NMR Study of MnSi under Pressure

C. Thessieu*, K. Ishida*, Y. Kitaoka*, K. Asayama*, G. Lapertot†

* Department of Material Physics, Faculty of Engineering Science,
Osaka University, Toyonaka, Osaka 560

† Centre d’Études Nucléaires de Grenoble, SPSMS, Grenoble, France
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Abstract

The magnetic phase diagram $T_c(P)$ of the weak itinerant helimagnetic compound MnSi is reviewed upon a Nuclear Magnetic Resonance (NMR) experiment. We present a systematic study on the evolution of the NMR spin echo signal at $T = 1.4$ K of the non-magnetic silicone sites, $^{29}$Si, up to 17.8 kbar. The pressure effect is interpreted as a weak variation of the local electronic spin polarization and the coexistence of magnetic and non magnetic Mn atoms under pressure. From the volume dependence of $f_0(P)$, we show notably that a local magnetic order remains above the critical pressure of $P_c = 14.8$ kbar.

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

A lot of studies have shown that in the strongly correlated electronic systems (the heavy fermions systems with the 4f and 5f electrons as well as the transition metals with the wider energy band of the 3d electrons), the ground state is very sensitive to the inter-atomic distance between the magnetic atoms. Indeed the mechanisms are different depending on the system, the tune of the unit cell lattice parameters, the electronic bandwidth and the magnetic exchange energy by an external parameter $\delta$ (which can represent the chemical substitution, $x$, the applied pressure, $P$, or the applied magnetic field, $H$) give rises to a large variety of ground states (paramagnetism, magnetic order, superconductivity or spin glass state, ...). The magnetic phase diagram, $T_c(P)$, of the helimagnetic spin-polarised compound MnSi is well-known since a couple of years now and several authors have elucidated main of its macroscopic features. Under pressure the Curie temperature decreases monotonously from 30 K at ambient pressure down to 0 at the critical pressure of $P_c = 14.8$ kbar inducing a quantum critical phase transition (QCPT) from a spin polarised state to a paramagnetic state. However it remains at least two questions concerning the evolution of the magnetic properties of this compound under pressure. The first one concerns the evolution of the magnetization when the system approaches the quantum critical point. The second is related to the exact nature of the ground state just above $P_c$. We address the problem of the long range magnetic order disappearance under pressure from a microscopic standpoint in the case of a pure magnetic metal (i.e. without disorder effect induces by the chemical substitution). In this study we bring answers to these questions illustrating that the Nuclear Magnetic Resonance is a powerful tool to deal with the magnetic instability under pressure.

II. EXPERIMENTAL DETAILS

For this study a single crystal has been crushed in fine powder (grain sizes less than 20 $\mu$m) in order to ensure the penetration of the radiofrequency pulsed field. Our sample
has a residual resistivity ratio of 40 and a residual resistivity at low temperature of roughly 4 $\mu\Omega.cm$. Preliminarily, we have verified from electrical resistivity measurements (AC 4 terminals method) that the bulk sample exhibits the same magnetic features under pressure (e.g. variation of $T_c(P)$ and the disappearance of the magnetic order at 14.8 kbar) than the one reported in the literature\textsuperscript{1-3}. The pressure was generated by a "classical" clamp-type pressure cell composed of a Be-Cu outer and inner parts. The pressure transmitting medium was an equal proportion of fluorinert (type 77 and 70) which becomes solid at temperatures below 200 K but remains liquid at room temperature and in the pressure range of interest. Every cooling procedure was made slowly enough to avoid stresses in the transmitting medium and thus inhomogeneous pressure distribution around the sample. The pressure calibration at low temperature was based on the pressure shift of the superconducting transition temperature of a bulk tin sample measured by a 4 probes AC electrical resistivity measurement. The pressure has been changed in a very systematic way from ambient pressure to the highest pressure reached during this experiment (17.8 kbar) and we estimate that the accuracy in the relative pressure change is equal to $\pm 0.1$ kbar. The NMR experiment was performed under zero external field and at 1.4 K using a phase-coherent pulsed spectrometer. The NMR spectrum was obtained by plotting the spin-echo intensity as function of frequency with an 20 kHz interval. For each frequency the NMR conditions (mainly the pulses width whereas the radiofrequency field amplitude was kept constant) have been adjusted to maximize the spin echo amplitude.

III. RESULTS AND ANALYSIS

Previous NMR studies at ambient pressure have reported that in MnSi the $^{29}$Si NMR signal was observed around $f_0=19.880$ MHz in the helical magnetic state and at $T=1.4$ K\textsuperscript{5}. Below $T_c$, the magnetic moments at the Mn sites induces a polarization of the conduction electron (electrons of the s or p shells) at the Si sites. As a result the transferred hyperfine field acts on the Si nuclei through the Fermi contact term or the spin-dipole interaction.
From this experiment, the transferred hyperfine field was estimated to be 2.35 Tesla (ambient pressure) with the gyromagnetic ratio $^{29}\gamma = 8.4578 \text{ MHz/Tesla}$. Moreover the hyperfine coupling constant was calculated such as $A_{hf} = 2.35 \text{ Tesla} / 0.389 \mu_B = 60.42 \text{ kOe/} \mu_B$, which is compatible with the value obtained from the $\kappa - \chi$ plot in the paramagnetic state. This agreement assures that the Si NMR can probe the microscopic magnetic properties inherent to MnSi. By contrast, the Mn NMR spectrum below $T_c$ exhibits a very broad feature due to the combined effect of the helical magnetic structure and to a very large anisotropic hyperfine interaction. Thus the analysis of the magnetic properties under pressure from this nuclei standpoint is not an easy task. The Fig. 1 represents the pressure variation of $^{29}f_0$ at $T=1.4 \text{ K}$, from ambient pressure up to 16.1 kbar. $^{29}f_0$ decreases from 19.880 MHz at ambient pressure to 14.500 MHz for 16.1 kbar pressure above which the NMR signal disappears (for a pressure of 16.3 kbar no signal was observed). With increasing pressure, the full width at the half maximum (FWHM) increases from 200 kHz up to 1.4 MHz. It is noted that the general spectrum’s shape is not modified under pressure conserving a Gaussian-like shape.

In table I are summarized the pressure dependence of $T_c$, magnetization $M_s$ and $f_0$. The Curie temperature, $T_c$, is extracted from the anomaly observed in the $M$-$T$ curves (not shown in this paper), the saturated magnetization, $M_s$, from the Arrot plot. The hyperfine coupling constant, $A_{hf}$, is calculated such as $A^{Si}_{hf} = f_0/(\gamma \times M_s)$. There is a large contrast between the pressure effect on the Curie temperature and on the saturated magnetization or the frequency resonance and we underline that the theoretical variation predicted by the renormalized spin fluctuation theory $^{2} T_c \propto M_s^{3/2}$ is not well respected under pressure in the case of MnSi. The decrease in $M_s$ is scaled with the one of $f_0$. As a matter of fact, $M_s$ vs $^{29}f_0$ plotted with the pressure as an implicit parameter is fitted linearly and the hyperfine coupling constant is deduced to be volume independant.

Surprisingly we have observed the persistence of the spin echo signal even above $P_c=14.8 \text{ kbar}$ identified as the critical pressure at which the long range magnetic disappears from the bulk measurements. The fact that the Si NMR signal was observable at zero-field assures the persistence of a local magnetic order over $P_c$ from a microscopic point.
To estimate the spectrum intensity, the spin echo intensity was fitted by $I(\tau) = I(0) \exp(-2\tau/T_2)$ where $\tau$ is the time duration between the two pulses and $T_2$ the spin echo decay time and $I(0)$ is reported on the Fig. 2 as function of the frequency. The area of this spectrum has been calculated by fitting this latter with the superposition of two Gaussian functions. As a result it turns out that this area is proportional to the total fraction of Si nuclei included in magnetic domains. The integrated intensity of the NMR spectrum at 1.4 K decreases linearly in a range of 7.5 - 12.5 kbar followed by a weak pressure dependence up to 16.1 kbar. Unexpectedly exceeding 7.5 kbar, non-magnetic domains start to be induced by pressure. This results in a decrease of the NMR spectrum intensity. That is to say that the helicoidally ordered state is partially suppressed over 7.5 kbar to presumably change into paramagnetic. However above $P_c$, local helimagnetic domains survive which disappears abruptly at 16.1 kbar. For this pressure, the domain size is anticipated to become smaller than the spatial periodicity of the helical structure ($\lambda = 2\pi/Q$) leading to a shrinkage of domains.

IV. CONCLUSION

We have carried out a $^{29}$Si NMR experiment at zero-field in order to study the helicoidally ordered to paramagnetic quantum phase transition induced by applying pressure up to 17.8 kbar. The $^{29}$Si NMR frequency has revealed a very weak variation of the transferred hyperfine field and hence of the local magnetization on the Mn sites. Moreover the persistence of a $^{29}$Si NMR signal over the critical pressure of 14.8 kbar has proved that the magnetic order is not destroyed from a local point of view even though the phase transition is not identified macroscopically. In this note we do not deal with the physical origin of the local magnetic order persisting above 14.8 kbar. Recently Buzdin and Meurdesoif have demonstrated theoretically that in the peculiar case of a helimagnetic modulated phase the presence of local defects (impurities, dislocations and defects) play a keyrole in the formation
of unusual local magnetic order centered on these defects. At this stage, it is worth wondering if the persistence of a local magnetic order above $P_c$ is not the mark of the local defects. In the peculiar case of MnSi the influence of these local defects may play an important role.
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FIGURES

FIG. 1. The main figure compares the pressure effect on the Curie temperature $T_c$ (—) and the frequency resonance $f_0$ ((●) increasing $P$ and (○) releasing $P$). The inset presents the pressure effect on the resonance line of the $^{29}\text{Si}$ nuclei at $T=1.4$ K (Ambient, 3.65, 5.77, 7.57, 10.29, 12.99 and 15.64 kbar).

FIG. 2. Evolution of the resonance line intensity with the applied pressure. The plain line represents schematically the magnetic phase diagram of MnSi.
### TABLE I. Pressure dependence of macroscopic and microscopic quantities in MnSi.

| $P$[kbar] | $T_c$[K] | $M_s$[$\mu_B$/Mn] | $f_0$[MHz] | $A_{hf}^{^{29}\text{Si}}$[kOe/$\mu_B$] |
|-----------|-----------|-------------------|------------|----------------------------------|
| Ambient   | 29.07     | 0.389             | 19.880     | 60.42                            |
| 3.6       | 24.01     | 0.374             | 19.270     | 60.91                            |
| 6.6       | 19.60     | 0.359             | 18.504     | 60.94                            |
| 8.7       | 16.27     | 0.348             | 17.840     | 60.61                            |
| 10.2      | 13.50     | 0.342             | 17.340     | 59.94                            |