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ScienceDirect
Journal of Magnetism and Magnetic Materials 310 (2007) 766–767
Journal of magnetism and magnetic materials
www.elsevier.com/locate/jmmm

Inelastic neutron scattering measurements in a quadrupolar ordering compound YbSb

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Available online 30 October 2006

Abstract

Inelastic neutron scattering measurements were performed to study the excitation spectra of a quadrupolar ordering compound YbSb. We observed the three peaks (12.5, 17 and 21 meV) around $\Gamma_6-\Gamma_8$ excitations far above the quadrupolar ordering temperature (T_Q). This result suggests that the magnetic excitations have a dispersion rather than the splitting of Γ_8 states at high temperatures. We also observed the strong q dependence in q scans at energy $=$ 1 meV above T_Q. The observed q dependence above T_Q suggests that the theoretical approach beyond the molecular field approximation is required in order to understand the magnetic properties of Yb monopnictides.

PACS: 61.12.−q; 71.20.Eh

Keywords: Quadrupolar ordering; YbSb; Neutron scattering

1. Introduction

Yb monopnictides (YbN, YbP, YbAs and YbSb) have attracted much interest as they show heavy fermion characteristics such as a large γ value of specific heat and reduced antiferromagnetic ordered moments although the carrier concentration is only several percents per Yb atom. The antiferromagnetic ordering temperatures (T_N) of YbN, YbP and YbAs are 0.74, 0.41 and 0.49 K [1] while the estimated exchange interactions are of the order of 10 K [2]. In contrast to other Yb monopnictides, YbSb shows two successive phase transitions; an antiferromagnetic ordering at 0.32 K and another phase transition at 5 K [2]. On the other hand, neutron diffraction measurements did not observe any long range ordering down to 7 mK. Recent observations by specific heat and NMR measurements reveal the phase transition at 5 K is of a mixing-type quadrupolar ordering [3,4]. Thus, YbSb has attracted renewed interest of those compounds. A molecular field approximation including quadrupolar interactions demonstrates that the mixing-type quadrupolar ordering occurs even in the ground states which do not have a quadrupolar moment and the first excited states are at 150 K. The model calculations explained the magnetic properties of Yb monopnictides qualitatively, however, there are still some problems which have not been resolved. The splitting of the first excited states (Γ_8) of the crystal field splittings is the most anomalous property. Keller et al. proposed that the unexpected splitting is explained by the occurrence of a quadrupolar ordering around 80 K in YbN, YbP and YbAs [5]. Kuwahara et al. has observed the similar splitting in YbSb [6]. However, there is no other evidence of the quadrupolar ordering around 80 K. In this study, we concentrate on the excited states in YbSb to study the dynamical properties above the quadrupolar ordering temperature ($T_Q = 5$ K) and to clarify the origin of the unexpected splitting below 80 K. The powder samples of YbSb were prepared by a reaction of the constituent elements in an evacuated quartz tube.

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doi:10.1016/j.jmmm.2006.10.167

2. Results and discussions

The energy spectra below 4 meV were measured on the triple axis spectrometer C1-1(HER) at the JRR-3M of JAEA. Fig. 1 shows the results of q scans with a fixed $k_f = 1.5061\,\text{Å}^{-1}$ at $E = 1\,\text{meV}$. There is a strong q dependence of intensity in q scans at energy $= 1\,\text{meV}$ as shown in Fig. 1. The decrease of intensity in small q vectors is noticeable although we used powder samples. The q dependence develops with decreasing temperature below 20 K. The another peak at $h = 2.0$ is a contamination from the nuclear incoherent peak. The energy spectra can be interpreted as the Gaussian function (an incoherent peak at 0 meV) and a Lorentzian function (a quasi-elastic peak) convoluted by the energy resolution. The temperature factor and k_i/k_f factor are calibrated. The fitting of the energy spectra shows that the decrease of the intensity at $h = 0.6$ is ascribed to the decrease of the width of the Lorentzian function. Ohoyama et al. reported that q dependence of the quasi-elastic peak in powder samples and a single crystal of YbAs exist up to 20 K [7,8]. The q dependence was ascribed to the short range magnetic correlations of an ordering wave vector $[1, 0, \frac{1}{2}]$. We should note that the magnetic correlations still exist above $40T_N$ ($T_N = 0.49\,\text{K}$). Therefore the q dependence observed in this study in YbSb seems to be the common feature of the Yb monopnictides. The magnetic correlations survive up to 20 K, which is in contrast to the $T_N = 0.32\,\text{K}$ in YbSb. The measurements of the correlations below T_Q are in progress.

Fig. 2 shows the energy spectra between 1 and 27 meV of powder samples of YbSb at 20 and 100 K measured on the triple axis spectrometer 6G(TOPAN) at the JRR-3M research reactor of JAEA. Three peaks at 12.5, 17 and 21 meV around $\Gamma_6 - \Gamma_8$ excitations were observed. Two peaks are consistent with the previous results, however, in addition to these two peaks we found a new peak at 12.5 meV. This new peak develops below 80 K with decreasing temperature. Here, we note that the temperature where the intensity of the new peak starts to develop is similar to the quadrupolar ordering temperatures in YbN, YbP, YbAs which Keller et al. proposed although it is very difficult to understand that the localized states like Γ_8 quartet split into three. If we assume that a quadrupolar ordering occurs at 80 K, the molecular field approximations show that Γ_8 states should split into two. Furthermore, we found the small q dependence of intensity at the energies of three peaks. These results may offer a new interpretation of the long-standing mystery of Γ_8 states; there is a dispersion in Γ_8 states rather than the splitting. Rather large magnetic exchange interactions and/or quadrupolar interactions between Γ_8 states are suggested. Further investigations of the nature of Γ_8 states are urgently required.

The observed q dependence above T_Q and the suggested strong interaction between Γ_8 states require the theoretical approach beyond the molecular field approximation in order to understand the magnetic properties of Yb monopnictides.

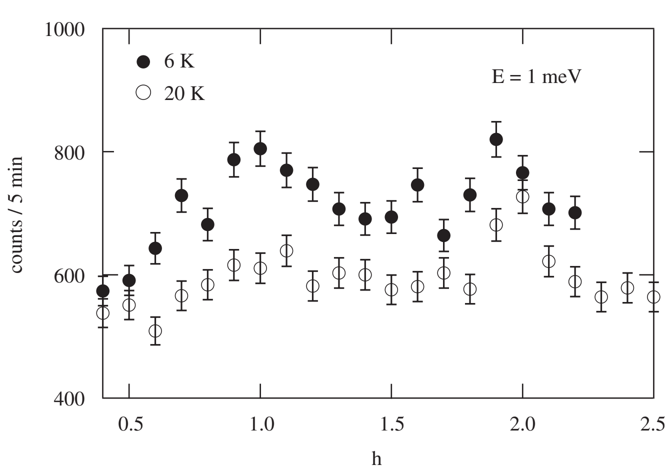

Fig. 1. The q scans at 1 meV with a fixed $k_f = 1.5061\,\text{Å}^{-1}$.

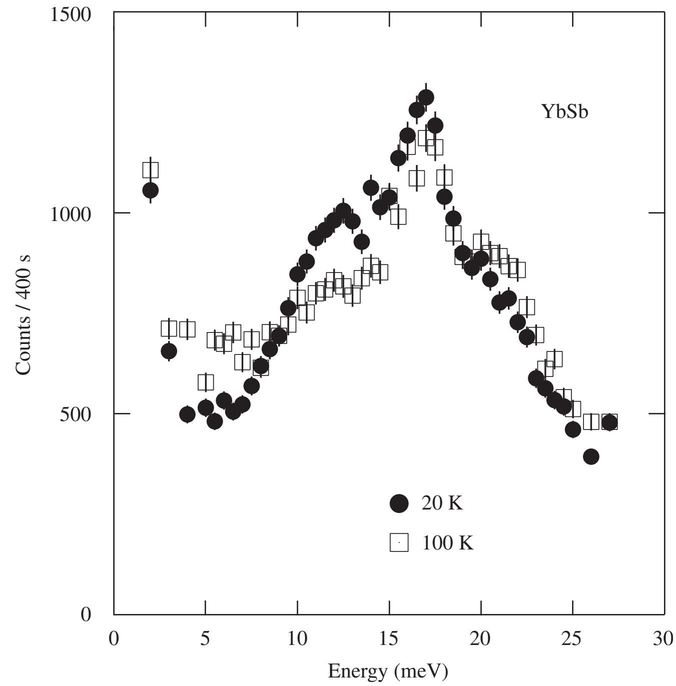

Fig. 2. Inelastic neutron scattering spectra measured at $q = (2.2, 0, 0)$.

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