



# Polarized neutron diffraction study in helical magnetic phases of MnP



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## ABSTRACT

MnP shows an interesting magnetic phase diagram under pressure up to  $\sim 7.5$  GPa, where magnetic long range order disappears and superconductivity appears. Polarized neutron analyses were applied to two different helical magnetic structures (helical-c and helical-b). Helicity was observed in the both structures. The two helical domain ratio becomes more unbalanced in the helical-b structure at a higher pressure, suggesting that the Dzyaloshinskii-Moriya interaction, which is enhanced by a local structural strain, could be related with the helical structures.

## 1. Introduction

MnP is a ferromagnetic metal at ambient pressure. It shows a ferromagnetic order below  $T_c \sim 290$  K followed by a helical order (helical-c) with the spins lying in the  $ab$  plane and the helical rotation propagating along the  $c$  axis below  $T_s \sim 50$  K at ambient pressure [1–3]. The Mn moment can be described with a localized spin model. Cheng et al. found superconductivity below  $\sim 1$  K under high pressures around 8 GPa [4]. Since MnP is the first superconductor in Mn-based materials, it has attracted much attention in terms of the pairing mechanism. The dominant magnetic interactions are ferromagnetic at ambient pressure. Therefore, ferromagnetic fluctuations might be related to the superconducting mechanism.

Cheng *et al.* also proposed a complex magnetic phase diagram below 7.5 GPa [4]. Matsuda *et al.* performed high pressure neutron diffraction study and determined the magnetic structures in the magnetic phases [5]. Both  $T_c$  and  $T_s$  are gradually suppressed with increasing pressure and the helical order disappears at  $\sim 1.2$  GPa. At intermediate pressures of 1.8 and 2.0 GPa, the ferromagnetic order first develops and changes to a conical or two-phase (ferromagnetic and helical-b) structure with the propagation along the  $b$  axis below a characteristic temperature. Above 2 GPa, a magnetic transition from paramagnetic to a helical magnetic order appears, which hosts the spins in the  $ac$  plane and the propagation along the  $b$  axis. This is the same structure as that of the  $ac$ -plane spin component (helical-b) in the intermediate pres-

ures. Muon-spin rotation studies also showed that the magnetic structure at higher pressures is the helical-b [6,7]. With increasing pressure, the helical transition temperature and the ordered moment decrease and the period of this  $b$  axis modulation becomes shorter [8].

We performed polarized neutron diffraction measurements in the two helical structures (helical-c and helical-b) of MnP. The helicity was observed in the two helical structures. The helical domain ratio was also determined, which gives useful information on the contribution of the Dzyaloshinskii-Moriya (DM) interactions to the helical structures.

## 2. Experimental method

The MnP single crystals used in the present study were grown by a modified Bridgman method from a stoichiometric mixture of Mn and P powders. Single crystals with dimensions of  $\sim 5 \times 3 \times 2$  mm<sup>3</sup> and  $1 \times 1 \times 3$  mm<sup>3</sup> were used for ambient and high pressure (1.8 GPa) experiments, respectively.

The full and half polarized neutron diffraction measurements on MnP at 0 and 1.8 GPa were performed on a triple-axis neutron spectrometer HB-1 installed at High Flux Isotope Reactor at Oak Ridge National Laboratory. For full polarization analysis, Heusler alloy (111) crystals were used as monochromator and analyzer. For half polarization analysis, Heusler alloy (111) crystal and Pyrolytic Graphite (002) crystal were used as monochromator and analyzer, respectively. The horizontal collimator sequence was 48'-80'-sample-80'-240' with

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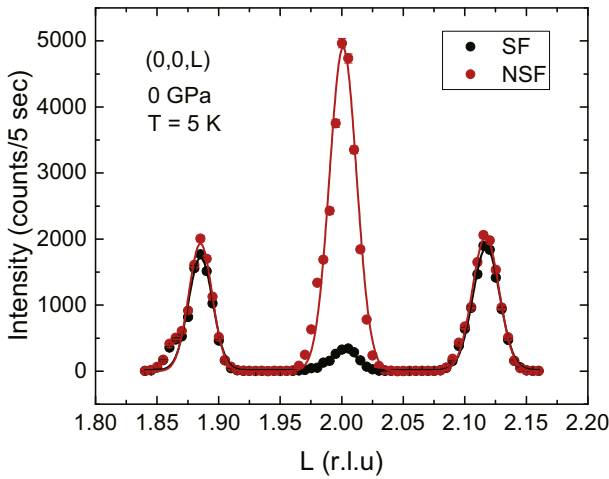


Fig. 1. Neutron diffraction intensities in the non-spin-flip (NSF) and spin-flip (SF) channels around the  $(0, 0, 2)$  Bragg reflection at 5 K and at ambient pressure, measured with the full polarization mode.  $(0, 0, 2)$  and  $(0, 0, 2 \pm \delta)$  peaks are nuclear and magnetic, respectively.

a fixed incident neutron energy of 13.5 meV. The contamination from higher-order beams was effectively eliminated using PG filters. A flipping ratio of  $\sim 15$  was observed at nuclear reflections. Single crystal was aligned in the  $(h0l)$  and  $(0kl)$  scattering zone for measurements at 0 and 1.8 GPa, respectively.

The high pressure of 1.8 GPa was generated with a self-clamped piston-cylinder cell (SCPCC) made of a Zr-based amorphous alloy [9]. Fluorinert was chosen as the pressure transmitting medium. The pressure inside SCPCC was monitored at room temperature by measuring the lattice constant of a comounted NaCl crystal.

### 3. Results and discussion

#### 3.1. Full polarization analysis

We first performed full polarization analysis at ambient pressure to confirm the helical-c structure which shows a small ellipticity [3,5,10]. As shown in Fig. 1, we observed slightly different intensities in the non-spin-flip (NSF) and spin-flip (SF) channels at the  $(0, 0, 2 \pm \delta)$  magnetic Bragg reflections. The NSF and SF intensities originate from magnetic components vertical to the scattering plane ( $M_{\perp}$ ) and perpendicular to  $\mathbf{Q}$  in the scattering plane ( $M_{\parallel}$ ), respectively. The easy plane of the helical spin structure is exactly perpendicular to  $\mathbf{Q}$  in the measurement. Therefore, the  $M_{\perp}$  and  $M_{\parallel}$  correspond to the spin components along the  $b$  axis ( $S_b$ ) and the  $a$  axis ( $S_a$ ), respectively. Therefore, the ratio of  $I_{\text{NSF}}/I_{\text{SF}} [= (M_{\perp}/M_{\parallel})^2]$  is expected to be 1 for isotropic circular helical structure. The observed ratio is 1.12 (5), which corresponds to  $S_b/S_a = 1.06(5)$ . This value is consistent with 1.05 ( $=\sqrt{1.091}$ ) obtained with a polarized neutron study in Ref. [10]. This value is also consistent with 1.23 ( $=1.73/1.41$ ) in Ref. [3] and 1.19 ( $=1.9/1.6$ ) in Ref. [5], which were obtained with unpolarized neutron studies.

The same analysis was performed in the helical-b structure at 1.8 GPa. Although the ferromagnetic component exists at this pressure, the incommensurate magnetic peaks can be explained by the helical-b structure [5]. Fig. 2(a) shows NSF and SF intensities at the  $(0, 1 - \delta, 1)$  magnetic Bragg reflection at 5 K. At  $(0, 1 + \delta, 1)$ , a powder line from the pressure cell is overlapped, which makes the accurate analysis difficult. The NSF and SF intensities originate from spin component along the  $a$  axis ( $S_a$ ) and the  $c$  axis spin component ( $S_c$ ) projected perpendicular to  $\mathbf{Q}$ , respectively. In this configuration, the easy plane of the helical spin structure is not perpendicular to  $\mathbf{Q}$ . Therefore, due to the neutron

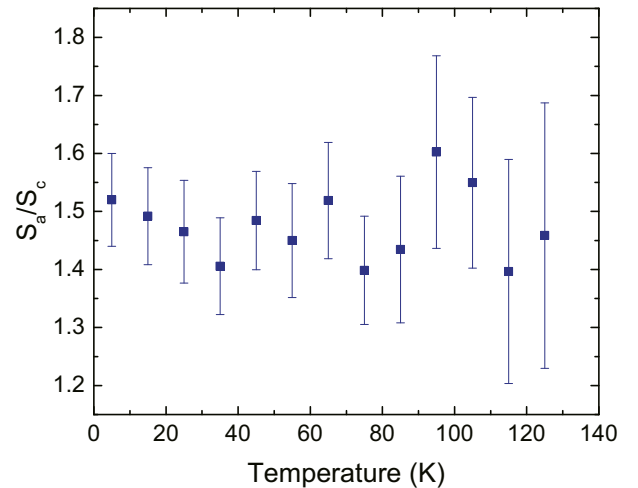
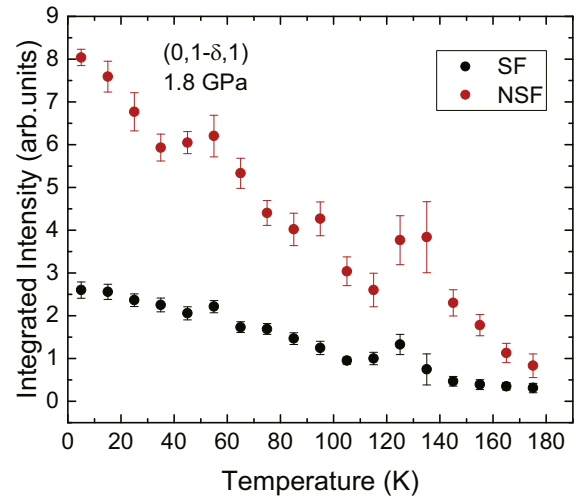
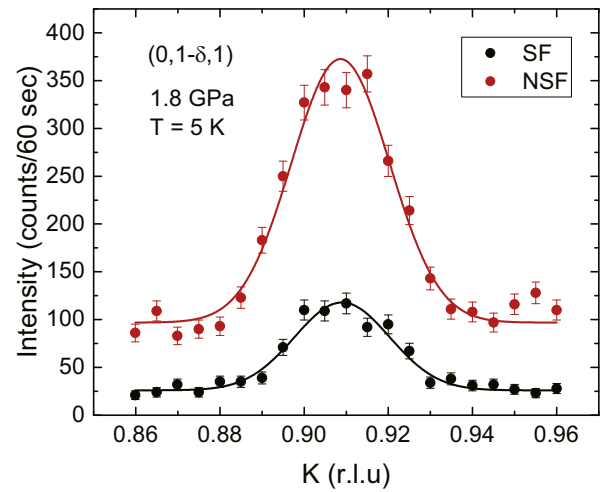


Fig. 2. (a) Neutron diffraction intensities in the NSF and SF channels at the  $(0, 1 - \delta, 1)$  magnetic Bragg reflection at 5 K and at 1.8 GPa, measured with the full polarization mode. Temperature dependence of the NSF and SF intensities measured at  $(0, 1 - \delta, 1)$  (b) and  $S_a/S_c$  (c).

polarization factor, ellipticity should be observed even in the isotropic helical structure.  $I_{\text{NSF}}/I_{\text{SF}}$  is expected to be 1.35 for isotropic circular helical structure. The observed ratio is 3.09 (23), which corresponds to  $S_a/S_c = 1.52(11)$ . This ratio is consistent with 1.4 ( $=1.5/1.1$ ) reported in Ref. [5]. The temperature dependence of the NSF and SF intensities was also measured, as shown in Fig. 2(b). The temperature dependence

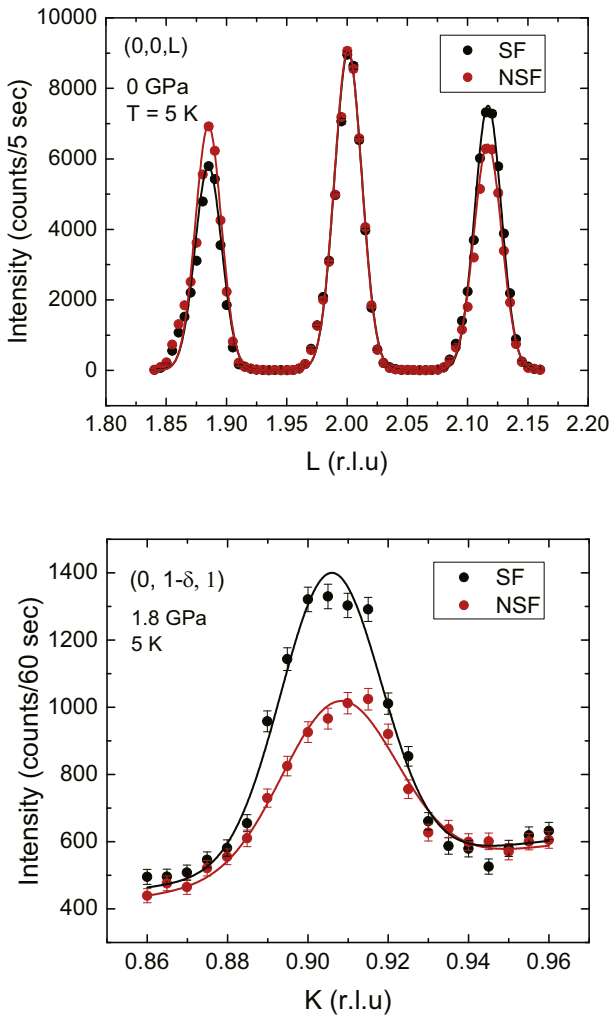


Fig. 3. Neutron diffraction intensities in the NSF and SF channels around the  $(0, 0, 2 \pm \delta)$  magnetic Bragg reflections at 5 K and at ambient pressure (a) and the  $(0, 1 - \delta, 1)$  magnetic Bragg reflection at 5 K and at 1.8 GPa (b), measured with the half polarization mode.

of  $S_a/S_c$  is plotted in Fig. 2(c). The ratio of  $\sim 1.5$  is almost temperature independent.

### 3.2. Half polarization analysis

Half polarization method is a technique to observe the helicity degree of freedom in magnetic materials. The cross section of one of the incommensurate magnetic Bragg peaks for incident neutrons with up and down spins is described as

$$\frac{d\sigma^\pm}{d\Omega} \propto m_\parallel^2 + m_\perp^2 \pm 2m_\parallel m_\perp (\mathbf{s}_N \cdot \mathbf{h}),$$

where  $m_\parallel$ ,  $m_\perp$ ,  $\mathbf{s}_N$ , and  $\mathbf{h}$  are spin components vertical to the scattering plane and perpendicular to  $\mathbf{Q}$  in the scattering plane, neutron spin vector, and helicity vector, respectively. By aligning the neutron spin parallel and antiparallel to  $\mathbf{h}$ , the magnetic intensities from two helical domains with clockwise and counter-clockwise helicities can be observed, respectively.

Fig. 3(a) and (b) show NSF and SF intensities measured around  $(0, 0, 2)$  and  $(0, 1 - \delta, 1)$  in the helical-c (ambient pressure) and helical-b (1.8 GPa) structures, respectively. At ambient pressure, NSF and SF intensities at  $(0, 0, 2 \pm \delta)$  are similar. This is very different from those expected from  $S_b/S_a \sim 1.1$ . The ratio of  $I_{SF}/I_{NSF}$  is expected to be  $\sim 15$ , which is close to the instrumental flipping ratio. As shown in Fig. 3(a),  $I_{NSF}$  is stronger than  $I_{SF}$  at  $(0, 0, 2 - \delta)$  and weaker than  $I_{SF}$  at  $(0, 0, 2 + \delta)$ . This indicates that the population of the two helical domains is slightly unbalanced. In the helical-b structure at 1.8 GPa, a larger difference was observed between the NSF and SF intensities. However,  $I_{SF}/I_{NSF} (\sim 1.4)$  is still much smaller than the value ( $\sim 15$ ) expected for a single helical domain structure, indicating that the ratio of the two helical domains is largely unbalanced ( $\sim 8 : 5$ ), which is estimated from the ratio of  $I_{SF}$  and  $I_{NSF}$ .

Series of successive phase transitions under pressure have been observed in MnP. One of the possible mechanisms is the increased DM interaction. Therefore, the observed unbalanced population may also be originated from the DM interaction, since a local structural strain enhanced by applied pressures can affect the DM interactions.

## 4. Summary

Full and half polarized neutron diffraction experiments were performed to further confirm the non-circular helicity in both the helical-b and the helical-c magnetic structures of MnP, which is consistent with the magnetic structures reported in the literature. The two helical domain ratio becomes more unbalanced at higher pressures, suggesting that the DM interaction, which is enhanced by a local structural strain, should relate with the helicity of the helical magnetic structures.

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