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## Spin-Waves in MnP

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Spin-waves in MnP have been measured by means of neutron inelastic scattering at various temperatures. Dispersion relation along *a*-axis exhibits anomalous wave vector and temperature dependences, while the quadratic q dependence was observed along both b and c-axis. The characteristic feature of the spin-wave dispersion relations agree qualitatively with those predicted on analogy with rare earth metals.

The 3d intermetallic compound MnP which is ferromagnetic between 291 K and 47 K and helimagnetic below 47 K has been considered as a localized spin system and the s-d model was applied to interpret the experimental results. Recently, however, it has been suggested that MnP should be a quasi-localized spin system from the band structure calculation<sup>1)</sup> and several experimental results; an enhancement of the paramagnetic moment  $(\mu_{\text{para}}=2.9\pm0.6\,\mu_{\text{B}}^{2})$  compared with the low temperature saturation moment  $(\mu_{sat} =$  $1.33 \pm 0.01 \,\mu_{\rm B}^{3}$ ), large  $\gamma$  value in the low temperature specific heat<sup>4)</sup> and photoelectron spectra.<sup>5)</sup> Spin-waves in MnP have already been measured in a small-q region at temperatures below 150 K by Tajima et al.<sup>6)</sup> Adopting a Heisenberg model they deduced nearest neighbour effective inter-planar exchange interaction  $(J_1)$  and next-nearest neighbour one  $(J_2)$  along the *a*-axis (the direction of propagation of screw in the helimagnetic state) and concluded that the ferro-screw transition at 47 K is caused by the slight change of the ratio of  $J_2/J_1$  with temperature. Furthermore with respect to the multicritical point or the Lifshitz point, measurements of spin-waves along the *a*-axis under magnetic field along the *b*axis were performed by Yoshizawa et al.<sup>7</sup> In this letter, we report ferromagnetic spin-waves of MnP along three principal axes at relatively

small-q region and discuss its characteristic feature comparing with a model dispersion postulated previously.<sup>8)</sup>

MnP has a orthorhombic, distorted NiAs structure with four Mn atoms and four P atoms in a unit cell. The lattice parameters are a=5.916, b=5.260 and c=3.173 Å at room temperature. In the ferromagnetic state spins are parallel to the *c*-axis, while in the helimagnetic state the screw propagation vector  $Q_h(=0.117a^*)$  is parallel to the *a*-axis.

For sample preparation, 99.999% P flakes and powdered 99.99% Mn were mixed and sealed in a quartz tube. The mixture was heated gradually and kept at 900°C for two days. Then single crystal of MnP was grown by Bridgman method and purified by zone refining. The ferromagnetic spin-waves (; at temperatures above 47 K) along the three principal axes have been measured by means of neutron inelastic scattering technique using triple-axis spectrometer TUNS installed at JRR-2, Tokai Establishment JAERI. The results together with data obtained by previous measurements<sup>6)</sup> are shown in Fig. 1. Examples of temperature dependence of constant-E spectra are shown in Fig. 2. Along the  $b^*$  and  $c^*$  direction the spin-wave dispersions show  $q^2$ dependence at small-q region, while along the  $a^*$  direction the dispersion shows unusual qdependence as shown already in refs. 1 and 2. The spin-wave stiffness constants at 150 K are 145 and 70 meVÅ<sup>2</sup> for the  $b^*$  and  $c^*$  direction

<sup>\*</sup> Deceased on 28 February 1986.

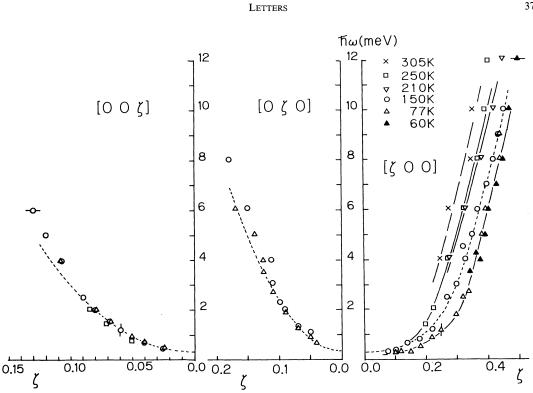


Fig. 1. Spin-wave dispersions along  $a^*$ ,  $b^*$  and  $c^*$  directions. The solid lines are guides to the eye. The dotted lines represent the relation of  $\hbar\omega_a = 86q_a^3 + \Delta$ ,  $\hbar\omega_b = 145q_b^2 + \Delta$  and  $\hbar\omega_c = 70q_c^2 + \Delta$  with  $\Delta = 0.35$  for  $a^*$ ,  $b^*$ and  $c^*$  directions respectively.

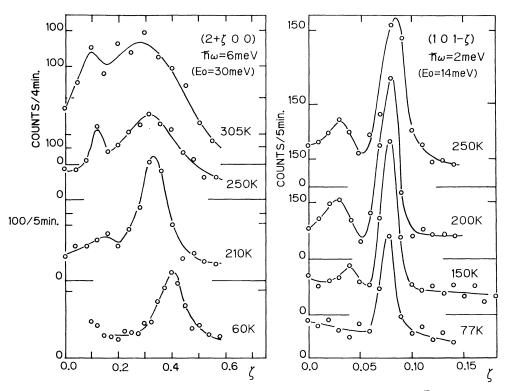


Fig. 2. Temperature dependence of spin-wave spectra obtained by constant-E scans.

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respectively. The dispersion along the  $a^*$  direction is roughly approximated by an expression of  $\hbar\omega_a = 86q_a^3$  at 150 K as indicated by dotted lines in Fig. 1. In contrast with usual ferromagnets, with increasing temperature the dispersion along the  $a^*$  direction is largely renormalized to higher spin-wave energy for the same  $q_a$  value in the measured q region and finally turns into the constant-E ridges above  $T_c^{9}$  as shown in Figs. 1 and 2. On the other hand, along the  $b^*$  and  $c^*$  direction dispersions are renormalized a little with temperature.

Previously, precise measurements of magnetization performed by Takase et al.<sup>8)</sup> revealed the anomalous temperature dependence of magnetization of MnP in the induced ferromagnetic phase under the external magnetic field along the c-axis. The temperature dependence of magnetization deviates from the  $T^{3/2}$  law and it was approximated to be proportional to  $T \ln T$  in a temperature range from 5 K to 140 K. In order to explain this unusual temperature dependence of magnetization they proposed a model of ferromagnetic spin-wave dispersion on the analogy with rare earth metals in which both ferromagnetic and helimagnetic state are realized. They assumed a flat dispersion in a smaller-q region and step-like increase in energy at  $q = Q_h$  along the *a*-axis and  $D_{\perp} q_{\perp}^2$ relation along the direction perpendicular to a-axis, that is,

$$\begin{split} &\hbar\omega_q(T=0\mathrm{K})=D_\perp q_\perp^2+f(q_a)+\varDelta\\ &f(q_a)=\begin{cases} 0 \quad 0\leq q_a\leq Q_h,\\ &\infty \quad Q_h\leq q_a, \end{cases} \end{split}$$

where  $\Delta$  is the energy gap at q=0. By fitting the calculated magnetization to the measured one, they obtained  $D_{\perp}$  to be 39 meVÅ<sup>2</sup> at 0 K. Although parameters of their simple modeldispersions do not agree with the measured ones, the several features of this model correspond to those obtained by present measurements; flat dispersion at low-energy region and steeper increase in energy according to  $q^3$ dependence of dispersion along the  $a^*$  direction and  $q^2$  dependences along the  $b^*$  and  $c^*$  directions. Furthermore their two-magnon renormalization calculations using this modeldispersion represent the tendency of unusual renormalization along the  $a^*$  direction mentioned before. Anomalous initial decrease in magnetization would be due to the low-energy flat portion of dispersion which appear in the  $a^*$  direction.

Within a localized spin model, such an anomalous dispersion can be reproduced by assuming long-ranged exchange interactions. Actually our preliminary calculation of spinwaves using an isotropic Heisenberg model within a two-sublattice model suggests that the exchange interactions are very long-ranged, to be more precise, more than sixth-neighbour exchange interaction parameters are necessary in order to reproduce the measured spin-wave dispersions. Therefore proper account of the itinerant nature of *d*-electrons beyond a localized model is much more important for understanding the magnetism of MnP. The behaviour of spin-waves in the whole Brillouin zone as well as its damping is important information and further measurements of spinwaves at larger-q,  $\omega$  region are in progress.

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