

Magnetization under High Pressure in MnSi

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Abstract

The magnetization $M(H)$ has been measured in the weakly helimagnetic itinerant compound MnSi under high pressure up to 10.2 kbar and high magnetic field up to 9 Tesla. We interpret the simultaneous decrease under pressure of the saturated magnetization, p_s , and the Curie temperature, T_c in the frame of the self-consistent renormalization theory (SCR) of spin fluctuations. From the analysis of the so-called Arrot-plot ($H/p[H, T]$ versus $p^2[H, T]$) and the respective volume dependence of p_s and T_c , we estimate the evolution of the characteristic spin fluctuation temperatures, T_0 and T_A when the system approaches its critical pressure, $P_c=15$ kbar, corresponding to the disappearance of the long range magnetic order at $T = 0$.

68.35.Rh, 74.25.Ha, 75.62.Fj, 75.30.Kz

I. INTRODUCTION

MnSi is a well-known weakly helimagnetic system which has given a lot of experimental supports to the Self Consistent Renormalized spin fluctuations theory (referred henceforth as the SCR theory) developed by Moriya and co-workers (for a general review see Ref. 1). At ambient pressure both macroscopic (electrical resistivity, magnetic susceptibility, magnetization)^{2,3} and microscopic properties (nuclear magnetic relaxation and neutrons scattering)^{4,5} are quantitatively well interpreted in the frame of a SCR treatment of the magnetic spin fluctuations. Although the SCR theory furnishes a phenomenological treatment of the long wavelength-low energy spin-fluctuations, it successfully explains the low value of the Curie temperature $T_c=29.4$ K, the T^2 low temperature thermal variation of the electrical resistivity $\Delta\rho = AT^2$, the high temperature constant value exhibited by the spin-lattice relaxation rate T_1 , and above all the puzzling Curie-Weiss like behaviour of the paramagnetic susceptibility, $\chi^{-1}(T) \propto (T - T_c)$. Magnetization measurements under pressure in this compound have already been reported in a previous study up to 5.2 kbar by Bloch et Al.⁷. However since that time several studies have shown the crucial role played by the hydrostatic pressure on the magnetic ground state of this compound⁸⁻¹⁰. In particular, the existence of a critical pressure, $P_c=15$ kbar, where the system undergoes a magnetic to non magnetic transition at $T = 0$ (Quantum Phase Transition) has been pointed out^{8,10}. Recently, in the vicinity of this quantum critical point, it has been interestingly shedded light on the fact that the disappearance of the long range helimagnetic order was accompanied by the occurrence of the so-called *Itinerant Electron Metamagnetic* transition under an external magnetic field¹¹. This first order magnetic phase transition is a common feature shown by other 3d itinerant magnetic systems, especially when the external temperature independent parameter tuning the quantum magnetic instability is the chemical substitution, x , ($Y(\text{Co}_{1-x}\text{Al}_x)_2$ ¹², $\text{Co}(\text{Se}_{1-x}\text{S}_x)_2$ ¹³) and more seldom the hydrostatic pressure, P , (CoS_2 ¹⁴). Although this magnetic phenomena has been largely enlighthened by various experimental works, its theoretical treatment is actually not unanimously recognized with notably the

ambiguity between the role played by the band structure effect at the Fermi level¹⁵ and the influence of an external parameter on the spin fluctuations spectrum¹⁶. In view of this unclear situation, it appears interesting to extend the measurements of the magnetization properties in this compound up to the highest pressure actually available with our technique (11 kbar).

II. EXPERIMENTAL DETAILS

For this experiment a single crystal of MnSi with parallelepiped shapes and typical dimensions of $3 \times 0.3 \times 0.5$ mm (total weight of 29.4 mg) has been measured. The measurements were performed using an He⁴ cryostat in a temperature range from 4.2 K to 60 K. The external magnetic field was supplied by a 9 Tesla superconducting magnet and the weak magnetic anisotropy of MnSi has permitted to apply the external field in an undefined crystallographic direction. The hydrostatic pressure was generated by a non-magnetic clamp-type pressure cell of small dimensions with a maximal enable pressure of almost 11 kbar. The original Ti-Cu alloyed composition of the clamp core added to a set of ceramic-type pistons give a small diamagnetic contribution to the background signal allowing absolute value measurements under pressure even in a strong external magnetic field. The principle of the measure is based on the oscillation of the whole pressure cell in a two pick-up coils system. The signal is analyzed by the extraction method. This experiment has been carried out at the Institute for Solid State Physics in the University of Tokyo.

III. RESULTS

The Fig. 1 gives the evolution of the isothermal magnetization process at $T = 4.2$ K, for magnetic field up to 9 Tesla and under different applied pressures. At ambient pressure the magnetic features are well understood as a rotation of the helical structure along the field direction ($H_1 \sim 1$ kOe) followed by the disappearance of the magnetic domains in an unique conical phase ($1 \text{ kOe} < H < 6.2 \text{ kOe}$) orientated in the field-direction. Above $H_2 = 6.2$ kOe,

where the abrupt kink observed in the curve $p[H]$ corresponds to the transition in an induced ferromagnetic state, we observe an unsaturated ferromagnetic behavior. All these features remain unchanged under pressure and it is noteworthy that the value of the characteristic fields, H_1 and H_2 , remain unchanged under pressure. The insert of the Fig. 1 shows the magnetization behavior at $P = 10.2$ kbar for the both way of sweeping the magnetic field. Below 1 kOe a tiny hysteresis is observable and clearly in this region the magnetization does not respond linearly to the applied field. However above 1 kOe this hysteresis disappears and the magnetization corresponds to the conical phase and the unsaturated behavior of the induced ferromagnetic state above H_2 . These facts ensured a tangible proof that the heli-magnetic structure of MnSi persists even under high pressure (at least up to 10.2 kbar) and that no new magnetic structure transition is induced by pressure. By intercepting the high field regime of the magnetization at $H = 0$, we obtain the value of the saturated magnetization p_s , which at ambient pressure exhibits a value of $0.39 \mu_B/\text{Mn}$ in accordance with previous results⁷. From the Fig. 1, we see that the pressure effect is to lower the value of the magnetization and in particular leads to a decrease of the saturated magnetization value at $T = 4.2$ K, $p_s[H = 0, 4.2 \text{ K}]$. In order to extract a precise quantitative variation of $p_s[P]$ we have presented on the Fig. 2 the so-called Arrot-plot $H/p[H, T]$ versus $p^2[H, T]$ which gives a direct way to obtain this value. The axis have been drawn in a reduced units system such as the external magnetic field is expressed in energy units $h = 2\mu_B H$ and the magnetization per magnetic atom ($p = 2\frac{M(T)}{N_0}$, N_0 number of magnetic sites), is expressed in $2\mu_B$ units. To obtain the saturated magnetization p_s for each pressure we have estimated the x -axis intercept of the high magnetic field linear extrapolation. The evolution of p_s , reported in the insert of the Fig. 2, is surprisingly weak with pressure and this contrasts with the strong decrease of the corresponding Curie temperature T_c . It is noteworthy to underline that even close to the critical boundary ($P_c = 14.8$ kbar) the saturated magnetization still exhibits a high value. Quantitatively under an applied pressure of 10.2 kbar, the value of the saturated magnetization is only modified from 0.39 to $0.34 \mu_B/\text{Mn}$ atom whereas T_c drops from 29.4 K to 13.4 K (cf. table I for the detailed values). The weak decrease of the magnetization has

been confirmed recently by zero-field NMR measurements under pressure⁶ showing the weak decrease of the transferred hyperfine field of the Mn sites with pressure. In a good agreement with previous authors⁷ we find that $d \ln p_s / dP = -1.27 \cdot 10^{-2} \text{kbar}^{-1}$ ($-1.15 \cdot 10^{-2} \text{kbar}^{-1}$ in the case the previous results of Bloch et Al.). The Fig. 3 shows the isothermal $p[H]$ curves around T_c for a pressure of 10.2 kbar. No metamagnetic transition either hysteretic process are observed around T_c , in accordance with the fact that the transition is still of a second order nature up to 12.5 kbar⁹.

IV. DISCUSSION

In the frame of the SCR theory, the magnetic spin fluctuations are treated in a self consistent way and integrated in the magnetic equation of state. The nature of the spin fluctuations is therefore characterized by a small set of parameters expressed as p_s , \bar{F}_1 , T_0 and T_A . The two first parameters are determined by the knowledge of the magnetic equation of state given by the Landau development of the magnetic free energy in the presence of an external magnetic field

$$-\frac{2(\alpha - 1)}{\rho} + \bar{F}_1 p^2 = \frac{2\mu_B H}{p} \quad (1)$$

where $\alpha = I\rho$, I is the intra-site coulombian interaction, ρ the density of states at the Fermi level, p the magnetization expressed in $2\mu_B$ units (the saturated magnetization at $T = 0$, p_s is with this convention such as $p_s = 2M(0)/N_0$) and the parameter \bar{F}_1 is the coefficient of mode-mode coupling between magnetic spin fluctuations. Consequently, it is possible upon magnetization measurements and the analysis of the Arrot-plot defined with our units systems such as $2\mu_B H/p[H, T]$ versus $p^2[H, T]$, to obtain quantitative values for these two parameters. The two remaining parameters T_0 and T_A are directly related to the imaginary part of the generalized dynamic susceptibility $\text{Im}\chi(q, \omega)$. Neutrons measurements on MnSi have clearly shown that in the weakly ferromagnetic limit, $\text{Im}\chi(q, \omega)$ has the lorentzian form in a small region of q and ω close to the ferromagnetic instability vector $q = 0$ and is expressed as:

$$\text{Im}\chi(q, \omega) = \frac{\chi(0, 0)}{1 + \frac{q^2}{\kappa^2}} \times \frac{\omega\Gamma_q}{\omega^2 + \Gamma_q^2} \quad (2)$$

with $\chi(0, 0)$ the static uniform susceptibility, Γ_q the spectral width of the magnetic SF excitations given by:

$$\Gamma_q = \Gamma_0 q(\kappa^2 + q^2) \quad (3)$$

κ is the inverse of the magnetic correlation length. This power spectrum of the magnetic spin fluctuations, $\text{Im}\chi(q, \omega)/\omega$ is in fact characterized by two temperatures scales, T_0 and T_A , representing respectively the dispersion along the ω -axis and the q -axis

$$\begin{aligned} T_0 &= \Gamma_0 q_B^3 / 2\pi \\ T_A &= \frac{q_B^2}{2\kappa^2} \frac{N_0}{\chi(0)} \end{aligned} \quad (4)$$

The effective boundary vector q_B is given by $q_B = (6\pi^2/v_0)^{1/3}$ (v_0 the volume per magnetic atoms). Usually these two temperatures scales are obtained upon microscopic measurements such as nuclear magnetic resonance (spin-lattice relaxation rate and Knight shift) for T_0 and neutrons diffusion measurements for T_A . The SCR theory¹⁷ predicts a number of interesting crossed-relations between these different parameters. For instance, the mode-mode coupling term can be remarkably expressed as function of the two temperatures scales, T_0 and T_A

$$\frac{\bar{F}_1}{k_B} = \frac{4}{15} \times \frac{T_A^2}{T_0} \quad (5)$$

T_0 and T_A can also be expressed in the following way as function of T_c , p_s and the ratio \bar{F}_1/k_B

$$\begin{aligned} T_0^{5/6} &= \frac{10.334}{p_s^2} \left(\frac{k_B}{\bar{F}_1} \right)^{1/2} T_c^{4/3} \\ T_A^2 &= \frac{38.737}{p_s^2} \left(\frac{\bar{F}_1}{k_B} \right)^{1/2} T_c^{4/3} \end{aligned} \quad (6)$$

From the following expressions, we readily see that upon macroscopic measurements such as magnetization and the Arrot Plot analysis, giving access to the ratio \bar{F}_1/k_B , p_s and the knowledge of the ordering temperature T_c ^{8,10}, we can estimate quantitatively the values of

the energy scale of the spin fluctuations spectrum. The different values obtained upon the Arrot-plot analysis and the Eqs. (4) (Γ_0) and (6) (T_0 and T_A) are listed in the table I. In the Fig. 4, we have compared the pressure effect on the characteristic temperatures in MnSi, T_c , T_0 and T_A . As the pressure increases and the system is driven towards its quantum critical point, the energies T_0 and T_A change in an appreciable way, indicating that the spectra of the magnetic excitations is largely modified by pressure due to a strong volume dependence of $\text{Im}\chi(q, \omega)$ in this compound. Consequently, we can suppose that from both sides of the critical pressure, the spatial distribution of $\text{Im}\chi(q, \omega)$ is drastically modified and that a discontinuous variation of T_0 and T_A is expected, corresponding to a modification of the magnetic spin fluctuations regime when crossing the critical boundary from the electronic spin-polarized state to the paramagnetic Fermi liquid. In particular the sizeable decrease of the energy dispersion of the spin fluctuations modes, Γ_0 , presented in the Fig. 5 indicates that these latter are more drastically over-damped at the approach of P_c where the long range magnetic order has been shown to disappear and the system remains paramagnetic at all temperature. Upon our experimental results we can estimate that the pressure effect on the damping rate of the magnetic spin fluctuations should be in the order of

$$\frac{\partial \ln \Gamma_0}{\partial P} = -8.6 \text{ kbar}^{-1} \quad (7)$$

On the other hand, the slope of the Arrot-plot is almost not modified by pressure (Fig. 2) indicating that \bar{F}_1 , the mode-mode coupling term, has only a weak pressure dependence. We deduce therefore that the pressure modified the spin-fluctuations spectra distribution but not the coupling strength between these latter. This scenario is based on thermodynamic measurements and must be completed by microscopic measurements under pressure. However, in view of these experimental results, it appears important to verify the accuracy of this analysis by additional measurements like neutrons inelastic diffusion experiment to study the evolution of T_0 or T_A under pressure.

V. CONCLUSION

The analysis presented in this paper tends to show that the evolution of the spin fluctuations spectra with pressure plays a key role in the occurrence of the Itinerant Electron Metamagnetic phenomenon as advanced recently by Takahashi and Sakai. However the band structure effects induced by pressure can not be readily analyzed from this measurement. Thus we do not exclude that such effects at the Fermi level, as postulated by Yamada, are not engaged in the Itinerant Electron Metamagnetism process. Our work mainly points out that probably a more refined theoretical treatment is needed in order to take into account the effects of both thermal and zero-point spin fluctuations modes on the magnetic ground state of the system. It seems to us that in MnSi the combined facts that there exists a well-pronounced difference in the volume dependence of the ordering temperature, T_c , and the saturated magnetization, p_s , and the fact that in this itinerant electron system the phenomenon of metamagnetism has been observed are strongly related. We believed that the physical origin is due to a strong volume dependence of the magnetic spin fluctuations spectrum in particular close to P_c .

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FIGURES

FIG. 1. Isothermal magnetization process at $T = 4.2$ K under different applied pressures. For each experiments, the pressure cell and the sample have been cooled down in zero-field. The insert shows the magnetization process at the maximal pressure obtained in this experiment $P = 10.2$ kbar close to T_c . The non-linearity of the magnetization at low field ($H \simeq 1$ kOe) is attributed to the persistence of the helical magnetic structure at $P = 10.2$ kbar.

FIG. 2. Typical Arrot plot $h/p - p^2$ at $T = 4.2$ K. The intercept of the high field linear-fit with the x axis gives a direct value of the saturated magnetization p_s . The evolution of p_s as function of the external pressure is reported in the insert and can be compared with the pressure dependence of the ordering temperature T_c

FIG. 3. Magnetization process for the maximal pressure obtained in this study (10.2 kbar). For temperatures around $T_c = 13.4$ K, no metamagnetic either hysteresis phenomena have been observed. All the curves are presented for both way of sweeping the magnetic field.

FIG. 4. Pressure dependence of the characteristic temperatures T_0 and T_A related to the imaginary part of the generalized susceptibility $\text{Im}\chi(q, \omega)$. The error bars are mainly due to the fitting error in the high field regime of the Arrot-plot.

FIG. 5. The damping rate of the magnetic spin fluctuations Γ_0 shows a strong decrease in the vicinity of the critical boundary ($P_c = 14.8$ kbar). The error bars are calculated from the error on T_0 .

TABLES

TABLE I. Characteristic temperatures T_0 and T_A of the generalized susceptibility $\text{Im}\chi(q, \omega)$ calculated upon the macroscopic measurements $p[H, P]$

Pressure[kbar]	T_c [K]	$p_s[\mu_B/\text{Mn}]$	\bar{F}_1/k_B [K]	T_0 [K]	T_A [K]	$\Gamma_0[k_B\text{\AA}^3]$
Ambient	29.07	0.389	9055	146	2230	292
3.6	24.01	0.374	9314	117	2025	234
6.6	19.60	0.359	8899	97	1804	195
8.7	16.27	0.348	8507	78	1576	155
10.2	13.50	0.342	8401	60	1384	121