Observation of the $T/(T-T_c)$ Divergence of the μ^+ Spin-Lattice Relaxation Rate in MnSi near T_c

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The spin-lattice relaxation time (T_1) of the interstitial μ^+ spin in the paramagnetic phase of a weak itinerant-electron helimagnet MnSi has been measured near the magnetic-ordering temperature in a 700-Oe longitudinal magnetic field. The observed temperature dependence of $1/T_1$ is represented very well by $T/(T-T_c)$ which was predicted on the basis of the self-consistent renormalization theory of spin fluctuations in itinerant-electron ferromagnets.

In this paper we report on the first successful observation of the divergence of the spin-lattice relaxation rate around T_c in a typical itinerant-electron magnetic system by using positive muons. Recent NMR[1,2] and neutron diffraction[3] studies of MnSi have revealed various interesting magnetic properties. MnSi is an intermetallic compound with the cubic B-20 crystal structure; it has been shown experimentally to order magnetically at $T_c \simeq 29$ K into a long-period (180 Å) helix. In an external magnetic field, the structure becomes progressively more conical, until above $H_c = 6.2$ kOe (at 4.2 K) it becomes ferromagnetic. Above T_c, the uniform susceptibility $\chi(0)$ shows a Curie-Weiss–like temperature dependence. However, despite the usual belief that such a behavior indicates the existence of localized moments, experimental results are all inconsistent with the localized-moment picture.[2] These rather unusual magnetic properties of MnSi, which could not be understood by the conventional random-phase-approximation (RPA) Hartree-Fock theory for itinerant-electron systems, were successfully accounted for when the self-consistent renormalization (SCR) theory on the magnetism of itinerant electrons, developed by Moriya and his collaborators,[4] was applied to MnSi[5]: We now understand that MnSi is an itinerant-electron weak helimagnet below T_c and an itinerant-electron paramagnet above T_c.

To check further the validity of the theory and our understanding of MnSi, it is of great importance to measure the temperature dependence of the spin-lattice relaxation time (T_1). Since MnSi has such a long period of helical structure, the temperature dependence of T_1 is expected to behave almost like that of an itinerant-electron ferromagnet. The SCR prediction for the itinerant-electron ferromagnet above T_c (under low external field) is[4]

$$\frac{1}{T_1} = 2\gamma_n^2 A_{\mathrm{hf}}^2 kT \sum_{\vec{q}} \frac{\chi_\perp''(\vec{q},\omega_0)}{\omega_0} \propto \frac{1}{T_{10}}\chi(0) \propto \frac{T}{T-T_c}, \tag{1}$$

where T_1 is related to the usual Korringa relation, and thus $1/T_{10} \propto T$, and $\chi(0)$ has a Curie-Weiss–like temperature dependence. This is qualitatively quite different from the result of the RPA Hartree-Fock theory, which predicts $1/T_1 \propto T/(T^2 - T_c^2)$.[4]

Yasouka *et al.*[2] measured the T_1 of ^{55}Mn via NMR in the temperature range 200–300 K and observed an almost temperature-independent spin-lattice relaxation time, $T_1 = 35 \pm 5$ μsec, qualitatively consistent with the SCR theory. Unfortunately, the relaxation time became too short for the NMR technique as the temperature was lowered; observation of the characteristic divergence of $1/T_1$ near T_c, a more explicit verification of the theory, was not possible by the NMR technique.

In the present work we used the positive muon (μ^+), a new interstitial magnetic probe, to observe the characteristic divergence of the spin-lattice relaxation rate near T_c. The μ^+, having different location and motions from NMR probes, can be considered complementary to NMR in many ways; it may, therefore, be able to yield information about spin fluctuations inaccessible to NMR techniques. We find μ^+ measurements especially attractive near T_c, since they can be carried out in a very low external field as opposed to the NMR case; the large external field required by NMR methods strongly suppresses the spin fluctuations to that the sharp divergence as predicted by Eq. (1) cannot be observed. It is also interesting to see if we can find any deviation from Eq. (1) near T_c, arising from the difference between the ferromagnetic and the helical ordering under a small external field (otherwise, MnSi will order ferromagnetically anyway).

Our recent measurement of the μ^+ Knight shift in paramagnetic MnSi using the muon-spin-rotation technique[6] showed that the μ^+ feels a unique magnetic field, independent of the crystal-axis orientation, and that the shift is directly proportional to the host magnetization with a hyperfine coupling constant of $A_{\rm hf}^{\mu} = -4.8 \pm 0.2$ kOe/μ_B. It was also observed that the relaxation time of the muon-spin-rotation signal (damping of the precession) becomes shorter as the temperature approaches T_c. We must be cautious when interpreting this result, because the damping of the precession signal results not only from the spin-lattice relaxation of the muon spin, but also from various dephasing processes such as caused by the inhomogeneous demagnetization field and nuclear dipolar fields.

In order to measure the spin-lattice relaxation time unambiguously, the "longitudinal-field" technique[7] was employed. In this method, an external field is applied along the initial muon spin direction, and the muon-decay positrons are detected by counter telescopes placed symmetrically at 0° (forward) and at 180° (backward) to the beam. The time-differential measurement of the forward-backward asymmetry yields the spin-lattice relaxation time, unaffected by any dephasing, as far as the external field is large enough to decouple the nuclear dipolar fields.

A single-crystal sphere of MnSi, 2.5 cm in diameter, was prepared from an ingot grown by the Czochralski method at the Institute for Iron, Steel and Other Metals, Tohoku University. The positive-muon beam, obtained from the M20 beam channel of TRIUMF, was collimated to 1.9 cm in diameter and stopped in the MnSi crystal. The forward and backward time spectra were recorded up to 16 μsec, in the temperature range between 29.7 and 285 K, and in longitudinal external fields of up to 700 Oe.

The zero-field data taken at 285 K, as shown in Fig. 1(a), clearly show a Gaussian-like relaxation follwed by recovery of asymmetry, which is characteristic of static random fields.[8] The width of the random fields estimated from this shape is about 4 Oe, consistent with the nuclear dipolar fields from ^{55}Mn, and a relatively small external field around 30 Oe was sufficient to decouple these random fields. This result clearly shows that the μ^+ is frozen at a site, and the observation of a unique frequency in our previous precession measurement indicates that all the muon sites are nearly magnetically equivalent. The low-field relaxation phenomena we studied will be described in a separate paper.[9]

We measured the longitudinal relaxation function $G(t)$, defined by

$$N(\theta,t) = N_0 \exp(-t/\tau_\mu)[1 + AG(t)\cos\theta], \qquad (2)$$

in a 700-Oe external field. At room temperature $G(t)$ was constant, showing that the relaxation time is too long to be determined, but at lower temperatures $G(t)$ became exponential as shown in Fig. 1(b), as a result of the transverse component of the fluctuating hyperfine fields created by itinerant-electron spins. The relaxation rate becomes greater as the temperature is lowered, and appears to diverge near T_c.

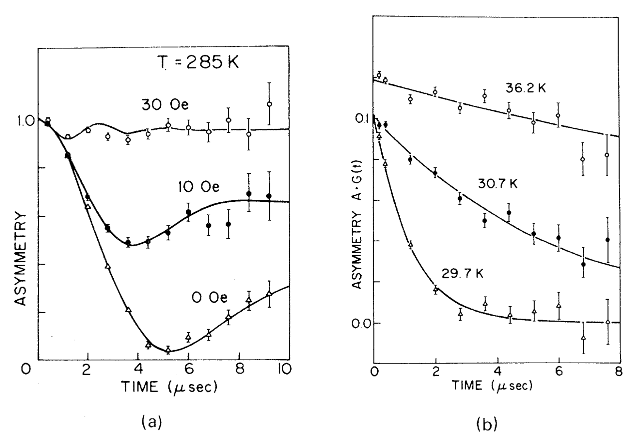

FIG. 1. (a) Longitudinal relaxation function $G(t)$ in MnSi at room temperature at low external fields. The data are fitted by the theoretical curves of Kubo and Toyabe (Ref. 8) in the static limit with a width $\Delta = 3.6$ Oe of random fields. (b) Typical forward-backward asymmetry of muon decay positrons from MnSi in a longitudinal magnetic field $H_0 = 700$ Oe at different temperatures near T_c.

If T_1 depends on temperature as predicted by Eq. (1), the T_1 should fall on a straight line when plotted against inverse temperature $1/T$. Such a plot, shown in Fig. 2(a), tells us that Eq. (1) does hold remarkably well throughout the temperature range studied. The best fit to the data by the formula

$$T_1 = T_1(\infty)(1 - T_c/T), \qquad (3)$$

yielded $T_1(\infty) = 153 \pm 3$ μsec and $T_c = 29.5 \pm 0.2$ K. The T_c thus obtained is in excellent agreement with the value obtained from the bulk magnetization measurement.

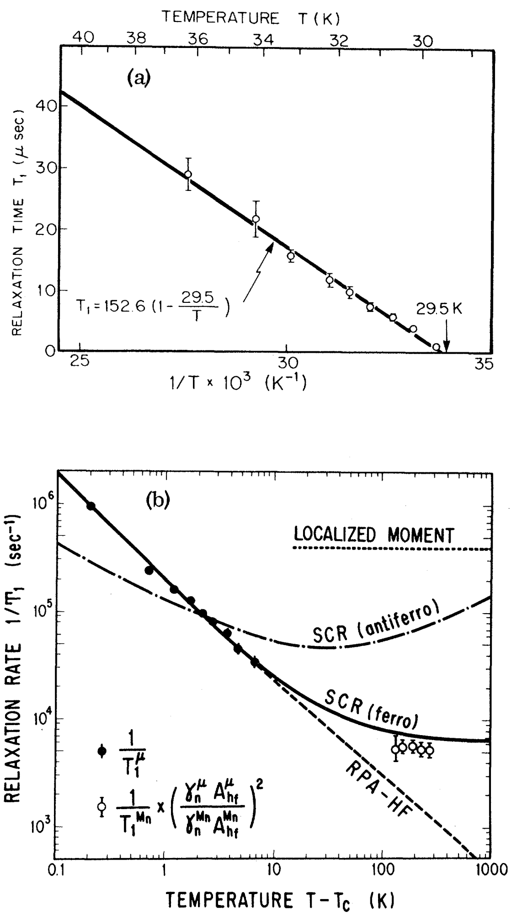

FIG. 2. (a) Spin-lattice relaxation time (T_1) of the muon spin in paramagnetic MnSi, observed with a 700-Oe longitudinal field, plotted against inverse temperature. (b) The present μ^+ data and the ^{55}Mn NMR data (Ref. 2) scaled according to Eq. (4) are plotted against $T - T_c$. The SCR prediction for itinerant-electron weak ferromagnets is drawn as a solid line, and the RPA Hartree-Fock prediction as a broken line. The localized-moment value estimated in Ref. 2 is scaled and presented as a dotted line. Also compared is the SCR curve for itinerant-electron weak antiferromagnets.

From Eq. (1) we expect a certain scaling relationship between the T_1 of ^{55}Mn and that of μ^+; namely,

$$T_1^{\mu}(\gamma_n^{\mu} A_{hf}^{\mu})^2 = T_1^{Mn}(\gamma_n^{Mn} A_{hf}^{Mn})^2, \qquad (4)$$

if the spin fluctuations of the itinerant d electrons in MnSi are identical both for the Mn and for the interstitial μ^+. A possible deviation from this relation may arise if there is a significant dipolar contribution to the local field as in the localized-moment case, since such a contribution is not included in A_{hf} obtained from the Knight shift, but nevertheless causes the spin-lattice relaxation. When the $\vec{q} \simeq 0$ (uniform) component of spin fluctuations plays the dominant role as in MnSi, we expect the scaling relation to hold. We scaled the ^{55}Mn NMR data with the parameters

$$A_{hf}^{Mn} = -(1.38 \pm 0.01) \times 10^5 \text{ Oe}/\mu_B,$$

$$\gamma_n^{Mn} = 2\pi \times 1.050 \times 10^3 \text{ (Oe sec)}^{-1},$$

$$A_{hf}^{\mu} = -(4.8 \pm 0.2) \times 10^3 \text{ Oe}/\mu_B,$$

$$\gamma_n^{\mu} = 2\pi \times 13.55 \times 10^3 \text{ (Oe sec)}^{-1},$$

and the results are shown with open circles in Fig. 2(b). The $T_1^{\mu}(\infty)$ value thus estimated from the scaling is 193 ± 30 μsec, which is fairly close to the extrapolated experimental value based on Eq. (3).

In Fig. 2(b), the best-fit SCR curve is drawn as a solid line together with results of other theories. The overall agreement between the SCR prediction and the experimental result is quite satisfactory. The RPA Hartree-Fock prediction, for instance, looks similar to the SCR curve near T_c, but its high-temperature behavior is totally inconsistent with the experimental results. The localized-moment picture, on the other hand, correctly leads to a temperature-independent behavior at higher temperatures, but the predicted rate[2] is orders of magnitude larger than the observed values.

We would also like to report that no field dependence of T_1 was observed between 200 and 700 Oe external fields at 30.2 K, and that the temperature dependence of T_1 followed Eq. (1) all the way down to 29.7 K, 0.2 K above T_c. Since the T_1 of helimagnets may deviate from Eq. (1) and become somewhat similar to the antiferromagnetic case near T_c [also shown in Fig. 2(b)], these results indicate that MnSi behaves as an itinerant-electron weak ferromagnet even at temperatures very close to T_c, and under such small external fields. They also indicate that the SCR theory does work in temperatures extremely close to the transition temperature.

To our knowledge, this is the first experimental work to show the divergent behavior of $1/T_1$ at T_c in weak itinerant-electron ferromagnetic systems, and we conclude that there is general agreement between the present experimental results and the SCR theory. We would also like to mention that the capabilities and usefulness of a new technique, muon-spin rotation especially the longitudinal-field method, have been fully demonstrated here, in a current problem of magnetism study inaccessible to conventional NMR techniques.

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Role of Local Plasmon Modes in Light Emission from Small-Particle Tunnel Junctions

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A resonancelike structure has been reported in the frequency spectrum of light emitted from small-particle tunnel junctions. Here we present a calculation of the frequency-dependent dipole moment and the radiation emission spectrum for a small particle located above a metal film. The dipole moment exhibits a resonance structure associated with a localized surface plasma mode at a frequency corresponding to the peak in the structure observed in the emission spectra.

Small metal particles deposited on an oxidized metal film emit light when driven by inelastic tunneling electrons. In this process, the particle-film system acts as an antenna and the inelastic tunneling electrons as a current generator. In a previous analysis, the induced dipole moment of the antenna was calculated within a quasistatic limit appropriate for frequencies well below the plasma resonance threshold.[1] Experimental studies of the intensity spectra from tunnel junctions formed by depositing gold particles onto an oxidized aluminum film exhibit a resonancelike structure near 650 nm (1.9 eV).[2] Here we report calculations which suggest that this structure arises from local plasma resonant modes of the gold-particle–aluminum-film system.

Figure 1 shows the geometry of the small-par-

