PHYSICAL REVIEW B VOLUME 50, NUMBER 17 1 NOVEMBER 1994-I

Time decay of the remanent magnetization in TDAE-C_{60}

R. Blinc, P. Cevc, D. Arčon, D. Mihailovič, and P. Venturini
J. Stefan Institute, University of Ljubljana, Ljubljana, Slovenia
(Received 7 March 1994; revised manuscript received 31 May 1994)

The time decay of the isothermal magnetization of tetrakis-dimethylaminoethylene (TDAE)-C_{60} after switching off a small external magnetic field has been studied between 20 and 4 K. Below T_c a remanent magnetization M_0—representing the fraction of the frozen out spins—that becomes unobservably small above 12 K has been found. The relaxing magnetization fraction component M^*, on the other hand, increases as T_c is approached from below. The time decay of the magnetization is rather slow below T_c and can be described by a stretched exponential form $M(t)=M_0+M^*\exp[-(t/\tau)^\alpha]$. The exponent α decreases from 0.14 at 4 K to 0.08 at 12 K. This shows that the spectrum of relaxation times slowly changes with temperature and becomes very wide as T_c is approached from below. No slowly relaxing component is found above T_c. The above features are characteristic of a spin-glass-like freezing process.

The nature of the "soft" ferromagnetic transition reported to occur in the C_{60} based charge-transfer compound TDAE-C_{60} (Refs. 1 and 2) (TDAE stands for tetrakis dimethylamino ethylene) at $T_c \approx 14.5$ K has attracted a great deal of interest.[3–7] The absence of a hysteresis in the magnetization curve and the unusual temperature dependence of the magnetic susceptibility[1,2] are not typical for ferromagnetic systems. The appearance of a broad and intense electron spin resonance (ESR) signal[4–8] below T_c suggests that at least some of the unpaired spins are ferromagnetically correlated. The fact that the local field below T_c—as seen from the shift in the center of the ESR line—is much smaller than expected for a ferromagnet with $T_c \approx 14.5$ K opens the possibility that we are dealing with a spin-glass-like state[9] or finite-size spin clusters rather than with bulk ferromagnetism.

To discriminate between these two possibilities we decided to check the dynamic magnetic properties of TDAE-C_{60} by measuring the relaxation of the isothermal magnetization after turning off the applied magnetic field. The time decay of the remanent magnetization in a spin glass[9] is known to differ dramatically from that of a ferromagnet. This is so as spin glasses and spin-glass-like mictomagnetic systems[9] are characterized below T_c by a wide distribution of relaxation times and a slow time decay of the remanent magnetization whereas the relaxation dynamics in ferromagnets is much faster. In spin glasses above T_c the entire magnetization responds rapidly to a change in the magnetic field whereas below T_c a significant fraction of the magnetization responds extremely slowly.[9] This phenomenon is absent in ferromagnets.

The experiment was performed by measuring the intensity of the electron spin resonance signal after switching off the applied external magnetic field. The ESR signal intensity is namely proportional to the static equilibrium magnetization $M^+ \propto M_0 \propto H_0$ so that the time decay of the ESR signal after the external magnetic field has been switched off measures the time decay of the remanent magnetization.[10] The TDAE-C_{60} sample was slowly cooled in zero magnetic field to a given temperature. Then a magnetic-field sweep was applied and the resulting magnetization was measured at a frequency $\omega_L/2\pi = 15$ MHz by observing the ESR signal.[8] The field sweep was stopped at 10 G. After equilibrium has been reached the external magnetic field has been switched off and the ESR signal at $\omega_L/2\pi = 15$ MHz was measured as a function of time.

The resulting isothermal magnetization decay curves have been found to be rather different above and below $T_c \approx 14.5$ K. After correcting for the finite decay time of the circuit providing the applied external magnetic field, no time persistent remanent magnetization or slowly relaxing magnetic component could be observed above T_c on the (1–500) s time scale of this experiment (Fig. 1). Below T_c, on the

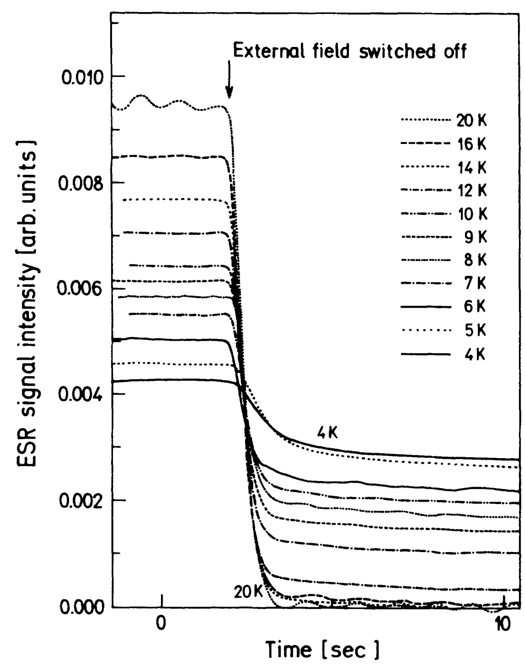

FIG. 1. Time dependence of the magnetization in TDAE-C_{60} after the external magnetic field has been switched off. The decay curves are shown for different temperatures above and below $T_c = 14.5$ K.

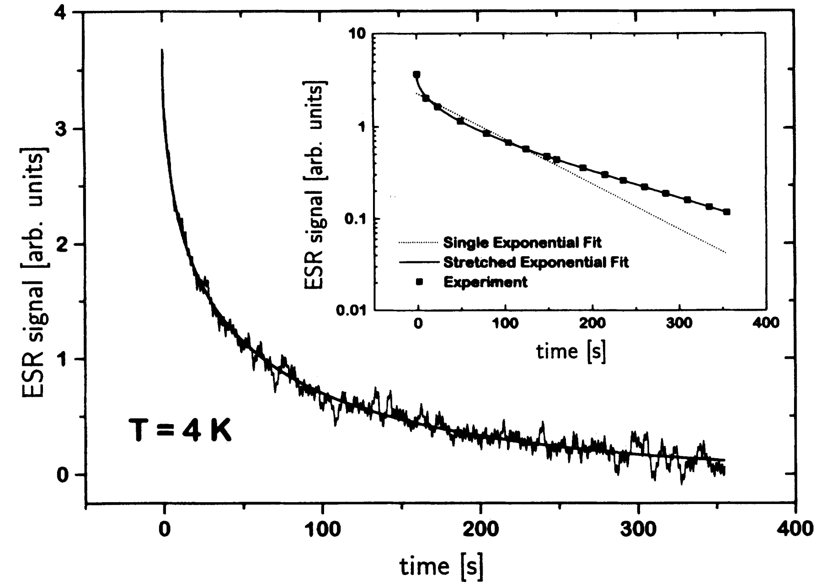

FIG. 2. Comparison between the observed time decay of the ESR signal intensity at $T=4$ K and $H=0$ after switching off the external magnetic field and the stretched exponential fit (solid line). The insert shows the comparison between the stretched exponential fit, the single exponential fit, and the digitalized experimental points.

other hand, both a slowly relaxing magnetization component and a remanent magnetization have been observed (Fig. 1). The magnetization decay curves cannot be described by a single exponential. They are, however, reasonably well represented by a stretched exponential type decay (Fig. 2),

$$M(t)=M_0+M^*\exp[-(t/\tau)^\alpha]. \quad (1)$$

The remanent magnetization M_0—representing the frozen out spin clusters—was found to decrease with increasing temperature and was too small to be observable above 12 K.

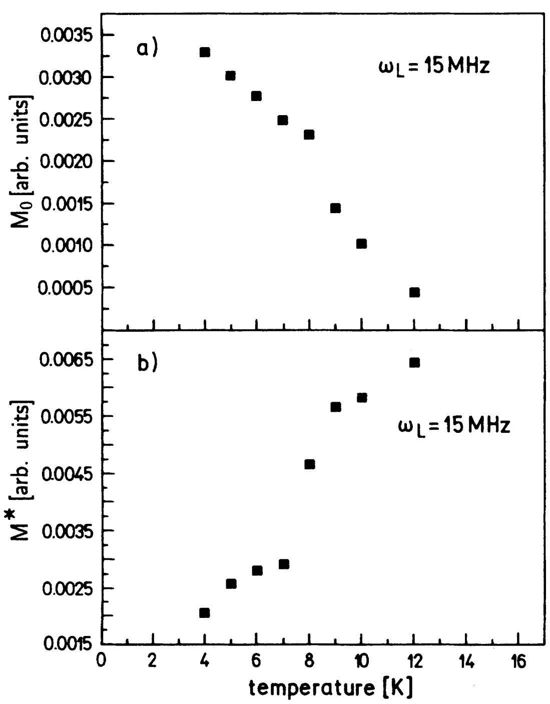

FIG. 3. Temperature dependences of (a) the remanent magnetization M_0 and (b) the slowly relaxing magnetization fraction M^* in TDAE-C_{60}.

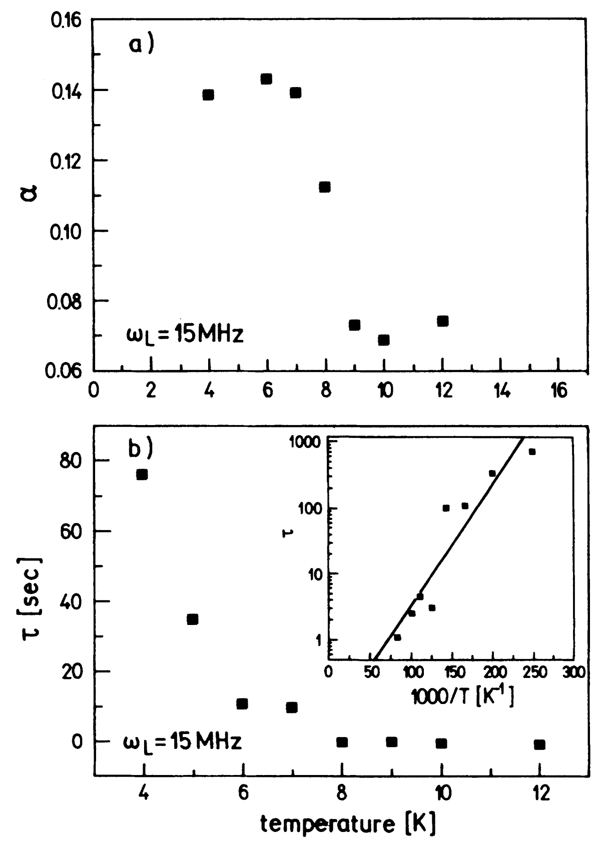

FIG. 4. Temperature dependences of (a) the stretched exponential exponent α and (b) correlation time parameter τ in TDAE-C_{60}. The inset (b) shows τ versus $10^3/T$.

Between 4 and 12 K it decreases by a factor of 6 or 7. The "unfrozen," i.e., relaxing, part of the magnetization M^*, on the other hand, increases with increasing temperature in the same interval by a factor of about 3 (Fig. 3). The difference in the temperature change of the magnitudes of M_0 and M^* shows that even below T_c a part of the magnetization relaxes too fast—i.e., $\tau < 10^{-2}$ s—to be observable in this experiment. Above T_c only the fast relaxing fraction persists.

The correlation time parameter τ—which is thermally activated—increases from about 1 s at 12 K to about 75 s at 4 K. In the same interval the stretched exponential exponent α increases from $\alpha=0.08$ to 0.14 (Fig. 4). The fact that the value of α decreases on approaching T_c from below shows that the spectrum of relaxation rates,

$$M(\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty} M(t)e^{-i\omega t}dt, \quad (2)$$

slowly changes with temperature and becomes very wide as $T \rightarrow T_c$.

The above dynamic features are characteristic of a spin-glass-like freezing process. A stretched exponential time decay of the remanenent magnetization was in fact predicted by a fractal cluster model to occur in spin glasses below T_c.[11] In contrast, in systems undergoing normal phase transitions of second-order-like classical ferromagnets or antiferromagnets, a rapid variation of a well-defined relaxation time occurs over many orders of magnitude as T_c is approached from below.

[1] P. M. Allemand, K. C. Khemani, A. Koch, F. Wudl, K. Hoczer, S. Donovan, G. Gruner, and J. D. Thompson, Science **253**, 301 (1991).

[2] P. W. Stephens, D. Cox, J. W. Lauher, L. Mihaly, J. B. Miley, P. M. Allemand, A. Hirsch, K. Holczer, Q. Li, J. D. Thompson, and F. Wudl, Nature **355**, 331 (1992).

[3] R. Blinc, J. Dolinšek, D. Arčon, D. Mihailovič, and P. Venturini, Solid State Commun. **89**, 487 (1994).

[4] P. Venturini, D. Mihailovič, R. Blinc, P. Cevc, J. Dolinšek, D. Abramič, B. Zalar, H. Oshio, P. M. Allemand, A. Hirsch, and F. Wudl, Int. J. Mod. Phys. B **6**, 3947 (1992).

[5] R. Seshadri, A. Rastogi, S. V. Bhat, S. Ramasesha, and C. N. R. Rao, Solid State Commun. **85**, 971 (1993).

[6] K. Tanaka, A. A. Zakhidov, K. Yoshizava, K. Okahara, T. Yamabe, and K. Yakushi, Phys. Rev. B **47**, 7554 (1993).

[7] K. Tanaka, A. A. Zakhidov, K. Yoshizawa, K. Okahara, T. Yamabe, K. Yakushi, K. Kikuchi, S. Suzuki, I. Ikemoto, and Y. Achiba, Phys. Lett. A **164**, 221 (1992).

[8] P. Cevc, R. Blinc, V. Eržen, D. Arčon, B. Zalar, D. Mihailovič, and P. Venturini, Solid State Commun. **90**, 543 (1994).

[9] K. Binder and A. P. Young, Rev. Mod. Phys. **58**, 801 (1986).

[10] A. Abragam, *The Principles of Nuclear Magnetism* (Clarendon, Oxford, 1961).

[11] A. P. Malozemoff and B. Barbara, J. Appl. Phys. **57**, 3410 (1985).