Correlated insulating ground state of $Ce_3Bi_4Pt_3$

S. H. Liu
Department of Physics, University of California, San Diego, La Jolla, California 92093-03219
(Received 22 February 2001; revised manuscript received 11 June 2001; published 18 October 2001)

In an earlier paper the author suggested a different ground state for $Ce_3Bi_4Pt_3$ consisting of localized dimers of *d* and *f* electrons. The stability of the state was studied under a restricted condition that the *d* band is inherently narrow. In this paper we show that the same ground state can be stabilized when the *d* band is broad provided that the combined effects of *d-f* hybridization and Coulomb interactions are considered. A narrow, hybridized conduction band emerges naturally. The calculated inelastic neutron scattering cross section bears close resemblance to the experimental data.

DOI: 10.1103/PhysRevB.64.184108 PACS number(s): 71.28.+d, 71.27.+a, 75.30.Mb, 72.80.−r

I. INTRODUCTION

The compound $Ce_3Bi_4Pt_3$ belongs to a class of solids known as ''Kondo insulators.''[1–3] The following is a summary of its basic properties. The temperature dependence of the electrical resistivity establishes that the material is an insulator, but the data do not conform to the Kondo behavior, nor does the plot of $\log \rho$ versus temperature yield a clear thermal activation behavior.[3,4] The Hall mobility is negative, which indicates electron conduction. The magnetic susceptibility follows a Curie-Weiss law above approximately 150 K. It peaks at 80 K, then falls to a low temperature limiting value of approximately one-half of the peak value after a sample dependent, low temperature Curie tail is subtracted.[5] The specific heat γ is low and sample dependent.[4,6] The authors concluded that γ vanishes for the pure material. Although the full electronic contribution to the specific heat curve has not been measured, Kwei *et al.*[7] have deduced a semiquantitative result from the thermal expansion data and the Grüneisen parameter. Their curve has the shape of a Schottky anomaly peaked at 50 K, and the entropy under the curve is roughly $R \ln 6$ per mole. The position of the peak is consistent with an energy gap in the electronic excitation spectrum of approximately 100–160 K.

Inelastic neutron scattering on a powdered single crystal sample of $Ce_3Bi_4Pt_3$ has revealed an energy gap in the electronic spectrum at small scattering angles.[8,9] At 2 K the magnetic scattering intensity vanishes for energy transfers smaller than 12 meV ($\simeq$140 K), rises sharply between 12 and 20 meV, then falls off steadily to zero around 65 meV ($\simeq$780 K). Above 100 K the gap structure gradually disappears. The data support an energy gap of approximately 140 K in the low momentum region of the Brillouin zone. The size of the gap is in good agreement with the value estimated from anomalous thermal expansion by Kwei *et al.*[7]

The origin of the gap has been attributed by Fisk *et al.*[1] to the hybridization gap, i.e. the gap in the band structure of a periodic spin fluctuation system in the low temperature coherent state.[10,11] There is an inconsistency in this picture, however, because Kwei *et al.* pointed out that the hybridization gap is an *indirect* gap, i.e., the gap between the upper band at the zone center and the lower band at the zone boundary. The direct gap at the band crossing point is expected to be much larger. Accordingly, they predicted that the gap could be seen by neutron magnetic scattering at large scattering angles.[7] The experiment of Severing *et al.* made it clear that the magnetic gap is identical to the thermodynamic gap, and is not an indirect gap.[8,9] Thus, the hybridization gap picture and the associated spin fluctuation theory are in conflict with neutron scattering results.

In a series of publications Irkhin and Katsnelson[12–14] showed that the *d-f* Coulomb interaction, the so-called Falicov-Kimball interaction,[15,16] can stabilize an energy gap in the one quasiparticle spectrum of intermediate valence materials such as SmS and YbB_{12}. They made a number of predictions of the physical properties of their model, but detailed data were not available for comparison. This line of inquiry was brought up to date by Duan *et al.*[17] who suggested that the low temperature phase is an excitonic insulator state with *p*-wave symmetry. The authors were able to fit the temperature dependence of the magnetic susceptibility above 120 K, and to explain the observed correlation between the susceptibility and the lattice expansion.[4] On the negative side, the theory predicts an indirect gap of 44 K, in disagreement with neutron scattering experiments.

The present author suggested in an earlier publication that the semiconducting gap in $Ce_3Bi_4Pt_3$ is not in the one-particle spectrum.[18] The argument is based on the results of a doping experiment of Canfield *et al.*[19] It was reported that partial replacement of Ce in $Ce_3Bi_4Pt_3$ by La yields an *n*-type semiconductor even though La has one fewer electron than Ce. Also, the impurity leaves the gap practically unchanged as seen from the position of the specific heat peak. To explain this unusual behavior the present author proposed that in the ground state the *d* electron of Ce forms a bound state with the *f* electron. With no *f* electron on the site where Ce is replaced with La, the correlated pair does not form. The *d* electron of La goes into the conduction band to provide electron conduction and specific heat γ. That the gap remain unaffected reflects the local nature of the correlated *d-f* pair. Thermal excitation also breaks up the local dimer state, sending one electron into the conduction band and another into a one-electron local state. Thus the gap is to be found in the two-particle excitation spectrum. It should be noted that the compound YbB_{12} has the normal doping behavior, namely, the Lu doped material is *n*-type semiconductor.[20,21] One must conclude that there is no universal theory for all Kondo insulators.

In Ref. 18 the dimer ground state was derived under the assumption that the *d* band is inherently narrow. Critics of this work argued that in a properly formulated theory the observed narrow conduction band should emerge as a result of the ground state correlation. The present paper takes a step in this direction. The theory developed in Sec. II assumes a broad *d* band and solves for the properties of the *d*-*f* dimer. The excitation spectrum is determined by the simultaneous promotion of two electrons, one into a one-electron local level and one into the conduction band. The inelastic neutron scattering cross section is calculated, and the result bears close resemblance to the data.

II. THEORY

The following Hamiltonian embodies all relevant interactions in the system ($\hbar = k_B = 1$):

$$H = \sum_{\vec{k}\sigma} \epsilon_k c^\dagger_{\vec{k}\sigma} c_{\vec{k}\sigma} + \sum_{j\sigma} \epsilon_f f^\dagger_{j\sigma} f_{j\sigma} + \frac{1}{\sqrt{N}} \sum_{\vec{k},j\sigma} (V_{\vec{k}} f^\dagger_{j\sigma} c_{\vec{k}\sigma} e^{i\vec{k}\cdot\vec{R}_j} + \text{H.c.}) + U_{ff} \sum_j f^\dagger_{j+} f_{j+} f^\dagger_{j-} f_{j-} + U_{dd} \sum_j c^\dagger_{j+} c_{j+} c^\dagger_{j-} c_{j-} + U_{df} \sum_{j\sigma} c^\dagger_{j\sigma} c_{j\sigma} \left(\sum_{\sigma'} f^\dagger_{j\sigma'} f_{j\sigma'} - 1 \right), \quad (1)$$

where

$$c_{j\sigma} = \frac{1}{\sqrt{N}} \sum_{\vec{k}} c_{\vec{k}\sigma} e^{i\vec{k}\cdot\vec{R}_j}. \quad (2)$$

The first term on the right-hand side of Eq. (1) is the energy of the conduction band with $0 \leqslant \epsilon_k \leqslant W$, W being the bandwidth. There is one $5d$ electron for each Ce atom. The second term is the energy of the localized *f* levels. For simplicity we consider only a Kramers doublet of *f* levels. The *V* term represents the *d*-*f* hybridization interaction; *N* is the number of Ce sites in the sample. The last three terms are various Coulomb interactions. The term involving U_{ff} denotes the Coulomb repulsion between two *f* electrons with opposite spins on the same site. It is so strong that it prevents the *f* level from more than singly occupied. The U_{dd} term is the Coulomb interaction between two electrons in the $5d$ state. The U_{df} term represents the attractive Coulomb interaction between an *f* hole and a *d* electron on the same site. It has been invoked by Falicov and coworkers to explain the α to γ phase transition of Ce,[15,16] and by the present author for the crossover from coherent to incoherent states in heavy fermion materials.[22]

In this Section we first show that it is possible to stabilize a one-electron local state in this model. This state is an integral part of the excited state spectrum of the system. We will then use the result to construct a trial wave function for the two-electron local state. The properties of the *f* electrons are analyzed for isolated sites because the interactions are basically local. Ultimately the ground state is coherent, but the effects of coherence is beyond the scope of this paper.

A. The one-electron local state

The effective Hamiltonian for the single site one-electron problem is

$$H_1 = \sum_{\vec{k}\sigma} \epsilon_k c^\dagger_{\vec{k}\sigma} c_{\vec{k}\sigma} + \sum_\sigma \epsilon_f f^\dagger_{j\sigma} f_{j\sigma} + \frac{1}{\sqrt{N}} \sum_{\vec{k},\sigma} (V_{\vec{k}} f^\dagger_{j\sigma} c_{\vec{k}\sigma} e^{i\vec{k}\cdot\vec{R}_j} + \text{H.c.}) + U_{df} \sum_\sigma c^\dagger_{j\sigma} c_{j\sigma} \left(\sum_{\sigma'} f^\dagger_{j\sigma'} f_{j\sigma'} - 1 \right). \quad (3)$$

The U_{ff} and U_{dd} terms are inoperative. The trial wave function for the one-electron local state is

$$\psi_{j\sigma} = a f_{j\sigma} + \sum_{\vec{k}} b_{\vec{k}} c_{\vec{k}\sigma} e^{i\vec{k}\cdot\vec{R}_j}. \quad (4)$$

The operator $\psi_{j\sigma}$ satisfies the Schrödinger equation $[H_1, \psi_{j\sigma}] = \omega_1 \psi_{j\sigma}$ where ω_1 is the energy of the one-electron state. In evaluating the commutator we encounter the following:

$$[H_1, f_{j\sigma}] = \epsilon_f f_{j\sigma} + \frac{1}{\sqrt{N}} \sum_{\vec{k}} V_{\vec{k}} c_{\vec{k}\sigma} e^{i\vec{k}\cdot\vec{R}_j} + U_{df} \sum_{\sigma'} c^\dagger_{j\sigma'} c_{j\sigma'} f_{j\sigma}.$$

The U_{df} term makes no contribution because the *d* band is empty in the insulating state. Similarly

$$[H_1, c_{\vec{k}\sigma}] = \epsilon_k c_{\vec{k}\sigma} + \frac{1}{\sqrt{N}} V^*_{\vec{k}} f_{j\sigma} e^{-i\vec{k}\cdot\vec{R}_j} + \frac{U_{df}}{\sqrt{N}} c_{j\sigma} \left(\sum_{\sigma'} f^\dagger_{\sigma'} f_{\sigma'} - 1 \right) e^{-i\vec{k}\cdot\vec{R}_j}.$$

In the U_{df} term we replace the *f* number operator by the expectation value zero. The left-hand side of the Schrödinger equation simplifies. By equating like terms we obtain

$$(\epsilon_f - \omega_1) a + \sum_{\vec{k}} b_{\vec{k}} \frac{V^*_{\vec{k}}}{\sqrt{N}} = 0 \quad (5)$$

and

$$(\epsilon_k - \omega_1) b_{\vec{k}} - \frac{U_{df}}{N} \sum_{\vec{k}'} b_{\vec{k}'} + a \frac{V_{\vec{k}}}{\sqrt{N}} = 0. \quad (6)$$

We find from Eq. (6) that

$$\sum_{\vec{k}} b_{\vec{k}}\left[1-\frac{U_{df}}{N}\sum_{\vec{k}}\frac{1}{\epsilon_k-\omega_1}\right]=-a\frac{V}{\sqrt{N}}\sum_{\vec{k}}\frac{1}{\epsilon_k-\omega_1},$$

where V is the isotropic part of $V_{\vec{k}}$ as discussed in Ref. 18. The equation for ω_1 takes the form

$$\epsilon_f-\omega_1=\frac{V^2}{N}\frac{\sum_{\vec{k}}\frac{1}{\epsilon_k-\omega_1}}{1-\frac{U_{df}}{N}\sum_{\vec{k}}\frac{1}{\epsilon_k-\omega_1}}.$$

We assume a rectangular d band with density-of-states ρ_0 and extending from 0 to W, then carry out the sums to obtain

$$\epsilon_f-\omega_1=\frac{\rho_0V^2\ln\left[\frac{W-\omega_1}{-\omega_1}\right]}{1-\rho_0U_{df}\ln\left[\frac{W-\omega_1}{-\omega_1}\right]}. \tag{7}$$

This equation reduces to

$$\omega_1=-\frac{W}{e^{1/\rho_0(V^2/(\epsilon_f-\omega_1)+U_{df})}-1}, \tag{8}$$

which can be solved by iteration. It is easy to show that $\omega_1<0$ and $\omega_1<\epsilon_f$.

The wave function coefficients are also solved from Eq. (6):

$$\sum_{\vec{k}} b_{\vec{k}}=-\frac{aV}{F\sqrt{N}}\sum_{\vec{k}}\frac{1}{\epsilon_k-\omega_1},$$

where

$$F=1-\frac{U_{df}}{N}\sum_{\vec{k}}\frac{1}{\epsilon_k-\omega_1}.$$

Therefore

$$b_{\vec{k}}=-\frac{aV}{F\sqrt{N}}\frac{1}{\epsilon_k-\omega_1}. \tag{9}$$

The normalization condition is

$$a^2+\sum_{\vec{k}} b_{\vec{k}}^2=1,$$

which reduces to

$$a^{-2}=1+\frac{V^2}{F^2N}\sum_{\vec{k}}\frac{1}{(\epsilon_k-\omega_1)^2}.$$

The f content of the state is a^2, and the d content is $1-a^2$.

We may define

$$d_{j\sigma}=\sum_{\vec{k}} b_{\vec{k}}c_{\vec{k}\sigma}, \tag{10}$$

where $b_{\vec{k}}$ is in Eq. (9). The operator $d_{j\sigma}$ satisfies the commutation rule

$$[d_{j\sigma},d_{j'\sigma'}^\dagger]=\delta_{jj'}\delta_{\sigma\sigma'}(1-a^2).$$

Then we can write the operator $\psi_{j\sigma}$ as

$$\psi_{j\sigma}=af_{j\sigma}+d_{j\sigma}. \tag{11}$$

This result will be used to construct the two-electron local level, which is our proposed ground state of the system.

B. Two-electron localized state

The two-electron problem is much more difficult to analyze. We must include the U_{ff} and U_{dd} terms so that the effective Hamiltonian becomes $H_2=H_1+U_j$ where

$$U_j=U_{ff}f_{j+}^\dagger f_{j+}f_{j-}^\dagger f_{j-}+U_{dd}c_{j+}^\dagger c_{j+}c_{j-}^\dagger c_{j-}. \tag{12}$$

The eigenvalue equations have been worked out in Ref. 22. They are difficult to solve in the broad band limit, and we will turn to a variational solution using a trial wave function constructed from the solution of the one-electron problem. We write

$$|\Phi_j\rangle=[\beta(f_{j+}^\dagger d_{j-}^\dagger+d_{j+}^\dagger f_{j-}^\dagger)+\gamma d_{j+}^\dagger d_{j-}^\dagger]|0\rangle, \tag{13}$$

where $d_{j\sigma}$ was defined in Eq. (10), $|0\rangle$ is the vacuum state, and β and γ are two variational parameters. The effect of infinit U_{ff} has been built into $|\Phi_j\rangle$ by leaving out doubly occupied f state. We denote the ground state energy by ω_0, and work out $\langle\Phi_j|H_2-\omega_0|\Phi_j\rangle=0$:

$$\begin{aligned}\langle\Phi_j|H_2-\omega_0|\Phi_j\rangle&=2\beta^2\sum_{\vec{k}} b_{\vec{k}}^2(\epsilon_f+\epsilon_k-\omega_0)+\gamma^2\sum_{\vec{k}\vec{k}'} b_{\vec{k}}^2b_{\vec{k}'}^2(\epsilon_k+\epsilon_{k'}-\omega_0)\\&+V\frac{4\beta\gamma}{\sqrt{N}}\sum_{\vec{k}\vec{k}'} b_{\vec{k}}b_{\vec{k}'}^2.+U_{dd}\frac{\gamma^2}{N^2}\sum_{\vec{k}_1\vec{k}_2\vec{k}_3\vec{k}_4} b_{\vec{k}_1}b_{\vec{k}_2}b_{\vec{k}_3}b_{\vec{k}_4}\\&-U_{df}\frac{2\gamma^2}{N^2}\sum_{\vec{k}\vec{k}'\vec{k}''} b_{\vec{k}}b_{\vec{k}'}b_{\vec{k}''}^2.\end{aligned}$$

The various terms can be evaluated by using results found in the one-electron calculation, chiefly

$$\frac{1}{\sqrt{N}}\sum_{\vec{k}} b_{\vec{k}}=-\frac{a}{V}(\epsilon_f-\omega_1),$$

and

$$\sum_{\vec{k}} b_{\vec{k}}^2=1-a^2.$$

All other sums can be worked out as combinations of these two. Without elaborating on the detail, we find

$$\langle\Phi_j|H-\omega_0|\Phi_j\rangle=A\beta^2+2B\beta\gamma+C\gamma^2,$$

where

$$A=2(1-a^2)(2\omega_1-\omega_0)+2(\epsilon_f-\omega_1),$$

$$B=2a(1-a^2)(\epsilon_f-\omega_1),$$

and

$$C=2a^2(1-a^2)(\epsilon_f-\omega_1)+(1-a^2)^2(2\omega_1-\omega_0)$$
$$+U_{dd}a^4\left[\frac{\epsilon_f-\omega_1}{V}\right]^4-2U_{df}a^2(1-a^2)\left[\frac{\epsilon_f-\omega_1}{V}\right]^2.$$

Minimization of the energy leads to

$$A\beta+B\gamma=0$$

and

$$B\beta+C\gamma=0. \tag{14}$$

The eigenvalue equation for ω_0 is given by

$$AC-B^2=0,$$

which is expanded into

$$(2\omega_1-\omega_0)^2+g(2\omega_1-\omega_0)+h=0, \tag{15}$$

where

$$g=(\epsilon_f-\omega_1)\frac{1+2a^2}{1-a^2}+u,$$

with

$$u=U_{dd}\frac{a^4}{(1-a^2)^2}\left[\frac{\epsilon_f-\omega_1}{V}\right]^4-U_{df}\frac{2a^2}{1-a^2}\left[\frac{\epsilon_f-\omega_1}{V}\right]^2$$

and

$$h=(\epsilon_f-\omega_1)^2\frac{2a^4}{(1-a^2)^2}+\frac{1}{1-a^2}(\epsilon_f-\omega_1)u.$$

The quadratic equation in Eq. (15) has two roots, and the larger one, denoted by x_1, determines the upper bound for $\omega_0=2\omega_1-x_1$.

The stability of the dimer state is determined by considering its excited modes. The lowest set of excitation modes break up the pair so that one electron goes into the one-electron local state and the other into the band. The two-electron state is stable against this excitation provided that $\omega_0<\omega_1$, because the bottom of the band is defined as 0. The dimer state can also be excited into uncorrelated d and f electrons, but the conditions that $\omega_1<\epsilon_f$ and $\omega_1<0$ put these modes at higher energies. Numerical calculation shows that the two-electron state is stable for sufficiently strong U_{df} and relatively weak U_{dd}. The insulating energy gap is given by $\Delta=\omega_1-\omega_0$.

Finally we normalize the two-electron wave function by imposing

$$2\beta^2(1-a^2)+\gamma^2(1-a^2)^2=1, \tag{16}$$

where the ratio β/γ is solved from Eq. (14). The f content of this state is $2\beta^2(1-a^2)$ while the d content is $2\beta^2(1-a^2)+2\gamma^2(1-a^2)^2$.

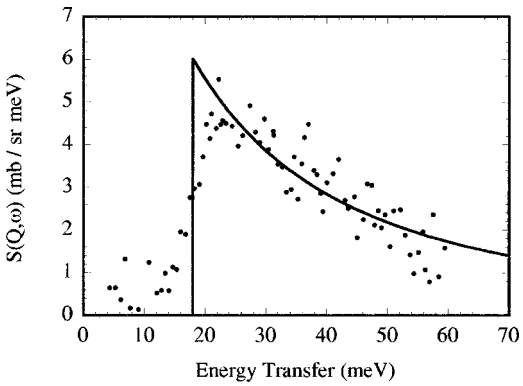

FIG. 1. The calculated neutron inelastic scattering cross section compared with the data taken at 2 K (Refs. 8 and 9).

C. Excitation spectrum

The elementary excitations of this system consist of breaking up the ground state dimer and moving one member into the one-electron local level $\psi_{j\sigma}$ and the other into the band. Therefore, we must obtain the spectrum from studying a two-electron response function. As an example we evaluate the dynamical susceptibility, which connects directly to the inelastic neutron scattering experiment. As shown in Ref. 18, the magnetic moment of a site j is given by $M_j=\mu(f_{j+}^\dagger f_{j+}-f_{j-}^\dagger f_{j-})$, where μ is an effective moment which depends on the composition of both the ground state and the excited state. The neutron scattering cross section is given by

$$S(\omega)=\pi\mu^2\sum_{\vec{k}\sigma}|\langle c_{\vec{k},-\sigma}\psi_{j\sigma}|f_{j+}^\dagger f_{j+}-f_{j-}^\dagger f_{j-}|\Phi_0\rangle|^2$$
$$\times\delta(\omega+\omega_0-\omega_1-\epsilon_k). \tag{17}$$

The matrix element is worked out by using the wave functions in Eqs. (11) and (13)

$$S(\omega)=2\pi\mu^2a^2\beta^2\frac{a^2\rho_0V^2\theta(\omega-\Delta)}{F^2(\omega-\Delta-\omega_1)^2}, \tag{18}$$

where $\Delta=\omega_1-\omega_0$ is the energy gap. The result for Δ shows clearly that the energy gap is not determined by the one-particle spectrum alone.

In Fig. 1 we show a calculated spectrum superimposed on the neutron data taken at 2 K.[8,9] The theoretical curve was calculated using the following parameters: $\epsilon_f=0, V=0.05W, U_{dd}=U_{df}=0.1W$, and the band width $W\approx1$ eV is the overall energy scale chosen to fit the size of the gap. Aside from W and the vertical scale, we have made no special effort to fit the data, because the theoretical curve is found to be quite insensitive to the model parameters. The theory does not include the experimental resolution function whose effect is to soften the leading edge at the gap as seen in the data. The gap edge is also softened by the fact that in the coherent state the one-electron level may be dispersive due to hybridization with the d band.[23] There seems to be some evidence that the gap is slightly dispersive.[24] We will investigate this problem in the future.

We conclude our discussion by reiterating the improvement made in this work over the author's previous publication cited as Ref. 18. The main point of the earlier paper was to show that a two-electron ground state can be stable under the condition of sufficiently narrow conduction band and sufficiently strong U_{df}. The analysis did not allow a determination of the neutron scattering spectrum. Instead, we *postulated* a simple triangular spectrum and showed how one can make a complete phenomenological analysis of the thermodynamic and transport properties by using this information. In this paper we have removed the restrictive condition of narrow band and *derived* the neutron scattering spectrum from first principles. Without repeating the phenomenological analysis, we feel certain that the spectrum in Fig. 1 should give equally good account of the bulk properties because they are determined largely by the low energy part of the spectrum.

ACKNOWLEDGMENTS

The author is indebted to Professor P. S. Riseborough, Dr. A. J. Arko, and Professor P. A. Alekseev for helpful communications. He also wishes to thank Professor M. B. Maple and Professor L. J. Sham for their hospitality.

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