usual features, a rapid increase at low temperatures, followed by a maximum and a smooth decrease towards higher temperatures, commonly observed in magnetic systems where *only* TA processes contribute to the relaxation. In such systems, H_f goes to zero when T tends to zero. This is not at all observed in Fig. 3, where the tendency of H_f towards zero, initiated at $\sim$15 K, breaks down abruptly below $\lesssim$9 K, from where H_f increases slightly, taking a constant value below $\sim$5 K. This anomalous behavior of H_f at low temperatures again indicates the change of the relaxation mechanism, from pure TA to a quantum transitions regime. In Fig. 4 we plot the average TA volume $v(T)$ obtained from Eq. (2). It shows an exponential decrease from high temperatures down to $\sim$8 K, below this temperature decreasing linearly to zero. This can be understood considering Eq. (2) and the temperature dependence of H_f, i.e., $H_f(T)\propto M_s(T)k_BT/\chi_{\rm irr}(T)$.[21,23] Thus, for TA processes $v(T)\propto\chi_{\rm irr}(T)/[M_S(T)]^2$. From our magnetization measurements we know that, below $\sim$60 K, M_s has little temperature variation. In the inset of Fig. 1 we see that $\chi_{\rm irr}$ is temperature independent below $\sim$7 K. This implies an average TA volume independent of T below $\sim$7 K. However, as a consequence of the little thermal variation of H_f below $\sim$8 K, certainly smoother than k_BT, v decreases linearly below $\sim$8 K (see Fig. 4 inset), according to Eq. (2). Such contradiction represents another way to show the nonthermal nature of the relaxation below $T_Q \sim 8$ K.

We can reasonably assume that the low-temperature TA volume, v ($T=10$ K), coincides with the average volume v_Q involved in the QTM processes below T_Q, thus $v_Q \approx 3.2\times10^3$ Å^3. This volume contains $\sim$100 Tb^{3+} ions, so a mesoscopic number of spins are involved in an individual tunneling event. In the language of a continuous field of spin rotations, employed by Chudnovsky[13] to describe macroscopic states of a random magnet, and to define a QTM variable, our volume v_Q corresponds to the volume δ^3 of a metastable spin configuration, or magnetic soliton of size δ ($\sim$9 Å for a-Tb_2Fe), which minimizes the energy of the system. As it has been pointed out,[9,13] the exchange interaction determines the tunneling "particle" in a random magnet. Thus, it is interesting to compare v_Q with the short-range ferromagnetic correlation length R_f. For strong RMA systems $R_f \sim R_a$,[17] where R_a is the short-range structural order correlation length. In our sample, from the determined x-ray-diffraction patterns we obtain that $R_a \sim 9.8$ Å (see Ref. 9 for details). Within this length (first and second neighbors) the Tb^{3+} moments should be nearly parallel. Moreover, according to the value of δ, they should remain rather parallel up to the first three shells of neighbors (15-Å scale). Note that the length

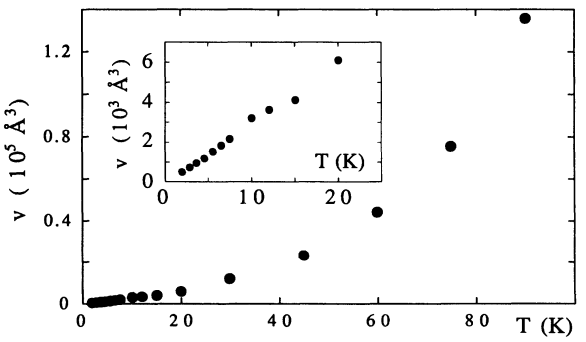

FIG. 4. Thermal variation of the average activation volume in a-Tb_2Fe obtained from Eq. (2). The calculated volume below 10 K has no physical meaning.

$\xi(\sim 56$ Å), deduced from SANS experiments[15] for a-Tb_2Fe is a measure of the medium-range magnetic order,[25] and two spins located a distance ξ apart are almost uncorrelated. Hence, it is R_f, rather than ξ, that determines the size of the metastable spin configuration.

It could be argued that since for TA processes, a distribution $P(U) \propto U^{-1}$ gives rise to a temperature-independent magnetic viscosity,[26] the behavior observed below T_Q for a-Tb_2Fe could be due not to QTM but to purely TA processes. We think that this is not our case, because in an amorphous sample the distribution of R_a values, which is responsible for the distribution of activation volumes and, hence, of $P(U)$, is expected to be a regular function, nondiverging at low R_a values. Moreover, the constancy of the magnetic correlation length below the transition temperature suggests the exclusion of any anomalous behavior of ν_Q at low temperatures, which could originate the plateau in the magnetic viscosity.

Finally, it is also worth mentioning that, although in the Tb-transition-metal amorphous alloys the magnetoelastic coupling is very strong,[27] it has been shown[28] that the magnetoelastic dissipation causes practically no reduction in the tunneling rate for QTM in magnetically ordered systems. In disordered magnets the magnetoelastic dissipation can be stronger[13] but, conversely, the dissipative interaction with conduction electrons should be less important in amorphous alloys than in crystalline systems due to the increased resistivity in disordered systems. In fact, we think that QTM accounts for the temperature-independent magnetic viscosity below ~ 8 K in a-Tb_2Fe.

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