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Inelastic Neutron Scattering Study on Crystal Field Excitations in PrMg$_3$

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Abstract. We have studied the crystalline-electric-field excitation spectrum in PrMg$_3$ with non-Kramers doublet $\Gamma_3$ ground state by inelastic neutron scattering experiments on powder and single crystal samples. Experimental results have revealed the development of a dispersive structure in the $\Gamma_3$-$\Gamma_4$ and the $\Gamma_3$-$\Gamma_5$ excitations below 50 K. Moreover, the excitation spectra of the $\Gamma_3$-$\Gamma_4$ transition were found to consist of two peaks, resulting in the two branches in dispersion relation. While one branch shows a relatively strong $q$-dependence, the other branch is almost dispersionless. The dispersion of the former branch is considered to result from the magnetic dipole exchange interaction, but the origin of the excitation of the latter branch is not clear at present.

1. Introduction
Cubic compound including non-Kramers rare-earth ion are of special interest since a doublet $\Gamma_3$ ground state with only multipole degeneracies, i.e. with no magnetic dipole but electrical quadrupole and magnetic octupole degeneracies, is possibly realized due to a crystalline-electric-field (CEF). PrMg$_3$ with the cubic Fe$_3$Al-type structure has been identified by neutron studies as the $\Gamma_3$ ground state system [1], [2]. Moreover, its relatively large CEF, the splitting $\sim 50K$ between the ground and the 1st excited states, is a favorable condition for the investigation of pure multipolar system. Recent study on the low temperature properties has revealed no cooperative phase transitions but a broad anomaly in the specific heat measurements [3]. It is suggested that the observed anomaly arises from a quenching of the multipole degrees of freedom of the $\Gamma_3$ ground state by forming a strongly correlated state through the hybridization of with the conduction electrons.

In this paper, we report the results of the inelastic neutron scattering experiments on the powder and the single crystal of PrMg$_3$. Galera et al. [2] reported a noticeable dispersion in the
excitation spectra of the $\Gamma_3$-$\Gamma_4$ transition at 8 K for a polycrystalline sample. Our experimental results have revealed the development of a dispersive structure in the $\Gamma_3$-$\Gamma_4$ and the $\Gamma_3$-$\Gamma_5$ excitations below 50 K, and two branches in the dispersion relation. We discuss the origin of the dispersion in terms of exchange interaction.

2. Experimental detail

Single crystal of PrMg$_3$ was prepared by the Bridgman method with Mo crucible sealed in a high vacuum. The powder sample was obtained by grinding the single crystals. Inelastic neutron scattering experiments on the powder and the single crystal were performed using the inverted-geometry time-of-flight spectrometers LAM-D at KENS, KEK, Japan, and the IN20 triple axis spectrometer at the Institute Laue Langevin, ILL, France, respectively.

3. Experimental results and discussion

Shown in figure 1 are the powder inelastic scattering spectra at several temperatures. The observed excitations and the resultant crystal field level scheme are consistent with the previous results [1], [2]. The lower-energy excitation around $E = 4.8$ meV corresponds to the transition from the ground state $\Gamma_3$ to the 1st excited state $\Gamma_4$, and the higher one around $E = 15.2$ meV from $\Gamma_3$ to the 3rd excited state $\Gamma_5$. The 2nd excited state is the singlet $\Gamma_1$, which has no matrix element with $\Gamma_3$. At higher temperatures, a peak appears around $E = 7.8$ meV, which corresponds to the transition from the thermally populated $\Gamma_4$ to $\Gamma_1$. As shown in the inset of figure 1, the $\Gamma_3$-$\Gamma_4$ line-width is found to increase with the decrease of the temperature below 50 K. On the other hand, the $\Gamma_3$-$\Gamma_5$ line-width decreases when the temperature decreases and becomes constant from 10 K down to the lowest temperature. Note that the half-width at half-maximum (HWHM) of the $\Gamma_3$-$\Gamma_5$ transition at lowest temperature reaches $\approx 0.8$ meV, a value much larger then the experimental resolution $\approx 0.4$ meV.

![PrMg$_3$ Inelastic Neutron Spectra](image)

**Figure 1.** (Color online) Temperature variation of inelastic neutron spectra of PrMg$_3$. The inset shows the temperature dependence of half-width at half-maximum (HWHM) of the peaks around 4.8 meV and 15.2 meV, corresponding to the $\Gamma_3$-$\Gamma_4$ and $\Gamma_3$-$\Gamma_5$ transitions, respectively.

To obtain detailed information about the origin of the broadening of the excitation peaks at low temperatures, we performed inelastic scattering experiment on the single crystal along
the three representative directions Γ-X (figure 2), Γ-K-X and Γ-L of the cubic Brillouin zone. A strong \( q \)-dependence was observed, as shown in figure 2. Moreover, the excitation spectra are found to consist of two peaks, as shown in figure 3(a). The \( q \)-dependence of the two peaks, which is obtained by least square fitting with double gaussian functions, is plotted in figure 3(b). Collecting the peak positions of these two peaks at each \( q \), we obtain the dispersion relation reported in figure 3 (b). The peak with the stronger intensity and with the sharper width, corresponding to the closed circles in figure 3 (b), shows a relatively strong \( q \)-dependence. The inelastic spectra do not show any significant differences between \( T = 70 \) mK and 1.5 K although a broad peak was observed in the specific heat measurement at \( T \approx 0.8 \) K [3].

Figure 2. The constant-\( Q \) scan spectra at \( T = 70 \) mK along the Γ-X direction. The solid lines represent the fitting results.

Figure 3. (a) Inelastic spectrum at \( Q = (0 \ 0 \ 0.1) \) and \( T = 70 \) mK. The solid line represents fitting result which consist of two peak components (Dashed lines). (b) (Color online) Inelastic scattering intensity map and dispersion curves in PrMg\(_3\) at \( T = 70 \) mK. The open and closed circles denote the peak positions determined by the least square fitting to the constant-\( Q \) scan profiles. The solid curves denote calculated results (see text).

The total dispersion reaches about 2 meV, which corresponds to the full-width of the lower-energy excitation observed in the powder neutron inelastic scattering. Thus, the increase of the line-width with decreasing the temperature shown in the inset of the figure 1 arises from the development of the dispersive structure in the excitation curve. We have performed inelastic scattering experiments also on the Γ\(_3\)-Γ\(_5\) transition only along the Γ-L direction. We observed a relatively smaller dispersion \( \sim 1 \) meV with the same \( q \)-dependence (not shown) compared with that of the Γ\(_3\)-Γ\(_4\) transition. The double peak structure could not be confirmed within the experimental resolution. The broad line-width of the Γ\(_3\)-Γ\(_5\) transition observed in the powder experiment at low temperatures is also attributed to the development of dispersion.

The dispersion curve reported in figure 3 (b) shows that the Γ\(_3\)-Γ\(_4\) transition reaches its energy minimum at the L point for \( Q = (1/2 \ 1/2 \ 1/2) \), which is the propagation wave vector of
the magnetic structure in the isomorphous compound of NdMg3 [6]. Moreover, the energy of the total dispersion is comparable with the Curie-Weiss temperature, $\theta_p = -36$ K, obtained from the temperature dependence of the magnetic susceptibility [2]. These results invoke that a magnetic dipole exchange interaction make an important role in the formation of the dispersion. The dispersion relation of the excitations with the coupling treated in random phase approximation is given as $\omega^2 = \Delta[\Delta+2\alpha^2 J(q)]$, where $\omega$, $\Delta$, $\alpha$ and $J(q)$ are the excitation energy, the transition energy between the CEF states, the matrix element and the exchange interaction, respectively [4]. The strong dispersion, observed experimentally, for the $\Gamma_3-\Gamma_4$ excitation can be reproduced by taking account of up to 4th nearest neighbor (n.n.). In the figure 3(b), the solid line represents the dispersion calculated with $\alpha^2 J_n = 1.5$ meV (1st n.n.), 0.35 meV (2nd n.n.) and -0.4 meV (4th n.n.). The negative sign of 4th n.n. interaction, corresponding to a ferromagnetic-type interaction, is reasonable since the 4th n.n. locates twice as far as the 1st n.n. in the (100) direction [5]. About twice difference in the dispersion width between $\Gamma_3-\Gamma_4$ and $\Gamma_3-\Gamma_5$ transitions can be explained by the above dispersion relation, where the dispersion widths are in proportional to $\alpha^2$, which are obtained as $|\langle \Gamma_3 \cdot \Gamma_4 \rangle|^2 = 9.3$ and $|\langle \Gamma_3 \cdot \Gamma_5 \rangle|^2 = 4$ for $\Gamma_3-\Gamma_4$ and $\Gamma_3-\Gamma_5$ transitions, respectively.

While the dispersion with a strong $q$-dependence is considered to result from the magnetic dipole exchange interaction, the origin of the broad excitation almost without $q$-dependence is not clear. Recent NMR and X-ray diffraction measurements at low temperatures have revealed no evidences of a distortion [7]. The only one Pr ion crystallographic site in the paramagnetic phase results in the only one degenerated pure magnetic mode, i.e. longitudinal and transverse modes are degenerated. Recent theoretical study has shown an appearance of the double peak structure of the inelastic spectrum in term of the doublet degeneracy of the ground state and multipole exchange interactions under cubic point symmetry [8]. In order to clarify the origin of the two components of the excitation, the more detail experiments such as studies of the magnetic field dependence and $Q$-dependence of the inelastic spectrum, are necessary and in preparation.

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