Field Dependence of the Muon Spin Relaxation Rate in MnSi

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Muon spin rotation/relaxation measurements have been performed in the itinerant helical magnet MnSi at ambient pressure and at 8.3 kbar. We have found the following: (a) the spin-lattice relaxation rate $1/T_1$ shows divergence as $T_1T \propto (T - T_c)^\beta$ with the power β larger than 1 near T_c; (b) $1/T_1$ is strongly reduced in an applied external field B_L and the divergent behavior near T_c is completely suppressed at $B_L \geq 4000$ G. We discuss that (a) is consistent with the self-consistent renormalization theory and reflects a departure from "mean-field" behavior, while (b) indicates selective suppression of spin fluctuations of the $q = 0$ component by B_L.

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Manganese monosilicide (MnSi), a magnetic system with itinerant electrons, orders magnetically at a temperature $T_c = 29.5$ K into a helical magnetic structure with a long period of 180 ± 3 Å and a small staggered magnetic moment $M_0 = 0.4\mu_B$ per Mn at $T = 0$ K [1]. In a magnetic field, the magnetic structure transforms progressively from helical to conical to ferromagnetic, as shown in Fig. 1(a). Above 6.2 kG (at 4.2 K) the system behaves like a ferromagnet having a relatively large high field susceptibility [3]. In the paramagnetic state the magnetic susceptibility χ obeys a Curie-Weiss law up to 400 K [4].

This system has been extensively studied as a prototype of itinerant weak ferromagnets which follow predictions of the self-consistent renormalization (SCR) theory developed by Moriya, Kawabata, and co-workers [5]. The SCR theory overcomes shortcomings of earlier mean field (Stoner-Slater) theory and random phase approximation, and successfully explains the Curie-Weiss behavior of χ, the ordering temperature, and other properties of the metallic ferro- and antiferromagnets, such as Fe, Ni, and $ZrZn_2$. In a crossover from the magnetism of localized moments to that of itinerant electrons, MnSi is located in a "very itinerant" region where the magnetic moment size M_0 and T_c are very small, while strong spin fluctuations extend to a rather high energy scale. Recently, Pfleiderer *et al.* [6] found that, in applied pressure above $p_c = 14.6$ kbar, MnSi becomes a correlated paramagnet.

Previously, Hayano *et al.* [7] performed muon spin relaxation (μSR) measurements in MnSi and reported divergence of the spin-lattice relaxation rate $1/T_1$ observed above T_c in an applied longitudinal field B_L of 700 G, following $T_1T \propto (T - T_c)$. This behavior is consistent with the prediction of the SCR theory in a wide temperature region above T_c. Kadono *et al.* [8] confirmed this result in subsequent μSR studies, which elucidated muon-nuclear double relaxation effects.

In this Letter, we report our new μSR measurements on a single crystal of MnSi under longitudinal magnetic fields $B_L \leq 6$ kG in ambient and applied pressure. We discovered the following: (a) a strong dependence of $1/T_1$ on B_L; (b) a departure of T_1T from the linear behavior in $(T - T_c)$ at temperatures very close to T_c. We will discuss these results in terms of (a) different

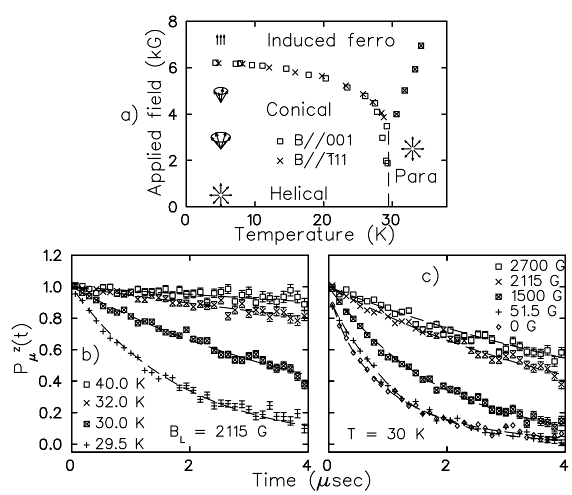

FIG. 1. (a) Field-temperature phase diagram of MnSi [2]. (b) The muon spin polarization function $P^z_\mu(t)$ observed in MnSi at ambient pressure with $B_L = 2115$ G. (c) The field dependence of $P^z_\mu(t)$ at $T = 30$ K.

effects of B_L on uniform and helical spin fluctuations, and (b) departure from "mean-field" behavior of χ near T_c.

We performed zero field (ZF) and longitudinal field (LF) μSR measurements at the M20 and M9 Channels of TRIUMF, Vancouver, Canada. A positive muon beam, polarized along the flight direction, was stopped in a specimen, and histograms of the muon decay positrons, as a function of the residence time t of μ^+ in the crystal, were recorded by using forward/backward counters. In ambient pressure, we used a beam of momentum of $\sim$28 MeV/c and a single crystal specimen of $8 \times 8 \times 2$ mm^3, with the largest surface perpendicular to the cubic crystal axis placed along the beam direction. For measurements at 8.3 kbar, we mounted a larger single crystal in a piston-cylinder pressure cell, in which Daphne oil is compressed to produce uniform hydrostatic pressure, and used a beam of momentum of $\sim$110 MeV/c directed perpendicular to the cubic crystal axis.

Since positrons are emitted preferentially along the muon spin direction, the muon spin polarization function P_μ^z can be obtained from the forward/backward asymmetry in positron histograms. In the paramagnetic state of ferro- or antiferro-magnets P_μ^z is usually given as a product of the Kubo-Toyabe function, $G_{KT}(t)$, which describes the effect of nuclear dipolar fields, and an exponential function, $\exp(-t/T_1)$, due to electron spin fluctuations. Application of LF with $B_L \geq 50$ G would "decouple" nuclear dipolar fields, making $G_{KT}(t) \approx 1$. In the ordered state P_μ^z consists of an oscillating signal representing muon spin precession around the static internal field, added to the exponentially decaying nonoscillating signal. These two signals appear with the amplitude ratio of 2:1 in ZF-μSR using powder samples, while the ratio changes in single crystal samples and/or in LF-μSR [9].

In MnSi, previous ZF-μSR studies [8] found two different precession frequencies, indicating two different muon sites with different magnitudes of internal fields. Because of limited statistics, however, it is difficult to decompose P_μ^z observed above T_c into a sum of two exponential functions having different decay rates. Thus, previous results in Refs. [7,8] were analyzed by using a single exponential relaxation function for the electron spin contributions. We follow this approach in the analysis of the present data, which allows direct comparison of the T_1 results with the previous measurements. Level crossing resonance with nuclear quadrupole oscillation was found in MnSi around $B_L = 100$ G [10]. We carefully avoided this B_L in the present study.

We performed temperature scans for $B_L = 0$–6000 G and magnetic field scans at $T = 30$, 31, and 32 K. Figure 1(b) shows the time spectra above T_c for $B_L = 2115$ G. The depolarization rate increases with decreasing T, reflecting the slowing down of spin fluctuations. We also observed a pronounced dependence of the time spectra on B_L, as shown in Fig. 1(c). With increasing field, the signal relaxes more slowly. For the case of fast spin fluctuations with the rate $\nu \gg \gamma_\mu B_L$ ($\gamma_\mu = 2\pi \times 13.54$ MHz/kG), the relaxation rate is $1/T_1 = \omega^2/\nu$, where $\omega = \gamma_\mu B_{int}$ is the Larmor precession frequency of the muon spins around the instantaneous local magnetic field, B_{int}. To estimate ω, we used $\omega = 2\pi f$, where $f = 28.07(7)$ MHz and $f = 12.249(6)$ MHz are the precession frequencies at 6 K [8]. The resulting spin fluctuation rate for the minimal $T_1 = 0.38$ μs is in the range 2.25–12.4 ns^{-1}, which is much higher than the Larmor precession frequency $\omega_L = \gamma_\mu B_L = 0.5$ ns^{-1} for $B_L = 6$ kG. Therefore, the observed field dependence of $P_\mu^z(t)$ close to T_c is not due to the Zeeman level splitting of muons, but it reflects the change of the electronic spin fluctuation rate caused by B_L.

In Fig. 2(a) we present the relaxation rate $1/T_1$ as a function of temperature and the applied field B_L. As seen in the figure, a magnetic field of 4000 G completely suppresses the critical behavior, but for magnetic fields up to 2700 G we still observe critical behavior near T_c.

According to the SCR theory [5], the muon spin relaxation time T_1 is related to the uniform susceptibility through the relations

$$\frac{1}{T_1} = \frac{\hbar \gamma_\mu^2 A_{hf}^2}{2\pi T_A} \frac{3t}{2y}, \tag{1}$$

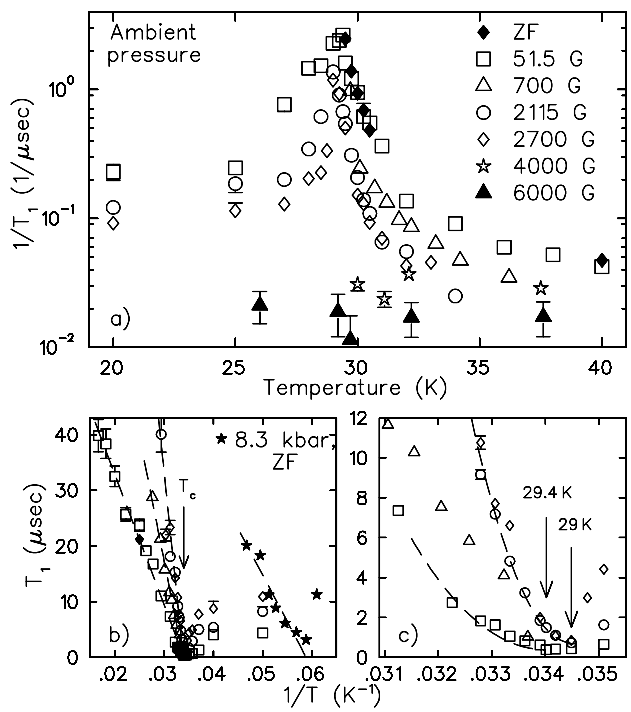

FIG. 2. (a) Temperature dependence of the relaxation rate $1/T_1$ in MnSi at ambient pressure for $B_L = 0$–6000 G. The results for $B_L = 700$ G were taken from [7]. (b) A plot of T_1 versus $1/T$ for the results in MnSi at ambient pressure and under $p = 8.3$ kbar (star symbol). (c) The relaxation time versus inverse temperature plot around $T_c \approx 29.5$ K, together with the fits to $T_1 T \propto (T - T_c)^2$ shown by the broken line.

where $y = 1/(2T_A\chi)$ is the reduced inverse susceptibility, $t = T/T_0$ is the reduced temperature, and A_{hf} is the muon hyperfine coupling constant. T_0 and T_A characterize the energy width of the dynamical spin fluctuation spectrum and the width of the distribution of static susceptibility in the q space, respectively [11]. In the temperature region where χ obeys a Curie-Weiss law, we expect T_1T to depend linearly on $(T - T_c)$, which leads to a linear relationship when T_1 is plotted against the inverse temperature $1/T$.

Figure 2(b) shows such a plot of T_1 vs $1/T$. In the paramagnetic phase, the relaxation time depends linearly on the inverse temperature in a wide temperature region under each applied magnetic field. The slope of the line fitted to these data points increases with increasing applied field up to $B_L = 2115$ G. The results reported by Hayano *et al.* [7] for $B_L = 700$ G are consistent with the trend found in our measurements.

Close to the critical temperature, the uniform susceptibility of itinerant systems deviates from the Curie-Weiss law, and instead follows $1/\chi \propto (T - T_c)^2$ [5], which results in $T_1T \propto (T - T_c)^2$. There is also an intermediate region between the high temperature region and the region around T_c where $1/\chi \propto (T^{4/3} - T_c^{4/3})$, which is a weakly superlinear dependence of $1/\chi$ on temperature [5]. As T approaches T_c, the spin fluctuation modes with small wave vectors and low energies become predominant in the very itinerant systems, such as MnSi. This feature causes the superlinear behavior of χ and T_1T in the critical region.

Improved experimental conditions allowed us to define the sample temperature with sufficient accuracy (± 0.02 K) around T_c to study this behavior. Figure 2(c) shows the plot of T_1 versus $1/T$ around T_c, together with a fit to the $(T - T_c)^2$ dependence, with $T_c = 29.4$ and 29.0 K at 51.5 and 2115 G, respectively. The T_1 vs $1/T$ plot shows nonlinear behavior also in the results obtained under 8.3 kbar, as shown in Fig. 2(b).

The magnetic field dependence of T_1 was not noticed or considered in earlier μSR studies of MnSi [7,8]. Because the helical magnetic structure has such a long period, we first look into theoretical predictions for a weak ferromagnet. We calculate the expected values of $T_1(B_L)$ in MnSi using the most recent approach of the SCR theory [12] with the parameters $T_c = 29.5$ K, $T_0 = 231$ K, $T_A = 2080$ K, and $F = 3520$ K, where F represents the renormalized coupling constant between the spin fluctuation modes and can be determined from the Arrott plot.

The result of these calculations, shown in Fig. 3(a), indicates that even a magnetic field as small as 700 G would have suppressed the divergent behavior of $1/T_1$ due to critical fluctuations. Comparisons of Figs. 2(a) and 3(a) clearly demonstrate that the observed divergent behavior of $1/T_1$ in 700 G $\leq B_L \leq$ 2700 G cannot be explained by the SCR theory for a weak ferromagnet.

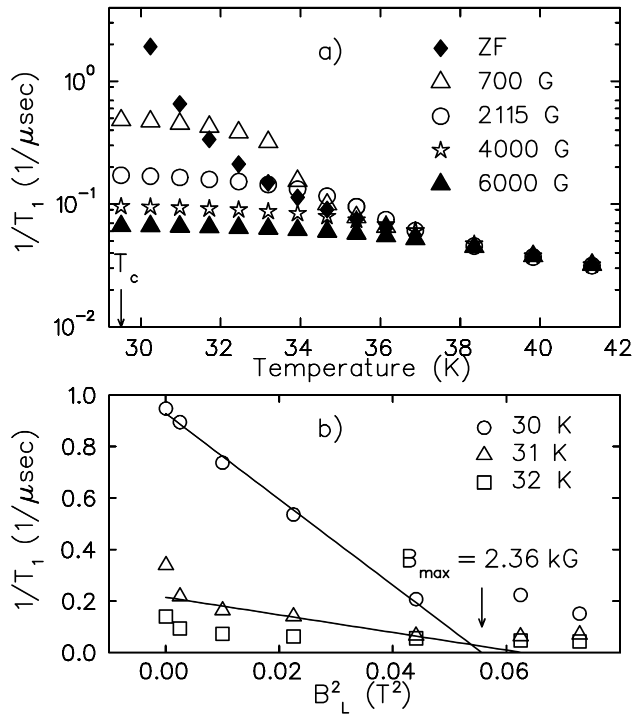

FIG. 3. (a) Temperature and magnetic field dependence of the relaxation rate calculated using the SCR model for a weak ferromagnet. (b) The measured relaxation rate at 30–32 K plotted versus the square of the applied magnetic field.

To explain this behavior we propose a simple model which takes into account two distinct contributions to muon spin relaxation: (a) the $q = 0$ component of spin fluctuations along the direction of the external magnetic field and (b) the spin fluctuations of the remaining helical component in the plane perpendicular to the magnetic field. Accordingly, the muon spin relaxation rate in MnSi can be expressed as $1/T_1 = (1/T_1)_{\text{parallel}} + (1/T_1)_{\text{perp}}$. In view of $1/T_1 \propto \omega^2/\nu$ and $\omega = \gamma_\mu B_{\text{int}}$, the first term should be proportional to the component S_z^2 of Mn spin fluctuations parallel to the magnetic field applied along the z direction and the second term proportional to $S_{x,y}^2$. Here we assume that $S_z^2 \propto B_L^2/B_{\max}^2$ and $S_{x,y}^2 \propto (B_{\max}^2 - B_L^2)/B_{\max}^2$, where $B_{\max}$ denotes the value of B_L required to eliminate the helical component of critical spin fluctuations.

In the ordered state at $T = 4.2$ K, the external field $B_L = 6.2$ kG is sufficient to eliminate the helical component, aligning all the spins ferromagnetically, as shown in Fig. 1(a). Although it is difficult to have an accurate *a priori* estimate for $B_{\max}$ in the paramagnetic state, we expect that $B_{\max}$ is of the order of magnitude of 6.2 kG. According to Ref. [13], lack of inversion symmetry in an itinerant ferromagnet could create a long-period helical spin density wave. This might explain the helical spin fluctuations in MnSi persisting above T_c.

Since the critical behavior due to ferromagnetic spin fluctuations is suppressed above $T = 30$ K at

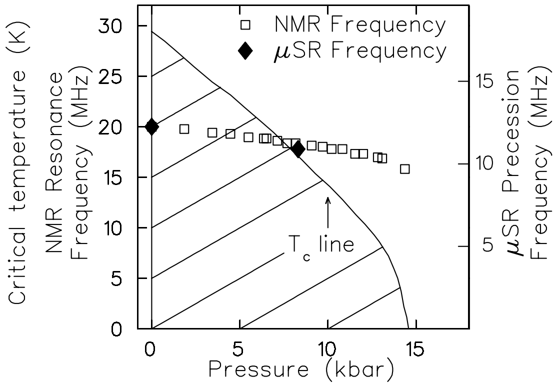

FIG. 4. Pressure dependence of the NMR frequency and T_c [14] in MnSi. The present μSR result at 5 K is plotted with the rhombic symbol together with the lowest temperature precession frequency obtained by Kadono *et al.* [8].

$B_L \geq 700$ G, as shown in Fig. 3(b), we assume that $(1/T_1)_{parallel}$ is negligibly small above $T = 30$ K. Then, the observed value of $1/T_1$ would solely reflect the contribution from helical spin fluctuations, leading to $1/T_1 \propto (B_{max}^2 - B_L^2)$.

In Fig. 3(b) we show the observed relaxation rate $1/T_1$ as a function of B_L^2 at 30, 31, and 32 K. The results at 31 and 32 K are not quite conclusive, but the magnetic field dependence of $1/T_1$ at 30 K confirms the above prediction. From the linear fit we obtained $B_{max} = 2.36$ kG. The fit for the data at 31 K is also consistent with this value of B_{max}, except for the lowest field point. As can be seen in Fig. 1(a), the magnetic field of 2 to 3 kG near T_c would alter the ordered state from helical to ferromagnetic. Therefore, the value of B_{max} obtained in our experiment seems reasonable. This result indicates that even in the paramagnetic state above T_c, the system "knows" to which ordered structure it will transform with decreasing temperature for a given value of B_L. The application of uniform field B_L suppresses the spin fluctuation of the $q = 0$ component selectively, while the helical critical fluctuations survive up to $B_L = B_{max}$.

Another interesting issue of the SCR theory is the crossover from itinerant ferromagnet to a correlated paramagnet. Since MnSi embodies this crossover in applied pressure, extensive studies have been performed by magnetic susceptibility, resistivity [6], and NMR [14] measurements to explore regions near the quantum critical point at p_c. Figure 4 shows the published results for the pressure dependence of T_c and of M_0 estimated from the ^{29}Si NMR frequency [14]. The use of single crystal specimens in μSR would assure homogeneity of the applied pressure, while powder specimens of NMR could be subject to possible inhomogeneity of pressure.

In the ordered phase, we measured the muon precession frequency under zero magnetic field. The precession disappears completely at 17 K, where T_1 showed minimum in Fig. 2(b). Our result of $T_c = 17$ K for 8.3 kbar is consistent with $T_c(p)$ in Fig. 4. We distinguished two precession frequencies. The results for the higher frequency have a large systematic error due to limited statistics and small precession amplitude compared to the large background signal from the pressure cell. In Fig. 4, we plot our results of the lower frequency under 8.3 kbar at $T = 5$ K, together with the value at ambient pressure. Our results confirm earlier NMR results and show that magnetic moment M_0 decreases much more slowly with increasing pressure, compared to the reduction of T_c. This result might be related to the first-order nature of the phase transition at p_c [6,14].

In conclusion, we presented new sets of μSR measurements in MnSi and elucidated the effect of the applied field on the critical behavior observed via the relaxation rate $1/T_1$. We also found near T_c a departure of $1/T_1$ from the linear behavior $T_1T \propto (T - T_c)$ and confirmed the very small pressure dependence of the magnetic moment in the ordered phase.

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[1] Y. Ishikawa, K. Tajima, D. Bloch, and M. Roth, Solid State Commun. **19**, 525 (1976).
[2] Y. Ishikawa, G. Shirane, J. A. Tarvin, and M. Kohgi, Phys. Rev. B **16**, 4956 (1977).
[3] E. Fawcett, J. P. Maita, and J. H. Wernick, Int. J. Magn. **1**, 29 (1970).
[4] H. Yasuoka, V. Jaccarino, R. C. Sherwood, and J. H. Wernick, J. Phys. Soc. Jpn. **44**, 842 (1978).
[5] T. Moriya, *Spin Fluctuations in Itinerant Electron Magnetism*, Springer Series in Solid State Sciences Vol. 56 (Springer, Heidelberg, 1985).
[6] C. Pfleiderer, G. J. McMullan, S. R. Julian, and G. G. Lonzarich, Phys. Rev. B **55**, 8330 (1997).
[7] R. Hayano *et al.*, J. Phys. Soc. Jpn. **49**, 1773 (1980).
[8] R. Kadono, T. Matsuzaki, T. Yamazaki, S. R. Kreitzman, and J. H. Brewer, Phys. Rev. B **42**, 6515 (1990).
[9] M. Takigawa *et al.*, J. Phys. Soc. Jpn. **49**, 1760 (1980).
[10] R. Kadono *et al.*, Phys. Rev. B **48**, 16 803 (1993).
[11] A. Ishigaki and T. Moriya, J. Phys. Soc. Jpn. **65**, 3402 (1996).
[12] A. Ishigaki and T. Moriya, J. Phys. Soc. Jpn. **67**, 3924 (1998).
[13] O. Nakanishi, A. Yanase, A. Hasegawa, and M. Kataoka, Solid State Commun. **35**, 995 (1980).
[14] C. Thessieu, Y. Kitaoka, and K. Asayama, Physica (Amsterdam) **259-261B**, 847 (1999).