Observation of an itinerant metamagnetic transition in MnSi under high pressure

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Magnetization measurements have been performed on single-crystalline MnSi under high pressures up to 1.54 GPa and high magnetic fields up to 10 T in the temperature range from 1.7 to 60 K. The magnetization process exhibits no hysteresis in the pressure region up to 1.34 GPa. At $P = 1.54$ GPa, we have clearly observed a metamagnetic transition around $B = 0.1$ T with hysteresis in the magnetization process at low temperatures below 6 K. From the experimental results, we have determined the magnetic phase diagram of MnSi for $P = 1.54$ GPa.

INTRODUCTION

The intermetallic compound MnSi has the $B20$-type crystal structure with a lattice parameter of 4.558 Å. At ambient pressure, MnSi in zero magnetic field has a helical spin structure with a long period of 180 Å along the [111] direction in the ordered state below the temperature $T_c = 29$ K.[1–3] The helical spin structure originates from the presence of an antisymmetric interaction called the Dzyaloshinski-Moriya interaction[4,5] together with the main ferromagnetic interatomic exchange.[6,7] In a magnetic field larger than 0.15 T, a conical spin structure is stabilized. With increasing field, the cone angle becomes small and an induced ferromagnetic state is realized at $B = 0.62$ T for $T = 1.4$ K.[1–3] The spontaneous magnetic moment of $0.4\mu_B$/Mn is much smaller than the effective magnetic moment of $1.4\mu_B$/Mn determined in the paramagnetic region.[8,9] The magnetic susceptibility above T_c obeys the Curie-Weiss law.[8,9] These results suggest that MnSi is a typical weak itinerant helical magnet.

The application of high pressure suppresses the magnetism of MnSi and the value of T_c decreases with increasing pressure. The magnetic order disappears at the critical pressure of $P_c \approx 1.5$ GPa.[9–14] Although the magnetic transition at T_c is second order at low pressure, a crossover from the second-order to a first-order transition was observed at $P \approx 1.2$ GPa by means of ac susceptibility measurements.[14] The temperature dependence of the susceptibility exhibits a broad maximum just above T_c in the first-order region $1.2 < P < 1.5$ GPa. In the paramagnetic region $P > 1.5$ GPa, a broad maximum was also observed. This indicates that MnSi above P_c is a nearly ferromagnetic exchange-enhanced Pauli paramagnet. Similar magnetic behavior under high pressure was found in itinerant ferromagnets $Co(S_{1-x}Se_x)_2$,[15–18] where itinerant electron metamagnetism, that is, a first-order field-induced transition from the paramagnetic to the ferromagnetic state in the ground state, was observed in the paramagnetic pressure region. These experimental results suggest that the itinerant metamagnetic transition may appear in MnSi under high pressure above P_c. Typical itinerant metamagnetic transitions were found in the Co-based Laves phase compounds $Y(Co_{1-x}Al_x)_2$ (Refs. 19 and 20) together with paramagnetic $Co(S_{1-x}Se_x)_2$.[15–17] Every compound with the metamagnetic transition exhibits a broad maximum in the temperature dependence of the susceptibility. The metamagnetic transition and the broad maximum of susceptibility are considered to originate from a peculiar shape of the density-of-states curve in the vicinity of the Fermi level. Recently, theoretical studies of the metamagnetic transition have been developed based on a spin-fluctuation model.[21–25]

In some Kondo antiferromagnets such as $CePd_2Si_2$,[26,27] $CeIn_2$,[29,28] and Ce_7Ni_3,[30,31] the antiferromagnetic ordering was found to be suppressed by the application of high pressure, and the ordering temperature becomes zero at the critical pressure. In the vicinity of the critical pressure, the physical properties are quite different from those predicted by the Fermi-liquid theory.[10,27,28] In MnSi, the resistivity was also found to show a non-Fermi-liquid behavior around P_c. Therefore, the magnetic behavior of MnSi around P_c is of much interest as in Kondo antiferromagnets. Many experimental studies have been performed under high pressure to examine the magnetic behavior of MnSi around P_c by means of the measurements of resistivity,[10–13,32] ac susceptibility,[14,33–37] NMR,[38] and magnetization.[39] Recent data on the ac susceptibility reported by Thessieu and co-workers and Pfleiderer suggest that a field-induced transition may exist around $B = 0.4$ T at a pressure of 1.55 GPa.[33–37] Although Thessieu and co-workers tried to measure the magnetization process at high pressures up to 1 GPa,[39] nobody, to our knowledge, has directly measured the magnetization process near P_c. In this study, we have performed magnetization measurements of MnSi with a single-crystalline sample in magnetic fields up to 10 T at high pressures up to 1.54 GPa (just above P_c) to confirm the appearance of a field-induced metamagnetic transition.

EXPERIMENT

A single crystal of MnSi was grown by a Czochralski technique with the stoichiometric composition in an argon atmosphere. As starting materials, we used manganese of

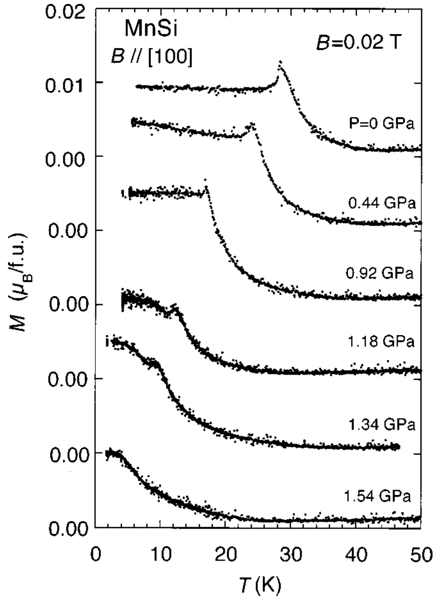

FIG. 1. Temperature dependence of the magnetization of MnSi at various pressures in a magnetic field $B=0.02$ T along the [100] axis.

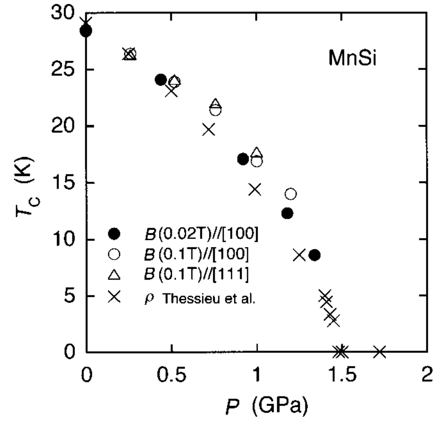

FIG. 2. Pressure dependence of the Curie temperature of MnSi in magnetic fields 0.02 T along the [100] direction and 0.1 T along the [100] and [111] axes. The crosses indicate the data of Thessieu *et al.* obtained from resistivity measurements (Ref. 13).

99.99% purity and silicon of 99.999% purity. The crystal was confirmed to have the cubic $B20$-type structure without secondary phases by powder x-ray diffraction with Cu $K\alpha$ radiation. The lattice parameter was estimated to be 4.561 Å. In order to check the sample quality, we measured the resistivity using a standard four-probe technique at ambient pressure in the temperature range from 4.2 to 290 K. The values of residual resistivity $\rho(T\rightarrow 0)$ and ratio $\mathcal{R}=\rho(T=290\,\mathrm{K})/\rho(T\rightarrow 0)$ are 2 $\mu\Omega$ cm and 130. The principal axes of the single crystal were determined by the x-ray backscattering Laue technique. The crystal was then cut into a small pillar with a size of $3\times 1\times 1$ mm^3 along the a axis.

The magnetization was measured in the pressure range up to 1.54 GPa using an extraction-type magnetometer with a nonmagnetic high-pressure clamp cell made of nonmagnetic CuTi alloy in magnetic fields up to 10 T produced by a 20-T superconducting magnet.[40] Here, we have analyzed the magnetization curves by considering a demagnetizing effect based on a spheroid approximation.[41] Although the pressure produced by the original cell is limited to 1.3 GPa, in this study, the maximum pressure was increased to 1.6 GPa by means of autofrettaging.[42] The produced pressure around 4 K, $P_{4\,\mathrm{K}}$, is calibrated by means of the Meissner effect of Sn for which the pressure dependence of the superconducting transition temperature is known to high accuracy.[43] The value of $P_{4\,\mathrm{K}}$ increases linearly with the load pressure P_{RT} applied at room temperature and is given by $P_{4\,\mathrm{K}}=1.10\times P_{\mathrm{RT}}-0.325$ GPa in the pressure region up to 1.6 GPa. In the experimental temperature range between 1.7 and 50 K, the change of the pressure produced in the high-pressure cell is negligibly small.

RESULTS AND DISCUSSION

We show in Fig. 1 the temperature dependence of the magnetization of MnSi at various pressures in a magnetic field $B=0.02$ T in the [100] direction. The magnetization was measured in an increasing temperature scan. The value of the transition temperature T_c, which is estimated to be 28.5 K at $P=0$ GPa, decreases with increasing pressure. It is noticed that the M-T curve shows a sharp peak and no hysteresis around T_c in the low-pressure region $P<1.18$ GPa, which are characteristics of a typical second-order ferromagnetic transition. In the high-pressure region $P>1.18$ GPa, the peak becomes broad. Pfleiderer *et al.* measured the ac susceptibility χac of MnSi in an ac magnetic field of $\sim$0.1 mT under high pressure. They found a crossover from the second-order to the first-order transition at about 1.2 GPa and the disappearance of magnetism at $P_c=1.46$ GPa.[14] The observed χac-T curve has a sharp peak at T_c in the second-order region $P<1.2$ GPa. In the first-order region $1.2<P<1.46$ GPa, however, they observed no sharp peak at T_c. The behavior of χac around T_c is consistent with that of the magnetization shown in Fig. 1. The χac-T curve in the paramagnetic temperature range has a broad maximum around 12 K for $P>1.2$ GPa, the position of which is weakly pressure dependent.[14] However, we cannot observe the broad maximum of the susceptibility at high pressures of 1.34 and 1.54 GPa. Pfleiderer found that the broad maximum is hidden by a small amount of magnetic impurities.[44] He used a single crystal $[\rho(T\rightarrow 0)=0.35\ \mu\Omega$ cm and $\mathcal{R}=248]$ investigated in a de Haas–van Alphen study for ac susceptibility measurements.

Figure 2 shows the pressure dependence of the transition temperature of MnSi in magnetic fields of 0.02 T and 0.1 T along the [100] direction and 0.1 T along the [111] direction. The experimental data obtained from electrical resistivity measurements by Thessieu *et al.*[13] are also plotted for comparison. There is no difference between the values of T_c for the [100] and [111] directions measured in a magnetic field of 0.1 T. However, the data for $B=0.1$ T deviate more from those of Thessieu *et al.* especially near P_c than those for $B=0.02$ T. This result indicates that T_c is sensitively affected by the magnetic field near P_c and is increased by a magnetic field 0.1 T. The initial pressure dependence of $T_c(P<0.5\,\mathrm{GPa})$ is estimated to be $d\ln T_c/dP=-0.38\,\mathrm{GPa}^{-1}$ from these data. Since the bulk modulus of MnSi is $\kappa=|d\ln V/dP|^{-1}=7.35\times 10^{-3}$ GPa at 50 K and is nearly con-

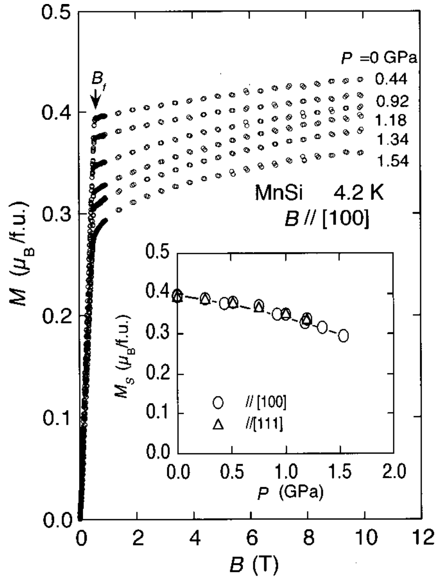

FIG. 3. Magnetization curves of single crystalline MnSi for 4.2 K at various pressures up to 1.54 GPa along the [100] axis in magnetic fields up to 10 T. The inset shows the pressure dependence of the magnetic moment M_S in the induced ferromagnetic state at 4.2 K.

stant in the experimental temperature range 4.2–50 K,[45] we can estimate the volume dependence of T_c to be $d\ln T_c/d\ln V=52$. This value is in good agreement with previous data.[9] As the pressure approaches P_c the critical temperature T_c decreases more abruptly.

Figure 3 shows the magnetization curves of single-crystalline MnSi for 4.2 K at various pressures up to 1.54 GPa in magnetic fields up to 10 T along the [100] axis. The magnetization curve for $P=0$ GPa exhibits a sharp kink at $B_f=0.55\,\mathrm{T}$ and the magnetization increases linearly with field. The critical field B_f corresponds to a magnetic transition from the conical state to the induced ferromagnetic one at ambient pressure. All the magnetization curves are not completely saturated even in a magnetic field of 10 T. At high pressures $P>0.92\,\mathrm{GPa}$, the magnetization increases nonlinearly above B_f. We determined the magnetic moment M_S in the induced ferromagnetic state from the extrapolation of the value of magnetization above B_f to zero field. The pressure dependence of M_S is shown in the inset of Fig. 3. There is no anisotropy for M_S. The value of M_S is $0.39\mu_B/\mathrm{Mn}$ at $P=0$ GPa and decreases gradually with increasing pressure up to 1.54 GPa, although the disappearance of magnetism occurs at $P_c=14.6\,\mathrm{GPa}$. We estimated the initial pressure dependence of M_S for $P<1.0\,\mathrm{GPa}$ to be $d\ln M_S/dP=-0.122\,\mathrm{GPa}^{-1}$ and obtained the volume dependence of M_S $d\ln M_S/d\ln V=16$ using the same value of the bulk modulus. This value is also consistent with previous data.[9,39]

We show in Fig. 4 the magnetization processes for 4.2 K in the low-field region at various pressures. The magnetization for $P=0$ GPa increases nonlinearly with magnetic field up to $B_c=0.1\,\mathrm{T}$ and then nearly linearly up to $B_f=0.55\,\mathrm{T}$. The critical field B_c corresponds to a magnetic transition from the helical state to the conical one.[1–3] As already described, the induced ferromagnetic state is realized above

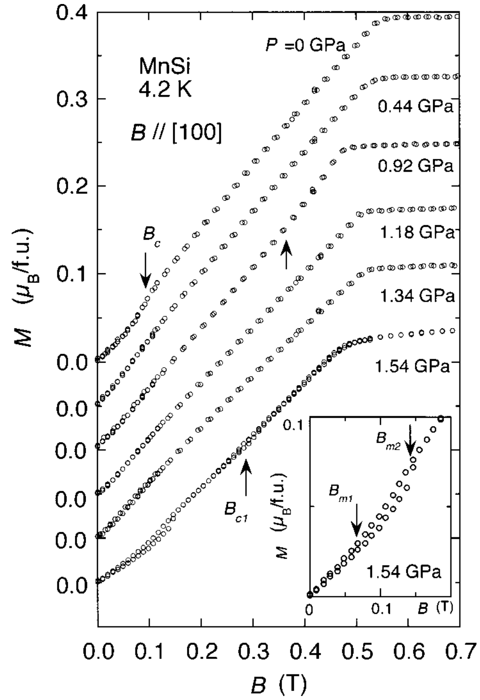

FIG. 4. Magnetization curves of MnSi for 4.2 K at various pressures up to 1.54 GPa along the [100] axis in magnetic fields up to 0.7 T. The inset shows the magnetization curve at 1.54 GPa in magnetic fields up to 0.2 T.

B_f. With increasing pressure, the magnetic transition at B_c becomes less sharp. It is interesting to note that some transition occurs at 0.92 GPa in the field region between B_c and B_f. In the pressure region $P\leqslant 1.34\,\mathrm{GPa}$, the magnetization curve has no hysteresis. At 1.54 GPa, however, a new transition appears with a hysteresis of about 0.02 T in a low-field region between $B_{m1}=0.07\,\mathrm{T}$ and $B_{m2}=0.14\,\mathrm{T}$, as shown in the inset. Furthermore, we found another field-induced transition at a higher field $B_{c1}=0.28\,\mathrm{T}$, which also has hysteresis. In order to show this transition more clearly, we indicate in Fig. 5 the magnetization process measured in increasing fields. The induced ferromagnetic state is realized at about 0.48 T. The quantitative shape of the magnetization curves implies that the wavelength of the spin structure in

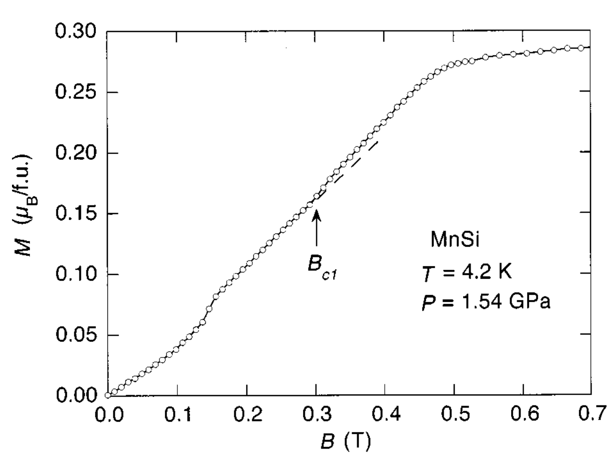

FIG. 5. Magnetization curve of MnSi for 4.2 K at 1.54 GPa measured in increasing fields. The transition at B_{Cl} can clearly be seen.

the helix or conical states is not dramatically changed as a function of pressure, because the system apparently is not clearly driven toward antiferromagnetism with increasing pressure.

Thessieu *et al.* and Pfleiderer suggested from ac susceptibility measurements that a metamagnetic transition with small hysteresis occurs around 0.4 T at 1.55 GPa.[33–35] They pointed out that the origin of the hysteresis could be either itinerant electron metamagnetism or the field-induced onset of the helical phase.[33,35,37] For the latter, the transition between the helical and the conical phase at ambient pressure implies first order. The metamagnetic transition reported by them may correspond to the second transition, which we found at 0.31 T.

Regarding the first metamagnetic transition between B_{m1} and B_{m2}, we believe that this transition occurs just above transition temperature near the critical pressure. For CoS_2 under 0.77 GPa, Goto *et al.* also showed an itinerant-electron metamagnetic transition with small hysteresis at $\sim$0.1 T just above transition temperature (117 K), whereas the transition temperature did not vanish.[15] In addition, the results of the χac-T measurements in the vicinity of the critical pressure suggest that MnSi in zero field at 1.54 GPa is an exchange-enhanced Pauli paramagnet.[14] Therefore, we consider that the transition with hysteresis observed at 1.54 GPa, which can be seen clearly in the inset of Fig. 4, is an itinerant metamagnetic transition. Typical itinerant electron metamagnets exhibit a sharp transition from the paramagnetic to the ferromagnetic state in magnetic fields, such as CoS_2 under 0.77 GPa.[15] We consider that the transition in MnSi at 1.54 GPa occurs from the paramagnetic to a conical state since the Dzyaloshinski-Moriya interaction exists. Since the pressure is slightly higher than the critical pressure P_c, the metamagnetic transition field will be very low.

It should be mentioned that recent studies of the zero-field NMR for MnSi are claimed to show the remaining of the local moments above the critical pressure.[34] However, the results of our dc magnetization and the previous χac-T measurements[14] suggest that there is a nonmagnetic state of MnSi in zero field above P_c. Therefore, the local moments reported by the NMR may be due to disorder of the powder samples and/or strong pressure gradients.

In order to examine the magnetic behavior of MnSi near P_c, we measured the magnetization curves at various temperatures and the M-T curves at various magnetic fields under a high pressure of 1.54 GPa. Figure 6 shows the magnetization curves at low temperatures from 1.7 to 20 K. With increasing temperature, the metamagnetic transition appearing between B_{m1} and B_{m2} is smeared and the hysteresis of the transition disappears at 7 K. The transition field to the induced ferromagnetic state B_f decreases slightly with increasing temperature, and the transition becomes broad above 10 K.

The temperature dependence of the magnetization for 1.54 GPa at various fields is shown in Fig. 7. We determined the position of T_c from the inflection point on the M-T curve. The transition has no hysteresis around T_c, which indicates that it is second order in these magnetic fields. Using the data of B_{m1}, B_{m2}, B_{c1}, B_f, and T_c, we determined the magnetic phase diagram of MnSi for 1.54 GPa in the B-T plane, as shown in Fig. 8. From the above discussion,

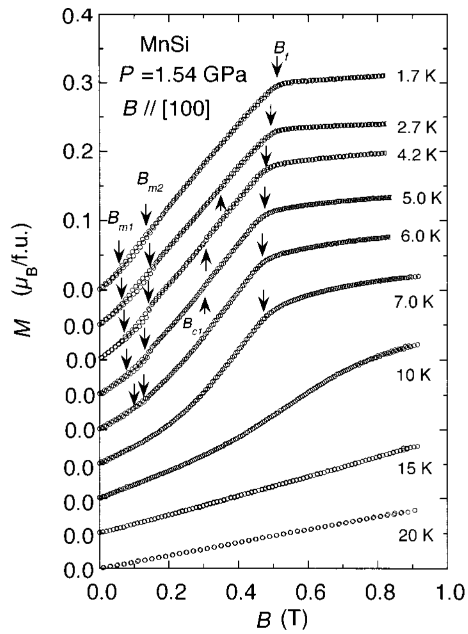

FIG. 6. Magnetization curves of MnSi along the [100] axis for 1.54 GPa in magnetic fields up to 1 T at various temperatures from 1.7 to 20 K.

the four magnetic phases are considered as follows: $M1$ is a paramagnetic phase, $M2$ is a conical phase, $M3$ is a new magnetic phase, and $M4$ is an induced ferromagnetic phase. The obtained phase diagram substantially corresponds to the one deduced from ac susceptibility measurements at 1.55 GPa.[30,33] However, we determined exactly the metamagnetic transition region from direct magnetization measurements at high pressure.

Here, we briefly discuss our results using a theory of itinerant electron metamagnetism at finite temperature based on a spin-fluctuation model.[21–25] According to the theory, the equation of state for the itinerant electron system with the metamagnetic transition is written in the ground state (T $=0$ K) as

$$B(M)=aM+bM^3+cM^5, \tag{1}$$

where a is the inverse susceptibility. The coefficients b and c are expressed in terms of the density of states (DOS) and its

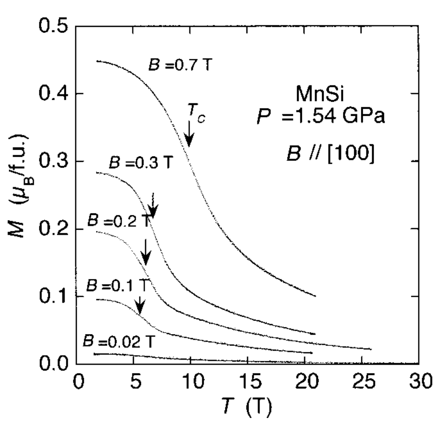

FIG. 7. Temperature dependence of the magnetization of MnSi for 1.54 GPa in various magnetic fields along the [100] axis.

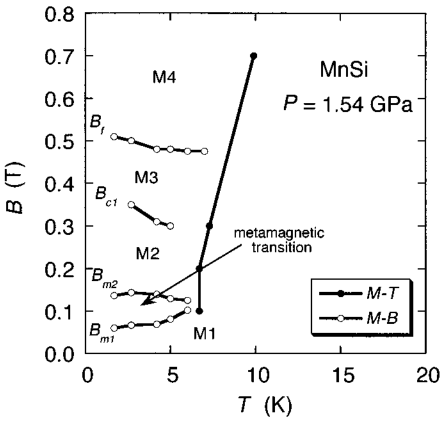

FIG. 8. Magnetic phase diagram of MnSi determined from magnetization measurements at 1.54 GPa.

derivatives at the Fermi energy E_F.[21] In this theory, the condition $a>0$, $b<0$, and $c>0$ is assumed. At finite temperature $(T>0)$, the coefficients a, b, and c are renormalized by thermal spin fluctuations and become temperature dependent, $a(T)$, $b(T)$, and $c(T)$, where $a(0)=a$, $b(0)=b$, and $c(0)=c$. Moriya determined the magnetic phase diagram of the system.[23] In the region of $ac/b^2<\frac{3}{16}$, ferromagnetism is stabilized. It should be noticed that this ferromagnetic condition ($a>0$, $b<0$, $c>0$, and $0<ac/b^2<\frac{3}{16}$) contradicts the Stoner condition for the appearance of ferromagnetism ($a<0$). The ferromagnetic transition is second order for $ac/b^2<\frac{5}{28}$, while it is first order for $\frac{5}{28}<ac/b^2<\frac{3}{16}$. Yamada[24] pointed out that just above T_c in the first-order ferromagnetic region, a metamagnetic transition appears, and the paramagnetic susceptibility exhibits a broad maximum. In the region of $\frac{3}{16}<ac/b^2<\frac{9}{20}$, the ground state is paramagnetic and itinerant electron metamagnetism occurs. The susceptibility in this region shows also a broad maximum. No metamagnetic transition appears for $ac/b^2>\frac{9}{20}$.[24]

The equation of state for MnSi cannot be described with Eq. (1) because the Dzyaloshinski-Moriya interaction exists. However, we consider that the magnetic behavior of MnSi under high pressure can be explained by the above theory because the main ferromagnetic interaction among the Mn moments is larger than the Dzyaloshinski-Moriya interaction. Since the magnetic transition at T_c is second order at ambient pressure and changes into the first-order one at 1.2 GPa, the value of ac/b^2 for MnSi is thought to be slightly smaller than $\frac{5}{28}$. The application of high pressure makes the $3d$ band wider, resulting in the increase of the parameter a (the decrease of the susceptibility) due to the decrease of DOS at E_F. Since the changes of b and c are smaller than that of a, the value of ac/b^2 increases with pressure. In the first-order transition region of $1.2<P<1.5$ GPa, the value of ac/b^2 is in the range of $\frac{5}{28}<ac/b^2<\frac{3}{16}$, where the transition temperature T_c is expected to be abruptly reduced by the application of high pressure.[23] In fact, T_c in the first-order pressure region is rapidly decreased and becomes zero around 1.5 GPa as shown in Fig. 2.[10,12–14] The pressure effect on T_c is extremely large. Above 1.5 GPa, the value of ac/b^2 is considered to become slightly larger than $\frac{3}{16}$ and the field-induced metamagnetic transition occurs. The observation of a broad maximum of the susceptibility in the region $P>1.2$ GPa is also consistent with the theory.

Recently, Yamada and Terao theoretically studied the magnetic properties of MnSi under high pressure on the basis of the electronic-structure calculation and the spin-fluctuation theory.[46] The obtained results are as follows: (1) the maximum of susceptibility is nearly independent of pressure, (2) the first-order ferromagnetic transition occurs in a narrow range of pressure just below the critical pressure for the disappearance of ferromagnetism, (3) a metamagnetic transition appears in a narrow range of the lattice constant near the critical lattice constant, which corresponds to the critical pressure. These results are qualitatively consistent with the present and previous studies.[45] However, the calculated magnetization processes for $T=0$ K near the critical lattice constant shows a sharp metamagnetic transition from the paramagnetic to the ferromagnetic state, which is in disagreement with our experimental data (from the paramagnetic to a conical state). This is because the calculation does not take into account the Dzyaloshinski-Moriya interaction.

In summary, we have performed direct magnetization measurements on single-crystalline MnSi under high pressures and high magnetic fields. At 1.54 GPa, we have clearly observed a metamagnetic transition around 0.1 T with hysteresis at low temperatures below 7 K. From the obtained results, we have determined the magnetic phase diagram of MnSi for 1.54 GPa just above the critical pressure.

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