ANOMALOUS MAGNETIC EXCITATIONS IN YbSb*

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We performed the inelastic neutron scattering experiment on a powder sample of YbSb in order to clarify the nature of the unusual non-magnetic phase transition at 5 K. The two broad magnetic peaks at ~13 meV and ~18 meV were observed, as reported by the previous inelastic neutron experiment. The clear difference of these magnetic excitation spectra between the paramagnetic phase and the ordered phase may indicate the change of the $4f$ electronic states due to the phase transition at 5 K. Furthermore a new magnetic excitation below ~3 meV, which is common feature in Yb-monopnictudes, was found.

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The Yb-monopnictudes YbX (X = N, P, As, Sb) have been attracting much interest because of their unusual magnetic properties. All YbX are semimetals with a cubic NaCl type crystal structure. The Yb ion in these compounds is trivalent with the $4f^{13}$ configuration. The $J = 7/2$ multiplet of Yb^{3+} splits into two doublets (Γ_6 and Γ_7) and one quartet (Γ_8)

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in the cubic crystal field (CF). From inelastic neutron scattering experiments [1,2], the CF level scheme of YbX is reported to be Γ_6 -Γ_8 -Γ_7, where the Γ_6 Kramers doublet is the CF ground state and the Γ_8 quartet is the first excited state with a large excitation energy of the order of 10 meV. In the Yb-monopnictudes, there are relatively few studies on YbSb because of the difficulty in the preparation of the samples. From Mössbauer spectroscopy measurement [3], it was reported that YbSb undergoes two successive phase transitions: an antiferromagnetic (AF) ordering at 0.32 K and a mysterious ordering at 5 K, where the phase transition at 5 K is considered to be an AF ordering with strongly reduced ordered moments or an antiferroquadrupolar (AFQ) ordering. On the other hand, neutron diffraction measurement on YbSb has not detected any long-range magnetic ordering down to 7 mK [4]. Recent specific heat measurement on a new high-quality sample of YbSb under magnetic field has reconfirmed the presence of the two phase transitions at 0.5 K and 5 K [5]. The specific heat measurement also revealed that the transition temperature at 5 K increases by applying magnetic field. This behavior is well known in the AFQ ordering systems, such as CeB_6 [6]. Contrary to the Γ_8 ground state in CeB_6, the Γ_6 doublet has not any quadrupolar degree of freedom. However the off-diagonal matrix element of quadrupole between Γ_6 and Γ_8 is large. Therefore, the AFQ ordering induced by the inter-site quadrupolar interactions may be possible in spite of the Kramers doublet ground state system. To clarify the nature of the unusual non-magnetic phase transition at 5 K, we performed the inelastic neutron scattering experiment on YbSb.

A powder sample of YbSb was prepared by a prereaction of the constituent elements in a vacuum sealed quartz tube at temperatures up to 700°C. About 10 g of the fine powder was filled under helium gas atmosphere into an Al holder and this holder was mounted in a ^{4}He cryostat. The neutron scattering experiment was done on the chopper spectrometer HET at the ISIS spallation neutron source. Background signals from the Al holder and the ^{4}He cryostat were subtracted by measurement of an empty holder in the ^{4}He cryostat.

Figure 1 shows the energy spectra at 10 K in the paramagnetic phase and at 2.4 K in the ordered phase at a scattering angle about 19.0°. The two broad magnetic peaks at ~13 meV and ~18 meV were observed, as reported by the previous experiment [2]. The two broad peaks may be correspond to the CF transition from the Γ_6 ground state to the Γ_8 first excited state which splits into two doublets. Such a double peak spectrum was also observed in YbN [1,2]. The reason for the splitting of the Γ_8 quartet as well as the broadness of the width of these peaks is not yet clear. On the other hand, the slightly but clear difference between the paramagnetic phase and the ordered phase was observed. In the ordered phase, the intensity around

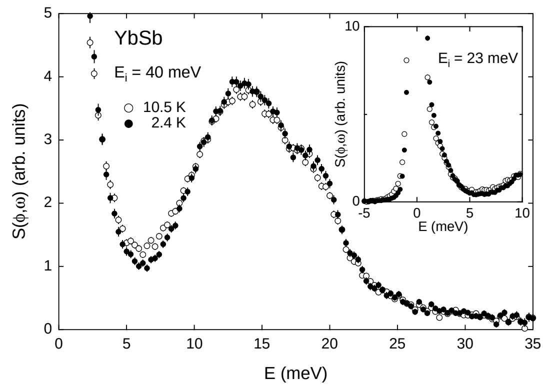

Fig. 1. Energy spectra of YbSb at 10 K (∘) and 2.4 K (•) with an incident energy of 40 meV. Inset shows energy spectra with an incident energy of 23 meV.

∼11 meV decreases, while the intensity around ∼13 meV – ∼18 meV increases. This may indicate the change of the $4f$ electronic states due to the phase transition at 5 K. Furthermore a new magnetic excitation below ∼3 meV was found, as shown in the inset of Fig. 1. This quasi-elastic scattering spectrum was also observed in other YbX (X = N, P, As) [7], so that this is common feature in all Yb-monopnictudes. The temperature dependence of the integrated intensities in the energy spectra with the incident energy of 40 meV is displayed in Fig. 2. This temperature dependence may also suggest that the $4f$ electronic states change in the ordered phase, although onset of increase or decrease of these intensities is about 7 K, slightly higher than the transition temperature of 5 K.

In Ref. [5], it is reported that Γ_3 quadrupole $O_0^2 = 3J_z^2 - J(J+1)$ is the best candidate for the order parameter of the phase transition at 5 K. Thus, let us examine whether this hypothesis reproduces the observed enhancement of the intensity around ∼13 meV – ∼18 meV in the ordered phase. Assume that the O_0^2 quadrupolar ordering occurs at 5 K. Since the Γ_6 doublet CF ground state, $|\Gamma_6^\pm\rangle = \sqrt{\frac{5}{12}}|\pm\frac{7}{2}\rangle + \sqrt{\frac{7}{12}}|\mp\frac{1}{2}\rangle$, mixes with only two states of Γ_8, $|\Gamma_8^\pm\rangle = \sqrt{\frac{7}{12}}|\pm\frac{7}{2}\rangle - \sqrt{\frac{5}{12}}|\mp\frac{1}{2}\rangle$, within the mean field approximation in this case, the ground state $|0^\pm\rangle$ and the excited state $|1^\pm\rangle$ in the ordered phase become $|0^\pm\rangle = \alpha|\Gamma_6^\pm\rangle + \beta|\Gamma_8^\pm\rangle$ and $|1^\pm\rangle = \beta|\Gamma_6^\pm\rangle - \alpha|\Gamma_8^\pm\rangle$, respectively. By calculating the transition matrix elements $|\langle 0^\pm|J_z|1^\pm\rangle|^2$, we can get the reasonable parameter set, $\frac{\sqrt{35}}{6} < \alpha < 1$ or $-\frac{1}{6} < \alpha < 0$ ($\beta > 0$), which qualitatively reproduces the present experimental result. For further check on this hypothesis, a precise X-ray diffraction experiment and a neutron diffraction experiment under magnetic field may be needed.

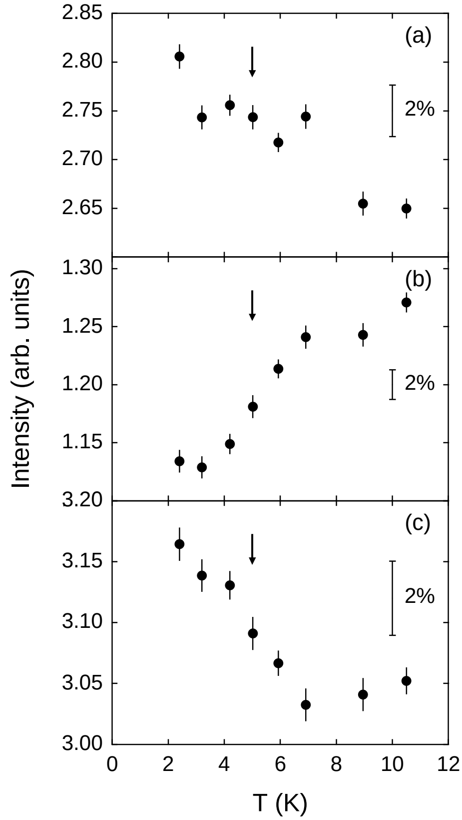

Fig. 2. Temperature dependence of the integrated intensities in the energy spectra of YbSb with the incidentenergy with 40 meV (a) between 1 meV and 3 meV, (b) between 3 meV and 11 meV and (c) between 11 meV and 21 meV. Arrows denote the phase transition temperature of 5 K.

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