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TITLE EVIDENCE FOR A "COHERENCE" GAP IN $Ce_3Bi_4Pt_3$

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Evidence for a "Coherence" gap in $Ce_3Bi_4Pt_3$

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In this paper we examine the physical properties of the f-electron system $Ce_3Bi_4Pt_3$. In the majority of cases f-moment compounds are metallic, and, depending upon the relative strength of competing magnetic interactions, have a superconducting, paramagnetic, or antiferromagnetic ground state. In rare instances f-electron compounds have a narrow-gapped insulating ground state instead. This gap is thought to result from hybridization between conduction and f electrons. We have measured the magnetic susceptibility, low-temperature specific heat, resistivity, Hall effect, and thermoelectric power of $Ce_3Bi_4Pt_3$ and find that this material is non-metallic at all temperatures. Isostructural lanthanum substitution for cerium suppresses the gap and produces transport and thermodynamic behavior that is characteristic of a relatively heavy Kondo impurity system. Further, the energy scale at which coherence develops is roughly equal to the energy gap in this compound. This suggests that the gap in $Ce_3Bi_4Pt_3$ appears to stem from the same requirements of lattice periodicity necessary for coherence to manifest itself in metallic systems. Hence, there is strong evidence that the insulating behavior displayed by $Ce_3Bi_4Pt_3$ results form the presence of a Kondo coherence-induced gap.

Evidence for a "Coherence" Gap in $Ce_3Bi_4Pt_3$

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We present the results of electronic transport, magnetic susceptibility, and thermodynamic measurements on single crystals of the cubic compound $Ce_3Bi_4Pt_3$. The susceptibility displays a broad peak centered at T_{max} = 80K, which in Ce compounds is usually indicative of mixed valence and moderately heavy-electron behavior. Both the electronic transport properties and the low temperature specific heat indicate that $Ce_3Bi_4Pt_4$ is non-metallic with a transport energy gap of roughly 35K. We argue that this temperature is close to that scale over which coherence should develop in a periodic metallic system with T_{max} = 80K. Resistivities typical of a Kondo-impurity system are realized when the compound is alloyed with moderate amounts of lanthanum. Further, lanthanum substitution increases the low-temperature specific heat to a value consistent with the single-ion energy scale determined from the magnetic susceptibility.

An extensive research effort has been carried out during the past decade in an attempt to elucidate the low temperature properties of correlated f-electron systems.[1] Unlike the situation at high temperatures where the f-moments are uncorrelated, these moments form a correlated Kondo lattice with a renormalized band structure at low temperatures. This gives rise to two possible ground state configurations. In the majority of cases f-electron compounds are metallic at low temperatures, and, depending upon the relative strengths of competing magnetic interactions, have a superconducting, antiferromagnetic, or paramagnetic ground state. In rare instances (notably SmB_6,[2,3] YbB_{12},[4-6] and $CeFe_4P_{12}$[7]) f-electron materials have a narrow-gapped insulating ground state. This gap is believed to result from hybridization between f and conduction electrons;[8] consequently, it frequently is referred to as a *hybridization* gap. In this brief report we examine the magnetic, electronic transport, and thermodynamic properties of the f-electron compound $Ce_3Bi_4Pt_3$.[9] As with the materials listed above, $Ce_3Bi_4Pt_3$ displays non-metallic behavior at all temperatures. Lanthanum substitution for Cerium suppresses the gap and produces transport and thermodynamic behavior that is characteristic of a moderately heavy Kondo impurity system. We argue that the insulating ground state stems from lattice periodicity and that the gap value is comparable to the temperature scale on which "coherence" should develop in a metallic system.

The magnetic susceptibility χ measured on single crystals of both $Ce_3Bi_4Pt_3$ and $La_3Bi_4Pt_3$ is shown in Fig. 1. The

susceptibility for $Ce_3Bi_4Pt_3$ shows Curie-Weiss behavior at high temperatures (with a moment nearly equal to the J=5/2 free Ce ion value) and a broad maximum centered at T_{max} = 80K (the low temperature Curie tail is somewhat sample dependent, indicating that it is probably an extrinsic effect). The broad, low temperature maximum is typical of moderately heavy-electron cerium compounds.[10] Based both on the susceptibility data and on Bethe-Ansatz calculations,[11] wherein the electronic contribution to the specific heat (γ) is found to be inversely proportional to T_{max}, one would expect $Ce_3Bi_4Pt_3$ to be a moderately heavy-electron metal with a low temperature electronic specific heat of $\gamma \approx 75$ mJ/mole-Ce K^2.

The resistivity, Hall coefficient, and thermoelectric power (TEP) of $Ce_3Bi_4Pt_3$ are depicted in Fig. 2. The temperature dependence of all three quantities are characteristic of a *non-metallic* small-gap system rather than that of a metal. The resistivity shows activated behavior above 100K that is characterized by an energy gap Δ/k_B = 35K (3 meV). The Hall coefficient and TEP can also be fit by activated expressions with similar gap values. Below 50K all three quantities show deviations from activated behavior which we attribute to extrinsic effects. These transport data are qualitatively similar to those exhibited by the narrow-gapped f-electron compounds SmB_6[2,3] and YbB_{12}.[5] A full account of electronic transport in $Ce_3Bi_4Pt_3$ will be presented in greater detail elsewhere.[12]

Low-temperature specific heat measurements (Fig. 3a) indicate that the electron contribution to the specific heat is quite small in $Ce_3Bi_4Pt_3$ (γ = 3.3 mJ/mole-Ce K^2), far below that predicted from the value of T_{max}. This value is also three times smaller than the low-temperature electronic contribution to the specific heat in the metallic analog $La_3Bi_4Pt_3$, where γ = 10 mJ/mole-La K^2. Moreover, given the presence of extrinsic low-temperature effects in both the electronic transport and the magnetic susceptibility, the low-temperature γ in the Ce compound is probably not intrinsic; in the absence of extrinsic effects γ should approach zero in $Ce_3Bi_4Pt_3$.

The effects on the specific heat and resistivity when $Ce_3Bi_4Pt_3$ is alloyed with non-magnetic lanthanum are also presented in Fig. 3. The specific heat data indicate that replacing nominally 14% of the cerium with lanthanum acts to increase γ to ~75 mJ/mole-Ce K^2, while the broad peak in the magnetic susceptibility data (not shown) moves downward in temperature only slightly to T_{max} = 65K. Further, the resistivity at this doping level no longer shows the extreme low temperature increase that occurs with pure $Ce_3Bi_4Pt_3$ but is characteristic of a single-ion Kondo system. Hence, moderate substitution of La for Ce acts both to destroy the energy gap and to push the system into the single-ion regime.

A Bethe-Ansatz solution[11] to the Coqblin-Schrieffer model[13] of a magnetic impurity with arbitrary orbital degeneracy in a free-electron metal indicates that γ and T_{max} are related by

$$\gamma = \frac{(\nu-1)\pi k_B N_A}{6(4T_{max})}, \qquad (1)$$

where $\nu = 2J+1$ is the orbital degeneracy, k_B is the Boltzmann constant, N_A is Avogadro's number, and $4T_{max}$ is the system's characteristic single-ion temperature. In Fig. 4 we compare Eqn. 1 ($\nu = 6$) with χ and specific heat data obtained from six lanthanum alloys of $(Ce_{1-x}La_x)_3Bi_4Pt_3$ (with nominal compositions of x = 0, 1.5, 7, 14, 25, and 50%). As discussed early, at x=0 the data are in severe disagreement with the scaling prediction, while with increased lanthanum substitution the data asymptotically approach the single-ion scaling line. For x above roughly 10% the data straddle the line.

The metallic ground state in heavy-electron compounds generally is attributed to the development of coherence among the periodically placed resonately scattering "Kondo-impurities". The temperature scale on which coherence develops has been suggested from 1/N expansion theory to be related to the single-ion temperature by $T_{coh} = T_o/2J+1$,[14] where J is the orbital degeneracy of the f-atom. Taking the single-ion scale to be $T_o \sim 4T_{max}$ and J=5/2 for Ce^{3+} gives $T_{coh} = 50K$, which is close to the estimate for Δ/k_B. A similar argument can be made[15] for YbB_{12} in which T_{coh} is also approximately equal to Δ/k_B and in which modest alloying with La increases γ close to the Bethe-Ansatz prediction. These observations, together with the variation of γ and $\rho(T)$ with La substitution, suggest that the gap in $Ce_3Bi_4Pt_3$ is made possible by

the same requirement of lattice periodicity necessary for coherence to manifest itself in metallic systems.

In summary, Ce-sublattice periodicity appears to be an essential requirement for the formation of a gap in $Ce_3Bi_4Pt_3$. Once disorder is introduced, single-ion effects dominate the magnetic, thermodynamic, and transport properties. From data available, we tentatively associate the gap with coherence resulting from interactions among the lattice of Kondo impurities.

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Figure Captions

Fig. 1. The magnetic susceptibility of $Ce_3Bi_4Pt_3$ (solid curve) and $La_3Bi_4Pt_3$ (dashed line) as measured in a 0.1 Tesla field. The vertical axis is normalized per mole of either Ce or La for the respective materials (the susceptibility for $La_3Bi_4Pt_3$ has been multiplied by 10).

Fig. 2. Electronic transport in $Ce_3Bi_4Pt_3$. The resistivity and the Hall coefficient below 100K are plotted in (a) and (b), respectively. The thermoelectric power below 300K is depicted in (c).

Fig. 3. (a) C/T versus T^2 below 7K for $(Ce_{1-x}La_x)_3Bi_4Pt_3$. The vertical axis is normalized per mole of Ce for the x=0 and 0.14, and per mole La for x=1.

(b) Normalized resistance for $(Ce_{1-x}La_x)_3Bi_4Pt_3$ plotted versus temperature for x=0, 0.14, and 1.

Fig. 4. γ versus $1000/T_{max}$ as determined from low-temperature specific heat and magnetic susceptibility measurements for six different lanthanum substitution levels (the number attached to each point indicates the nominal percent La concentration). The vertical axis has been normalized per mole Ce. The solid line is the theoretical prediction from Eqn. 1 with a degeneracy ν = 6 (corresponding to J = 5/2).

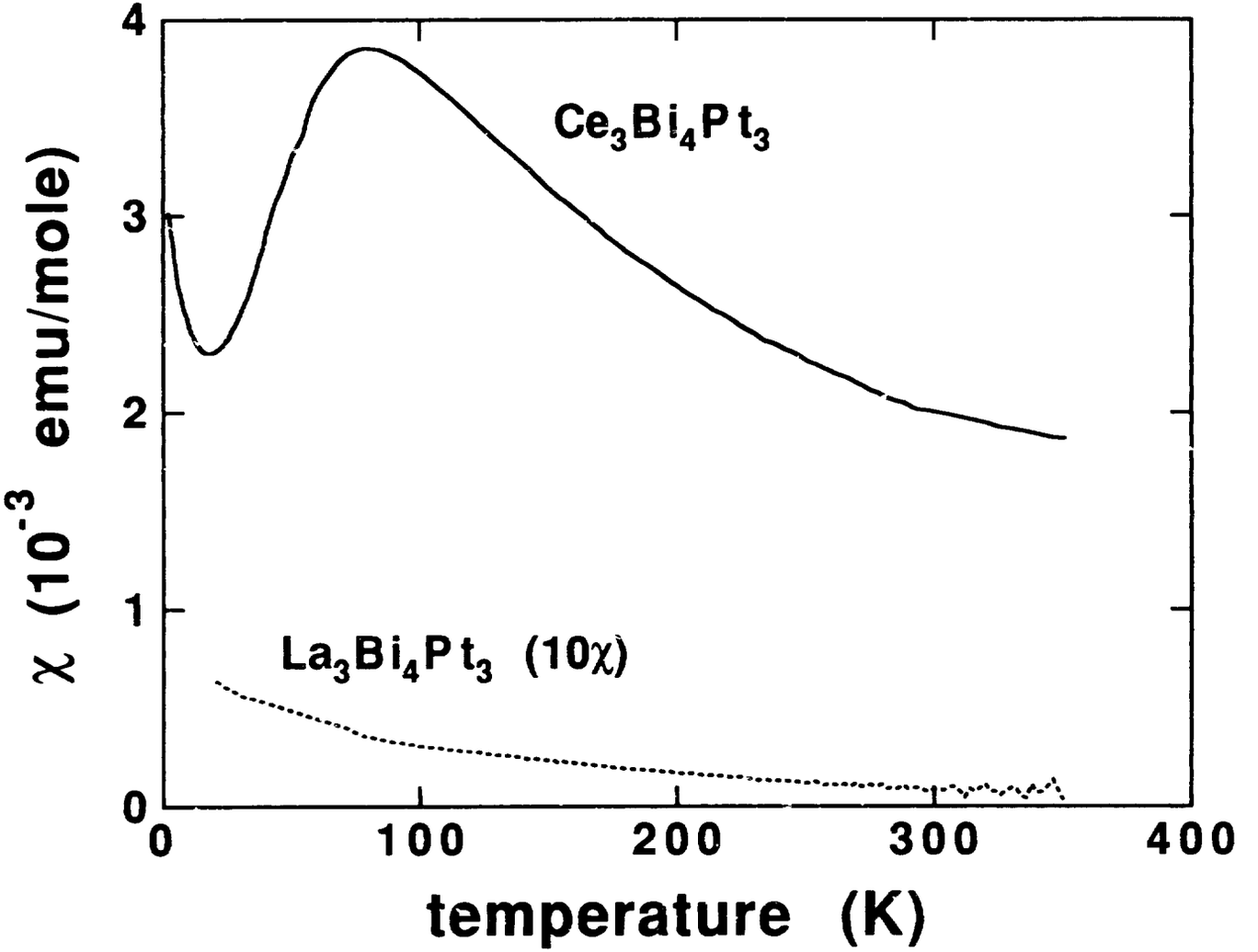
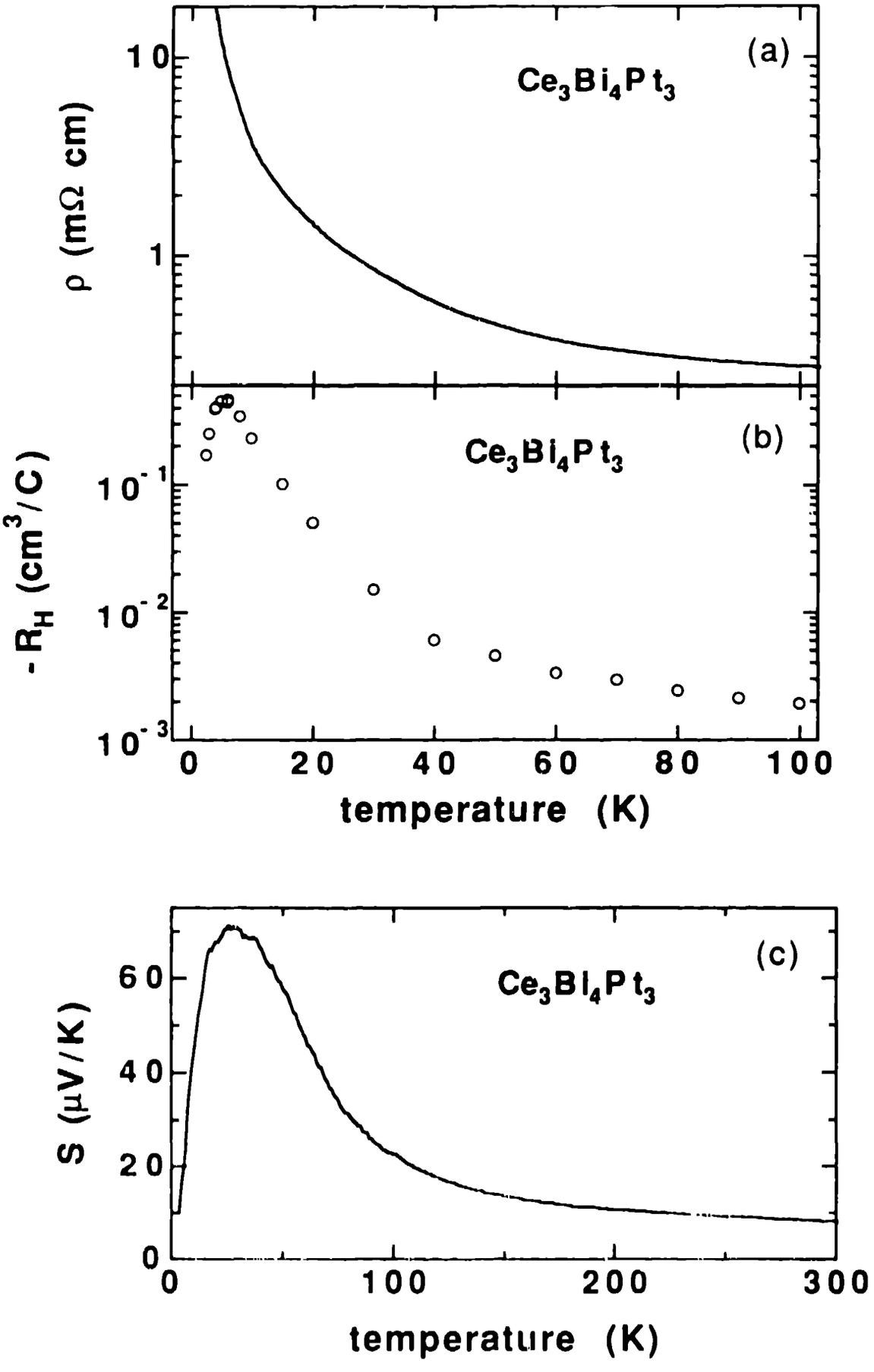


Fig. 1. Hundley et al.


(a)
$Ce_3Bi_4Pt_3$
ρ (mΩ cm)
10
1
(b)
$Ce_3Bi_4Pt_3$
$-R_H$ (cm^3/C)
10^{-1}
10^{-2}
10^{-3}
0
20
40
60
80
100
temperature (K)
(c)
$Ce_3Bi_4Pt_3$
S (μV/K)
60
40
20
0
0
100
200
300
temperature (K)

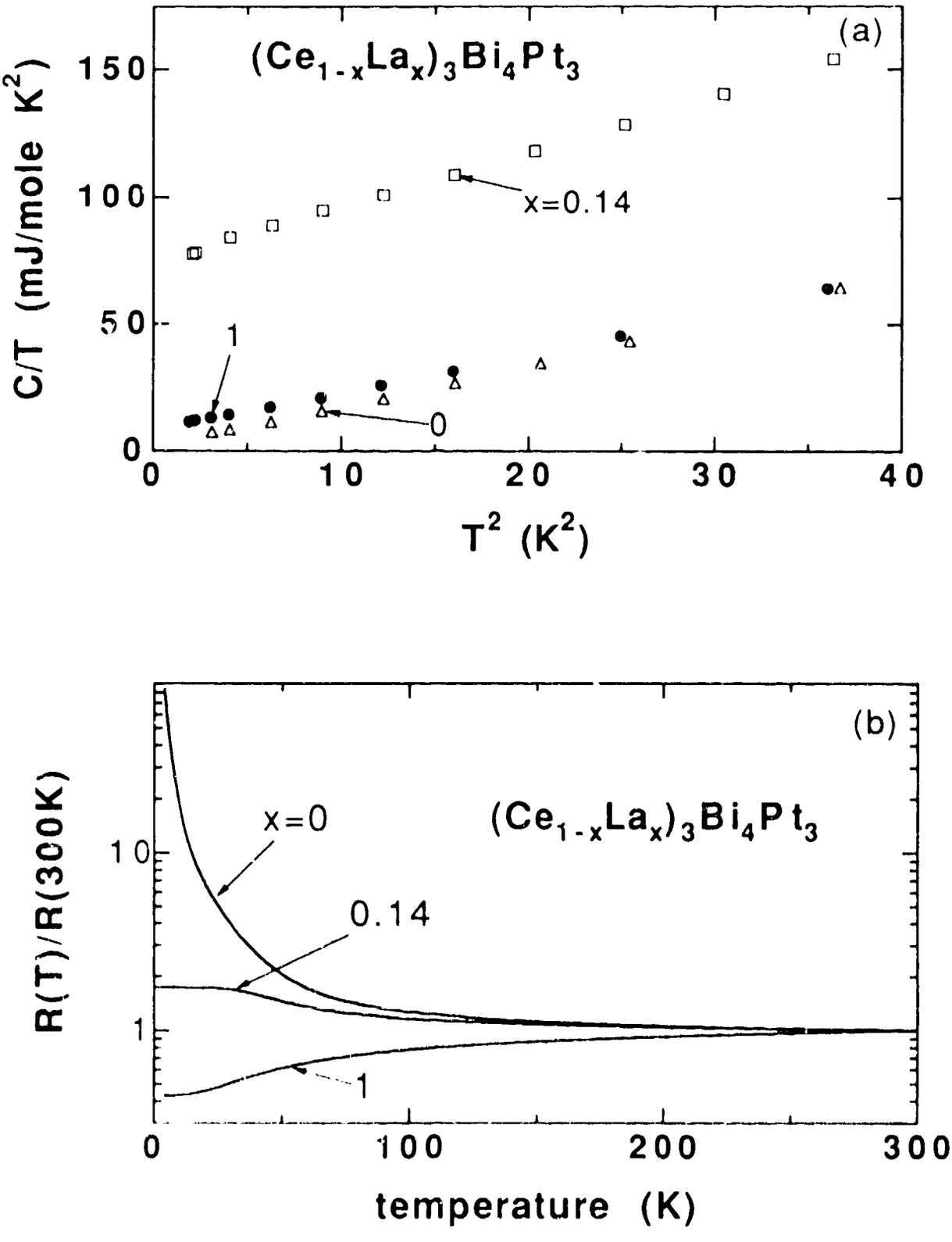


Fig. 3 Hundley et al.

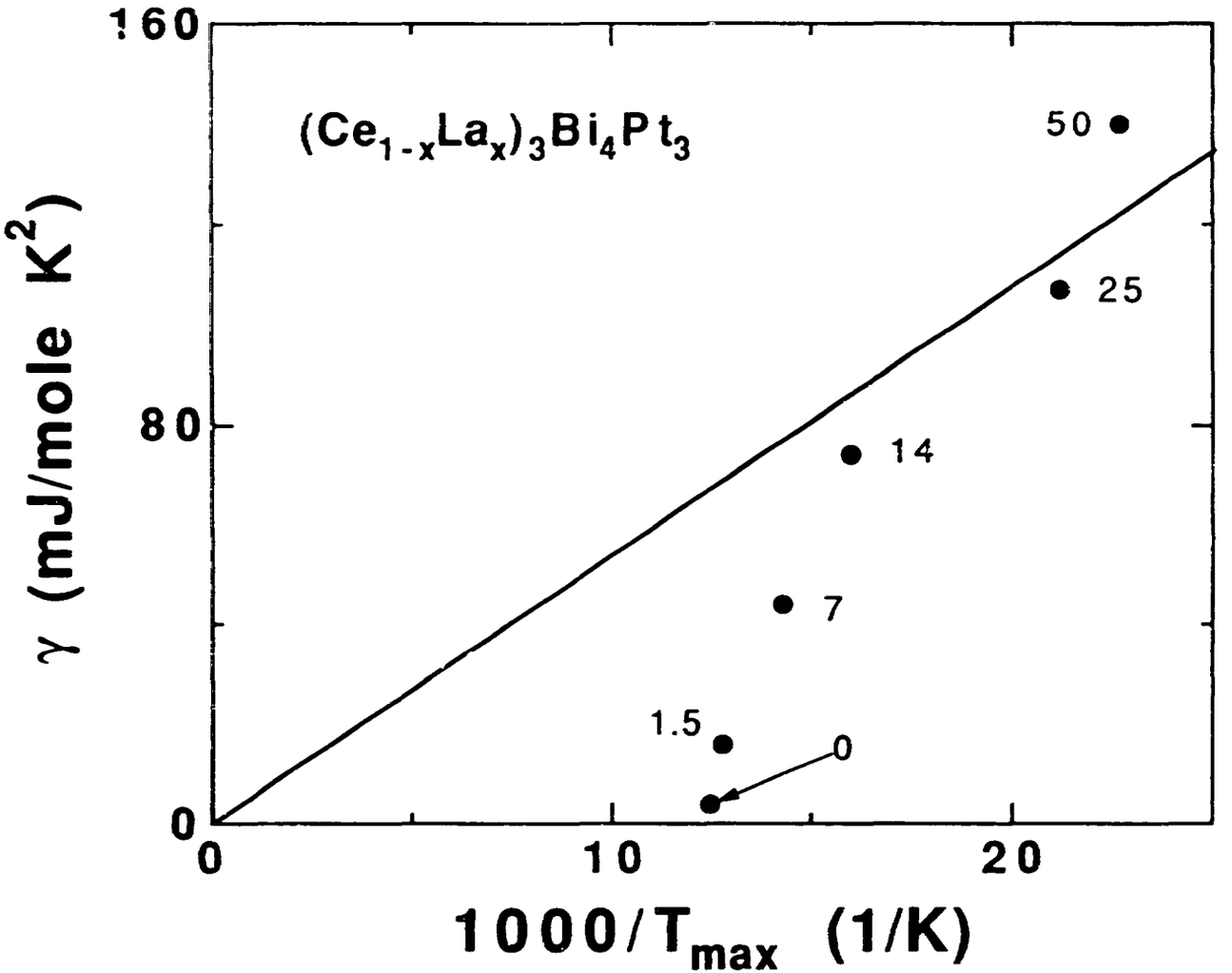


Fig. 4 Hundley et al.