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Partial magnetic order in the itinerant-electron magnet MnSi

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Abstract. MnSi is an itinerant ferromagnet with a long-wavelength helical modulation of the spin structure. Macroscopic measurements suggest that the ordering temperature T_{c} is reduced with increasing pressure from $T_{\mathrm{c}} = 30$ K at $p = 0$ to zero at the critical pressure $p_{\mathrm{c}} = 14.6$ kbar. Resistivity measurements show that MnSi enters a non-Fermi liquid state at p_{c}, which remains to be understood. Neutron scattering techniques have been used to investigate the magnetic structure at and above p_{c}, i.e. triple-axis spectrometry and small angle neutron scattering. Surprisingly, sizeable quasi-static moments were found to survive to pressures considerably above p_{c}. They are, however, organized in a highly unusual way such that the magnetic Bragg reflections are sharp in the longitudinal direction but are very broad in the transverse direction, implying a partial magnetic order that was never seen before.

Keywords. High pressure; magnetic phase diagram; helical order.

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1. Introduction

MnSi is an itinerant ferromagnet with a relatively low ordered moment (0.4 μ_{B}). Due to the lack of inversion symmetry of the cubic B20 crystal structure of MnSi, the Dzyaloshinski–Moriya interaction leads to long wavelength helical modulation (≈40 lattice constants). At ambient pressure, the transition temperature is about 30 K. T_{c} as seen in the susceptibility and resistivity measurements decrease significantly with pressure and is thought to vanish at a critical pressure $p_{\mathrm{c}} = 14.6$ kbar [1]. That is to say, the magnetic behavior is expected to undergo a magnetic quantum phase transition at precisely this pressure. Interestingly, resistivity measurements have shown an extended regime of non-Fermi liquid (NFL) behavior at pressures above p_{c} leading to speculations about a close relationship between the magnetic properties and the unusual temperature dependence of the resistivity [2,3]. However, the magnetic properties of MnSi at elevated pressures were not well-characterized because they were only studied by macroscopic measurements. Neutron scattering techniques have promised to give deeper insight because of their microscopic probe. Indeed, the neutron experiments revealed magnetic properties which were totally unexpected with the macroscopic measurements. In particular, it was found that sizeable quasi-static magnetic moments survive to pressures far above p_c but are arranged in a highly unusual way. The neutron scattering techniques used for this study and the salient results will be described in the following sections. For further information, the reader is referred to ref. [4].

2. Experimental techniques

The samples were single crystals of dimensions $3\times3\times4$ mm^3 having a high degree of crystalline perfection, including a very small mosaic spread (<0.2°). They were pressurized inside a clamped cell made of Cu–Be with fluorinert as the pressure transmitting medium. A small piece of Sn was introduced into the pressure chamber as pressure gauge: the pressure obtained after cooling the cell to low temperatures was deduced from the superconducting transition temperature T_c of the Sn specimen and the established relationship of T_c versus pressure. The maximum pressure attainable in this pressure cell is about 20 kbar. The pressure range explored in the experiments carried out so far was 0 kbar $< p <$ 18.5 kbar.

The first series of experiments was performed on the 4F-triple-axis spectrometer located at the ORPHEE-reactor of the Laboratoire Léon Brillouin at Saclay. This spectrometer is fed by a cold source, which enabled us to use long-wavelength neutrons ($\lambda = 0.45$ nm) for the sake of a good resolution both in energy (50 μeV) and in momentum transfer (<0.01 Å^{-1}). A triple-axis spectrometer was chosen to distinguish between static order and dynamic order, i.e. fluctuations. The main drawback of such a spectrometer for the study in point is the fact that the momentum resolution is high only when compared to the total momentum transfer $Q \approx \tau = (110)$ but rather moderate when compared to the length of the propagation vector of the magnetic order in MnSi (≈ 0.04 Å^{-1}). A better resolution in momentum transfer can be achieved by using a small angle scattering instrument albeit at the expense of having no energy analysis. The second series of measurements was indeed carried out on the V4 small angle scattering instrument of the Hahn-Meitner-Institut using neutrons of wavelength $\lambda = 6$ Å. This instrument has the option of installing a horizontal cryomagnet, which turned out to be essential for the measurements at pressures above the critical pressure p_c. All measurements were performed with a He^4-cryostat allowing us to cool the sample to $T_{\rm min} = 1.5$ K.

3. Experimental results

The very first experiments were carried out at ambient pressure as a test for the later experiments at elevated pressures. The results were in complete agreement with the published data, i.e. we found a transition temperature $T_c = 30$ K and a magnetic propagation vector of $q_m = 0.038$ Å^{-1} pointing along the (1 1 1) direction of the crystal lattice. Subsequently, we performed measurements at $p = 14.3$ kbar which is only slightly lower than the critical pressure $p_c = 14.6$ kbar deduced from macroscopic measurements. A strong magnetic signal appeared on cooling at $T =$

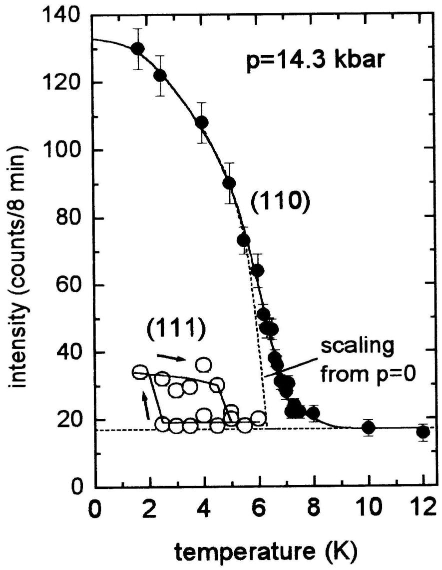

Figure 1. Temperature dependence of the magnetic scattering intensity observed in the (1 1 0) and (1 1 1) directions, respectively, at a pressure slightly below the critical pressure p_c.

7 K (figure 1) along the (1 1 0) direction, which is much higher than expected from the established phase diagram. This is also in contrast to what was expected from low pressure measurements showing an ordering along (1 1 1), which appeared as well but with a smaller intensity and at a lower temperature as shown in figure 1. An ordering along (1 1 0) appears particularly surprising in view of the fact that theory predicts minima of the crystal field potential either for (1 1 1) or (1 0 0) but not for (1 1 0).

The magnetic peaks were found to be resolution-limited when scanned along the longitudinal direction indicating a magnetic correlation length of at least 1200 Å (figure 2a). On the other hand, the peaks were much broader than the experimental resolution in the transverse direction (figure 2b) indicating a considerable degree of disorder. We note that the peaks did NOT sharpen on cooling. On the contrary, they were found to widen further because additional intensity showed up along the (1 1 1) direction. This extra intensity along (1 1 1) showed hysteresis in agreement with hysteretic behavior observed in measurements of the magnetic susceptibility for pressures above 12 kbar [1]. In other words, a small volume fraction of the crystal showed a behavior in line with expectations from the macroscopic measurements whereas the bulk of the crystal appeared to order at a much higher temperature. As stated above, the large width of the magnetic peaks in the transverse direction indicates that this particular order is incomplete. However, energy scans revealed that the magnetic peaks were resolution-limited (figure 2c), i.e. the partial order observed at this pressure is static on the time-scale of the neutron measurements.

Additional measurements at a somewhat higher pressure (16 kbar) revealed qualitatively similar results albeit with a somewhat lower magnetic ordering

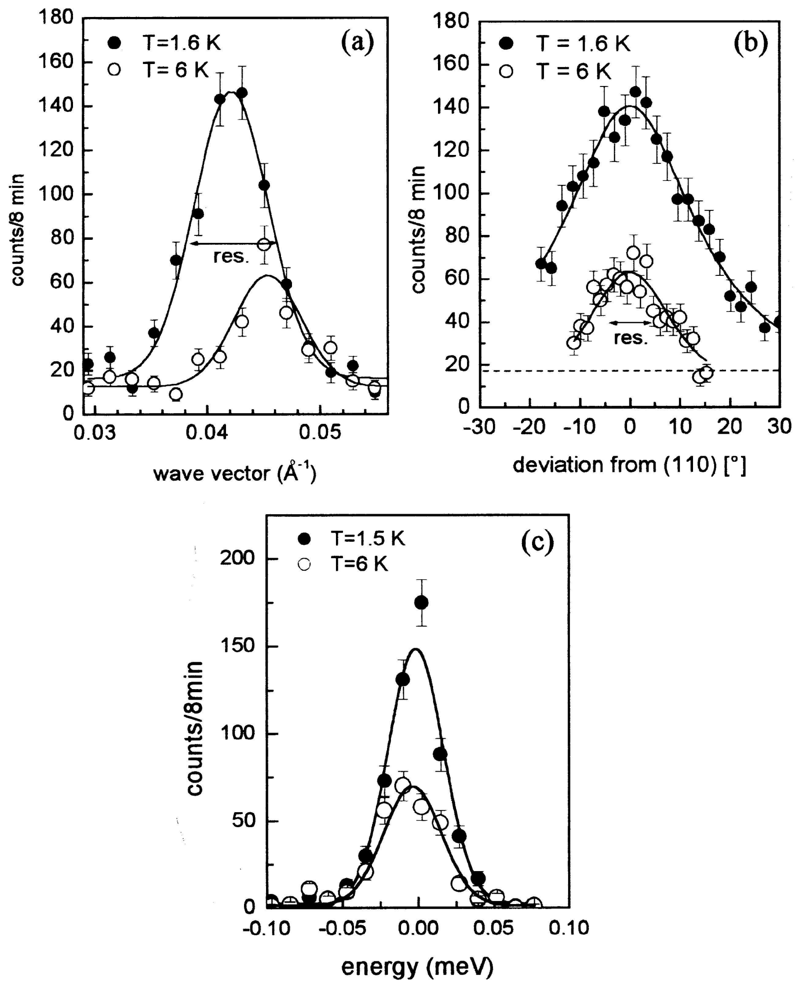

Figure 2. Neutron scattering scans through the magnetic Bragg peaks in the (1 1 0) direction at $p = 14.3$ kbar in the longitudinal direction (**a**) and in the transverse direction (**b**) as well as in energy (**c**).

temperature, i.e. $T_c = 5$ K. After increasing the pressure to $p = 18.5$ kbar, only a weak magnetic signal was found. Subsequent measurements for the same pressure were performed on a small angle scattering diffractometer instead of on a triple-axis spectrometer. Here, a clear magnetic signal was found only after applying a transverse magnetic field of $B = 0.4$ T. This finding indicated a field-induced magnetic order. However, after reducing the magnetic field strength to zero, a sizeable magnetic signal remained when the temperature was kept below 3 K (the full hysteresis loop is shown in figure 3). In addition, the reduction of the magnetic peak intensity on reducing the magnetic field from $B = 0.4$ T to zero could at least partly be attributed to a concurrent increase of the magnetic mosaic spread. We

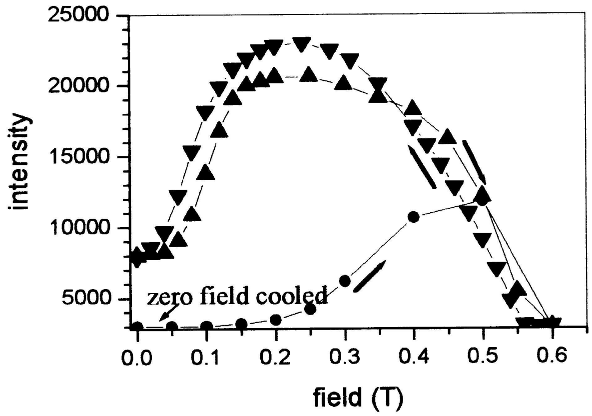

Figure 3. Field dependence of the magnetic Bragg intensity observed on a small angle scattering instrument at $p = 19$ kbar and $T = 1.5$ K. The intensity was found to peak at a wavevector $q = 0.04$ Å^{-1} corresponding to a periodicity of the magnetic helix of about 150 Å which is very similar to the corresponding value observed at ambient pressure. Note that a magnetic field of 0.6 T is known to be sufficient to change the character of the magnetic order from helical to ferromagnetic, which explains the loss of intensity at high magnetic fields.

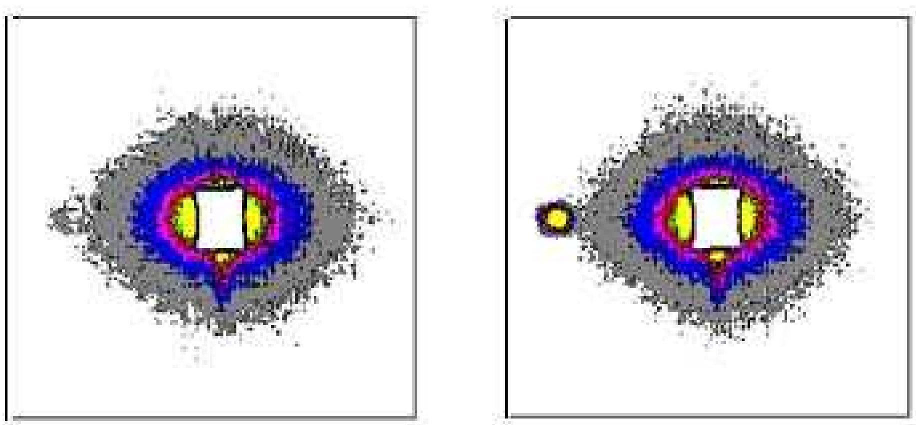

Figure 4. Small angle scattering patterns observed for two different orientations of the neutron spin relative to the magnetic propagation vector at $T = 1.5$ K with a transverse magnetic field of $B = 0.4$ T. The strong scattering at the centre comes from the walls of the pressure cell, whereas the spot close to the left-hand border in the right-hand panel is due to magnetic scattering. A residual of this spot is also seen in the left-hand panel due to the finite flipping ratio of the instrument.

conclude from these observations that there is a spontaneous magnetic order even at a pressure of about 4 kbar above the critical pressure deduced from macroscopic measurements. The main role of the applied magnetic field is to align the magnetic propagation vector parallel to the field direction. This alignment survives after turning off the magnetic field but not completely so: obviously, there is a strong tendency for directional disorder of the magnetic propagation vector at such high pressures. In contrast, the length of the magnetic propagation vector was found to be very well-defined corresponding to a correlation length in the longitudinal direction of at least 2000 Å.

The similarity of the intensity patterns observed at ambient and at high pressures strongly suggested a very similar character of the magnetic order at all pressures. This conjecture was confirmed by additional measurements with polarized neutrons. As can be seen in figure 4, the magnetic Bragg peak intensity is completely suppressed on flipping the neutron spin direction relative to the magnetic propagation vector apart from a small residual associated with the finite flipping ratio (≈22) of the instrument. We note that the helical character of the magnetic order at ambient pressure was demonstrated in a similar way.

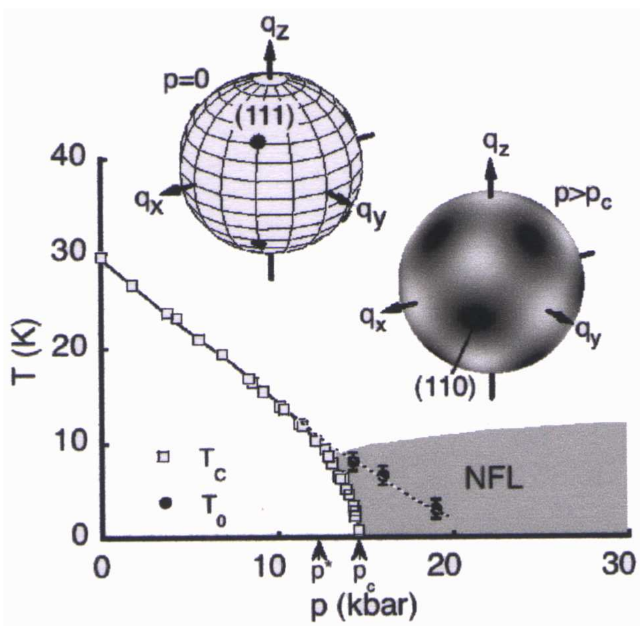

Figure 5. The magnetic phase diagram of MnSi. The T_{c} values and the T_0 values were deduced from macroscopic measurements and from neutron experiments, respectively. The T_0 values refer to the partial magnetic order described in the text. The area shown in gray is the region where non-Fermi liquid behavior was observed in resistivity measurements. The diagram shows the distribution of the magnetic scattering intensity for full magnetic order observed at ambient pressure (left sphere) and for the partial magnetic order observed for $p > p_{\mathrm{c}}$, respectively.

4. Conclusions and outlook

Neutron scattering experiments revealed that the magnetic phase diagram of MnSi is much richer than anticipated from macroscopic measurements. In contrast to expectations from the accepted phase diagram, the ordered moment does not vanish at the so-called critical pressure $p_c = 14.6$ kbar but survives to pressures which are several kbar higher than p_c. Extrapolating the critical temperatures observed by neutron scattering to higher pressures suggests that the ordered moment will vanish only for $p > 20$ kbar (figure 5). The order observed for $p > p_c$ is closely related to the one observed at ambient pressure in that it is helical with nearly the same periodicity. Further, it appears to be long-range, i.e. the longitudinal correlation length is at least 200 nm. On the other hand, this particular magnetic order is highly unusual since there is a considerable degree of disorder as to the direction of the magnetic propagation vector. A broad angular distribution was found around the (1 1 0) direction, i.e. a direction which is not expected to be favored by the crystal field in cubic symmetry.

Neutron scattering experiments have played a decisive role in exploring the unique magnetic properties of MnSi since many years [5,6]. Further progress was achieved by developing the technique for high pressures and low temperatures. We will (i) further increase the pressure to about 20 kbar and (ii) extend the temperature range to even lower temperatures, i.e. $T = 0.3$ K by using a He^3-cryostat in the near future. The application of a transverse magnetic field will again be of primary importance, likewise the use of polarized neutrons. All these requirements can be fulfilled on a modern small-angle neutron diffractometer like the V4 instrument at the Hahn-Meitner-Institut, Berlin.

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