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## Strong electronic correlations in a new class of Yb-based compounds: $\text{YbXCu}_4$ ( $X = \text{Ag, Au, Pd}$ )

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A new series of heavy-electron Yb compounds  $\text{YbXCu}_4$  with  $X = \text{Ag, Au, or Pd}$  are reported which have electron effective masses as high as  $\sim 60m_e$ , where  $m_e$  is the mass of the free electron. Whereas the compounds with  $X = \text{Au and Pd}$  order magnetically below 1 K, the compound with  $X = \text{Ag}$  is nonmagnetic. The magnetic susceptibility and specific heat of  $\text{YbAgCu}_4$  can be accounted for by the Bethe-ansatz solution of the Coqblin-Schrieffer impurity model without any adjustable parameters.

The remarkable physical properties of compounds of rare earth and actinide elements which are associated with the hybridization between conduction-electron and localized  $f$ -electron states continue to attract the attention of experimentalists and theoreticians alike. A significant amount of effort has been expended in characterizing exemplary  $f$ -electron materials and developing theoretical models with which their extraordinary properties can be compared.<sup>1,2</sup> Compounds of the rare-earth elements Ce and Yb are especially well suited for this purpose, since the  $4f$  shell of Ce and Yb can accommodate at most one electron or hole, respectively, which considerably simplifies relevant theoretical models.

In this paper we report transport, thermal, and magnetic measurements on a new series of Yb compounds with the formula  $\text{YbXCu}_4$ , where  $X = \text{Ag, Au, or Pd}$ . These compounds appear to be nearly ideal materials for testing theoretical models since their crystal structure has cubic symmetry, the Yb  $4f$  shell contains approximately one hole, and interactions between the Yb magnetic moments are relatively weak. Our results reveal that the compounds with  $X = \text{Au and Pd}$  order magnetically below 1 K, whereas  $\text{YbAgCu}_4$  has a nonmagnetic ground state. Moreover, the magnetic ground state in  $\text{YbAuCu}_4$  and the nonmagnetic groundstate in  $\text{YbAgCu}_4$  both develop out of a heavy Fermi liquid characterized by an effective mass of  $\sim 50m_e$ , where  $m_e$  is the mass of the free electron. We have found that the magnetic susceptibility  $\chi$  as a function of temperature  $T$  and the value of the coefficient  $\gamma$  of the specific heat  $C = \gamma T$  of the nonmagnetic compound  $\text{YbAgCu}_4$  can be described by the Bethe-ansatz solution of the Coqblin-Schrieffer Hamiltonian<sup>3</sup> for angular momentum  $J = \frac{7}{2}$ , which is appropriate for the Hund's rule groundstate multiplet of trivalent Yb, without any adjustable parameters.

The polycrystalline  $\text{YbXCu}_4$  samples were prepared in three ways: by melting the pure elements in (1) an argon arc furnace on a Cu hearth, (2) a BeO crucible, or (3) a

sealed Ta tube, in order to minimize losses of the volatile Yb. The physical properties of the samples were not influenced by the different cooling rates involved in these three techniques. X-ray powder diffraction and metallographic analyses yielded no evidence of impurity phases. In contrast to  $\text{YbCu}_5$ , which has the hexagonal  $\text{CaCu}_5$ -type crystal structure and in which Yb is divalent,<sup>4</sup> the three  $\text{YbXCu}_4$  compounds with  $X = \text{Ag, Au, and Pd}$  have the cubic  $\text{AuBe}_5$  structure (space group  $C15b$  or  $F-43m$ ) and Yb appears to be trivalent, or nearly so. The values of the lattice parameters are 7.0696, 7.0519, and 7.0396 Å for  $X = \text{Ag, Au, and Pd}$ , respectively. The Yb-Yb distance in these materials is of the order of 5 Å, a relatively large value which seems to favor a magnetic and/or heavy-electron ground state as proposed for the actinides by Hill.<sup>5</sup> Electrical resistivity  $\rho$  measurements were made with a self-balancing four-wire impedance bridge operating at a frequency of 16 Hz between 80 mK and 295 K. The data below 1.2 K were taken in a conventional <sup>3</sup>He-<sup>4</sup>He dilution refrigerator. The magnetic susceptibility  $\chi$  was measured with an SHE superconducting quantum-interference device (SQUID) magnetometer in a magnetic field of 1 T between  $\sim 2$  and 300 K. Heat-capacity  $C$  measurements were carried out in a semiadiabatic <sup>3</sup>He calorimeter between 0.45 and 16 K.

Normalized electrical resistivity  $\rho(T)/\rho(295 \text{ K})$  versus  $T$  data between 80 mK and 295 K for the three  $\text{YbXCu}_4$  compounds, displayed in Fig. 1, exhibit different types of behavior for each of the three substituent  $X$  elements. Little temperature dependence is observed for all three samples down to  $\sim 80$  K. However, below  $\sim 60$  K, the resistivity of  $\text{YbAgCu}_4$  falls rapidly and follows a  $T^2$  power-law dependence between  $\sim 1$  and 28 K which is characteristic of Fermi-liquid behavior or spin fluctuations. In contrast, the resistivity of  $\text{YbAuCu}_4$  increases with decreasing temperature below  $\sim 100$  K and displays two maxima, one at  $\sim 20$  K and a second one at  $\sim 1.1$  K (see inset of Fig. 1). The drop in  $\rho(T)$  below this second

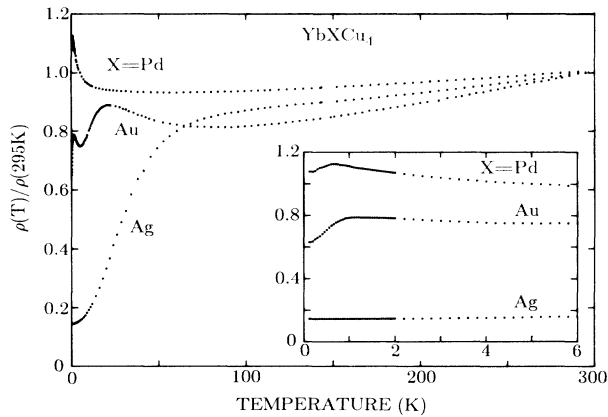


FIG. 1. Normalized electrical resistivity  $\rho(T)/\rho(295 \text{ K})$  vs temperature  $T$  for  $\text{YbXCu}_4$  compounds with  $X=\text{Ag}$ ,  $\text{Au}$ , and  $\text{Pd}$ . Shown in the inset are the curves below 6 K. The presence of a maximum at 1.1 K for  $X=\text{Au}$  and at 0.7 K for  $X=\text{Pd}$  is associated with freezing out of magnetic scattering due to the onset of magnetic order.

peak is due to the onset of magnetic ordering (probably antiferromagnetic) as verified by specific-heat measurements which are discussed below. The drop in  $\rho(T)$  below  $\sim 0.7 \text{ K}$  for the compound  $\text{YbPdCu}_4$  is also associated with magnetic order. In these latter two compounds  $\rho(T)$  seems to exhibit Kondo-like behavior with a typical minimum located around 85 K for the Au-based sample and  $\sim 60 \text{ K}$  for the Pd-based material. At room temperature, the electrical resistivity of these compounds ranged between 30 and  $100 \mu\Omega \text{ cm}$ .

The types of  $\rho$  versus  $T$  curves displayed by the  $\text{YbXCu}_4$  compounds could be associated with a temporal admixture of divalent and trivalent Yb (valence fluctuations) which are thought to be appropriate for compounds like  $\text{YbAl}_3$ ,  $\text{YbCu}_2\text{Si}_2$ ,  $\text{YbInAu}_2$ , and  $\text{YbCuAl}$ ,<sup>6</sup> or to the transition from a single-impurity Kondo regime at high temperature into a coherent Kondo lattice at low temperature. It is interesting to note that the presence of a Kondo-like resistivity maximum in Yb systems such as  $\text{YbAuCu}_4$ , discussed here, or in  $\text{YbCuAl}$ , which passes from a high-temperature localized to a low-temperature Fermi-liquid regime in the vicinity of 30 K [with  $\gamma(0) \sim 255 \text{ mJ/mol K}^2$ ], is less common than in Ce systems. The origin of this asymmetry of behavior at the two ends of the lanthanide series is not understood.<sup>6</sup>

The inverse magnetic susceptibility  $\chi^{-1}$  versus  $T$  for the  $\text{YbXCu}_4$  compounds is shown in Fig. 2. For  $\text{YbAuCu}_4$  and  $\text{YbPdCu}_4$ , the  $\chi^{-1}$  versus  $T$  plots are linear between room temperature and about 30 K [i.e.,  $\chi(T)$  follows a Curie-Weiss law  $\chi(T) = N\mu_{\text{eff}}^2/3k_B(T - \Theta)$ , where  $N$  is the number of ions which carry magnetic moments] and show a slight deviation from linearity below  $\sim 30 \text{ K}$ . The values of the respective Curie-Weiss temperatures  $\Theta$  and effective magnetic moments  $\mu_{\text{eff}}$ , which are close to the  $4.54\mu_B$  value expected for a free  $\text{Yb}^{3+}$  ( $J = \frac{7}{2}$ ) ion, are summarized in Table I together with the extrapolated values of  $\chi(T)$  at 0 K.

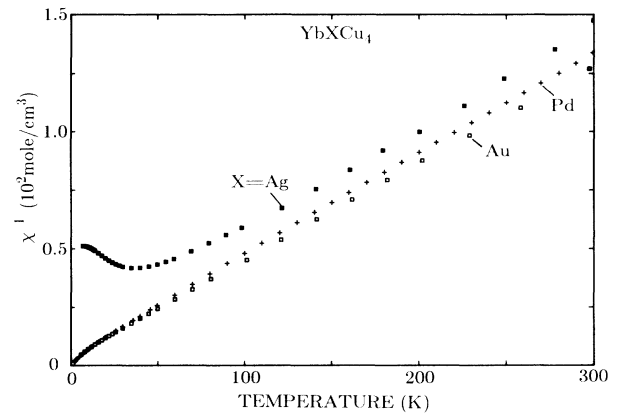


FIG. 2. Inverse magnetic susceptibility  $\chi^{-1}$  vs  $T$  for  $\text{YbXCu}_4$  compounds with  $X=\text{Ag}$ ,  $\text{Au}$ , and  $\text{Pd}$ , measured in a magnetic field  $\mu_0 H = 1 \text{ T}$ .

Shown in Fig. 3 are  $\chi(T)$  data for  $\text{YbAgCu}_4$  which follow a Curie-Weiss law between room temperature and  $\sim 100 \text{ K}$  (see Fig. 2) and then pass through a maximum at  $\sim 35 \text{ K}$  before reaching a constant value at low temperature. This behavior of  $\chi(T)$  is rather interesting and similar to that reported for  $\text{YbCuAl}$ .<sup>7</sup> Rajan<sup>8</sup> has calculated  $\chi(T)$  and the specific heat  $C(T)$  in zero magnetic field for angular momentum values from  $J = \frac{1}{2}$  to  $\frac{7}{2}$  by solving the exact thermodynamic equations of the Coqblin-Schrieffer impurity model with the Bethe-ansatz. Calculations based on the Coqblin-Schrieffer model<sup>8,9</sup> have been applied successfully by Hewson *et al.*<sup>10</sup> to  $\text{YbCuAl}$  which suggests that this compound is a good example of a Kondo lattice. Kondo-lattice behavior is expected for compounds containing rare-earth ions with integral valence such as  $\text{Ce}^{3+}$  (e.g.,  $\text{CeAl}_3$ ),  $\text{Sm}^{3+}$  (e.g.,  $\text{SmB}_6$ ), or  $\text{Yb}^{3+}$  which have a ground-state multiplet and tendency towards mixed valence, and where the antiferromagnetic exchange coupling between the rare-earth ion and the conduction electron spins dominates over the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction. Such compounds are usually paramagnetic at  $T = 0 \text{ K}$  or have very low ordering temperatures which appears to be the case for the new  $\text{YbXCu}_4$  compounds discussed herein.

As shown in Fig. 3 the numerical results of Ref. 8 (solid line in the figure) provide an excellent description of the  $\chi(T)$  data for  $\text{YbAgCu}_4$ . The theoretical curve is based on only one parameter, the characteristic temperature  $T_0$  which is given by  $T_0 = \nu(\nu^2 - 1)g_J^2\mu_B^2/24\pi k_B\chi(0)$ , where  $\nu$  is the degeneracy of the ground state,  $g_J$  is the Landé  $g$  factor of the Hund's-rule ground-state multiplet with angular momentum  $J$ , and  $\chi(0)$  is the experimental value of the susceptibility at zero temperature. The value  $T_0 = 167.5 \text{ K}$  used to generate the theoretical curve in Fig. 3 was calculated from this relation using  $\chi(0) = 1.96 \times 10^{-2} \text{ cm}^3/\text{mol}$  for  $\text{YbAgCu}_4$  and  $\nu = 2J + 1 = 8$  for  $\text{Yb}^{3+}$ .

Specific heat  $C$  versus  $T$  data are displayed in Fig. 4 for the three compounds with  $X=\text{Ag}$ ,  $\text{Au}$ , and  $\text{Pd}$ . The large peak in  $C(T)$  at the respective temperatures of  $\sim 0.8 \text{ K}$

TABLE I. Summary of magnetic susceptibility and specific-heat data for  $\text{YbXCu}_4$  ( $X=\text{Ag,Au,Pd}$ ).

Material	$\mu_{\text{eff}}$ ( $\mu_B$ )	$\Theta$ (K)	$\chi(T)$		$\gamma(0)$ (mJ/mol K <sup>2</sup> )	$\Theta_D$ (K)	$C(T)$		$T$ ( $S=R \ln 2$ ) (K)
			$\chi(0)$ (cm <sup>3</sup> /mol)	$T_{\text{max}}$ (K)			$T_N$ (K)	$m^*$ ( $m_e$ )	
YbAgCu <sub>4</sub>	4.28	-31	$1.96 \times 10^{-2}$	34	245±6	129±1		60	17.2
YbAuCu <sub>4</sub>	4.39	-8.8	$2.75 \times 10^{-1}$				0.6		5.6
YbPdCu <sub>4</sub>	4.33	-13.5	$1.74 \times 10^{-1}$		200	138	0.8	48	8.9

and  $\sim 0.6$  K for the specimens with  $X=\text{Au}$  and  $\text{Pd}$  is presumably associated with the anticipated long-range antiferromagnetic order. The absence of an anomaly in  $C(T)$  for  $\text{YbAgCu}_4$  confirms that this compound does not exhibit any magnetic order, at least down to 0.45 K. The plots of  $C/T$  versus  $T^2$  presented in Fig. 5 reveal large  $\gamma(0)$  values of  $\sim 245$  mJ/mol K<sup>2</sup> for  $\text{YbAgCu}_4$  and  $\sim 200$  mJ/mol K<sup>2</sup> for  $\text{YbPdCu}_4$ . For this latter compound, a linear extrapolation of the  $C/T$  versus  $T^2$  data to  $T=0$  to estimate  $\gamma(0)$  seems reasonable in spite of the magnetic contribution at low  $T$ . In the case of  $\text{YbAuCu}_4$ , there does not seem to be a reliable way of determining  $\gamma(0)$  even if a fairly large value were to be expected. The values of  $\gamma(0)$ , the Debye temperature  $\Theta_D$ , the Néel temperature  $T_N$ , the effective mass  $m^*$  and the temperature at which the entropy  $S$  reaches  $R \ln 2$  are summarized in Table I. The effective mass was estimated from the relation  $m^* = \hbar^2 k_F^2 \gamma(0) / \pi k_B (Z/\Omega)$  where  $k_F = (3\pi^2 Z/\Omega)^{1/3}$ , with  $Z=4$  the number of  $4f$  holes per unit cell (assuming Yb to have a valence 3+) and  $\Omega$  the volume of the cubic unit cell.

According to Ref. 8, the zero-temperature, zero-field limit of the specific heat is given by  $C(T \rightarrow 0)/T = \gamma = (\nu - 1)\pi k_B / 6T_0$ . Using the value  $T_0 = 167.5$  K determined above from the zero-temperature limit of the magnetic susceptibility  $\chi(0)$ , we obtain a value of 182 mJ/mol K<sup>2</sup> for  $\gamma$  which is in reason-

able agreement with the experimental value of  $\sim 245$  mJ/mol K<sup>2</sup>.

The reason that the Coqblin-Schrieffer model for  $\text{Yb}^{3+}$  impurities provides such a good description of  $\chi(T)$  and  $\gamma$  for  $\text{YbAgCu}_4$  seems to be that the intersite Yb-Yb interaction (presumably Néel) temperatures of the  $\text{YbXCu}_4$  compounds with  $X=\text{Au}$  and  $\text{Pd}$ . A general trend which seems to emerge is that the Néel temperatures  $T_N$  of Yb compounds are, on the average, smaller than those of Ce compounds, which, in turn, appear to be smaller than those of U materials [i.e.,  $T_N(\text{Yb}) < T_N(\text{Ce}) < T_N(\text{U})$ ]. In fact, all of the Yb-based systems which are known to order antiferromagnetically have  $T_N < 1$  K [e.g.,  $\text{YbIr}_2$ ,<sup>11</sup>  $\text{YbPd}$ ,<sup>12</sup>  $\text{YbX}$  ( $X=\text{N,P,As}$ ) (Ref. 13) and  $\text{YbXCu}_4$  ( $X=\text{Au,Pd}$ )] with the exceptions of  $\text{YbBe}_{13}$  ( $T_N = 1.28$  K) (Ref. 14) and  $\text{Yb}_3\text{Pd}_4$  ( $T_N = 3$  K).<sup>15</sup> In the Ce and U compounds,  $T_N$  is usually closer to 10 K [e.g.,  $\text{CeAl}_2$  ( $T_N = 3.5$  K),  $\text{UCd}_{11}$  ( $T_N = 5$  K) or  $\text{U}_2\text{Zn}_{17}$  ( $T_N = 9.7$  K)].

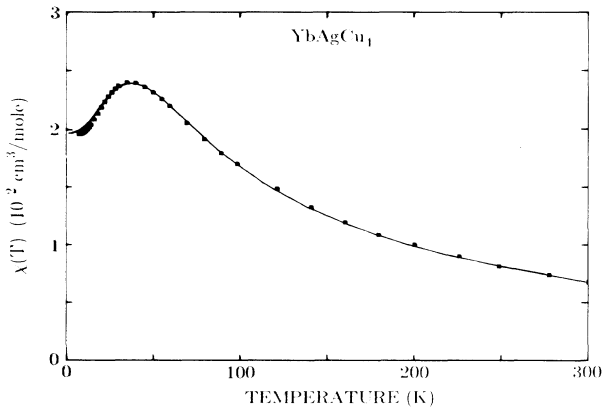


FIG. 3. Magnetic susceptibility  $\chi$  vs temperature for  $\text{YbAgCu}_4$ . The solid line is the calculated susceptibility  $\chi(T)$  scaled to the zero temperature value, using the results of Ref. 8 for the  $J = \frac{7}{2}$  Coqblin-Schrieffer model.

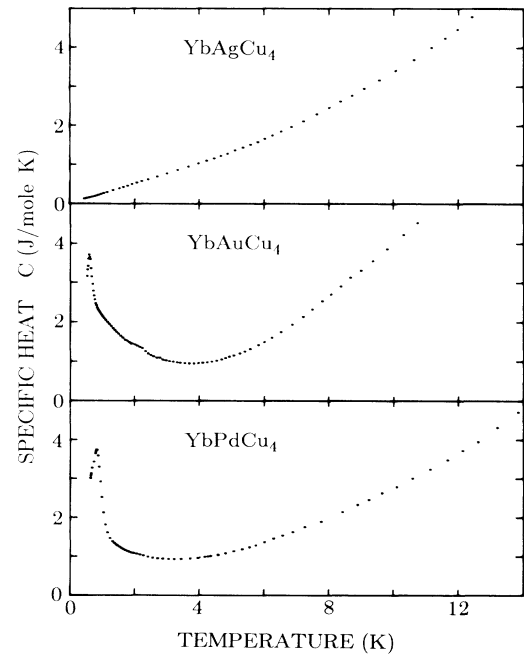


FIG. 4. Specific heat  $C$  vs temperature for  $\text{YbXCu}_4$  compounds with  $X=\text{Ag, Au, and Pd}$ .

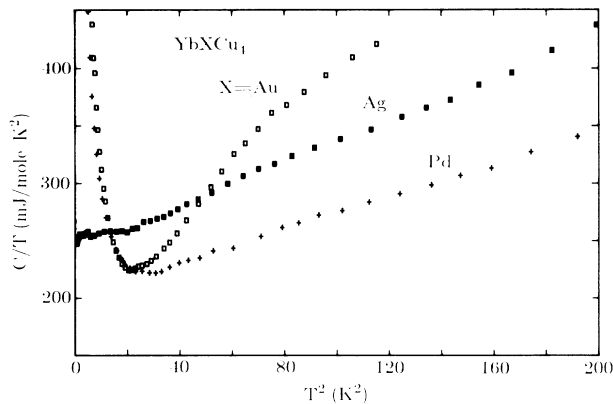


FIG. 5. Specific heat  $C$  divided by temperature  $T$  vs  $T^2$  for  $\text{YbXCu}_4$  compounds with  $X=\text{Ag, Au, and Pd}$ .

Another reason which makes the present family of Yb compounds interesting is the rather low entropy associated with the ground state which is expected to be a magnetic  $\Gamma_6$  or  $\Gamma_7$  doublet. It is apparent from Table I that for both  $\text{YbAuCu}_4$  and  $\text{YbPdCu}_4$  an entropy of  $S=R \ln 2$  is not recovered until the temperature is raised from 0 K to about 10 times  $T_N$ , or in other words, only about 30–40% of  $R \ln 2$  is released by the magnetic transition. Similar behavior has been reported for  $\text{YbX}$  compounds with  $X=\text{N, P, and As}$  (Ref. 13). It seems to be a general observation in magnetically ordered heavy-electron systems that the highly correlated electronic ground state removes a sizable amount of the entropy over a scale of the order of the Kondo temperature  $T_K$ . Since  $T_K$  is usually larger than  $T_N$ , only a small fraction of the expected magnetic entropy  $R \ln 2$  is recovered at  $T_N$ . The competition between the magnetic ground state and the nonmagnetic heavy-electron ground state has been addressed by several authors,<sup>16</sup> but the criteria for the occurrence of magnetic order in a heavy-electron system remains an unresolved problem for all Ce, U, and Yb materials.

In conclusion, we have shown that the hexagonal  $\text{YbCu}_5$  compound, which is a nonmagnetic normal metal, can be transformed into a face-centered-cubic structure by

replacing one of the Cu atoms with an X atom of Ag, Au, or Pd, yielding dramatic changes in its electronic properties. In particular, the three  $\text{YbXCu}_4$  compounds exhibit strong electronic correlations at low temperature with high  $\gamma$  values and large effective masses  $m^*$ . Whereas  $\text{YbAuCu}_4$  and  $\text{YbPdCu}_4$  order magnetically below 1 K,  $\text{YbAgCu}_4$  does not display any cooperative phase transition. The excellent description of the temperature dependence of the magnetic susceptibility  $\chi(T)$  and the electronic specific-heat coefficient  $\gamma$  of  $\text{YbAgCu}_4$  by the Bethe-ansatz solution of the Coqblin-Schrieffer model seems to prove that the conduction electron-impurity spin-exchange interaction is much larger in this compound than the intersite interaction and any possible crystalline electric field terms in the effective Hamiltonian.

Finally, it is interesting to note that the original motivation for this study was based on some previous work performed in the Ce-Cu and U-Cu phase diagrams. For example, the hexagonal compound  $\text{CeCu}_5$  orders magnetically at low temperature and has a rather low electronic specific-heat coefficient,<sup>17</sup> whereas, in contrast, the two neighboring compounds  $\text{CeCu}_4$  (Ref. 17) and  $\text{CeCu}_6$  exhibit heavy-electron behavior.<sup>18</sup> Another interesting case consists of the magnetically ordered  $\text{UCu}_5$  and  $\text{UAgCu}_4$  systems in which the formation of a heavy-electron state has also been reported.<sup>19</sup> The present study clearly demonstrates that new efforts should be made to include Yb-based compounds in the fascinating and rapidly growing family of heavy fermion systems.

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