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Crystal Field Levels in YbBiPt

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Abstract

The new massive-electron system YbBiPt has been studied by means of neutron inelastic scattering at temperatures between 3.3 and 77K, using the crystal-analyser spectrometer LAM-40. Crystal field excitations have been observed at approximately 6 meV and at very low energy. There is also significant quasielastic scattering and the higher-energy level softens slightly with increasing temperature. While these observations are in qualitative agreement with the bulk susceptibility measurements, our data suggest that the first excited crystal-field level is much lower in energy than had been deduced from bulk measurements.

1. Introduction

The cubic heavy-fermion compound YbBiPt was discovered in 1991 by Canfield et al. [1,2] and has very large linear specific-heat coefficient $\gamma = 8J\ mol^{-1}\ K^{-2}$. It has previously been characterised by means of electrical-resistivity, magnetoresistance, ac and dc magnetic-susceptibility and specific-heat measurements, and in order to explain these data, it was suggested that the cubic crystal field splits the $4f_{7/2}$ Yb^{3+} Hund's rule state into two doublets Γ_7 and Γ_6 and a quartet Γ_8, in that order, with the two doublets split by approximately 1meV. The main purpose of the work reported here has been to clarify this situation by observing the crystal-field levels directly using neutron scattering. YbBiPt forms in the $C1_b$ or MgAgAs structure type (space group $F\bar{4}3m$), which is a common Heusler-alloy structure[3], and in which some uranium-based ternaries like UNiSn[4] form. The Yb atom lies on the same site as Mn or U in these compounds, with point group symmetry $\bar{4}3m$.

2. Method

The sample was made by growth in a bismuth flux, as reported previously[1,5]. 9.6g of polycrystalline material was sealed under helium gas in a thin-walled aluminum tube and mounted within a helium cryostat. Neutron powder diffraction analysis showed the presence of a small amount of elemental bismuth in the sample, presumably from the flux. The cryostat was mounted in the time-of-flight crystal-analyser spectrometer LAM-40 at KENS, the spallation neutron source of the Japanese National Laboratory for High-Energy Physics (KEK). This spectrometer[6] views a solid-methane moderator and employs 7 large-solid-angle pyrolytic-graphite focussing analysers, each with its own beryllium filter and detector. The final energy is 4.6 meV. The analyser arms were positioned at scattering angles of 8°, 24°,40°, 56°,72°, 88° and 104°, though in our analysis we have only used the four higher-angle detectors, because of contamination from the cryostat walls. The instrumental

resolution has a width of 0.35 meV FWHM, with some asymmetry due to the pulse se shape of the incident neutrons, and one can measure in neutron energy loss down to 0.3meV with insignificant influence from incoherent inelastic scattering.

3. Results

The raw data were normalised to the monitor count, corrected for the wavelength variation of the incident spectrum and the k_f/k_i term in the cross section, and the resultant resolution-broadened scattering functions $S(Q,\omega)$ measured at 3.3, 10, 30 and 77K are shown in Figure 1. At low temperature, there is a clear excitation at 5.7 meV, and it softens to 5.1 meV with temperature at 77K. Its intensity decreases with temperature, indicating that it is magnetic in origin and not a phonon. No other strong magnetic peaks were observed above 6 meV. In addition, there is very significant quasielastic scattering, which we were unable to fit using a single gaussian (or Lorentzian) response function. However, we can obtain reasonable fits to the observed spectra with a two-component quasielastic response, one with a width roughly twice that of the resolution function and the other ten times broader. The second component is quasielastic to within 0.05 meV. Gaussian lineshapes were assumed in all cases and the fits are shown in Figure 1. We tried Lorentzian lineshapes in some cases, but the agreement with observation was always worse. In all subsequent discussion, we assume the imaginary part of the generalised susceptibility to have the following form:

$$\mathrm{Im}\,\frac{\chi(Q,\omega)}{\hbar\omega} = c_1\, e^{-(\hbar\omega/\sigma_1)^2} + \sum_{i=2,3} c_i\, e^{-(\hbar(\omega \pm \omega_i)/\sigma_i)^2} \qquad (1)$$

where the first term is the true quasielastic response, and the second two terms are inelastic, with $\hbar\omega_2 \approx 0$ and $\hbar\omega_3 \approx 6$ meV. While the quasielastic linewidth hardly changes with

temperature, the two inelastic excitations broaden monotonically as temperature is increased. There is also an indication, in Figure 1, of some magnetic scattering at about 8 meV. But it is very weak, too weak for the published matrix elements[7,8], and at this point, we have little idea as to its origin.

4. Discussion

Our data clearly indicate that the quasielastic spectra are more gaussian-like in character, than Lorentzian-like. This means that the spin fluctuations are mainly due to inter-site spin-spin correlations, rather than single-site spin relaxation. In other words, there are strong (para)magnetic correlations at least up to 30K. From the width of the broad quasielastic component, we estimate that the correlation energy is approximately 20K. Secondly, within our model of a three-component gaussian-lineshape response function, we can integrate with respect to energy to obtain the susceptibility $\chi(Q)$. As we do not yet have an absolute intensity normalisation, we have scaled our result to the bulk measurement[2] at 77K, and the results are shown in Figure 2. If this scaling is correct, the agreement is quite good. While our analysis is preliminary, it does seem that there are three components in the spectrum, and it is natural to associate these with the three crystal field states of the Yb^{3+} ion in a cubic potential, Γ_6, Γ_7 and Γ_8. The interesting thing is that the lower two states are degenerate, or almost so (to order 0.1 meV). Thus, the ground state is either four-fold degenerate (Γ_6 and Γ_7) or six-fold degenerate (Γ_8 and Γ_6 or Γ_7). The point charge model would give Γ_7, Γ_8 and Γ_6, in that sequence[9] and the ratio of our observed intensities for the 6 meV excitation (at 3.3 and 77 K) is in good agreement with a 6-fold degenerate ground state, i.e. Γ_7 with Γ_8 directly above. Considering the fact that there is no magnetic long-range order above 0.4K, in spite of strong magnetic correlations with characteristic energy 20K or so, we conclude that there is a strong Kondo effect. Qualitatively, it seems that this strong Kondo suppression together with the high multiplicity of the ground state gives rise to

the giant enhancement of γ. We are currently in the process of performing a complete quantitative analysis simliar to that made on other cubic Yb compounds[7,8], and this will be the subject of a subsequent publication.

Acknowledgements

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Figure Captions

Figure 1. Observed time-of-flight spectra from YbBiPt, taken at 3.3, 10, 30 and 77K. The solid lines depict fits using the three-component gaussian model described in the text.

Figure 2. Magnetic susceptibility of YbBiPt as obtained from neutron scattering intensity (open circles) and as reported in Ref. 2 (solid line). The neutron data are scaled to the bulk measurement at 77K.

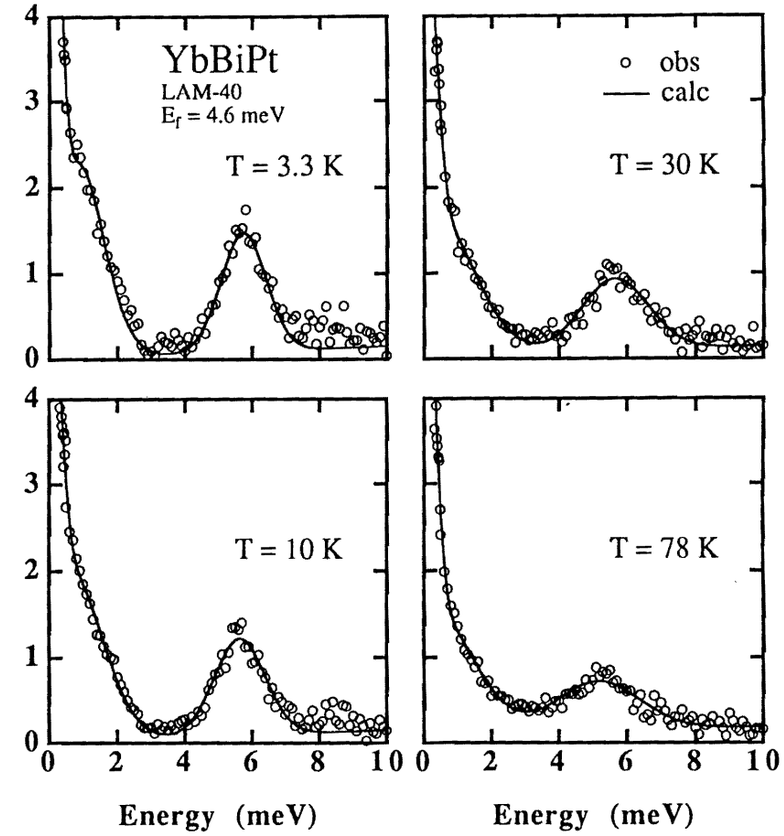

Fig. 1

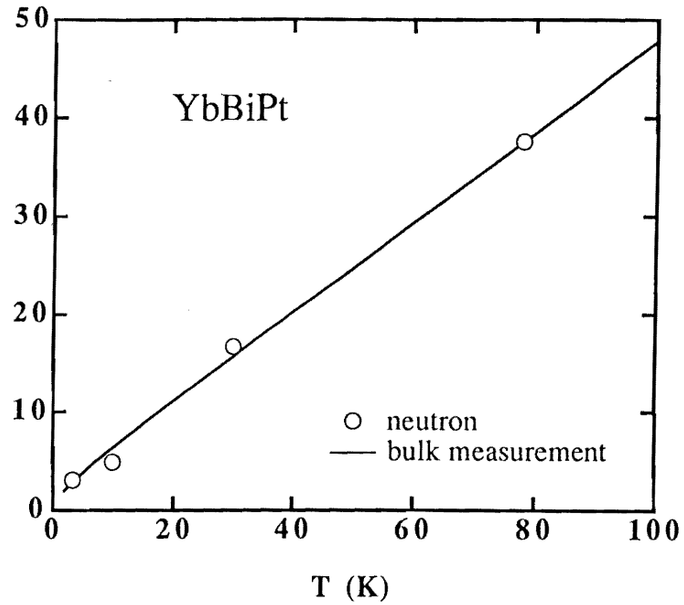
YbBiPt
neutron
bulk measurement
0
10
20
30
40
50
0
20
40
60
80
100
T (K)

Fig. 2