Longitudinal Spin Relaxation of μ^+ in Paramagnetic MnO

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The temperature dependence of the longitudinal relaxation time of positive-muon spin in paramagnetic MnO has been determined by measuring the μ^+-e^+ forward/backward asymmetry. The relaxation time, $T_1 = 2.00$ μsec at 124 K can be accounted for in terms of exchange-fluctuating Mn^{++} atomic dipolar fields; the increase of T_1 with temperature shows that the relaxation is further slowed by fast diffusional motion of the μ^+.

In this paper we report on the first observation of longitudinal spin relaxation of positive muons in magnetic material, which showed a surprisingly fast diffusion of positive muons in MnO that competes even with the exchange frequency of atomic spins in its paramagnetic phase. The positive muon (μ^+) has been applied extensively as a probe for studying transition metals, and has revealed various interesting features of the internal fields at the μ^+, several of which are especially relevant to conduction-electron polarization[1]; however, few studies have been made of magnetic insulators. The usual technique, called positive-muon spin rotation (μ^+SR), detects the precession of the muon spin in a transverse magnetic field. From the precession frequency, the local field felt by μ^+ can be determined; and from the damping of the precession amplitude, the relaxation time T_2 of the muon spin can be obtained. Nearly free μ^+ precession was observed in preliminary μ^+SR studies on MnO at 293 and at 198 K[2]; the net paramagnetic shift was very small compared with the Lorentz field, which means that the spin relaxation in the paramagnetic phase is caused by atomic dipolar fields. A possible temperature dependence of the spin relaxation was suggested. The damping of the precession amplitude is often caused by the dephasing due to inhomogeneous fields in the sample and/or trapping of μ^+ by defects or impurities. The precession method is therefore inappropriate for studies of the longitudinal relaxation of the muon spin.

We have established a simple and reliable method for measuring the longitudinal relaxation time T_1 of the muon spin. This method was used to study paramagnetic MnO in the temperature range between 124 and 364 K. Details of the technique will be described elsewhere. It is similar to "longitudinal field" methods used earlier[3] but includes several systematic improvements.

A single crystal of MnO of approximate dimensions $1.0\times1.0\times2.5$ cm^3, produced by Nakazumi Crystal Co., was suspended in vacuum, attached to the copper cold finger of a liquid-nitrogen Dewar. Two Chromel-Alumel thermocouples were used to measure the target temperature; one of their outputs was compared with a reference and the difference amplified and fed back to regulate the current of a heater wound around the cold finger. This gave effective control of the target temperature. The muon beam, obtained from the M20 channel of TRIUMF, was collimated to 2.5 cm in diameter. The muon-decay positrons were detected by two sets of counter telescopes, placed symmetrically at 0° (forward) and at 180° (backward) to the beam, and the μ-e decay time spectra were recorded up to 8 μsec.

These spectra were corrected for constant background. We then calculated the time-differential ratio of the forward and the backward spectra as follows:

$$R(t)=\frac{B_0\exp(-t/\tau_\mu)[1+AG(t)]}{F_0\exp(-t/\tau_\mu)[1-AG(t)]}$$

$$\simeq\alpha[1+2AG(t)], \qquad (1)$$

where $\alpha\equiv B_0/F_0$, τ_μ is the muon lifetime, A is the initial asymmetry, and $G(t)$ represents the spin relaxation. Such a simple procedure works excellently, as shown in Fig. 1. From these curves, we can readily tell that the relaxation function $G(t)$ is exponential and that the relaxation time depends strongly on temperature. The instrumental asymmetry coefficient, α, was obtained by fitting the low-temperature runs (124

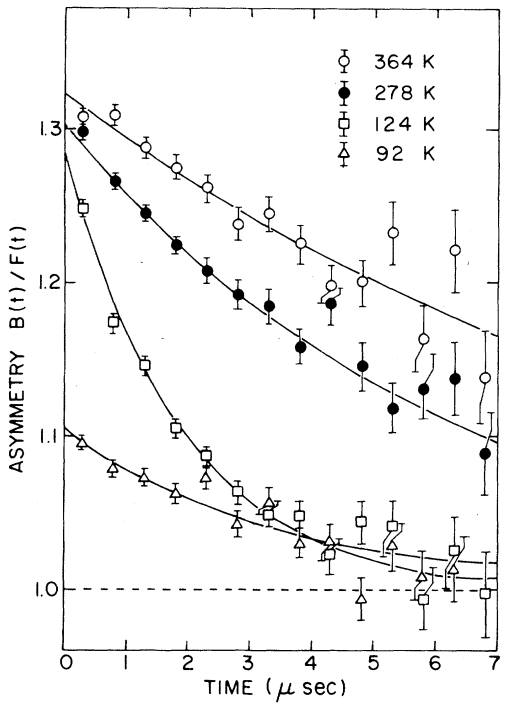

FIG. 1. Forward-backward asymmetry of muon decay positrons from MnO at various temperatures. Corrections for the instrumental asymmetry coefficient, α, were made as described in the text. Solid curves represent best fit to the function $[1+A\exp(-t/T_1)]/[1-A\exp(-t/T_1)]$. The 92-K data (ordered phase) is also shown for comparison.

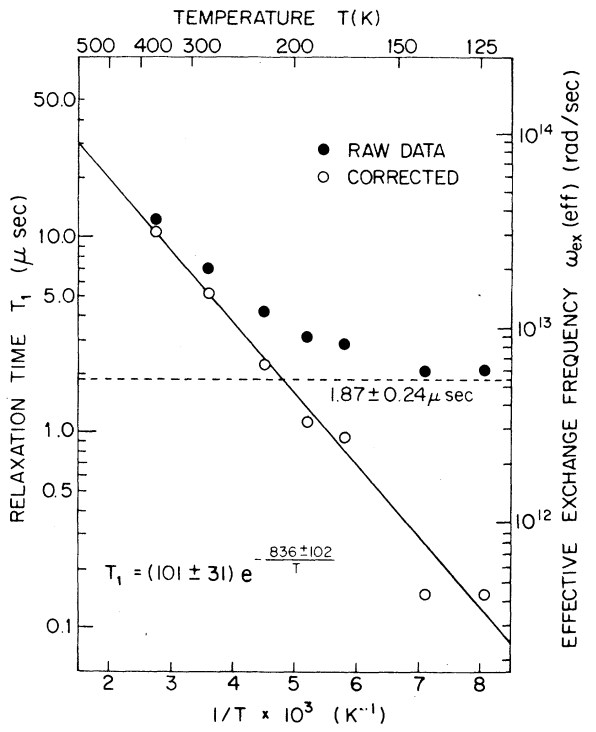

FIG. 2. Longitudinal relaxation time (T_1) of the muon spin in paramagnetic MnO vs inverse temperature. Open circles represent data points after subtraction of the temperature-independent exchange contribution, thus giving the diffusion frequencies $\sqrt{\frac{1}{2}\pi}/\tau_D$ (the scale shown on the right), which are expressed by $\tau_D = \tau_D{}^0 \exp(E_a/kT)$, with $\tau_D{}^0 = (4.2 \pm 1.4) \times 10^{-15}$ sec and $E_a = 72.0 \pm 8.8$ meV.

and 141 K) where the relaxation time is much shorter than the observation-time range. The value thus obtained was then used to fit high-temperature data taken under otherwise identical conditions, with longer relaxation times. In the following, we discuss three points briefly.

(I) *Spin relaxation.*—The exponential shape of $G(t)$ excludes static dipolar relaxation. MnO is a NaCl-type ionic crystal with a lattice constant $a = 4.44 \times 10^{-8}$ cm. Although we are not sure about the μ^+ location in the crystal, we assume that the μ^+ occupies the interstitial site surrounded by four Mn^{++} ions and four O^{--} ions. Let us examine the role of the atomic dipolar field from neighboring Mn^{++} spins which fluctuate with a characteristic exchange frequency ω_{ex}. In the limit of random fluctuation, the relaxation time, according to the theory of Moriya,[4] is given by

$$T_1 = \frac{1}{\sqrt{2\pi}} \left(\frac{d^3}{2\sqrt{2}\gamma_\mu g_e \mu_B} \right)^2 \frac{3}{S(S+1)} \omega_{ex}, \qquad (2)$$

where $\gamma_\mu = 2\pi \times 1.3553 \times 10^4$ rad/sec G and d is the distance between the μ^+ and the nearest-neighbor Mn^{++} ions. Ignoring possible distortion of the nearest-neighbor lattice due to the presence of μ^+, we insert $d = \sqrt{3}a/4 = 1.92 \times 10^{-8}$ cm and $\omega_{ex} = 4.9 \times 10^{12}$ rad/sec [5] into Eq. (2), and obtain $T_1 = 1.68$ μsec. After performing a lattice summation, we obtain

$$T_1 = 1.54 \ \mu\text{sec}. \qquad (3)$$

The low-temperature data yielded a relaxation time very close to this estimate; but at higher temperatures, the relaxation time becomes almost an order of magnitude longer. What accounts for this strong temperature dependence of T_1?

Suppose the μ^+ is diffusing in the crystal. If it "hops" faster than the exchange frequency, the field fluctuation frequency felt by the μ^+ spin will be effectively increased. It is reasonable to assume that the ω_{ex} in Eq. (2) should be replaced by

$$\omega_{ex}(\text{eff}) = \omega_{ex} + \sqrt{\tfrac{1}{2}\pi}/\tau_D, \qquad (4)$$

where τ_D is the μ^+ hopping time. If the diffusion obeys the Arrhenius law in this temperature region, τ_D should take the form

$$\tau_D = \tau_D^0 \exp(E_a/kT), \quad \tau_D^0 = (\tfrac{1}{2}a)^2/6D_0, \qquad (5)$$

where E_a is the activation energy, and D_0 is the diffusion coefficient.

To test this model, we generated an Arrhenius plot of T_1 as shown in Fig. 2. The raw data points for $\ln T_1$ vs $1/T$ do not fall on a straight line; however, if we fit the data assuming that there is a temperature-independent minimum value of the relaxation time [as predicted by Eq. (4) for $\tau_D \to \infty$], we obtain a good fit using a constant term $T_1(0) = 1.87 \pm 0.24$ μsec. This value is a little larger than the theoretical value (3), $T_1 = 1.54$ μsec, for rigid lattice without muon diffusion. It may indicate a local lattice expansion of about 3%.

From the slope of the line, the activation energy of the diffusion was determined to be

$$E_a/k = 836 \pm 102 \text{ K or } E_a = 72.0 \pm 8.8 \text{ meV}, \qquad (6)$$

which is considerably larger than typical activation energies in metals. Presumably, the mechanism of the diffusion is different between metals and ionic crystals. The preexponential factor is determined to be $\tau_D^0 = (4.2 \pm 1.4) \times 10^{-15}$ sec, giving $D_0 = (2.0 \pm 0.7) \times 10^{-2}$ cm^2 sec^{-1}. We emphasize that the absolute scale of τ_D is determined here in comparison with the known spin exchange frequency ω_{ex} without knowledge of the absolute strength of the dipolar field at the μ^+. This surprisingly short hopping time may imply that the equilibrium location of the μ^+ is shifted from the body center toward the O^{--} ions and that the μ^+ undergoes fast diffusion among the four possible positions in a cell.[6]

(II) *Initial asymmetry in the ordered phase.*—MnO is antiferromagnetic below the Néel temperature, 122 K. The result of a similar measurement performed at 92 K is plotted in Fig. 1 for comparison. We notice that the initial asymmetry (asymmetry at $t = 0$) is $\frac{1}{3}$ of that for the paramagnetic-phase data, which is consistent with the fact that there is a nonvanishing dipolar field at the tetrahedral interstitial site in antiferromagnetic MnO. Fourier analysis of the data did not show any precession frequency below 100 MHz. This fact seems to be consistent with the expected diffusion time of 0.19 nsec at 92 K, extrapolated in the Arrhenius plot of Fig. 2. Further measurments of μ^+ spin precession and relaxation in the ordered phase should be interesting.

TABLE I. Summary of the longitudinal relaxation time (T_1) and the initial asymmetry A of μ^+ in MnO at various temperatures.

T (K)	$10^3/T$ (K^{-1})	T_1 (μsec)	A
91.8[a]	10.89	3.68 ± 0.44	0.044 ± 0.002
124.1	8.06	2.00 ± 0.07	0.117 ± 0.003
124.1[b]	8.06	2.87 ± 0.15	0.115 ± 0.002
140.9	7.10	2.01 ± 0.08	0.118 ± 0.002
171.8	5.82	2.79 ± 0.05	
192.1	5.21	3.00 ± 0.15	0.120 ± 0.002
220.2	4.54	4.06 ± 0.20	
278.0	3.60	6.84 ± 0.14	0.125 ± 0.001
278.0[b]	3.60	7.67 ± 0.32	0.126 ± 0.001
363.6	2.75	12.00 ± 0.50	0.131 ± 0.001

[a] Antiferromagnetic phase of MnO.
[b] With an 800-Oe longitudinal field.

(III) *Temperature dependence of the initial asymmetry.*—A careful look at the initial asymmetry reveals that it increases monotonically as the temperature is raised. We applied an 800-Oe longitudinal field at 124 and 278 K to see whether this effect is field dependent. The initial asymmetry was unchanged within error. (See Table I.) However, we observed that T_1 becomes longer when the field is applied both at 124 and 278 K. We await the results of systematic studies of the field dependence of T_1 (and possibly of the initial asymmetry) before attempting to interpret these effects.

In summary, it has been demonstrated that the longitudinal-field method is effective for studying the longitudinal relaxation of the muon spin. The relaxation rate of the muon spin in paramagnetic MnO is shown to be caused by the surrounding Mn^{++} dipoles, and is reduced by the diffusion of the μ^+ as well as by exchange fluctuations.

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[6]The shortest hopping time $\tau_D{}^0$ ever observed is 10^{-11}–10^{-12} sec for bcc iron: N. Nishida, R. S. Hayano, K. Nagamine, T. Yamazaki, J. H. Brewer, D. M. Garner, D. G. Fleming, T. Takeuchi, and Y. Ishikawa, Solid State Commun. 22, 235 (1977).

Pressure-Induced Phase Transformation in Nontransforming V_3Si

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The dependence on hydrostatic pressure of the superconducting transition temperature T_c of a nontransforming V_3Si single crystal is determined to 45 kbar with use of a newly developed pressure cell. A clear break in the slope dT_c/dP at 32 kbar supports the existence of the pressure-induced structural phase transition predicted by Larsen and Ruoff.

High-temperature superconductivity, occurring, for example, in A-15 compounds like Nb_3Ge, Nb_3Sn, or V_3Si, is coupled with structural phase instabilities and other anomalous normal–solid-state properties.[1,2] In recent years considerable theoretical and experimental effort has been directed towards a better understanding of this interrelation. High-pressure techniques are especially well suited for testing theory because it allows a well-defined continuous variation of parameters using only a single sample.

One of the most interesting of such anomalous normal-state properties is the drastic reduction of the elastic shear modulus $c_s = \frac{1}{2}(c_{11} - c_{12})$ with decreasing temperature observed, for example, in V_3Si.[3] In so-called "transforming" crystals the lattice softening is sufficient to lead to a cubic-to-tetragonal phase transformation[4] at a temperature $T_L \approx 21$ K only slightly higher than that for the onset of superconductivity $T_c \approx 17$ K; in "nontransforming" crystals, on the other hand, the reduction in c_s is less dramatic and no phase transformation occurs. Pressure studies on "nontransforming" V_3Si crystals are of interest for the following reasons: (1) In "transforming" V_3Si T_L and T_c approach each other under pressure[5] and meet, allowing a study of superconductivity in both tetragonal and cubic phases, consecutively. To investigate accurately the interference of this structural transformation with superconductivity, knowledge of the behavior of superconductivity in a "nontransforming" crystal is essential. (2) Larsen and Ruoff[6] have shown that in "nontransforming" V_3Si c_s decreases with pressure at low temperatures; they predict the onset of a lattice transformation at pressures near 30 kbar. Previous investigations[7] to 20 kbar have found no evidence for a phase transition.

Using a newly developed hydrostatic pressure cell for electrical resistivity measurements, we have determined the pressure dependence of T_c to 45 kbar for two V_3Si single crystals. These crystals are of the "nontransforming" type as evidenced by low-temperature x-ray diffraction studies[8]; in addition, there is no sign from 18 to 30 K of the slight kink in the temperature dependence of the resistivity which is believed to mark the cubic-to-tetragonal transition.[9] The two V_3Si single crystals are cut from the same host crystal and have residual resistance ratios (RRR) of ~14 and $T_c = 16.6$ K with 10%-90% transitions widths $\Delta T_c = 0.015$K.