Spin Waves in Ferromagnetic Phase of MnP

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Abstract. Inelastic neutron scattering experiments on an intermetallic compound, MnP, were performed by using a chopper spectrometer as well as triple axis spectrometers. Spin waves were observed in the ferromagnetic phase in the entire Brillouin zone along the $a^*$- and $b^*$-axes. The observed dispersion relations of spin waves were well described by an isotropic Heisenberg interaction adding a single ion anisotropy with two sub-lattices.

An intermetallic compound, MnP, is ferromagnetic below $T_C$, and transforms into a proper screw spiral phase at $T^* \approx 46.2$ K for the present sample [3]. The crystal structure of MnP is the distorted NiAs-type structure with the lattice constants, $a = 5.917 \text{ Å}$, $b = 5.260 \text{ Å}$, and $c = 3.173 \text{ Å}$ at room temperature [1]. In the ferromagnetic phase, the easy axis of the magnetization is the $c^*$-axis and the hardest one is the $a^*$-axis. In the proper screw spiral phase, the spin rotates in the $b^*$-$c^*$ plane with a propagation vector of $0.117a^*$ [2, 4]. Very recently, a canted antiferromagnetic structure with weak ferromagnetic magnetization along the $b^*$-axis in the ferromagnetic phase and an alternatively tilted helimagnetic structure below $T^*$ were found [5].

In order to understand the origin of these complicated properties of MnP, the microscopic interactions should be determined by observing spin waves [6, 7]. However, these early studies are limited to observations on the low energy ($E$) and low momentum ($q$) region. It was suggested that the emergence of the proper screw spiral phase is caused by a delicate balance between the nearest and the next-nearest neighbor exchange interactions [6]. Subsequently, it was reported that the spin-wave dispersion relation in the ferromagnetic phase exhibits an unusual $q$ dependence along the $a^*$-axis, while the conventional $q^2$ law was observed along the $b^*$- and $c^*$-axes [6, 7].

It is essential to determine the spin dynamics in the entire Brillouin zone, in order to elucidate the microscopic origin of magnetism in MnP. As a first step of this study, we have determined the spin wave dispersion relations in the entire Brillouin zone along the $a^*$- and $b^*$-axes in the

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Figure 1. Typical excitation spectra in MnP along the $b^*$-axis, after background subtraction (a). Spin-wave spectrum with a fitted curve (solid line) (b). The observed peak positions are located at the crossing points between the scan locus (dashed line) and the spin-wave dispersion curves (solid lines, from Fig. 3) within the resolution (c).

ferromagnetic phase of MnP, by using a chopper spectrometer installed at a pulsed neutron source as well as triple axis spectrometers at a steady state neutron source [3].

Inelastic neutron scattering experiments were performed on the High Resolution Chopper Spectrometer (HRC), which is installed at the Materials and Life Science Experimental Facility (MLF) of the Japan Proton Accelerator Research Complex (J-PARC) [8], also on the triple axis spectrometers, LTAS and TAS-1, which are installed at the JRR-3M reactor in the Tokai Institute of the Japan Atomic Energy Agency.

On the HRC, the monochromatic neutron beam with the energy of $E_i$ is incident upon the sample, and the energy transfer $E$ is determined by the time-of-flight (TOF) of the detected neutron. The single-crystal sample of MnP was mounted with the $c^*$-axis vertical, and the experiments were performed at temperatures $T = 60$ and $300$ K. Empty scans were also performed by removing the sample crystals, in order to estimate the background. Figure 1(a) shows the observed energy spectra at $\phi = 18.9^\circ$ with $E_i = 89.4$ meV and $\psi = 108.9^\circ$ on the HRC, where the background was subtracted and the intensities were divided by the temperature factor, $n + 1 = [1 - exp(-E/k_BT)]^{-1}$, with $k_B$ the Boltzmann constant. $\phi$ and $\psi$ are the scattering angle and the crystal angle between the $a^*$-axis and the incident neutron beam, respectively. In this setup, the detector element at $\phi = 18.9^\circ$ scans (2$\theta$) [3]. The energy resolution was determined to be $\Delta E = 5.0$ meV by measuring the energy width (full width at half maximum) of the incoherent elastic scattering. Although excitation peaks were observed at $T = 60$ K, some peaks remained even at $T = 300$ K, which might be due to phonons. The energy spectrum subtracting the spectrum at $T = 300$ K from that at $T = 60$ K was assumed to be a spin-wave spectrum, and the peaks were fitted with Gaussians with a resolution width of $\Delta E = 5.0$ meV, as shown in Fig. 1 (b). The peak positions are located at the crossing points between the scan locus and the dispersion curve, as shown in Fig. 1 (c). To obtain the dispersion relations of the spin waves in the entire Brillouin zone for the $a^*$- and $b^*$-axes, we chose scans with various sets of $E_i$ and $\psi$ [3]. The peak positions determined in this way are plotted in Fig. 3 in the reduced zone scheme.

Because anomalous properties along the $a^*$-axis were reported [6, 7], we also measured excitations along the $a^*$-axis using triple axis spectrometers. The LTAS spectrometer was used
for the energy region below 2 meV with $\Delta E = 0.19$ meV, and the TAS-1 was used up to 35 meV with $\Delta E = 1.5$ meV, both measurements were performed with a constant-$q$ scan. Spin-wave peak positions obtained on these spectrometers at $T = 60$ K are plotted in Fig. 3. The peak positions at $T = 77$ K, which were previously reported [6, 7], were also plotted in Fig. 3.

\[ H = - \sum_{nn'jj'} J_{nn'jj'} S_{nj} S_{n'j'} - D \sum_{nj} (S_{nj}^z)^2, \]

where $S_{nj}$ is the spin of a Mn atom located at the $n$th unit cell and the $j$th sub-lattice in a unit cell ($j = 1, 2$). We took into account of exchange interactions between Mn pairs up to the 6th neighbors, as discussed in a theory presented before [10]. The eigenvalues of the Hamiltonian in Eq. (1) lead the two dispersion relations of spin waves: acoustic and optical modes.

The dispersion curves were well fitted to the calculated curves with the parameters of $J_1 = 0.37 \pm 0.14$, $J_2 = -0.65 \pm 0.10$, $J_3 = -0.06 \pm 0.13$, $J_5 = 0.26 \pm 0.10$, $J_6 = 0.64 \pm 0.08$, and $D = 0.10 \pm 0.02$ meV, as shown in Fig. 3. Note that $J_3$ will be determined by experiments along the $c^*$-axis. Along the $a^*$-axis, the acoustic and optical modes are separated from each other, but, along the $b^*$-axis, these two modes resonate with each other at around $k = 0.2$ rlu (reciprocal lattice unit) and switch roles, but the two modes are not crossed. As shown in Fig. 3 (c), the low energy part in the $a^*$-axis for the presently-improved dispersion relation was also well fitted to the present model, where the dispersion curve along the $a^*$-axis shows a minimum at around $h = 0.14$ rlu. This complicated $q$ dependence is caused by the displacement parameter $u$, because the minimum disappeared for $u = 0$. Note that such minimum was previously observed at $T = 77$ K [6], as shown in Fig. 3 (c), and that the dispersion relation along the $a^*$-axis shows the $q^3$ dependence at $T = 150$ K [7]. The fitted curve for the acoustic mode at $T = 60$ K was lower than the data at $T = 77$ K, this is consistent with the reported $T$ dependence [6, 7]. In the previous study, the dispersion relation along the $b^*$-axis was determined to be $E(k) = D_b k^2 + E_0$ with $D_b = 145$ meV at $T = 150$ K almost independently on $T$ [7]. In fact, the spin-wave positions at $T = 77$ K are on the low energy part of the fitted acoustic mode for the data at $T = 60$ K at present, as shown in Fig. 3 (a). By using the present parameters, the stiffness constant was estimated to be $D_b = 170 \pm 31$ meV [3]. The new value is in approximate agreement with the previous value. Observed spin-wave peak positions are well explained by the two modes. In fact, two spin-wave peaks were clearly observed at $h = 0.5$ rlu on the TAS-1.
Figure 3. Dispersion relation of spin waves in the ferromagnetic phase of MnP along the \( a^* \)- (a) and \( b^* \)-axes (b) at \( T = 60 \) K, with calculated curves, acoustic (solid line) and optical (dashed line) modes. The bars on the marks represent statistical errors along the scan directions. Low energy part along the \( a^\ast \)-axis (c). The spin-wave peaks at \( T = 77 \) K, which were reported previously [6, 7], are also plotted.

In summary, we successfully observed spin waves in the ferromagnetic phase of MnP along the \( a^* \)- and \( b^* \)-axes in the entire Brillouin zone up to the zone boundary, by using the HRC, LTAS and TAS-1. The dispersion relations were well described by an isotropic Heisenberg model adding a single ion anisotropy with two sub-lattices, and the anomalous dispersion relation along the \( a^* \)-axis was found to be caused by the atomic displacement from the primitive lattice. The previous studies suggested that it is caused by an exchange competition [6], or attributed to itinerant nature of \( d \) electrons [7, 11]. The recent structural study suggested the existence of the Dzyaloshinsky-Moria interaction [5]. Further investigations are required to resolve this problem.

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