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LETTER TO THE EDITOR

ON THE NON-THERMAL RELAXATION TIME IN THE MAGNETIC QUANTUM TUNNELING

MARIAN APOSTOL[1]

Centre de Recherches sur les Très Basses Températures, CNRS, BP 166 X, 38042 Grenoble-Cedex 9, France

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The domain-wall concept is employed to derive the relaxation time of the macroscopic quantum tunneling in small magnetic particles as a function of temperature and magnetic field. The result is used to analyze the recently reported experimental data.

The relaxation rate of the magnetization has recently been measured[1,2)] in small, weakly interacting ferromagnetic particles of $Tb_{0.5}Ce_{0.5}Fe_2$ of mean diameter ≈ 15 nm, at low temperatures (from 10 K down to ≈ 50 mK) and for relatively high magnetic fields (≤ 0.6 T). The measurements of the frequency-dependent magnetic susceptibility of small-size (≈ 7 nm) antiferromagnetic particles of a natural protein[3)] revealed a sharp peak at the resonance frequency $\approx 10^3$ kHz, at very low temperatures (≈ 30 mK) and low magnetic fields. Similar results on the relaxation time have also been reported[4,5)] in Fe/Sm magnetic multilayer systems. These data raise the interesting question of magnetic quantum tunneling over a macroscopic scale[6−9)] in the limit of low temperatures. The non-thermal relaxation of the magnetization, corresponding to the quantum tunneling between two locally-stable states, is described in the present paper by the equivalent picture of

[1] On leave from the Department of Theoretical Physics, Institute of Atomic Physics, Magurele-Bucharest MG-6, PO Box MG-35, Romania.

the domain-wall motion, as to allow for the finite-size effects (domain-wall width and particle-size length scales). Together with the thermal relaxation of the magnetic excitations of the domain wall, this enables one to derive the relaxation time of the magnetization as a function of both the temperature and the magnetic field. The result is used to discuss the experimental data mentioned above.

As soon as the size d of the magnetic particles becomes comparable to the lattice constant a, the magnetization is never saturated across the particle, i.e. there is no single domain over the whole size of the particle[10]. The deviation of the magnetization from its saturated value across the particle size may be viewed as a domain wall, whose width may vary between the extreme value of the lattice constant a (extremely narrow domain wall), as in the case of strong anisotropy and non-linearity of the magnetic interactions[11], and a value which may be much larger than the particle size d, as in the case of very thin particles[3]. Within this picture, the quantum tunneling of the magnetization between two locally-stable states (as, for example, between two axes of easy magnetization) amounts to the motion of the domain wall along a distance of the order of the particle size d. In the presence of the magnetic field the energy of the domain wall is a slowly decreasing function $\omega(H)$ which saturates at high values of the field H, as indicated also by the saturation of the sample magnetization[1-5]. Consequently, above a threshold field H_0, which may be taken as the maximum coercitive field, one may approximate by $\omega(H) \cong \omega(H_0) \equiv \omega_0$. Having the energy fixed in this way one can easily estimate the velocity of the domain wall, as being of the order $v \sim \omega_0 a$, where a is taken for the domain-wall width. Generally, this velocity has to be reduced by the probability p of the domain-wall motion across the particle-size length d, as for including the pinning effects, for example. A free motion of the domain wall corresponds to $p = 1$, while the complete pinning of the domain wall corresponds to $p = 0$. The relaxation time of the magnetization is now readily obtained within this picture, as

$$\tau_0 = d/v \approx d/(ap\omega_0)\,. \tag{1}$$

It corresponds to the non-thermal relaxation (Casimir limit in the phonon physics), and is what one usually computes in the quantum tunneling theory[12-18]; when the (real) space motion of the magnetization is included. One should emphasize at this point that (1) is nothing else but the rewriting of the non-thermal relaxation time of the quantum magnetic tunneling in terms of the domain-wall motion. Pursuing this picture further, one should notice that at lower values of the magnetic field the energy $\omega(H)$ begins to differ sensibly from ω_0. This deviation can be obtained by expanding $\omega(H)$ in powers of $\delta(1/H) = 1/H - 1/H_0$ and retaining, to the first approximation, only the first term of the series,

$$\delta\omega = \omega_1 \delta(1/H) = \omega_1(1/H - 1/H_0)\,, \tag{2}$$

where ω_1 is the derivative of ω_0 with respect to $1/H$ at $H = H_0$. The additional contribution to the energy given by (2) may be viewed as corresponding to the "elementary excitations" of the domain wall, i.e. the excitations associated with the small deviations of the magnetization from its domain-wall equilibrium value. They are delocalized waves, obeying the Bose-Einstein statistics, with a group velocity $v \approx \omega_0 a$ (to the first approximation), and, therefore, with a thermal mean-free path

$$\Lambda \approx a/n_{\rm th} \approx a{\rm e}^{\omega_1 \delta(1/H)T}, \tag{3}$$

where, again, a has been taken for the domain-wall width. The corresponding thermal relaxation time is now easily obtained as

$$\tau_{\rm th} = \Lambda/v \approx \frac{1}{\omega_0}{\rm e}^{\omega_1 \delta(1/H)T}. \tag{4}$$

The usual Matthiessen rule,

$$\frac{1}{\tau} = \frac{1}{\tau_0} + \frac{1}{\tau_{\rm th}}\,, \tag{5}$$

can be employed now, for τ_0 given by (1) and $\tau_{\rm th}$ given by (4), to interpolate between thermal and non-thermal regimes. One obtains, in this way, the relaxation time of the magnetization

$$\ln(1/\tau) = \ln\omega_0 + \ln\left[ap/q + {\rm e}^{-\omega_1\delta(1/H)T}\right], \tag{6}$$

which will be used below to discuss the experimental data of magnetic tunneling.

First, one should notice that $\ln(1/\tau)$ given by (6) corresponds, as a function of $\delta(1/H)$, to a bundle of straight lines for H close to H_0 (low values of $\delta(1/H)$) all of them passing through the fixed point of coordinates $(1/H_0, \ln\omega_0)$, and having the slopes $-\omega_1/T$; this part of $\ln(1/\tau)$ corresponds to the thermal regime (when $ap/d \ll 1$), and is currently obtained in the experiments[1,2,5]. All these thermal straight lines terminate, according to (6), with a horizontal tail $1/\tau = \omega_0 ap/d$ corresponding to $\omega_1\delta(1/H)/T \gg 1$, i.e. at very low temperatures and towards weaker magnetic fields. The presence in the experimental data of this horizontal tail (or, equivalently, a plateau in $1/\tau$) is an indication of non-thermal regime corresponding to the quantum tunneling. The bundle of thermal strength lines reported in Refs. 1 and 2 is fitted by (6) for $1/H_0 = 1.5$ T^{-1}, $\omega_0 = 10^9$ s$^{-1} \cong 10^{-2}$ K and $\omega_1 = 73.5$ KT. A refined analysis of the low-temperature data of Refs. 1, 2 and 5 would enable one to decide whether or not these data exhibit the non-thermal horizontal tail, indicative of magnetic tunneling.

This tail is clearly exhibited by the frequency-dependent magnetic susceptibility reported in Ref. 3. Indeed, the resonance frequency of the magnetic tunneling corresponds to $1/\tau$ given by (6). In the limit of vanishing magnetic field and zero temperature (≈ 30 mK), (6) reduces to the non-thermal contribution $1/\tau_0 = \omega_0 ap/d = 10^3$ kHz. This frequency increases with increasing magnetic field[3], such that, for $H \to \infty$ one obtains from (6) $1/\tau \equiv \omega_0(ap/d + 1)$, a result which should be equated to $\approx 10^4$ kHz from Ref. 3. From these two equations one easily obtains $\omega_0 \simeq 9\times10^3$ kHz and $ap/d \simeq 0.1$. According to the data in Ref. 3, the width of the domain wall in these antiferromagnetic protein particles is larger than the particle size, therefore $a/d \geq 1$, whence one gets $p \leq 0.1$, which is consistent with the probability of this parameter ($0 < p < 1$). Actually, p may be lower, as we underestimated the value of ω_0 by assuming that the resonance frequency 10^4 kHz corresponds to H going to infinity. Nevertheless, one should emphasize that the experimental H-dependence of the resonance frequency reported in Ref. 3 for the temperature $T \approx 30$ mK is quite well followed by the theoretical dependence predicted by (6). The ω_1 parameter in (6) is difficult to be estimated from these experimental data (Fig. 2a in Ref. 3), since (6) is highly sensitive to the magnetic field; in order to get a reliable value of ω_2, the accuracy of the experimental data should be sensibly improved.

In conclusion, one should add two further remarks. First, the relaxation time given by (6) predicts a cross-over temperature

$$T_c = \frac{\omega_1}{\ln(d/ap)}\,\delta(1/H)\,, \tag{7}$$

which depends on the magnetic field and separates the thermal from non-thermal regime. According to (7), the non-thermal behaviour can be obtained at higher temperatures for lower magnetic fields; however, from (6) one can see that in this case the relaxation time is extremely long. In order to have the relaxation time values within the "experimental window" one should look for non-thermal effects at higher magnetic fields, in which case the cross-over temperature is extremely low. In any case, the value of this temperature depends on the parameter values ω_1, d/ap which are specific to each experimental situation. Finally, one remarks that the non-thermal relaxation time τ_0 given by (1) tends to infinity for the bulk sample, i.e. the magnetization gets frozen in this case, as expected.

References

1) C. Paulsen, L. C. Sampaio, B. Barbara, D. Fruchart, A. Marchand, J. L. Tholence and M. Uehara, Phys. Lett. A **161** (1991) 319;

2) C. Paulsen, L. C. Sampaio, B. Barbara, D. Fruchart, A. Marchand, J. L. Tholence and M. Uehara, Phys. Rev. Lett., submit. (1992);

3) D. D. Awschalom, J. F. Smyth, G. Grinstein, D. P. DiVicenzo and D. Loss, Phys. Rev. Lett. **68** (1992) 3092;

4) L. L. Balcells, X. X. Zhang, F. Badia, J. M. Ruiz, C. Ferrate and J. Tejada, J. Mag. Mag. Mat. **109** (1992) L 159;

5) X. X. Zhang, L. L. Balcells, J. M. Ruiz, J. L. Tholence, B. Barbara and J. Tejada, J. Phys. Cond. Mat. **4** (1992) L 163;

6) A. Caldeira and A. J. Leggett, Ann. Phys. **149** (1983) 374;

7) A. J. Leggett, S. Chakravarty, A. T. Dorsey, M. A. P. Fisher, A. Garg and W. Zwerger, Rev. Mod. Phys. **59** (1987) 1;

8) E. M. Chudnovskry and L. Gunther, Phys. Rev. Lett. **60** (1988) 661;

9) E. M. Chudnovskry and L. Gunther, Phys. Rev. B **37** (1988) 9455;

10) A. Aharoni, IEEE Trans. Mag. **27** (1991) 4775;
11) J. J. Niez, P. G. Averbuch, J. C. Fanton and J. Lajzerowicz, J. Phys. C **11** (1978) 3715;
12) B. Barbara, G. Fillion, D. Gignoux and R. Lemaire, Solid State Commun. **10** (1972) 1149;
13) M. Enz and R. Schilling, J. Phys. C **19** (1986) L 711;
14) A. Garg and G. H. Kim, Phys. Rev. Lett. **63** (1989) 2512;
15) I. V. Krine and O. B. Zaslavskii, J. Phys. Cond. Mat. **2** (1990) 9457;
16) B. Barbara and E. M. Chudnovskry, Phys. Lett. A **145** (1990) 205;
17) P. C. E. Stamp, Phys. Rev. Lett. **66** (1991) 2802;
18) A. Garg and G. H. Kim, Phys. Rev. B **45** (1992) 12921.

O TOPLINSKOJ VODLJIVOSTI IDEALNOG KRISTALA

MARIAN APOSTOL

Centre de Recherches sur les Très Basses Températures, CNRS, BP 166 X, 38042 Grenoble-Cedex 9, France

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Koristeći koncept domenskih zidova određeno je relaksaciono vrijeme za makroskopsko kvantno tuneliranje u malim magnetskim česticama kao funkcija temperature i magnetskog polja. Rezultati su iskorišteni za analizu najnovijih rezultata mjerenja.