LOW TEMPERATURE SPIN DYNAMICS IN TMMC FROM NUCLEAR SPIN-LATTICE RELAXATION RATE

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Measurements of proton spin-lattice relaxation rate in $(CH)_4NMnCl_4$ (TMMC) in the temperature range (1.2–4.2 K) and for different magnetic fields (0.085–2 T) for both $H \parallel c$ and $H \perp c$ are presented and compared with theoretical calculations based on Villain theory for damped paramagnons in 1D magnets. The implication of the disagreement found between theory and experiment is briefly discussed.

The nuclear spin-lattice relaxation rate, T_1^{-1} of protons and other nuclei in one-dimensional (1D) antiferromagnet $(CH_3)_4NMnCl_4$ (TMMC) probes the low frequency part of the spectral density of the electronic spin fluctuations. The behavior of T_1^{-1} in the temperature interval 5–300 K, where the spin dynamics is dominated by the onset of short range order correlations, has been satisfactorily described [1,2] in terms of an extension of Moriya's magnetic relaxation theory to the linear chain system using the exact results for static two-spin-correlation functions.

At low temperature e.g. $T < 5$ K, but still above the 3D transition temperature, TMMC is characterized by a pronounced 1D short range order and the spin dynamics is dominated by damped paramagnons and soliton-like excitations [3]: both are very sensitive to the magnitude of the external magnetic field and its direction with respect to the chain axis c. The results of T_1^{-1} in high fields (H > 2 T and $H \perp c$) were shown [4] to be consistent with a nuclear relaxation rate driven by the interaction with an ideal soliton gas. On the other hand, measurements in low magnetic fields (H < 2 T, $H \parallel c$ and $H \perp c$) reported previously were not interpreted [5]. We present here new low field data aimed at gaining information about the spin dynamics in TMMC for a magnetic field range and for an orientation for which the soliton's description is not directly applicable.

The results of proton spin-lattice relaxation rate are shown in figs. 1a, b and figs. 2a, b.

An explanation of the results in terms of nuclear relaxation driven by interaction with solitons is unlikely. In fact for fields H < 1 T the width of the π-solitons in TMMC is expected to be so large as to invalidate any description of the spin-dynamics in terms of a dilute gas of solitons [6]. Furthermore, for $H \parallel c$, one does not expect the presence of solitons, at least of the Sine–Gordon type for which the theory has been developed in TMMC [6,7]. We thus turn to the description for the relaxation rate in terms of interaction with the paramagnons. The two-magnon and three-magnon relaxation process can be ruled out because they yield a temperature dependence of T_1 opposite [8] to the one observed experimentally.

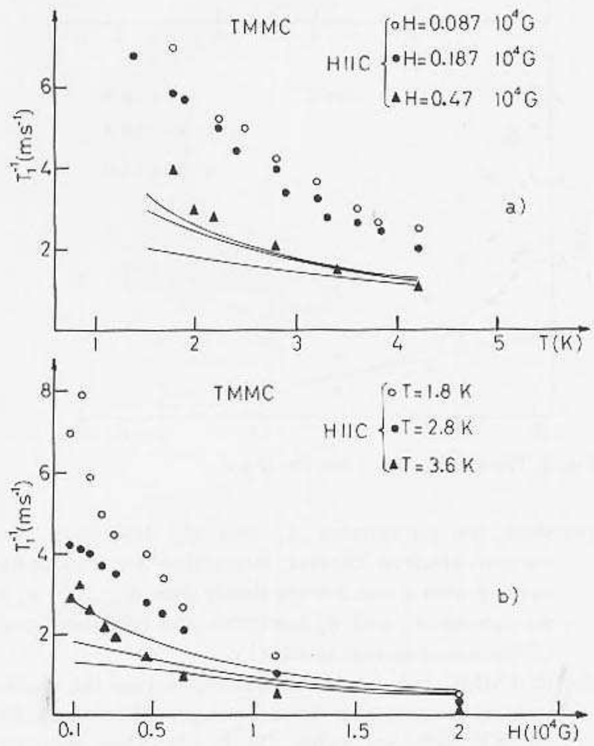

Fig. 1. Proton relaxation rate for $H \parallel c$: (a) T_1^{-1} vs. T; (b) T_1^{-1} vs. H. The solid lines are theoretical best fit using eqs. (2), (3), (4) and normalizing the curve to the lowest experimental point.

The direct one-magnon process remains the only effective mechanism in presence of almost gapless and overdamped spin-waves.

By using the weak collision approach we can write [1]:

$$\frac{1}{T_1} = \sum_q \left[A_q S_{\pm}(q, \omega_L) + B_q S_z(q, \omega_L) \right], \tag{1}$$

where ω_L is the nuclear Larmor frequency and ±, z refer to the spin components perpendicular and parallel to the external magnetic field, respectively.

We proceed now to make some simplifying assumptions:

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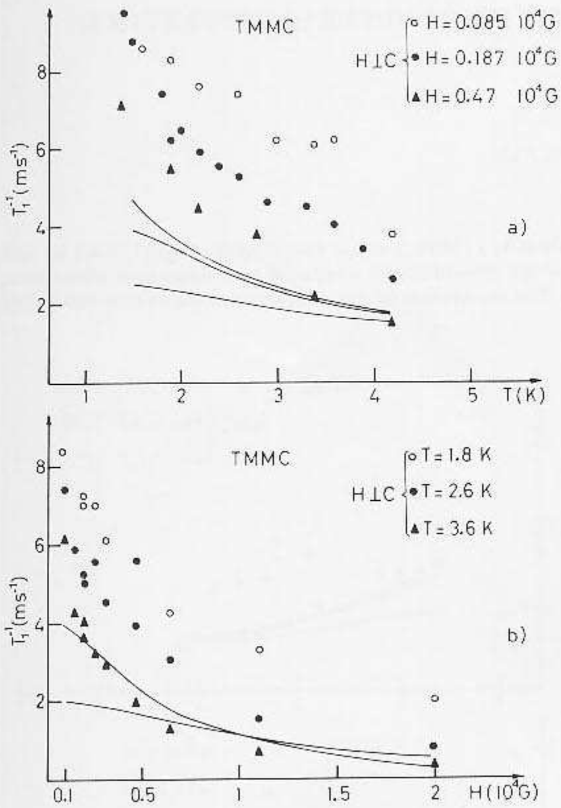

Fig. 2. The same as fig. 1 but for $\boldsymbol{H} \perp \boldsymbol{c}$.

(a) since the parameters A_q and B_q describing the nuclear–electron dipolar interaction are functions varying with q much more slowly than $S_{\pm, z}(q, \omega_L)$ we can set A_q and B_q constants and take them out of the q-summation in eq. (1);

(b) in TMMC the dipolar interaction within the chain induces a cross-over from isotropic Heisenberg to an XY behavior below 20 K [2]. Thus we can neglect in eq. (1) the fluctuations for the spin out of the plane perpendicular to the c-axis;

(c) we write for $S(q, \omega) = S(q, 0)\, f_q(\omega)$ where the first factor is proportional to the static susceptibility and the second is a relaxational function whose shape depends on the model adoped: in general it is a Lorentzian, centered at $\omega = 0$ for diffusive modes or overdamped paramagnons and at $\omega = \omega(q)$ for underdamped ones.

According to Villain's theory [9], well defined paramagnons are present in TMMC only for $q > K$, where K is the inverse correlation length. The resulting expressions of T_1^{-1} which we use to fit the data are (only the leading term is explicitated):

$$\frac{1}{T_1} \propto \int_0^K \mathrm{d}q \frac{K}{K^2 + q^2} \frac{\Gamma}{\Gamma^2 + (cq + \omega_e)^2} + \ldots \quad (\boldsymbol{H} \parallel \boldsymbol{c}), \tag{2}$$

$$\frac{1}{T_1} \propto m_\parallel \int_0^{K_\parallel} \mathrm{d}q \frac{K_\parallel}{K_\parallel^2 + q^2} \frac{\Gamma_\parallel}{\Gamma_\parallel^2 + (cq)^2 + \omega_e^2}$$

$$+ m_\perp \int_0^{K_\perp} \mathrm{d}q \frac{K_\perp}{K_\perp^2 + q^2} \frac{\Gamma_\perp}{\Gamma_\perp^2 + (cq^2) + \omega_e^2} \quad (\boldsymbol{H} \perp \boldsymbol{c}). \tag{3}$$

For $\boldsymbol{H} \parallel \boldsymbol{c}$ the dispersion curve for paramagnons is gapless i.e. $\omega(q) = cq$ while for $\boldsymbol{H} \perp \boldsymbol{c}$ a gap is introduced by the magnetic field i.e. $\omega(q)^2 = (cq)^2 + \omega_e^2$ ($\omega_e = \gamma_e H = 1.34 H$). The temperature and field dependence of K, $K_\parallel$ and $K_\perp$ (expressed in Kelvin) are given for TMMC by [10]:

$$K = \frac{K_B T}{4JS^2 a_0} = 1.24\ T$$

$$K_\perp = K(1 - H^2/T^2) \quad K_\parallel = K(1 + H^2/T^2) \tag{4}$$

and $m_\perp = 1 + H^2/2T^2$, $m_\parallel = 1 - H^2/2T^2$, where the field is in kelvin * and $\parallel$, $\perp$ refer to the direction with respect to H. For the damping of the paramagnons Villain's theory gives $\Gamma = cK$ (and similarly for $\Gamma_{\parallel, \perp}$), c being the paramagnons' speed. This relation seems to be supported by neutron scattering results at H = 0 [11].

The theoretical behaviors obtained from eqs. (2), (3), (4) are compared with the experiments in figs. 1 and 2. No consistent fit of the data can be obtained using a linear relationship between Γ and K, the best fit beeing obtained for $\Gamma = \frac{1}{4} cK$.

For $\boldsymbol{H} \parallel \boldsymbol{c}$ the strong field dependence of T_1^{-1} could be due to the presence of a narrow central peak component with $K \gg \Gamma \lesssim \omega_e$.

For $\boldsymbol{H} \perp \boldsymbol{c}$ one should conclude that the relationship between Γ and K is given by a general dynamic-scaling law $\Gamma(H)/\Gamma(0) \sim [K(H)/K(0)]^z$ where $z = 1$ for $H = 0$ [11] while for $H \neq 0$, $z > 1$. It is likely that the field dependence of z may be related to the existence of broad solitons not directly responsible for the nuclear relaxation.

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* In all the equations the field H expressed in kelvin is related to the field H in tesla by the formula $H = M_B S \mathrm{H} / 2 K_B$.

[1] D. Hone, C. Scherer and F. Borsa, Phys. Rev. B9 (1974) 965.
[2] D. Hone and A. Pires, Phys. Rev. B15 (1977) 323.
[3] F. Borsa, M.G. Pini, A. Rettori and V. Tognetti, in: Magnetic Excitations and Fluctuations eds. S.W. Lovesey, U. Balucani, F. Borsa and V. Tognetti (Springer, Berlin, 1984) p. 2.
[4] J.P. Boucher and J.P. Renard, Phys. Rev. Lett. 45 (1980) 486.
[5] F. Borsa, J.P. Boucher and J.C. Jug, J. de Phys. 39 (1978) C6-724.
[6] K.M. Leung, D. Hone, D.L. Mills, P.S. Riseborough and S.E. Trullinger, Phys. Rev. B21 (1980) 4017.
[7] H.J. Mikeska, J. Phys. C13 (1980) 2913.
[8] T. Goyo, Phys. Rev. B28 (1983) 6347.
[9] J. Villain, J. de Phys. 35 (1974) 27.
[10] J.M. Loveluck, J. Phys. C12 (1979) 4251. F. Borsa, J.B. Boucher and J. Villain, J. Appl. Phys. 3 (1978) 49.
[11] M. Steiner, J. Villain and C.B. Windsor, Advan. Phys. 25 (1976) 87.