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A. OYAMADA and T. SUZUKI

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Spin Dynamics of YbX (X=N, P, As)

K. Ohoyama [a], M. Kohgi [a], T. Otomo [b], T. Osakabe [a], A. Oyamada [a] and T. Suzuki [a]

[a] *Department of Physics, Tohoku University, Aramaki Aoba Sendai 980, Japan*

[b] *Institute for Materials Research, Tohoku University, Katahira Aoba Sendai 980, Japan*

Abstract

The neutron quasi-elastic scattering (QES) spectra for YbX (X=N, P, As) show a quite common feature independent of X. The Kondo effect almost completely suppresses the magnetic ordering, which indicates that T_K is comparable to T_{RKKY} of about 20K. The crossover of QES spectra from Gaussian-like to Lorentzian-like form occurs with the increase of temperature.

Keywords: Neutron quasi-elastic scattering
Spin fluctuations
Dense Kondo effect
YbN, YbP, YbAs

Contact Address: K. Ohoyama
Institute for Materials Research,
Tohoku University
2-1-1 Katahira Aoba-ku Sendai 980, Japan
Tel: 001-81-22-215-2037
Fax: 001-81-22-215-2036

1. Introduction

YbX (X= N, P and As), which crystallizes in the NaCl structure, is a semimetallic compound with low carrier density (a few percent per formula). At low temperatures around 0.5 K, YbX undergoes antiferromagnetic long range ordering [1]. However, the saturation moment and entropy at the Néel temperature are considerably smaller than the expected values of the crystal field ground state doublet. It also shows an anomalously enhanced specific heat around 5 K. These are considered to be the characteristic features of the Kondo effect. We have studied the low carrier Kondo effect in this system by means of neutron inelastic and quasi-elastic scattering. Some results of the inelastic scattering work were reported elsewhere [2] [3], and the neutron quasi-elastic scattering (QES) experiments on these compounds are presented here.

2. Experimental

We performed neutron quasi-elastic scattering experiments on powder samples of YbX (X=N, P and As) on both the crystal analyzer type spectrometer, LAM-D, at the spallation neutron source KENS in National Laboratory for High Energy Physics (KEK), and the triple axis spectrometer, TOPAN, at the JRR-3M in Japan Atomic Energy Research Institute (JAERI). LAM-D uses a fixed final neutron energy of about 4.5 meV. This condition facilitates the high resolution experiments in lower energy transfer. The samples were the same as those used in the inelastic scattering work.

3. Results and Discussions

Figure 1 shows QES spectra of YbN at a scattering angle of 35° using LAM-D at 14 K (upper portions) and room temperature (lower portions). Figures 1(a) and (b) show the same observed spectra and results of different methods of analysis, as described below. The observed spectra (open circles) were corrected for the background, absorption and wavelength dependence of incident neutron flux. The results of the least squares fit of the QES spectra to calculated line shapes are also shown in Fig.1 (chain lines). The QES spectra are fitted to either a Gaussian

(Fig.1(a)) or a Lorentzian (Fig.1(b)) line shape convoluted with the instrumental resolution. The dotted lines correspond to the incoherent elastic scattering parts of the spectra and indicate the energy resolution as well. The dependence of the intensity on the temperature cannot be discussed here since the experimental conditions for the spectra at 14 K and room temperature are not identical. As seen in the figures, the QES spectra at lower temperatures below 50 K are explained well by the Gaussian line shape, whereas, at higher temperatures, the Lorentzian line shape gives somewhat better agreement than the Gaussian one. The QES spectra of YbP and YbAs show exactly the same features.

The important feature of the crossover from the Gaussian line shape to the Lorentzian one with the increase of temperature is seen in the plot of the temperature dependence of the reliability parameter, called the R-factor of fitting. The R-factor is defined as $\sqrt{\sum_i (I_i^{obs} - I_i^{cal})^2 / \sum_i I_i^{obs}}$, where $I_i^{obs(cal)}$ indicates the intensity at the i-th channel for an observed (calculated) spectrum. Figure 2 shows the temperature dependence of the R-factors obtained by the Gaussian (open circles) and Lorentzian (closed circles) fitting for the three compounds. The lower dashed lines indicate statistical minimum values of the R-factors. It is quite remarkable that the behavior of the crossover is common for these compounds. Furthermore, the transition point of about 150 K is also the same for the compounds.

In Fig.3, the linewidths of the QES spectra (HWHM) of YbN (circles), YbP (squares) and YbAs (triangles) determined from the fitting are plotted against temperature. Open and closed symbols indicate the results of the Gaussian and Lorentzian fittings, respectively. The data below 10K were taken with the triple axis spectrometer TOPAN with incident neutron energy of 14 meV. The results observed with TOPAN are consistent with those observed with LAM-D at higher temperatures. The results of the Lorentzian fitting below 50 K and those of the Gaussian fitting above 200 K are not shown in Fig.3, because the QES spectra cannot be explained well by these line shapes. The observed linewidths of the QES spectra of these three compounds are almost the same, and increase with temperatures up to 50 K. They are almost constant or even decrease slightly above 150 K.

We have also measured the momentum transfer (Q) dependence of QES spectra of YbN at 10 K and room temperature and of YbP at 1.5 K

and room temperature using TOPAN for $1.1\ \text{Å}^{-1} \le Q \le 1.7\ \text{Å}^{-1}$. Figure 4 shows the Q dependence of the linewidth (a), HWHM, and integrated intensity (b) of YbN at 10 K. The dotted line in Fig.4(b) indicates the Q dependence of the square of the magnetic form factor f(Q) of free Yb^{3+} ion [4]. It is concluded that the linewidth and integrated intensity are nearly independent of Q. We also observed the Q-independent feature of QES spectra of YbP at 1.5 K. No appreciable Q dependence of the QES spectra of either compound was observed at room temperature. Therefore, the spatial magnetic correlations in these compounds might not be strong enough to be detectable in powder samples.

Now, we discuss the magnetic fluctuations for the ground state Γ_6 in Yb monopnictides. The c-f mixing effects for the ground states are concluded to be common in Yb monopnictides, since the QES spectra are essentially the same for these three compounds. The spin fluctuations at low temperatures are governed by the RKKY-type interatomic exchange interactions since the observed QES spectra of the YbX compounds are explained well by the Gaussian function rather than the Lorentzian one below about 50 K. The characteristic energy of the exchange interactions in these compounds is estimated to be about 20 K from the linewidth at about 50 K. The big difference between the actual Néel temperatures (~0.5 K) and the exchange energy (~20 K) arises in the strong suppression of the magnetic ordering by the Kondo effect. It is also concluded that the characteristic energy T_K in the YbX compounds is about the same value due to the complete suppression of the magnetic ordering. This value is about four times larger than the value previously estimated from the anomalous peak of the specific heat based on the impurity Kondo model [5].

The gradual change of the spectral line shape from the Gaussian-like function to the Lorentzian-like one with increasing temperature indicates that the single-site relaxation process is not appreciable at low temperatures, but becomes important at higher temperatures. The Lorentzian linewidth of 10 K at room temperature is relatively small compared with those of the typical dense Kondo materials such as CeB_6, $CeAl_2$ or $CeCu_2Si_2$ [6]. However, if the fact that the carrier density is very low is taken into account, it should be noted that the c-f mixing effect in the YbX system is rather strong.

A more detailed study of the QES spectra using single crystal samples is now in progress.

Acknowledgements

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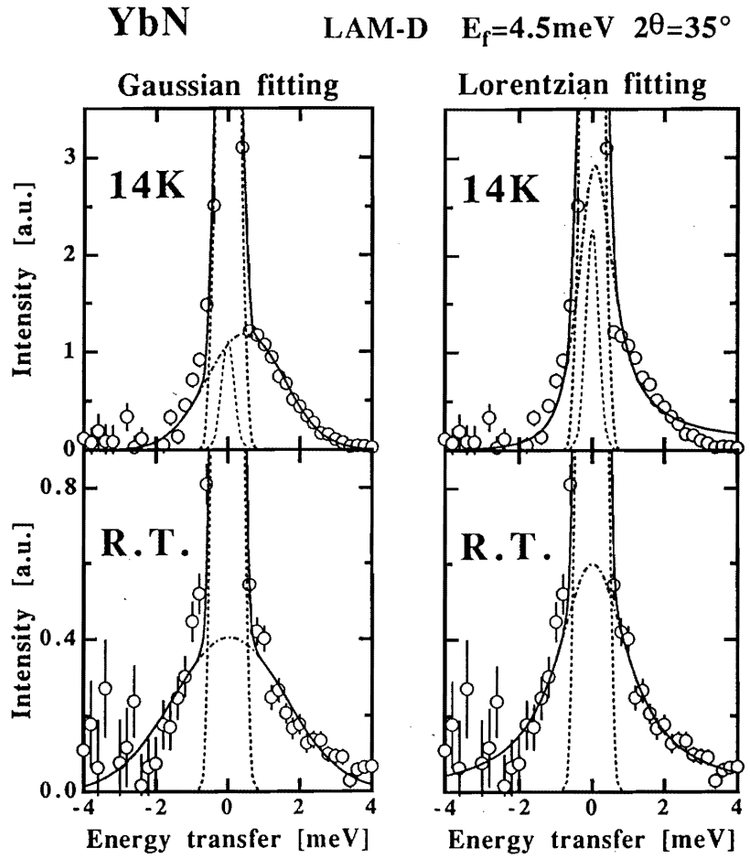
Figure Captions

Fig.1 QES spectra of YbN at $2\theta=35°$ using LAM-D with $E_f=4.5$ meV at 14 K and room temperature. The solid lines indicate the Gaussian (a) and Lorentzian (b) spectra convoluted with the instrumental resolution. The chain lines and dotted lines show the quasi-elastic and incoherent elastic parts of the spectra, respectively.

Fig.2 R-factors for the Gaussian (open circles) and Lorentzian (closed circles) fits as a function of the temperature. The lower dashed lines indicate the statistical minimum values of the R-factors.

Fig.3 Temperature dependence of the quasi-elastic linewidth (HWHM) of YbN (circles), YbP (squares) and YbAs (triangles). The open and closed symbols represent the Gaussian and Lorentzian linewidths, respectively.

Fig.4 Q dependence of the linewidth (a) and integrated intensity (b) of the QES spectra of YbN at 10 K using TOPAN with $E_i=14$ meV. The dotted line indicates the square of the magnetic form factor of free Yb^{3+} ion [4].


YbN
LAM-D E_f=4.5meV 2θ=35°
Gaussian fitting
Lorentzian fitting
14K
14K
R.T.
R.T.
Intensity [a.u.]
Intensity [a.u.]
Energy transfer [meV]
Energy transfer [meV]

(a) (b)

Fig.1

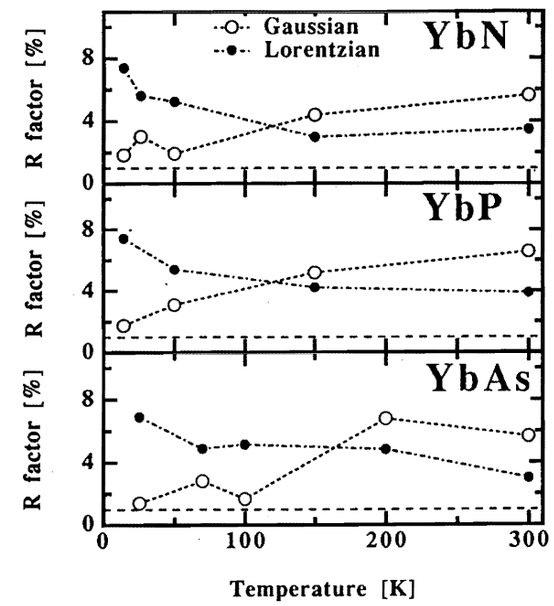

Gaussian
Lorentzian
YbN
YbP
YbAs
R factor [%]
Temperature [K]

Fig.2

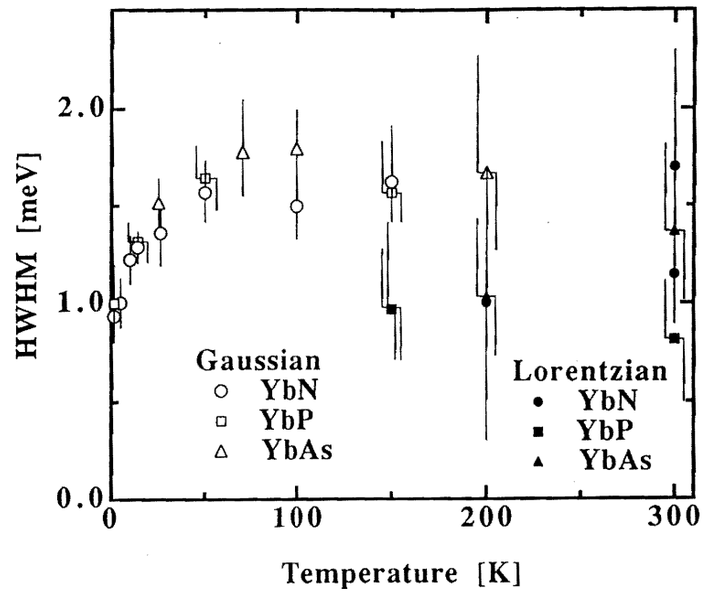

HWHM [meV]
2.0
1.0
0.0
0
100
200
300
Temperature [K]
Gaussian
YbN
YbP
YbAs
Lorentzian
YbN
YbP
YbAs

Fig.3

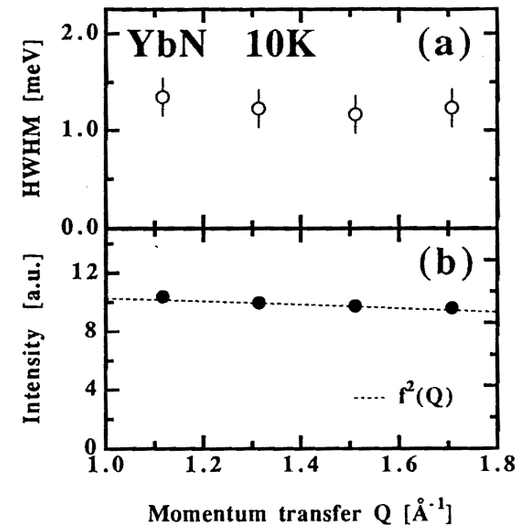

YbN 10K
(a)
HWHM [meV]
2.0
1.0
0.0
(b)
Intensity [a.u.]
12
8
4
0
f²(Q)
1.0
1.2
1.4
1.6
1.8
Momentum transfer Q [Å⁻¹]

Fig.4