Quantum relaxation in the low-temperature magnetic susceptibility of the amorphous alloy a-Tb_2Fe

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The ac magnetic susceptibility of melt spun ribbons of the strong-random magnetic-anisotropy amorphous alloy a-Tb_2Fe has been measured from room temperature down to 0.10 K. In the temperature region $4<T<300$ K the system behaves as a random anisotropy sperimagnet, with freezing temperature $T_f=160$ K. In-phase χ' and out-of-phase χ'' components of the susceptibility deviate from the classical linear temperature dependence to a temperature-independent value below a crossover temperature $T_Q\approx 3$ K. The low-temperature ''plateau'' behavior supports the existence of magnetic quantum tunneling relaxation. The tunneling entities are spin drops which comprise an average number $n\approx 11$ spins.
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I. INTRODUCTION

Amorphous alloys $R_{1-x}Fe_x$ of rare earths (R) and iron have been characterized as systems with random magnetic anisotropy (RMA).[1] Quantum tunneling of magnetization (QTM) processes have been reported to occur both in weak[2] and strong[3] RMA systems. In those works it was found that magnetic viscosity becomes temperature independent below a crossover temperature T_Q. On the other hand, attempts to observe the crossover behavior in amorphous systems by means of a different experimental technique, such as magnetic susceptibility, have been unsuccessful.[4]

The a-Tb_2Fe alloy is a good candidate in which QTM might be observable by means of magnetic susceptibility experiments since the crossover found at T_Q (≈ 8 K) in magnetic viscosity measurements is the highest found up to date in a RMA magnet, to our knowledge.[5] Our aim has been to check whether the crossover would also be observed with ac magnetic susceptibility, and, if positive, which were the main differences in behavior with respect to the magnetic viscosity data. We have extended our magnetic measurements down to 0.10 K to ascertain any crossover in its temperature dependence. We have observed that the real and imaginary components of the ac susceptibility, χ' and χ'', become temperature independent below $T_Q\approx 3-4$ K, and consider this to provide further support for the existence of QTM in this compound.

If R is nonmagnetic, such as $R=$Y, no magnetic moment is present in Fe below a certain critical value, $x_c=0.4$.[6,7] Indeed, the alloy a-Y_2Fe is below this threshold and has no moment if no strong field is applied.[8] In contrast, in a-Tb_2Fe the Fe atoms do have a nonzero moment, thus the R-T interaction is capable of polarizing the $3d$ band of Fe.[1] Below the freezing temperature T_f and for predominant ferromagnetic interactions $J>0$, the ratio (D/J) determines the magnetic ground state of the amorphous material, where D stands for the RMA crystal-field strength and J is the main exchange interaction between the Tb^{+3} atoms.[1] For weak RMA, $D/J<1$, the average magnetization vector rotates smoothly along the sample so that the magnetic moments are correlated at relatively long distances. If we are in the strong RMA regime, $D/J>1$, magnetic moments lie along the respective local easy directions; therefore, the ferromagnetic correlation length is limited at most to the short-range structural correlation length.[9] In particular, for a-Tb_2Fe the values $J/k_B=1.53$ K and $D/k_B=8$ K were determined from the fit of the magnetization and anisotropic magnetostriction thermal dependences, thus $D/J=5.2>1$. Since $D/J>1$ and the R-T interaction is antiferromagnetic, then the a-Tb_2Fe system is a sperimagnet.[10]

The organization of our paper is as follows: first, experimental details are given, then a theoretical section introduces a model for the spin excitations involved and obtains predictions for χ' and χ'' above and below T_Q. The predictions are compared to experiment in the last section.

II. EXPERIMENTAL DETAILS

Magnetization and complex ac magnetic susceptibility measurements were carried out between 1.8 and 300 K with a commercial Quantum Design superconducting quantum interference device (SQUID) magnetometer. The sensitivity of the magnetometer is 10^{-7} emu. The field was applied parallel to the plane of the ribbons. The amplitude of the ac excitation was $h_0=1$ Oe. We varied the frequency between 0.01 Hz and 1 kHz. Before each measurement series the superconducting magnet remanent field in the center of the sample chamber was eliminated. The remaining field was reduced, with the sample at room temperature, to a very low value (0.01 Oe) applying a compensating field with the primary coil. Then, the sample was cooled down to the lowest measuring temperature. Nevertheless, irreproducibilities of the order of a 4% of the total signal were observed between different measurement series, whereas they were below 1% for data obtained after the same cooling down process.

Ac susceptibility data below 2 K were measured with a susceptometer set up on a ^{3}He-^{4}He Oxford Instruments dilution refrigerator; the lowest temperature reached was 0.10 K. A mutual inductance coil was wound on a glass-tube placed under the mixing chamber of the refrigerator, and the sample placed in one of the oppositely wound secondary coils. The

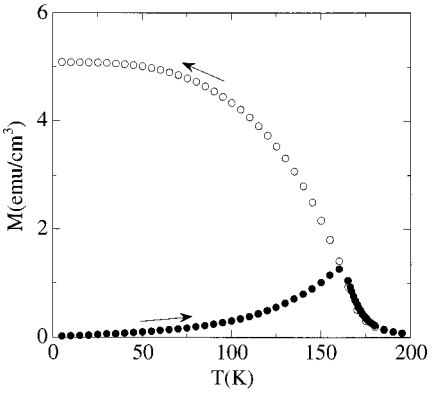

FIG. 1. Temperature dependence of the magnetization of the amorphous alloy a-Tb_2Fe; ●, measured after zero-field cooling (ZFC) ($H<0.05$ Oe); ○, measured after field cooling ($H=1.0$ Oe) (FC).

tube was filled with ^{3}He-^{4}He fluid to achieve thermal equilibrium of the sample with the mixing chamber. The exciting field amplitude was $h_0 \approx 3.6$ mOe and the frequency was 159 Hz. We used the temperature overlap region between 1.8 to 3 K of the magnetometer and susceptometer data to scale the latter measurements.

Amorphous ribbons of a-Tb_2Fe were prepared by melt spinning. The resulting material were strips 50 μm thick and several cm long. The amorphous structure of the samples was checked by x-ray diffraction. The SQUID magnetometer measurements were performed on a collection of 20 stacked ribbons. The measurements are given in the Gaussian system. The samples used in very low-temperature experiments consisted of about 40 ribbons glued with GE varnish. Thus its weight could not be determined accurately and the very low-temperature measurements are given in arbitrary units.

III. EXPERIMENTAL RESULTS

A. Magnetic transition

We first show in Fig. 1 the temperature dependence magnetization measurements at an applied field of 1 Oe. ZFC and FC stand for data measured after the sample was cooled in zero field and under the measuring field, respectively. The ZFC curve shows an acute anomaly at the freezing temperature $T_f = 160$ K, which indicates the onset of the sperimagnetic structure. Below T_f the ZFC and the FC curves strongly diverge, indicating that some of the irreversible magnetization processes last for times of the order or longer than the time elapsed between two experimental points $Z_{\rm exp}$; i.e., of the order of 100 s.

In-phase χ' and quadrature χ'' components of the complex susceptibility near T_f are shown in Fig. 2. Above T_f, χ' follows Curie's law and $\chi'' \approx 0$. Both components χ' and χ'' have an acute maximum, similar to that of a spin glass or to the one shown by other random anisotropy materials[11] at temperatures T_f' and T_f'', respectively, which increase as the exciting frequency increases. The onset of a nonzero χ'' is due to magnetic relaxation processes with characteristic relaxation times τ of the order of $\tau_{\rm exp}$, i.e., the same mechanism which gives rise to the difference between the ZFC and FC magnetization curves. These results indicate that the

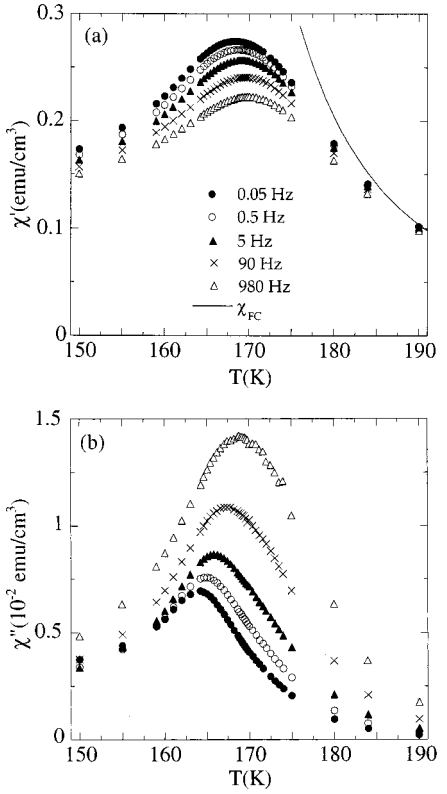

FIG. 2. Ac magnetic susceptibility of a-Tb_2Fe near the spin-freezing temperature T_f, measured at several frequencies; (a) real component χ'; (b) imaginary component χ'''. The full line in (a) represents the dc susceptibility measured after FC in a field of 1 Oe.

spectrum of relaxation times broadens dramatically when T approaches T_f, at least up to about 20 s.

B. Low-temperature region

The temperature dependence of χ' and χ'' below 10 K is shown in Fig. 3. Above 4 K, χ' and χ'' increase linearly for increasing temperature. The real component χ' deviates from linearity below 4 K, approximately, and increases slightly at lower temperatures. χ'' tends to a nearly temperature-independent behavior below 3 K, as well. In Fig. 4 we show our data down to 0.10 K, where the two sets of data, those obtained with the SQUID magnetometer and the dilution refrigerator susceptometer, have been combined. We note that the data tend to a ''plateaulike'' behavior below 4 K for χ' and 3 K for χ''. In fact, there is a slight temperature dependence; a small maximum of 3% shows up at about 1 K in both components. In Sec. IV we propose a simple model to explain this behavior.

However, without recourse to any model, the observed crossover behavior already allows us to establish that some irreversible magnetization processes exist which do not become blocked even at the lowest temperatures achieved ($T=0.10$ K), thus, these processes are not thermally activated. We remark as noteworthy that χ'' is nonzero; this feature had not been detected in any other previous random anisotropy amorphous system, though there is an antecedent in a high permeability amorphous ferromagnet.[12] On the other hand, in the random anisotropy system a-DyNi (in which $D/J<1$,

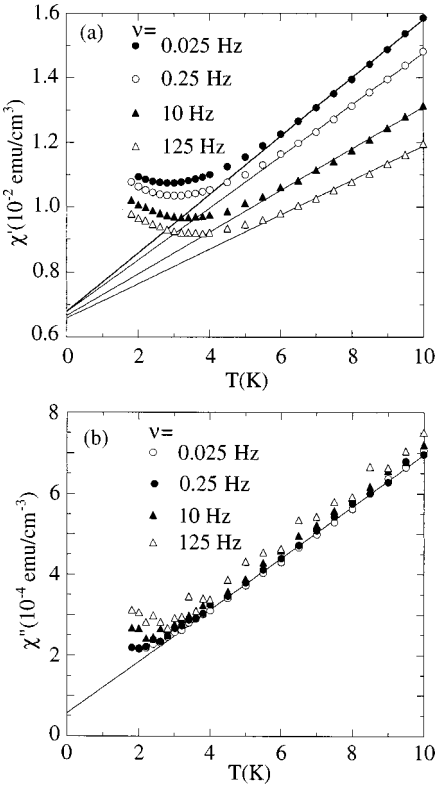

FIG. 3. Ac magnetic susceptibility of a-Tb_2Fe at low temperatures; (a) real component χ'; (b) imaginary component χ''. The full lines correspond to the best fit to the experimental data for $T>5$ K.

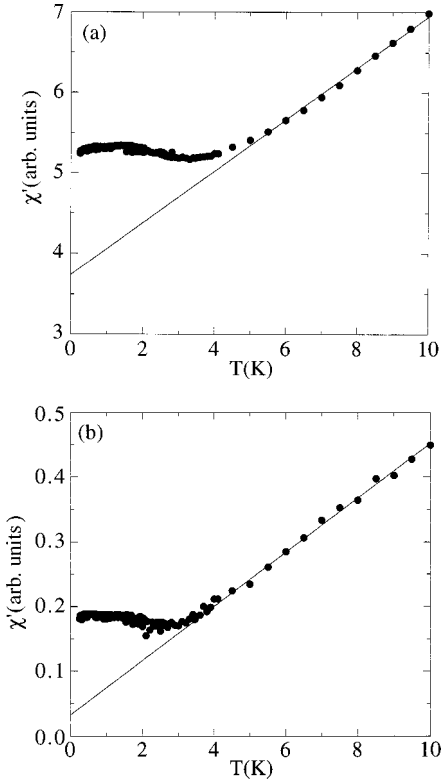

FIG. 4. Ac magnetic susceptibility of a-Tb_2Fe measured at the frequency $\nu=159$ Hz; (a) real component χ'; (b) imaginary component χ''. The full lines correspond to the best fit to the experimental data for $T>5$ K.

recently studied, it was found that χ'' decreases with temperature in the whole experimental range ($T>1.1$ K).[13]

At all temperatures studied, χ' depends on the logarithm of the exciting frequency ω (see Fig. 5), which implies that the relaxation time spectrum is nearly constant.[14] Moreover, it is remarkable to find that the well-known relation between χ' and χ''

$$\chi''=\frac{\pi}{2}\frac{\partial\chi'}{\partial(\ln\omega)} \tag{1}$$

holds in the temperature range where frequency-dependent data were collected ($1.8<T<20$ K) (see Fig. 6). In fact, $\partial\chi'/\partial(\ln\omega)$ also approaches a temperature-independent behavior below 3 K.

The most relevant direct conclusion from the present experiments is that there exist effective relaxation processes down to the lowest temperatures explored. We discuss below on the physical nature of these processes.

IV. ANALYSIS OF RESULTS

A. A magnetic drop model for spin relaxation in strong RMA systems

In this section we describe a simple model of drops of correlated spins as a first step to interpret our experimental results. The original model was introduced by Anderson, Halperin, and Varma to explain the low-temperature heat capacity of a glass,[15] and was developed later to calculate the susceptibility and magnetization of a random exchange spin glass.[16] Recently, the same model was applied to interpret the ac susceptibility of some ferromagnetic amorphous alloys[12] and of weak random anisotropy systems.[13]

The central hypothesis of the model consists of the assumption that magnetic excitations involve the reversal of groups of ferromagnetically correlated spins closely aligned along the local axis of the RMA. We shall denominate them drops. The boundary of each drop separates spins that are nearly orthogonal to each other; i.e., not transversally correlated because of the RMA [see Fig. 7(a)].[9] Such a configuration corresponds to a minimum of energy. A second minimum, which differs in energy by ΔE [see Fig. 7(b)], is reached when the spins within the drop flip as a whole, while maintaining the boundary, as the surrounding spins are not

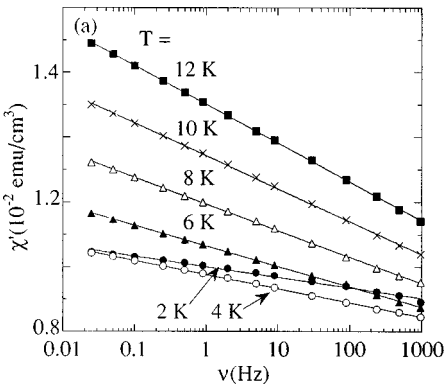

FIG. 5. Frequency dependence of χ' at low temperatures. Full lines in (a) are fits to a logarithmic dependence.

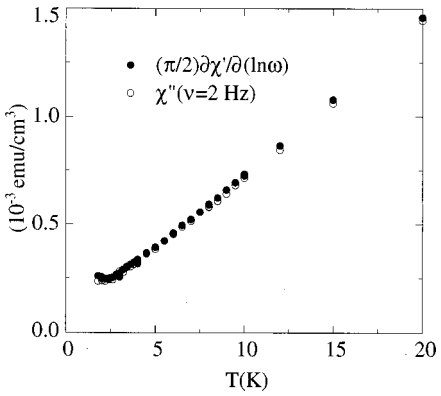

FIG. 6. Comparison between χ'' and the first derivative of χ' (scaled by $\pi/2$) with respect to the logarithm of ν [see text Eq. (1)], at low temperatures.

correlated on a scale larger than the drop size. U is the energy barrier to overcome classicaly when flipping from the first minimum to the second minimum [see Fig. 7(b)]. ΔE may be nonzero since the angle between the across boundary spins may vary. In fact, the average angle is an increasing function of the D/J ratio. However, this difference may be considered as very small with respect to U, because of the competing effects of the random anisotropy and exchange.[9] The low-energy excitations which involve these flippings determine the linear magnetic response of RMA systems at low temperatures. Their existence has been proven experimentally in the amorphous alloy a-DyNi.[17]

Since all the drop inner spins are correlated along the direction of the local axis of anisotropy, the energy barrier height U is proportional to the drop volume V, while ΔE may have an arbitrarily small value, even for relatively large drops. Since there is a large energy range of barriers the difference ΔE may also have a wide range of values. For a given U, there may be many quasidegenerate compatible drop configurations, with different values of ΔE, and vice versa. Therefore, it is reasonable to assume that there is a smoothly varying distribution of values for ΔE and U; i.e., a distribution $f(\Delta E,U)$ that varies smoothly in the energy interval k_BT.

In general, the relaxation time τ and the equilibrium susceptibility of one of these drops depend on ΔE and U. For

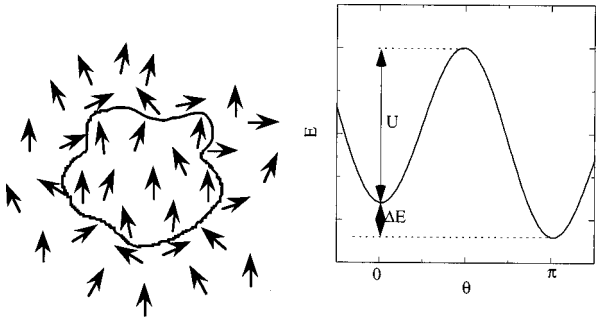

FIG. 7. (a) Schematic representation of a drop of transversally correlated ferromagnetically coupled spins. The line represents the boundary separating sites such that their local anisotropy axis is orthogonal. (b) Dependence of the magnetic energy as a function of angle θ, which describes the direction of alignment of the drop magnetic moment shown in (a).

simplicity sake, we shall consider each drop as a two-level system. If, moreover, $U \gg \Delta E$ the relaxation time is

$$\tau = \tau_0 \exp\left(\frac{U}{k_B T^*}\right), \tag{2}$$

where τ_0 is a characteristic time, T^* is equal to T if the process is thermally activated, and equal to T_Q if the drop flips by means of a tunneling process.

This tunneling process is probably assisted by phonons rather than due to ground-state resonant tunneling.[15,19] Indeed, the tunneling splitting of the ground state of a drop which contains approximately 11 spins (see last section) can be roughly estimated to be smaller than 10^{-12} K.[18] This quantity is much smaller than the smallest splitting possible $\Delta E \approx 10^{-2}$ K, imposed by the ac field. Under this condition the ground-state resonant tunneling channel is blocked.[19]

B. Calculation of the low-field magnetic susceptibility

The equilibrium parallel susceptibility for a two-level system of $+\mu$ and $-\mu$ magnetic moments separated by an energy ΔE, for a given U, may be written as

$$\chi_{\parallel}(T,\Delta E,U) = \frac{V\rho^2\mu^2}{k_B T \cosh^2(\Delta E/2k_BT)}, \tag{3}$$

where $\rho = n/V$ is the spin density, n is the number of spins in the volume V, and μ is the magnetic moment of a Tb^{3+} ion. On the other hand, the perpendicular component is zero. Thus $\chi_{eq} = \chi_{\parallel}/3$ is a monotonically decreasing function of $\Delta E/k_BT$, at a given temperature, becoming negligible above $\Delta E/4k_BT > 1$. It can be concluded that only the lower energy excitations ($\Delta E \leqslant 4k_BT$) give a significant contribution to the equilibrium magnetic susceptibility.

In contrast to this result, we note that all the excitation spectrum contributes to the remanent magnetization decay; i.e., independently of the value of ΔE, an excitation contributes to the magnetic viscosity if

$$k_BT^* \ln(1/\omega\tau_0) = U$$

with $T^* = T$ and $T^* = T_Q$ above and below T_Q, respectively. We conclude that viscosity is sensitive to larger energy excitations than ac susceptibility, thus the average value of ΔE is larger in magnetic viscosity experiments. In other words, ac susceptibility gives information on the metastable ground states of the system, while magnetic viscosity probes excited states.

When τ is of the order or longer than $\tau_{ex} = 1/\omega_{ex}$, the system cannot follow the excitation and the magnetic response is lower than the equilibrium susceptibility; i.e., a phase shift appears in the response giving rise to an imaginary component. The contribution to the susceptibility due to each drop may then be calculated with the Debye relaxation expressions:

$$\chi'(T,\omega,\Delta E,U) = \frac{\chi_{eq}}{1+(\omega\tau)^2} + \chi_{rev}, \tag{4a}$$

$$\chi''(T,\omega,\Delta E,U)=\frac{\chi_{\rm eq}\omega\tau}{1+(\omega\tau)^2}, \tag{4b}$$

where τ is given by Eq. (2). $\chi_{\rm rev}$ is the reversible susceptibility due to small rotations of the magnetization within the energy minimum; i.e., it is the limit of χ' for $\omega\to\infty$. To calculate the susceptibility of a given sample, one must add the contributions due to all excitations assuming that $f(\Delta E,U)$, the normalized distribution function, is a smooth function in the relevant energy region. Then

$$\chi'(T,\omega)=\chi_{\rm rev}+\int_0^\infty \frac{1}{1+(\omega\tau)^2}\,\tilde{\chi}(T,U)dU, \tag{5a}$$

$$\chi''(T,\omega)=\int_0^\infty \frac{\omega\tau}{1+(\omega\tau)^2}\,\tilde{\chi}(T,U)dU, \tag{5b}$$

where

$$\tilde{\chi}(T,U)=\int_0^\infty \chi_{\rm eq}(T,\Delta E,U)f(\Delta E,U)d(\Delta E). \tag{6}$$

Note that $\tilde{\chi}(T,U)$ describes the contribution to the equilibrium susceptibility due to all drops which have a definite size. Equations (5) are only valid if each spin is only involved in one of the excitations. This is a reasonable approximation since the magnetic exciting field is small and the perturbation it can provoke in the magnetization is much smaller than the saturation magnetization at high fields. These expressions are valid at any temperature.

Let us consider some simplifying conditions. On one hand, from Eq. (3) we know that the equilibrium susceptibility is practically negligible above $\Delta E=4k_BT$. On the other, if the distribution $f(\Delta E,U)$ is a smooth function of ΔE in the temperature region of interest, as argued above, we can consider $f(\Delta E,U)\approx f(0,U)$ and we can remove it from the integrand. The integral in Eq. (6) can be calculated analytically, and $\tilde{\chi}(T,U)$ becomes

$$\tilde{\chi}\cong\frac{2}{3}\,V\rho^2\mu^2 f(0,U), \tag{7}$$

which is independent of temperature.

Some simple expressions for χ' and χ'' can be obtained by substituting Eq. (7) in Eqs. (5), taking $q(U,\omega)=1/[1+(\omega\tau)^2]$ as a step function at $U=U_c$ and considering that $r(U,\omega)=q\omega\tau$ is nonzero only in the $\pm k_BT$ energy region around $U=U_c$:

$$\chi'(T,\omega)\cong\chi_{\rm rev}+\frac{2}{3}\,\rho^2\mu^2\int_0^{U_c}V(U)f(0,U)dU, \tag{8a}$$

$$\chi''(T,\omega)\cong\frac{\pi}{3}\,k_BT^*\rho^2\mu^2V(U_c)f(0,U_c), \tag{8b}$$

where $U_c=k_BT^*\ln(1/\omega\tau_0)$. The approximations introduced give as a result that χ'' maps out the function $V(U)f(0,U)$, with a resolution window of $2k_BT$. Besides, it is simple to verify that the ''$\pi/2$ rule'' [Eq. (1)] follows easily from Eqs. (8).

The temperature dependence of χ' and χ'' should reflect the relaxation mechanism which dominates at any given temperature:

(i) If the relaxation mechanism involves *thermal activated excitations*, $T^*=T$, then τ follows an Arrhenius law and increases exponentially for decreasing temperature [see Eq. (2)]. In that case, the progressive blocking of the irreversible processes leads, for decreasing temperature, to a decrease of χ' and a linear dependence of χ'' on temperature. In the $T\to0$ limit χ'' should be zero, while χ' should be equal to $\chi_{\rm rev}$.

(ii) If the flipping process takes place by means of QTM (phonon assisted or otherwise), T^* is a constant and τ becomes temperature independent. In that case, the temperature dependence of χ' and χ'' are due to $\tilde{\chi}(T,U)$ only. The temperature dependences of χ shows two interesting limits: (a) if $\Delta E\ll k_BT$ for all excitations then χ would follow a Curie law, identical to the equilibrium susceptibility of independent spins; (b) if the ideal condition $f(\Delta E,U)=$const is fulfilled in a broad range of ΔE (from $\Delta E=0$ to $\Delta E\gg k_BT$), as is our case, χ would be temperature independent. In the real system any small temperature dependence of χ' and χ'' reflects the actual dependence of $f(\Delta E,U)$ on ΔE near $\Delta E=0$.

We now assume that the dominant mechanism is thermally activated flipping above T_Q and QTM below this temperature. According to our previous discussion χ' and χ'' must decrease with decreasing temperature for $T>T_Q$ and become almost temperature independent [as much as $\tilde{\chi}(T,U)$] for $T<T_Q$. It is important to remark again that only the low-energy excitations contribute significantly to the susceptibility.

C. Comparison with experiments, discussion and conclusions

χ' and χ'' calculated with Eqs. (8) can be compared directly to the experimental results. The thermal activated region is identified by its linear temperature dependence above 4 K for χ', and 3 K for χ''. In turn, the QTM relaxation temperature region is identified by the ''plateau'' below those temperatures [see Figs. 4(a) and 4(b)]. We determine the crossover temperature more reliably from the χ'' data ($T_Q\approx3$ K), since this component is more directly related to the relaxation process [see Eq. (8b)]. It is considerably lower than $T_Q=8$ K, as determined from magnetic viscosity measurements. The difference between both values of T_Q can be explained, at least qualitatively, as follows: The resonant tunneling mechanism is blocked (not effective) when $\Delta E\gg\Delta\hat{E}$, where $\Delta\hat{E}$ is the tunnel splitting of the ground state in the resonant condition, i.e., assuming $\Delta E=0$. Therefore, the most likely alternative is MQT assisted by phonons.[15,20,21] Now, the phonon-assisted tunneling transition rate $1/\tau$ depends on $\Delta E(1/\tau\propto\Delta E^3)$ and, consequently, T_Q is also an increasing function of ΔE. This point, which had been missed in Ref. 13, allows us to understand the observation that T_Q obtained from the susceptibility measurements may be lower than that determined from the magnetic viscosity. Indeed, the ac susceptibility is sensitive just to the low-lying energy excitations, in contrast to the magnetic viscosity, where all the excitations are involved. Then, the average value of ΔE is small in the ac susceptibility experiments and,

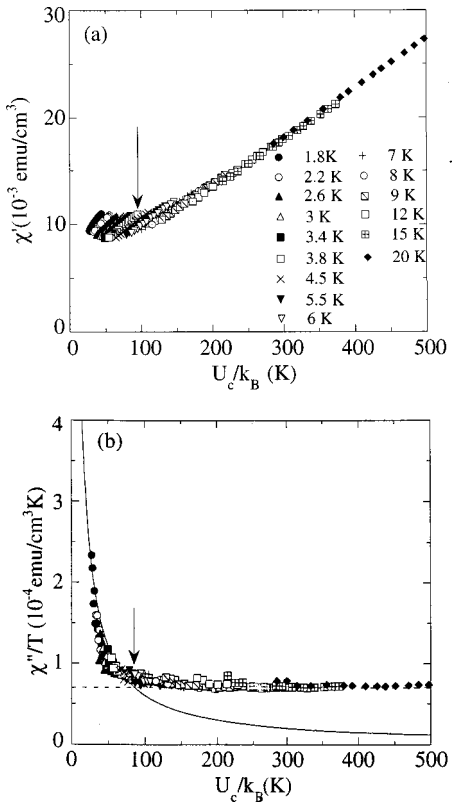

FIG. 8. Scaling of χ' as a function of the variable U_c/k_B (see text) of χ' (a) and χ''/T (b) measured at different temperatures and frequencies, for a common $\tau_0=10^{-10}$ s. In (b), (——), corresponds to U_c^{-1}, which intersects the experimental χ'' data at $U_c\approx 80$ K, while (--) depicts the theoretical prediction [see Eq. (8b)] for thermal activated relaxation when $Vf(0,U)=$const. The vertical arrow in (a) indicates the crossover temperature at which the scaling property breaks down ($T_Q\approx 3.8$ K). In (b), the arrow indicates the activation energy of the excitations which flips due to tunneling.

correspondingly, the observed crossover takes place at a lower temperature than in viscosity measurements.

The imaginary component χ'' contains information about the low-energy excitation distribution function $f(0,U)$. In the classical relaxation region, $T>T_Q$, χ''/T is approximately constant [see Fig. 8(b)], which means that, according to Eq. (8b), $Vf(0,U)$ is nearly independent of U. In contrast, below T_Q, Eq. (8b) predicts that $\chi''/T\propto U_c^{-1}$; i.e., nearly constant but not completely, as is observed experimentally [see Fig. 8(b)]. However, the discrepancy is less than 3%, so we believe that the approximation $f(\Delta E,U)\approx f(0,U)=$const is valid for the present a-Tb_2Fe compound. Thus, the crossover from the classic to the quantum regime takes place when $U_c=80$ K, as determined from χ''.

If we now apply the approximation $f(0,U)=$const above T_Q in Eq. (8a), we obtain that χ' should have a linear dependence on T and on $\ln(\omega)$, in excellent agreement with the experiments [see Figs. 3(a), 4(a), and 5]. Moreover, by extrapolation of this linear dependence down to $T=0$ K the value $\chi'(0)\approx 6.6\times 10^{-3}$ emu/cm^3 is deduced. This yields a good estimate of $\chi_{\rm rev}$ at all frequencies measured. This term corresponds to a reversible rotation of the spins within an energy well, without jumping to another well. It is difficult to compare this result with a theoretical prediction since detailed calculations exist only for the weak anisotropy case, that is, when $D/J<1$.[22] In contrast, in the present case of strong anisotropy the excursions of the spins relative to their preferent direction are expected to be small, then we may approximate $\chi_{\rm rev}$ by[23]

$$\chi_{\rm rev}\approx\frac{\rho\mu}{2H_k}, \tag{9}$$

where $H_k\approx 95$ kOe is the effective anisotropy field and $\rho\mu=1.5\times 10^3$ emu/cm^3 is the saturation magnetization.[5] Substituting these values we obtain $\chi_{\rm rev}\approx 7.9\times 10^{-3}$ emu/cm^3, in very good agreement with the experimental value.

From Fig. 6 we concluded that the ''$\pi/2$ rule [Eq. (1)]'' between χ' and χ'' is well satisfied. This fact reinforces the validity of Eqs. (8) and all approximations therein. More important, above T_Q, Eqs. (8) predict that χ' and χ''/T are functions of the scaling variable $T\ \ln(1/\omega\tau_0)$. Indeed, the χ' and χ''/T experimental data measured at different temperatures merge on a single line for $T>3$ K; i.e., the scaling relation is verified with the common value of $\tau_0\approx 10^{-10}$ s, as can be seen in Fig. 8(a). The loss of the scaling law occurs at the value $U_c\approx 100\pm 50$ K [see Fig. 8(a)].

On the contrary, it is not possible to scale the data below $\approx$3.5 K to the same line [Fig. 8(a)]. We can get further insight on the loss of the scaling property below 3 K by inspecting the frequency dependence of the isotherms depicted in Fig. 5. There it can be seen that χ' decreases for increasing ω and decreasing T, for $T>3$ K. However, for $T<3$ K, χ' still decreases for increasing ω but increases for a lower T [see Fig. 3(a)], thus the trend is inverted. Since the verification of the scaling condition below T_Q would imply that the trend be maintained, its inversion implies that the scaling condition is not obeyed. The violation of the scaling rule of the susceptibility below T_Q indicates that thermal activation is not the predominant relaxation mechanism. Thus, there is a definite crossover to a different relaxation regime below T_Q.

We note that the value $\tau_0=10^{-10}$ s obtained from the scaling is three orders of magnitude larger than that deduced in a similar analysis of the magnetic viscosity data.[24] This remarkable difference may be due to the fact that the magnetic susceptibility technique we have employed is sensitive to spin drops that interact weakly with their neighbors; i.e., for excitation energies ΔE very small (see Sec. IV A), in contrast to magnetic viscosity which is sensitive to larger excitations which involve larger interactions. It is known that interaction with other spins leads to an effective τ_0 value shorter than that for free spins when the Arrhenius law is applied to fit the data.[25] Thus we may expect that the weaker the interaction is the longer the τ_0 that may be found, as observed.

The number of spins that flip by tunneling effect may be then estimated equating the value of U_c at T_Q (which ranges from 80 to 150 K, as determined from χ' or χ'', respectively) with KV, taking $K=9\times 10^7$ erg/cm^3 from the Ref. 5, and solving for V. We obtain an effective drop diameter of $\sim 7\pm 2$ Å, and since the interatomic distance is $a\approx 4.5$ Å, this means that each excitation involves at most one spin and its nearest neighbors, that is, $n\approx 11$. Such a small number of correlated spins is in agreement with the fact that, in our limit of $D/J\gg 1$, spin-wave excitations are little populated

since the magnon spectrum becomes unstable for small enough k vectors.[9] The activated volumes deduced from susceptibility data are one order of magnitude lower than those determined from magnetic viscosity ($n \sim 100$),[5] probably because the characteristic time in our experiment is much shorter (by 2–6 orders of magnitude) and is sensitive only to the low-lying energy excitations. In conclusion, we are observing processes involving tunneling at an intermediate microscopic-mesoscopic scale.

In conclusion, we have proven that there exist low-lying energy excitations which allow the spin to approach equilibrium even at temperatures as low as $T = 0.10$ K. The observed low-temperature susceptibility can be explained within a model in which MQT between two quasidegenerate states of minimum energy is the predominant relaxation channel. The main interest of this work is that it arrives at the same conclusions as those derived from magnetic viscosity measurements,[5] with a different technique and time window. Consequently, the existence of MQT in a-Tb_2Fe is strongly supported.

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