Magnetic excitations in MnP

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Abstract. We performed inelastic neutron scattering experiments on a three-dimensional magnetic system MnP, which is a ferromagnetic inter-metallic compound below \( T_C = 291 \) K and transitions into a screw state at \( T^* = 47 \) K. Using newly developed high resolution chopper spectrometer (HRC) and triple axis spectrometer LTAS (for \( E = 0-2 \) meV), 6G (for \( E = 2-8 \) meV), and TAS1 (for \( E = 10-35 \) meV), we succeeded in observing the excitations from low energy to Brillouin zone center. We assumed isotropic 6 exchange parameters, and calculated dispersion relation using Heisenberg model for 2-sub lattices. We confirm that nearest neighbor (n.n) exchange parameters \( J_m \) are \( J > 0 \) (ferromagnetic interaction), the next nearest neighbor (n.n.n) exchange parameters are \( J < 0 \) (anti-ferromagnetic interaction). We conclude that the competition between n.n and n.n.n would be the cause of helimagnetic structure in MnP.

1. Introduction

Manganese phosphide MnP is a ferromagnetic intermetallic compound below \( T_C = 291 \) K, and it transforms into a double helix proper screw state at \( T^* = 47 \) K [1]. The crystal structure is a slightly distorted NiAs-type structure with the lattice parameters of \( a = 5.916 \) Å, \( b = 5.260 \) Å, \( c = 3.173 \) Å at room temperature. In the ferromagnetic state, the easy-axis of the magnetization is the c-axis. In the helimagnetic state, the spin rotates in the b-c plane with a propagation vector \( 0.117a^* \) along the a-axis. The precise structure model of screw spiral state in MnP is proposed to be Elliptic or Bunching state[2,3].

Recently, Yamazaki et al.[4] performed neutron diffraction experiment, and found that there is magnetic \( (\delta,1,0) \) reflection in helimagnetic phase, and they could not find \( (1+\delta,0,0) = (\delta,1,0)+(-1,1,0) \). (the \( \delta \) is the same period of magnetic propagation vector). They also reported the magnetic susceptibility measurement. From these experimental data, they concluded that anti-ferroic modulated magnetic structure should exist in MnP.

The conventional theory for helimagnetism cannot explain why the complicated magnetic structure is stable and a ferromagnetic intermetallic compound transforms into a proper screw spiral state. In order to elucidate the mechanism of the spiral state, the information of spin wave in the whole Brillouin zone is crucially important. We focus our attention to the spin wave dispersion of the ferromagnetic state in MnP. Spin waves in MnP have already been measured in a small-\( q \) region below 150K by Tajima et al.[5]. Adopting a Heisenberg model, they deduced
Figure 1. Purple and ash color atoms are Mn, P atoms respectively. Mn 4c cite in unit cell are Mn1(−x, −y, −z), Mn2(x, y, z), Mn3(−x + 1/2, y + 1/2, z), Mn4(x + 1/2, −y + 1/2, −z).

| Jm | J1 | J2 | J3 | J4 | J5 | J6 |
|----|----|----|----|----|----|----|
| rm(Å) | 2.70 | 2.82 | 3.18 | 3.92 | 4.17 | 4.27 |
| z   | 2   | 2   | 2   | 2   | 4   | 4   |

Table 1. The definition of exchange parameters, J₁, J₂, J₃, J₄ are the nearest neighbor Mn1-Mn2, Mn1-Mn3, Mn1-Mn1, Mn1-Mn4. J₄, J₅ are the next nearest neighbor Mn1-Mn3, Mn1-Mn2. We show here the distance between Mn-Mn atoms and coordination number.

the nearest neighbor effective inter-planar interaction (J₁) and next-nearest neighbor one (J₂) along the a-axis and concluded that ferro-screw transition at 47K is caused by the slight change of the ratio of (J₂)/(J₁) with temperature. Todate et.al.[6] also reported spin waves of MnP in ferromagnetic state along the three principal axes at relatively small-Å region. Dispersion relation along the a-axis exhibits anomalous wave vector and temperature dependence, while the quadratic q dependence was observed along the b-axis and the c-axis. MnP is extensively studied so far, however, it was limited to be focused on relatively low energy because of limitation of experimental condition.

2. Experimental
2.1. Inelastic neutron scattering on HRC
We constructed the High Resolution Chopper Spectrometer (HRC) at the BL12 beamline in the Materials and Life Science Experimental Facility (MLF) of the Japan Proton Accelerator Research Complex (J-PARC) in collaboration with the High Energy Accelerator Research Organization (KEK) and the University of Tokyo [7]. The HRC delivers high-resolution and relatively high-energy neutrons for a wide range of studies on the dynamics of materials. We successfully installed the DAQ system on the HRC. A total of 251 PSDs and 2 monitors were mounted on the spectrometer. The event data of the detected neutrons are processed in the DAQ system and visualized in the form of the dynamic structure factor. We verified the data analysis process by visualizing the excitations in one- and three-dimensional single-crystal magnetic systems in inelastic neutron scattering experiments[8]. We performed inelastic neutron
scattering experiments in three-dimensional (3D) magnetic systems to demonstrate excitation spectra with the DAQ system on the HRC. Using HRC, we examined the wide range of $S(Q, \omega)$ with high energy resolution conditions ($E_i = 30-350$ meV, $\Delta E/E_i = 0.03-0.06$), and succeeded in observing the excitations from low energy to nearly Brillouin zone center.

2.2. Inelastic neutron scattering on TAS’s

We also performed the neutron inelastic scattering experiments at triple-axis spectrometer LTAS, TAS-1, 6G JRR-3M reactor in JAEA (Tokai). Using triple axis spectrometer LTAS, we focus on relatively low energy magnetic excitation. Spin wave spectra were obtained by constant $Q$ scan, and spectra were fit to Gaussian. We plot peak center and the error, the errors are half maximum full-width. We performed constant $Q$ scan at $E_f$ fix mode ($E_f = 3.0$ meV). We found that the linear dispersion relation contribution of phonon, and the energy gap at $q = 0$ in the ferromagnetic state at 5K was estimated to be $(0.5 \pm 0.1)$ meV which is consistent value with the previously reported[5]. We performed the neutron inelastic scattering experiments at triple-axis spectrometer TOPAN (6G), for middle range energy measurement ($E = 2-8$ meV). The measurements are $E_i$ fixed = 13.5 meV condition. Data are fitted to $\hbar \omega = Aq^3 + \Delta$, and determined to be $A = 84.4$ meV $\Delta= 0.34$ meV which are also consistent values with the previously reported. We also performed the neutron inelastic scattering experiments at triple-axis spectrometer TAS-1, JRR-3M reactor in JAERI (Tokai) for high energy measurement ($E = 10-35$ meV). The measurements are done under $E_f$ fixed = 14.7 meV condition. The obtained data clearly showed that the other higher magnetic excitation exists in MnP. Using newly developed chopper spectrometer HRC and triple axis spectrometers LTAS, 6G and TAS-1, we succeeded in observing the two branches of magnetic excitations from low energy to Brillouin zone center.

3. Analysis and Discussion

In order to explain the 2 distinctive branch of magnetic excitation, we derive spin wave dispersion relation in the ferromagnetic state of MnP. We assumed on isotropic Heisenberg model has been taken acount of 6 exchange interactions between Mn-Mn, and calculation was made for 2-sub lattices.

$$H = \sum_{nn'jj'} J_{nn'jj'}S_{nj}^- \cdot S_{n'j'}^- - D \sum_{nj} (S_{nj}^z)^2$$
$$= \sum_{nn'jj'} J_{nn'jj'} [S_{nj}^- S_{n'j'}^- + \frac{1}{2} (S_{nj}^+ S_{n'j'}^- + S_{nj}^- S_{n'j'}^+)] + D \sum_{nj} (S_{nj})^2$$

where $n, n'$ denote unit cell (the number is $N$), $j, j'$ denote sub lattice in each of unit cells($j, j'=1-4$, the number of magnetic ion = 4N). The definition of exchange parameter is discribed in Fig1 and Table1. Here, $J_1, J_2, J_3$ and $J_6$ are nearest neighbor interaction between Mn-Mn within the unit cell of MnP, and $J_4, J_5$ are next nearest neighbor interactions. D is the axial anisotropy constant whose easy axis is along the $c$-axis. We have four dispersion relations $\lambda_1-4$, however we found that $\lambda_1$ and $\lambda_2$ are spurious branches because dynamical structure factor becomes 0, so we have 2 magnetic branches. Obtained dispersion relations along the $a$-axis are shown in Fig2.

Using newly developed high resolution chopper spectrometer HRC and triple axis spectrometers LTAS, 6G and TAS-1, we succeeded in observing the excitations from low energy to Brillouin zone center. We assumed isotropic 6 exchange parameters, and calculated dispersion
Figure 2. Calculated spin wave dispersion relation in MnP along the $a$-axis

Figure 3. Obtained spin wave dispersion relation in MnP along the $a$-axis

| $J_m$ | $J_1$ | $J_2$ | $J_3$ | $J_4$ | $J_5$ | $J_6$ | $D$ |
|-------|-------|-------|-------|-------|-------|-------|-----|
| $E$(meV) | 11.30 | 1.67  | 8.99  | -5.27 | -3.48 | 2.55  | 0.24 |

Table 2. Determined exchange parameters using model fitting.

relation using Hisenbert model for 2-sub lattices. We determined exchange parameters $J_m$ by model fitting to experimentally obtained dispersion relation along the $a$-axis. We found that nearest neighbor (n.n) exchange parameters $J_m$ are $J > 0$ (ferromagnetic interaction), the next nearest neighbor (n.n.n) exchange parameters are $J < 0$ (anti-ferromagnetic interaction), so that we concluded that the competition between n.n and n.n.n should be the cause of helimagnetic structure in MnP.

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