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Massive Electron State in YbBiPt

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Measurements of the electrical resistivity, magnetoresistance, ac and dc magnetic susceptibility, and specific heat on YbBiPt indicate this compound to be a *very* heavy-electron system. The low-temperature Sommerfeld coefficient of 8 J/K² (mole Yb) is not affected by a phase transition at 0.4 K. We suggest that the heavy-mass state in YbBiPt is unconventional in that it develops from Bloch states in an electronic subsystem with low carrier concentration.

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The rare-earth element Yb in its trivalent state is the $4f^{13}$, hole analog of trivalent $4f^{1}$ Ce. One expects to and does see many of the interesting Kondo-related properties in Yb compounds that are found in Ce materials [1]. Very large electronic specific-heat γ 's lof order 1 J/K² (mole Yb)] appear to be found less frequently, however, in Yb than in Ce compounds. This apparent asymmetry between Yb and Ce compounds has been the motivation for the work reported here.

YbBiPt is a face-centered-cubic material, crystallizing in the MgAgAs structure [2], sometimes referred to as the half-Heusler structure. In it, each component occupies a face-centered-cubic sublattice, respectively positioned along the body diagonal at (0,0,0) and $(\pm \frac{1}{4}, \pm \frac{1}{4}, \pm \frac{1}{4})$, with the pnictogen (Bi) in the unique (0,0,0) position, as we have verified by x-ray powderdiffraction intensity measurements for our compound. We prepared our material in the form of small single crystals via flux growth from molten Bi. The entire rareearth sequence *R*BiPt with the exception of the La and Pm members has been prepared [3], and the latticeparameter variation with rare earth suggests that Yb in this compound is essentially trivalent, the lattice parameter lying smoothly between those of TmBiPt and LuBiPt.

The RBiPt compounds, with the possible exception of R = Yb, appear to be low carrier concentration materials, with a gradual evolution from semiconducting towards semimetallic behavior as R is varied through the rareearth series [3]. NdBiPt, for example, has a negative temperature coefficient of electrical resistivity, whereas for LuBiPt $\partial \rho / \partial T$ is weakly positive. A low-temperature plot of the specific heat of LuBiPt in the form of C_P/T vs T^2 indicates an electronic contribution of $\gamma \leq 1$ mJ/K² (mole Lu). Chemically, we can see this as derived from the 3-5 valence matching of R-Bi, with Pt in the material having a full $5d^{10}$ shell. Our interest in this Yb compound stems, in part, from interest in the stability of the Yb moment in the vicinity of a metal-semiconductor transition.

The electrical resistivity is that of a metal, with characteristics similar to UPt₃ [4] (Fig. 1). Furthermore, C_P/T of YbBiPt rises to exceptionally large values in the vicinity of 1 K (inset, Fig. 2). A feature indicative of some kind of phase transition is evident at 0.4 K (Fig. 2), but this feature does not coincide with any apparent loss of electronic specific heat at lower temperature, such as is usually evidenced in heavy-electron systems below their Néel temperature [5]. Rather, C_P drops linearly towards zero between 0.2 and 0.09 K (the lowest temperature of measurement) with a slope corresponding to $\gamma = 8$ J/K² (mole Yb). As we explain below, this points to itinerant quasiparticles of extremely heavy mass.

The marked change in electronic properties between the Lu and Yb compounds cannot be ascribed to large valence differences between the R ions, and, in this context, it should be mentioned that fully relativistic local-



FIG. 1. Temperature dependence of the electrical resistivity of YbBiPt between 0.1 and 280 K.



FIG. 2. Low-temperature specific heat $C_P(T)$ of YbBiPt between 0.09 and 0.85 K. Inset: Same data as C_P/T vs T.

density-approximation band-structure calculations predict a gap of 0.05 eV at E_F for YBiPt and a metallic state for YbBiPt and LuBiPt [6]. Magnetic susceptibility measurements show Curie-Weiss behavior for YbBiPt, with a paramagnetic Curie temperature $\Theta = +2$ K and an effective moment $\mu_{eff} = 4.2\mu_B$ for the high-temperature data. This latter value is only slightly smaller than the expected full moment of $4.54\mu_B$ (Fig. 3). For T < 10 K, $\mu_{eff} = 3.6\mu_B$ and $\Theta = -0.8$ K. Below 1 K, the susceptibility has also been tracked via low-frequency and low-field ac measurements. These data (inset, Fig. 3) show a cusplike feature at 0.4 K, again indicative of some kind of phase transition, perhaps antiferromagnetic.

We now turn to the temperature evolution of the specific heat. An integrated entropy of $R \ln 2$ per mole Yb is reached at approximately 1 K. This indicates that the low-temperature phase transition involves only a doublet state resulting from a crystal-field splitting of the Hund's rule ground state of Yb³⁺. About half of the entropy of this doublet is lost above the phase-transition temperature. Note that we see no indication for any nuclear Schottky term in the specific heat above 0.09 K. Ytterbium is known for large hyperfine coupling [7], and the absence of such a contribution to the low-temperature specific heat points to a small, if any, internal field at the Yb nuclei induced by the 0.4-K phase transition. A remarkable feature is clearly the almost temperatureindependent specific heat between 0.4 and 1 K, implying that single-ion Kondo-type explanations can only be regarded as a crude approximation in this case.

The entropy at 20 K, with the lattice contribution of LuBiPt subtracted, is slightly less than R ln5. There is a broad maximum in C_P near 5 K, and the shape of the specific-heat curve is best fitted with the Γ_6 and Γ_7 doublets lying lowest and being separated by approximately 12 K. In several cubic compounds of Yb, inelastic neutron studies have found the remaining quartet state Γ_8 be-



FIG. 3. Inverse magnetic susceptibility $\chi^{-1}(T)$ of YbBiPt between 1 and 350 K. Inset: $\chi_{ac}(T)$ between 0.08 and 0.80 K.

tween the two doublets: in YbBe₁₃ at 45 K, above the ground-state doublet [8], and in YbAuCu₄ at 34 K, above the ground state [9]. Considering the specific-heat data, the possibility of the Γ_8 quartet lying between the doublets in this case is very unlikely. In our case, the magnitude of the low-field susceptibility at 1 K is closest to the value expected for the Γ_7 doublet, and thus we assume that the ground-state properties develop off this doublet. We will argue that the low-temperature state is a heavy-electron state with a characteristic temperature near 1 K.

We look now at the magnetic-field dependence of the properties of YbBiPt. The effect of external magnetic fields on the specific heat is exceptionally large [10]. At 0.6 K, a field of 5 T causes a reduction of C_P/T by a factor of 5, implying that T_K is of order 1 K or less [11]. This value is close to what one estimates from the magnitude of the low-T slope of the C_P -vs-T curve using the single-impurity relationship for T_K [12]. These data do not rule out that strong short-range magnetic fluctuations are responsible for $C_P(T)$, rather than heavy-electron effects, but other considerations presented below clearly favor the heavy-electron viewpoint.

Next, we address the temperature and field dependence of the electrical resistivity (Fig. 4). The temperature dependence is somewhat like that of UPt₃ [4], showing negative curvature at all temperatures, and no clear evidence of Kondo-type effects. We do note that there are weak features superimposed on the curve. They may be due to scattering processes involving crystal-field-split 4f-electron energy levels, a feature that is also often observed in Ce compounds. What is very intriguing is the strong positive field dependence of the electrical resistance at fields less than 0.2 T, followed by a negative field dependence. Both the positive and the negative magnetoresistances are temperature dependent, although not in quite the same way. The positive magnetoresistance followed by negative magnetoresistance for larger fields has



FIG. 4. Relative change of the resistivity $\rho(H)/\rho(0)$ of Yb-BiPt in external magnetic fields at 0.3 and 15.0 K. The solid lines are to guide the eye.

been seen in the heavy-electron compound CeCu₆ at very low temperatures [12], with a peak in the magnetoresistance at higher fields than observed here. The much lower characteristic field for YbBiPt no doubt reflects the considerably lower scale of T_K , and correspondingly a higher value γ in YbBiPt.

The physical properties of YbBiPt are seen to be consistent within a heavy-electron interpretation. A Wilson ratio $[W = \pi^2 k_{B\chi}^2(0)/\mu_{eff}^2(0)]$ of 2.64 is found using the low-temperature effective moment, a value consistent with a heavy-electron system with magnetic correlations. The large electronic specific-heat coefficient results from a very small T_K ; the monotonic decrease of ρ with decreasing temperature to below 1 K confirms that singleion Kondo features are distinctly modified. We believe that a T_K this small comes to manifest itself because the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction is equally weak as indicated by the small value of θ_P . Magnetic order is therefore not expected to preempt the heavy-electron state.

The Yb atoms sit on an fcc lattice, a lattice not favorable for simple antiferromagnetic structures. This means that the influence of RKKY-type interactions is reduced by what is essentially a geometrical factor, as well as possibly by the small carrier concentration in these materials discussed earlier. We can quantify this argument somewhat. The paramagnetic Curie-Weiss Θ for isostructural GdBiPt is -40 K, and for it $T_N = 9$ K. Using a de Gennes factor scaling $[T_N \propto (g_J - 1)^2 J(J+1)]$, the expected Curie-Weiss Θ for YbBiPt is -0.8 K, equal to that observed in the low-temperature data, and with the same ratio of T_N / Θ we would predict $T_N = 0.2$ K. These effects can conspire to allow the small Kondo scale to make itself evident. The smallness of the Kondo scale plus coherence effects can also prevent the Kondo features from dominating the electrical resistivity. One way to think about YbBiPt could be as a system in which Bloch states already exist at temperatures much higher than the Kondo scale, and that the interactions responsible for the heavy-electron ground state modify the band states in the vicinity of 1 K. The interesting point here is that particulars of crystal structure and electron count appear to "protect" the small Kondo scale, and that in this case the heavy-electron effects can develop out of a normal metallic state.

The enhancement of the electronic specific heat over that of LuBiPt is approximately 10^4 . One naturally wonders how the Kondo "compensation" can occur in a low-carrier-density material, and whether the large γ is connected with the proximity of the material to a semiconducting boundary. The point must be that, here, the many-body effects manifest themselves unconventionally, relative to the more usual, i.e., more metallic, heavyelectron systems and that the physics ought to be viewed from a new perspective.

Finally, we comment on some aspects related to the phase transition. Small-moment ordering is a distinct possibility, especially because the transition occurs in the coherent state and because all large-moment orderings known so far reduce γ , which is not the case here. It is also not apparent that these results can be those of a classically frustrated spin system on an atomically ordered lattice [13]. In particular, when such a system finally orders we see no reason why C/T would remain finite. Although the fcc lattice is in a loose sense frustrated, such spin lattices have no real problem in establishing longrange antiferromagnetic order. As in most heavyelectron systems the ground state of the Yb fconfiguration is a doublet, but it will not act as an ideal spin- $\frac{1}{2}$ system, due to the strong spin-orbital coupling: The doublet is anisotropic and the spin- $\frac{1}{2}$ quanta fluctuations can be expected to be suppressed significantly. Of course, microscopic studies will reveal more details about the low-temperature state of YbBiPt.

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