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Low-Energy Excitations and the Electronic Specific Heat of YbBiPt

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Neutron scattering data from the cubic heavy-fermion compound YbBiPt are presented, along with an interpretation in terms of strongly broadened crystal-field levels. To first order, the ground state is sixfold degenerate, consisting of a doublet (Γ_7) and quartet (Γ_8). Integration over these levels reproduces the observed linear specific-heat coefficient γ of 8 $J\,mol^{-1}\,K^{-2}$ to within $\pm 20\%$. At the lowest temperatures, an extra symmetry-forbidden splitting is observed.

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The discovery of a huge linear-temperature coefficient of specific heat $\gamma = 8\ J\,mol^{-1}\,K^{-2}$ in YbBiPt [1,2] has generated considerable excitement. This value is an order of magnitude more than in typical heavy-fermion materials [3] and three orders of magnitude greater than in normal metals. The origin of this huge specific-heat contribution is of central interest to the understanding of the electronic properties of heavy-fermion systems, and we have been carrying out inelastic neutron scattering experiments to probe the low-energy dynamics of this material directly. We find that the Γ_8 quartet and Γ_7 doublet crystal-field levels lie at low energies and are broadened strongly relative to their splitting by intersite exchange interactions. The calculated specific heat, based on these broadened magnetic excitations, accounts semiquantitatively for the enormous experimentally observed specific heat of this material.

YbBiPt forms in the face-centered cubic $C1_b$ or MgAgAs structure type in which all three atoms are located on sites with tetrahedral point-group symmetry $\bar{4}3m$. Of the four such sites in the space group $F\bar{4}3m$, one is vacant, two have such vacancies as nearest neighbors, and the fourth site is *unique* in that all the nearest-neighbor sites are occupied. In a recent crystallographic

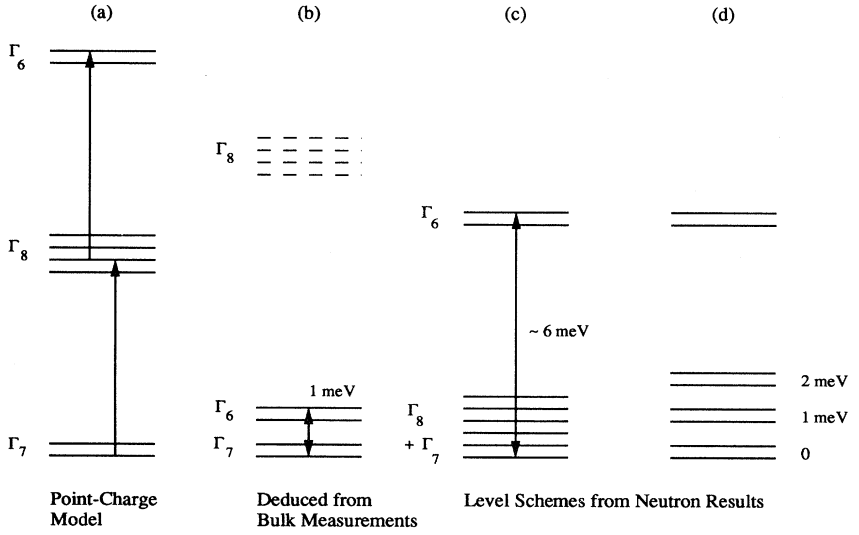

FIG. 1. Possible schematic crystal-field level schemes for YbBiPt: (a) The result for a point-charge calculation assuming $-3e$ on the Pt tetrahedron surrounding the Yb^{3+} ion. (b) The scheme proposed by Fisk *et al.* [2]. (c) The cubic scheme proposed in Ref. [8] and explained here. (d) The noncubic scheme invoked to explain the new data reported in this Letter. In YbBiPt, these levels are not sharp, but are strongly broadened if not overdamped.

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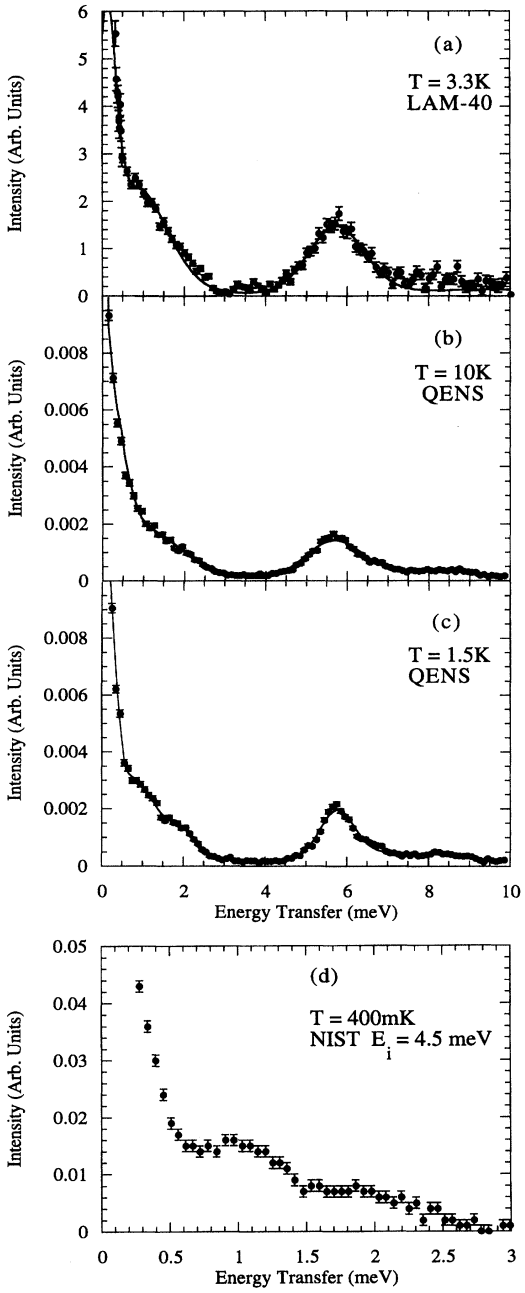

FIG. 2. Observed time-of-flight spectra from YbBiPt, taken at (a) 3.3 K on LAM-40 (see Ref. [8]), (b) 10 K and (c) 1.5 K on QENS, and (d) the sum of data taken at 0.45 and 0.35 K on the NIST time-of-flight spectrometer using $E_i = 4.5$ meV. The solid lines in (a) and (b) depict fits using a three-component Gaussian model described in the text.

study [4], we have shown that the Pt ion is on the unique site, in contrast to what had been reported previously [2]. The Yb ion therefore has only Pt ions as nearest neighbors. In a study of the whole RBiPt series [1], where R symbolizes the rare-earth element, the lattice parameter of YbBiPt showed no deviation from the general trend, and this is good evidence that Yb is in the Yb^{3+} state.

In all heavy fermions, the correlated state can be thought of as developing from a crystal-field eigenstate (a doublet in Ce-based heavy fermions). The subsequent many-body interactions with the conduction electrons broaden this underlying crystal-field state, giving a large continuous density of excited states down to zero energy. In YbBiPt, the cubic crystal field splits the eightfold degenerate $J = 7/2$ state of Yb^{3+} into two doublets, Γ_6 and Γ_7, and a quartet, Γ_8. Assuming a simple point-charge model, one arrives at a crystal-field scheme such as that shown in Fig. 1(a), where the levels are Γ_7, Γ_8, and Γ_6 in that order, and the total splitting is of order 12 meV. However, in order to interpret their original susceptibility data, Fisk *et al.* [2] invoked a crystal-field level at roughly 1 meV and proposed the level scheme given in Fig. 1(b). Their specific-heat results gave an integrated entropy of $R\ln 2$ by 1 K, indicating an electronic degeneracy of 2 below 0.1 meV, and $R\ln 5$ by 20 K.

We have conducted inelastic neutron scattering experiments using the LAM-40 spectrometer [5] at the Japanese National Laboratory for High-Energy Physics (KEK), the QENS spectrometer [6] at Argonne National Laboratory, and the Cold-Neutron Fermi-Chopper Spectrometer [7] at the National Institute of Standards and Technology (NIST). LAM-40 and QENS are time-of-flight crystal-analyzer spectrometers with quasielastic resolutions of 0.35 and 0.07 meV FWHM, respectively. The NIST experiments were performed in a ^{3}He cryostat using incident energies E_i of 4.5 and 2.7 meV, giving resolutions of 0.28 and 0.11 meV, respectively. The work on LAM-40, a preliminary account of which has been published previously [8], showed a clear crystal-field level just below 6 meV, with a more complicated response at low energies as shown in Fig. 2(a). Other than some weak magnetic scattering at around 8 meV, no further excitations were seen up to 20 meV. Although we show only the lowest temperature data at 3.3 K in Fig. 2(a), data were also collected at various temperatures up to 77 K. The temperature dependence of the intensity of the 6 meV level can be used to extract its degeneracy, g_1, relative to that of a hypothetical sharp ground-state level, g_0. The intensity of the 6 meV line is shown in Fig. 3(a), while the energies and widths obtained from the fits are shown in Figs. 3(b) and 3(c), respectively. The temperature dependence of the intensity can be fit assuming it to be proportional to the total ground-state occupancy n_0 given by

$$n_0 = \{1 + (g_1/g_0)e^{-\hbar\omega/kT}\}^{-1}. \quad (1)$$

We obtain $g_1/g_0 = 0.56 \pm 0.10$, which is clear evidence that the 6 meV level is a doublet and not a quartet. The level also softens slightly with increasing temperature, as shown in Fig. 3(b).

The entire spectrum can be fit fairly well assuming that the imaginary part of the dynamic susceptibility consists of three Gaussian components, two of which are

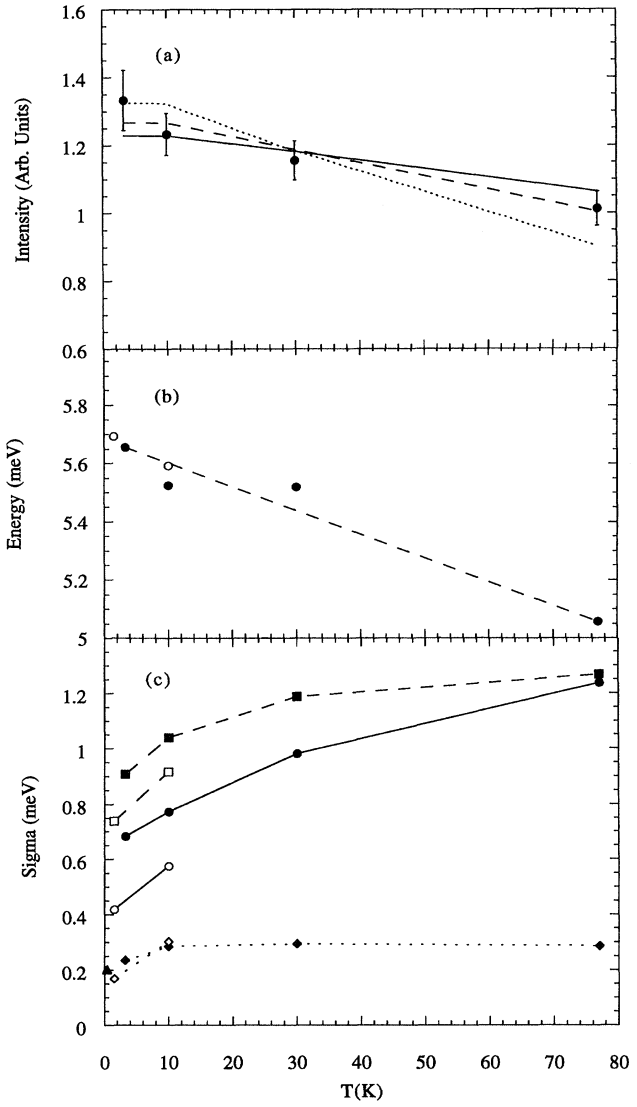

FIG. 3. The variations with temperature of (a) the intensity and (b) the energy of the crystal-field excitation just below 6 meV, and (c) the intrinsic widths of all three levels from fits to the data. In all three panels, the solid symbols represent data taken on LAM-40, while the open symbols represent data taken on QENS. In (a) the lines are all least-squares fits to the data: the solid line assumes ground-state degeneracy of 6, the dotted line, 4, and the dashed line is the best fit, giving $g_1/g_0 = 0.56$ as described in the text. In (c) the dotted line refers to the sharp quasielastic response (σ_1), the dashed line the broad quasielastic response (σ_2), and the solid line that of the inelastic line (σ_3) at ~6 meV. The triangle represents data taken at NIST, with $E_i = 2.7$ meV.

quasielastic and the third is the inelastic level with an energy of roughly 6 meV. All the data in Ref. [8], along with the QENS data shown in Figs. 2(b) and 2(c), are better fit by this model than any model consisting of Lorentzians, indicating that the normal single-site Kondo picture is inappropriate and that intersite correlations must be important. One quasielastic component is narrow and has a width of roughly 0.2 meV, which corresponds to a correlation temperature of 2 K, and the second quasielastic component is broader. Given these observations, the fact that the scattering below 2 meV must contain both the Γ_8 quartet and one of the doublets and the fact that the specific heat only gives an entropy of $R\ln 2$ by 1 K, we proposed [8] the level scheme given in Fig. 1(c). In this scheme, the sharp quasielastic response would belong to the Γ_7 doublet, while the broad quasielastic response would belong to the quartet. To a first approximation the ground state contains both, but they are hybridized with the conduction electrons in different ways. It is well known that the quartet is very sensitive to perturbations [9], and it is plausible that it is hybridizated more strongly. The temperatures dependences of the widths, which can be thought of as correlation or Kondo temperatures, are shown in Fig. 3(c).

However, the fits are not perfect, and there is clearly some structure within the "broad quasielastic" response, especially at low temperature. This is even clearer in the data shown in Fig. 2(d), some of which were taken at 350 mK and some at 450 mK. The intent was to look for differences above and below the 400 mK phase transition [1,10]. No differences were found and to improve statistics, the two data sets have been added. There are clearly two separate levels at roughly 1 and 2 meV, respectively. Similar data [11] have been observed in a single crystal of YbBiPt at Risø, and no dispersion of these levels has been seen so far. This splitting is not correlated with the 400 mK transition, as we see it at temperatures well above 400 mK. In the level scheme we propose above, it is now natural to assume that the fourfold degeneracy of the Γ_8 is lifted by a noncubic field, to give a total of four Kramers doublets as shown in Fig. 1(d). Interestingly, this analysis has led to a model with the 1 meV level originally proposed by Fisk *et al.* [2]. If these peaks are due to additional crystal-field splitting and not some more exotic many-body effect, they would be forbidden in cubic symmetry. In our crystallographic study [4], we searched very carefully for static noncubic displacements or distortions of the structure, as well as nonstoichiometric site occupancies which could give noncubic local symmetry, but found none.

Within either of the models shown in Fig. 1(c) or 1(d), it is straightforward to calculate the specific heat. Generalizing the normal formula [12] for a Schottky anomaly to take account of a continuum of excited states and assuming the temperature-independent three-component excitation spectrum corresponding to Fig. 1(c), together with the low-temperature linewidths and the appropriate crystal-field degeneracies, we obtain the C and C/T values shown in Fig. 4. The doublet and quartet contributions can be calculated separately, and they are shown as the dashed and dot-dashed lines, respectively. For such a simple model, the low-temperature C/T value of

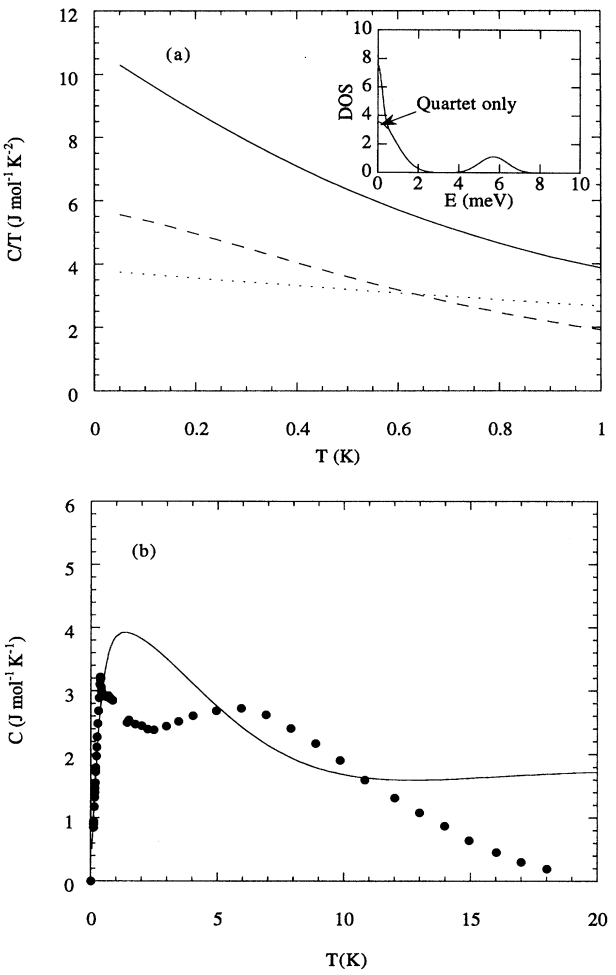

FIG. 4. C/T and specific-heat values calculated assuming the temperature-independent three-component line shape with $\sigma_1 = 0.2$ meV, $\sigma_2 = 0.9$ meV, and $\sigma_3 = 0.7$ meV. In (a) the solid line for C/T shows the full calculation, while the dashed line shows the specific heat due to the lowest sharp doublet alone, and the dotted line shows that due to the next four levels. The inset shows the corresponding density of excited states. In (b) the solid line is the full calculation, and the points are the experimental specific-heat data from Ref. [2].

10 J mol^{-1} K^{-2} is in remarkable agreement with the measured value of 8 J mol^{-1} K^{-1} [2], and roughly half of this comes from the broadened "sharp" doublet and may be considered "normal" heavy-fermion behavior. This is in very good agreement with the independent estimate of 5 J mol^{-1} K^{-2} for this component from an analysis of the resistivity associated with the 400 mK transition, assuming a spin-density wave model [10]. The other half originates from the crystal-field degeneracy at low energies. This model gives an entropy of $R\ln 2$ by 0.9 K and $R\ln 5$ by 9.2 K, in fairly good agreement with values derived from the bulk specific heat [2]. If one goes further and includes the split levels as separate Kramers doublets, the qualitative structure of the bumps at 0.5 and 6 K in the specific heat as a function of temperature can be reproduced, but the quantitative agreement is still not perfect.

In summary, we have identified the excitations responsible for the huge "electronic" specific heat in this extraordinary compound, and we propose a very simple explanation in terms of broadened crystal-field levels. YbBiPt is in many ways a prototypical heavy-fermion compound to study, because the hybridization is not so strong as to wash out the crystal-field levels completely, in contrast to many uranium-based heavy fermions, but yet it is still hybridized more than the cerium-based heavy fermions which show clearly separated levels. The remaining questions for the future concern the cause of the "forbidden" splitting shown in Figs. 2(c) and 2(d), and the exact nature of the 400 mK transition. In addition, it is remarkable that this huge amount of entropy is expressed at such low temperatures, without the compound developing conventional long-range magnetic order. But clearly, crystal-field degrees of freedom are active on the same energy scale as the correlation energy and any exchange energy. Finally, we note that excitation spectra fairly similar to ours have been observed by Marshall [13] in the isostructural compounds YbBiPd and YbSbPd, which are moderate heavy fermions.

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