EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

CERN-EP/82-13
2 February 1982

INVESTIGATION OF THE CRITICAL RELAXATION IN MnF_2 BY MUON SPIN ROTATION*)

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ABSTRACT

The magnetic relaxation in MnF_2 has been studied by means of Muon Spin Rotation. An increase was found close to T_N in the damping of the precession signal from positive muons implanted in a single-crystal sample. This is attributed to the critical slowing down of the antiferromagnetic spin fluctuations. An orientation-dependent shift in the signal frequency was also detected. The location of the muon in the lattice is tentatively determined.

Submitted to Solid State Communications

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*) Work supported by the Consiglio Nazionale delle Ricerche, Gruppo Nazionale di Struttura della Materia.

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We present here a study of a single crystal of MnF_2 by the Muon Spin Rotation (μSR) technique. MnF_2 is an antiferromagnetic insulator, with a critical temperature of 67.3 K, which has been extensively subjected to NMR [1,2] and neutron-scattering [3] measurements. Through these techniques both the static and the dynamic magnetic properties of the compound have been determined: the data agree very well with the Heisenberg model if a relatively modest anisotropy is introduced.

Our main aim in looking at the behaviour of implanted positive muons in this crystal was to determine the effect of the critical phenomena on this probe, which is also of inherent interest as a light isotope of hydrogen. As the muon, in principle, could reside at an interstitial site without forming a bond to the Mn atoms one can hope to see a small coupling to the critical electronic spin fluctuations around T_N, which would enable us to follow their divergence in a very large range of μSR linewidths ($0.05\ \mu s^{-1} \lesssim \lambda \lesssim 10\ \mu s^{-1}$). Recent measurements [4] on MnSi have already shown evidence of the usefulness of the technique in detecting similar phenomena.

The single crystal on which our measurements were made was grown at Cristaltec - Grenoble by the Bridgeman method and has the shape more or less of a cylinder (25 mm diameter by 35 mm height).

The μSR spectra were collected at the CJ1 beam line of the CERN Synchro-cyclotron in the conventional transverse field spectrometer described elsewhere [5] (with respect to the set-up used in ref. [5], the only noteworthy difference is the adoption of a new electromagnet, with a homogeneity of one part in 10^5 over the sample volume, provided by the Rutherford Appleton Laboratory). The sample was kept in a He flow cryostat -- belonging to the Polarized Target Group of CERN -- and stabilized to the chosen temperature within ±0.1 degree (this is actually an upper limit for the error bar). The free muon precession was observed in applied magnetic fields of 150 G and 1500 G and fitted to a Lorentzian decaying wave to yield the amplitude, the Lorentzian linewidth, the exact frequency, and the phase of the oscillation.

The results obtained for the amplitude parameter are shown in fig. 1. Very evident is a sudden drop below T_N: the value found for $T < T_N$ is completely accounted for by the fraction of muons stopping in the cryostat walls and in the sample holder, so that these same measurements were used to determine the "background" to be subtracted in the higher temperature runs. No other signal was detected in a Fourier transform of the μSR time-differential histogram at any temperature. The amplitude measured at $T > T_N$, fairly constant over the whole range, accounts for only 36% of the muons stopped in the sample. The missing fraction can be tentatively attributed to muonium formation, even if its characteristic frequencies do not show up: the coupling to both the ^{19}F nuclear spins and the Mn electrons, even in the limit of exchange narrowing, would be sufficiently strong to broaden these lines outside our resolution. In these conditions only longitudinal field "quenching" experiments [6] could reveal the presence of muonium; measurements of this kind are planned for the near future.

The most interesting information is yielded by the behaviour of the linewidths with temperature, which is shown in fig. 2. The decay time of the initial amplitude is fairly constant from room temperature down to $T - T_N = 1$ K at a value of 2.4 ± 0.2 μs. Approaching T_N it undergoes a sudden reduction that brings its value to 1.2 ± 0.15 μs at $T - T_N = 0.25$ K, which is the closest to T_N obtained in this series of data. The qualitative behaviour agrees perfectly with the NMR data [2] plotted in the same figure. We can thus attribute the two phenomena to the same cause, which has already been identified as the critical slowing down of the antiferromagnetic spin fluctuations. In terms of the approximate relationship between the observed linewidths, $\Delta\omega$, and the correlation time, τ_c, of fluctuations in the local field, ω_0/γ,

$$\Delta\omega = \tau_c \omega_0^2 \ .$$

We can deduce that τ_c is the same referred to the ^{19}F nuclei as to the μ^+, regardless of the muon's dynamics. This identity requires the local field at the interstitial site to be smaller than the one at the fluorine site by a factor of nearly 2.

Additional data on the frequency of the precession have been collected at the higher external magnetic field (to enhance the sensitivity). Although this part of the programme is at a preliminary stage we have found two interesting features: first of all a shift is measured, with respect to the Larmor frequency, which ranges from about 0.1% at room temperature to a little less than 1% at 67.55 K. An estimate of the dipolar contribution at various interstitial sites agrees with the order of magnitude of the observed shifts. The demagnetization field correction has been evaluated and is negligible by comparison. Secondly, the shift reverses its sign at different crystal orientations in the applied field. A more systematic determination of the angular dependence of the shift at various fields is in progress; the present information, however, is relevant to the understanding of the muon's localization as well as its behaviour in the critical region near T_N. It seems therefore reasonable to discuss the hypothesis that the muon sits in a well-determined interstitial site, where it could either be trapped or hop between equivalent sites.

The effect of antiferromagnetic fluctuations on the μ^+ damping is consistent with the occupation of an asymmetric position with respect to the two magnetic Mn sublattices (see fig. 3), while the existence of a shift and of its angular dependence requires that the muon occupies coherently sites with well-defined crystal line symmetry. Even a slow diffusion between non-equivalent sites would smear out both the above-mentioned effects.

It is interesting to note, furthermore, that a fast diffusion between crystallographically (but not magnetically) equivalent sites would also produce a motional narrowing of the different hyperfine fields experienced by the muon in the ordered phase. The result would still be seen as a precession at the Larmor frequency, as is the case in Cr [7], and this is in contradiction with the observed reduction of the signal amplitude below T_N.

It is possible to recognize the occurrence of intrinsic localization if one takes into account a small but significant component of ionic character of MnF_2. The muon being a positive point charge, almost unscreened in an insulator, one can conceive that it finds sufficiently deep electrostatic potential wells between two F^- ions. A good candidate for such a site would be the position at the centre of the upper face of the unit cell and its equivalent on the edge of the cell (marked α in fig. 3). The local field experienced in this position is of dipole-dipole nature, if we neglect contact hyperfine terms, and the resulting shift of the μ^+ precession frequency has to be positive when the field is along the c-axis and negative when the field is along the a-axis. This is indeed what is observed experimentally. Considering the F^- ionic radius it may not be possible to neglect contact hyperfine terms arising from hydrogen-type bonds between the μ^+ and the two fluorines. If this is the case, the hyperfine fields are anisotropic as for the dipole fields and the present data do not yield the magnitude and the sign of such a contribution. In addition, assuming the proposed site for the location of the muon, we can explain the disappearance of the signal below T_N for our detection system in transverse geometry. In fact, the hyperfine frequency would have opposite signs at the two magnetically non-equivalent positions so that it would give in-phase signals only in telescopes parallel or antiparallel to the initial beam polarization, $\underline{P_0}$ (which an external field would split into two lines). Measurements in a longitudinal arrangement at nearly zero field are therefore planned in order to find evidence supporting our hypothesis.

Experiments performed near T_N on a single crystal of $RbMnF_3$, whose magnetic properties are similar to those of MnF_2, do not show any "critical" damping of the μ^+ signal. By applying the same electrostatic localization model as for MnF_2 the most likely interstitial site for the muon in $RbMnF_3$ turns out to be symmetrically placed between four equidistant Mn atoms, evenly distributed between the two spin sublattices. This clearly does not predict any critical broadening of the μSR line in the same range of temperatures near T_N.

Summarizing, the positive muon experiences a detectable effect of the critical antiferromagnetic fluctuations in MnF_2. The magnitude of the internal field at the muon appears to be comparable to that measured at the ^{19}F nuclei. The orientation-dependent frequency shift indicates that the particle must sit at a specific interstitial site. Both these observations in the paramagnetic phase seem to agree with an intrinsic localization of the muon rather than with impurity (or defect) trapping mechanisms, because the latter would destroy the spatial coherence which is responsible for the observed results. The role of impurities and defects in the ordered phase, however, should still be studied. The Muon Spin Rotation technique has proved to be a useful tool for monitoring critical phenomena, which can be employed in conjunction with longitudinal and zero field relaxation studies [4] to yield a complete picture of the magnetic properties of antiferromagnetic insulators at an interstitial site.

Acknowledgements

We wish to thank Tapio Niinikoski for letting us use the Polarized Target refrigerator and Gilbert Coubra for all his efforts in preparing it. Ola Hartmann, Lars Olov Norlin and Roger Wäppling of the Uppsala Group have been of invaluable help during the experimental runs. We are indebted to CERN and to the SC machine staff in particular for their constant support. One of the authors (R.D.R.) was supported during his stay at CERN by a "A. Della Riccia" scholarship.

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Figure captions

Fig. 1 : Amplitude of μ^+ precession as a function of temperature. The curve is corrected for the beam polarization and for the μ^+ decay asymmetry.

Fig. 2 : Linewidth of the μ^+ precession from the sample as a function of temperature. Our data (▲) are compared with ^{19}F linewidths from ref. [2] (●), which have been normalized at high temperature $[\Delta\nu_\mu/\Delta\nu_F(T = 300\ K) \simeq 3]$.

Fig. 3 : Structure of the MnF_2 crystal: the orientation of the Mn spins is shown as for the antiferromagnetic phase. α is a possible site for the muon.

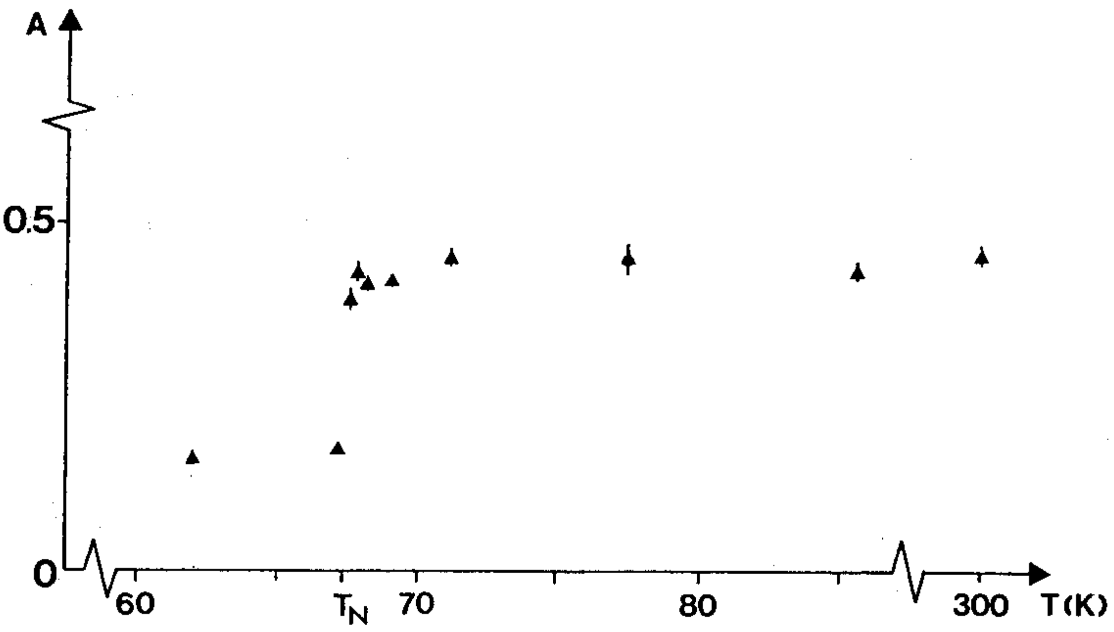

Fig. 1

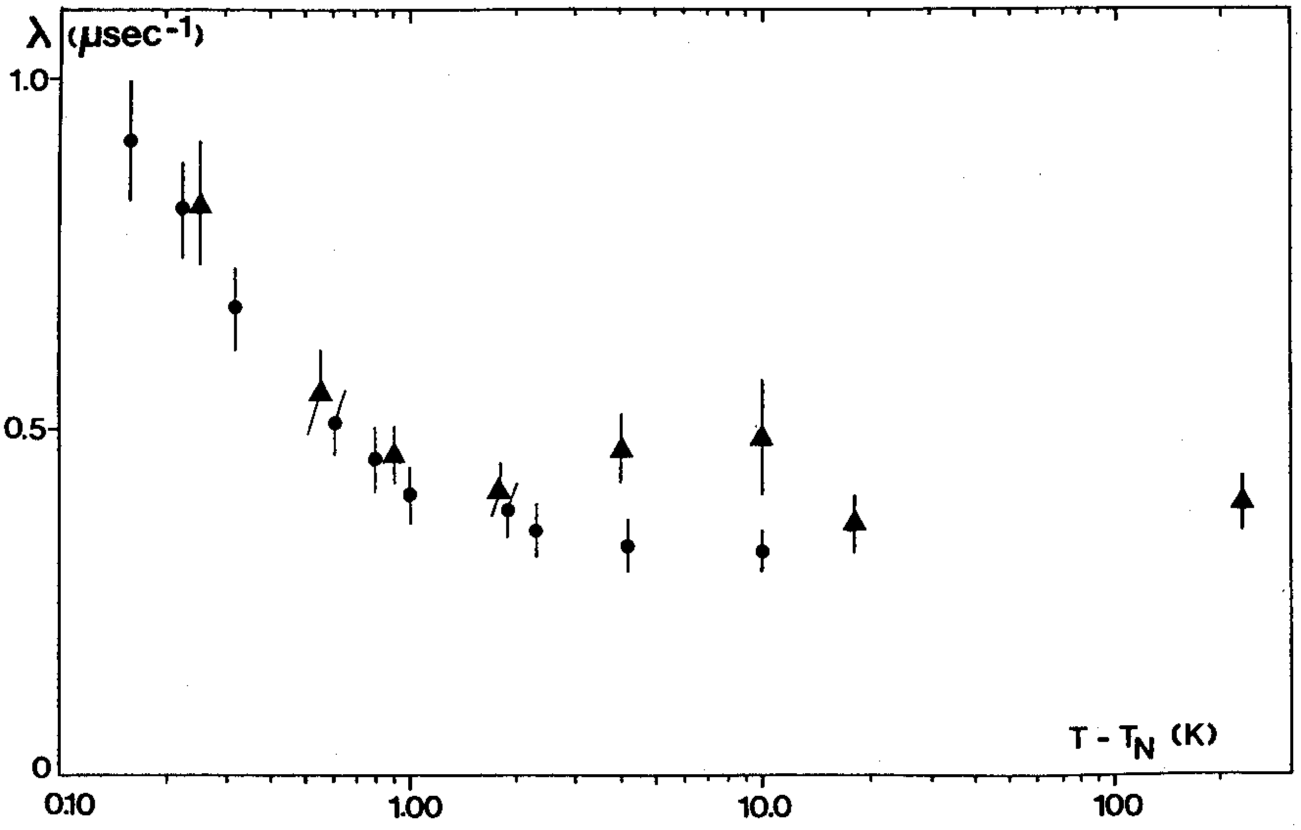

Fig. 2

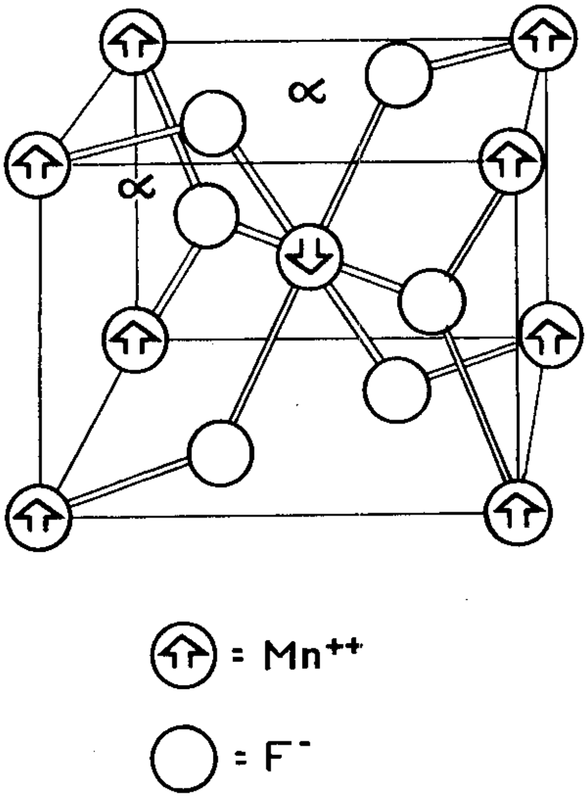

Fig. 3