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Journal

Journal of Magnetism and Magnetic Materials, 76-77(C)

ISSN

0304-8853

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Publication Date

1988-12-02

DOI

10.1016/0304-8853(88)90368-X

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Peer reviewed

INVITED PAPER

OBSERVATION OF A COHERENT QUASIPARTICLE BAND IN THE PERIODIC HEAVY FERMION SYSTEM CeCu_6

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The question addressed here is whether there is structure in the narrow renormalized quasiparticle bands due to coherence in a periodic heavy fermion lattice. In CeCu_6 the specific heat indicates that the Fermi temperature, T_F , is about 3 K. We find that for a crystal of CeCu_6 , well below T_F , the Hall constant shows changes in behavior at about 400 mK and again at 25 mK. This result is evidence that there is structure in the band on a scale smaller than T_F due to the periodicity of the Kondo lattice system.

On cooling through a characteristic temperature, T_F , heavy fermion systems develop a narrow renormalized quasiparticle band. This band is a Fermi liquid characterized by a Fermi temperature, T_F . Evidence of the narrow band is seen in the specific heat data of Amato et al. [1] for CeCu_6 which shows a dramatic increase in C/T below 10 K to a value of 1600 mJ/m-K². In a Kondo impurity model [2], this value corresponds to a Kondo temperature of about 3 K. For our concentrated Kondo system, 3 K characterizes the Fermi temperature of the renormalized band. The neutron quasielastic line width of 4 K found by Aeppli et al. [3] and by Walter et al. [4] is another measure of T_F , consistent with the specific heat determination.

Martin [5] suggested that in a periodic Kondo lattice there would be structure in the Kondo resonance due to periodicity. Bredl et al. [6] looking for evidence of this structure, found a maximum in C/T at about 0.5 K for both CeAl_3 and CeCu_2Si_2 . In substituted alloys, without perfect periodicity, the enormous values of C/T (of order 1 J/m-K²) remained but the maximum disappeared. They concluded that there was structure in the density of states for the pure periodic systems and that the Fermi level lay in a small minimum. In the case of CeCu_6 , a maximum is not seen [1] in C/T , so that if there is structure, the Fermi level is not in a minimum of the density of states.

Steglich et al. [7,8] found for both CeAl_3 and CeCu_2Si_2 that the thermopower changed sign from negative to positive at approximately the same temperature as the C/T maximum [6]. They at-

tributed this change to the same structure in the Kondo resonance that they had invoked for the C/T maximum. Positive thermopower peaks at 100 mK for CeAl_3 and at 200 mK for CeCu_2Si_2 imply structure on this scale as well. Flouquet et al. [9,10] reported peaks in the thermopower and thermal conductivity of CeAl_3 and concluded that there was a second temperature scale smaller than T_K produced by coherence.

In the temperature range above T_F , there is a strong Kondo like resistivity (fig. 1) due to incoherent scattering of individual Ce ions, similar to that observed by Sumiyama et al. [11–13], for Ce in LaCu_6 . In the region of T_F , where C/T rises rapidly, the resistivity falls to a very small fraction of its incoherent value as the Ce ions begin to scatter coherently. For temperatures $T \ll T_F$, the resistivity follows $\rho = AT^2$ as expected for electron–electron scattering in a Fermi liquid [14–17]. Furthermore, the coefficient A is approximately proportional to $(1/T_F)^2$ [18]. The T^2 behavior is only seen in the extreme degenerate limit for $T \leq 100$ mK. From these results alone, one might conclude that the low temperature behavior is given by just one temperature scale, T_F , but that it is necessary to have $T \ll T_F$ to observe the limiting Fermi liquid behavior.

We have reported previously the results of Hall and resistivity measurements on two single crystals of CeCu_6 for temperatures down to 30 mK [19–21]. In the incoherent scattering regime, above 30 K, the Hall effect is dominated by skew scattering and follows $\chi\rho_m$, the product of the magnetic susceptibility of the Ce ions and the resistivity due

Fig. 1. Resistivity of a single crystalline bar of CeCu₆ with current in the orthorhombic *b* direction. The solid line is the magnetic scattering remaining after subtraction of the resistivity of the reference compound, LaCu₆ [11]. The small resistance at low temperature is indicative of coherent scattering while the large resistance at the peak is from incoherent scattering. Figure from ref. [21].

to the magnetic scattering from the Ce ions. In the region of the resistivity maximum, as the resistivity breaks away from the incoherent impurity behavior so does the Hall constant break away from $\chi\rho_m$. In fact the Hall constant goes strongly negative as shown in fig. 2. The incoherent region and the transition to the coherent region have been discussed and relevant work cited in our previous publications [19–21]. Subsequently, Hall studies on Ce_xLa_{1-x}Cu₆ by Onuki and Komatsubara [11] have shown that with increasing La substitution, the Hall constant at 1 K changes from strongly negative, as we observe, to being strongly positive as expected for incoherent scattering. Recent theoretical calculations by Levy and Fert [22] show another contribution to the Hall effect in addition to the skew scattering. It is a side jump mechanism which they call an anomalous velocity effect. It may be as strong as the skew scattering for temperatures in the neighborhood of T_F . It is still a single site incoherent scattering effect. At present there is no theory for the Hall effect in the coherent regime.

The important issue addressed in this work is the possibility of fine structure in the quasiparticle band, due to coherence which is observable only below T_F . Our studies show that the Hall effect is particularly sensitive to such structure. Indeed, we see (fig. 2) that there is an abrupt change in behavior near 400 mK. In another sample [20] with different orientation, the change takes place at about 200 mK. Winzer [23] finds a negative extremum like ours at 500 mK for a polycrystalline sample of CeCu₆. In some other heavy fermion systems, such as CeAl₃, CeRu₂Si₂, and UAl₂ and UPt₃, Lapierre et al. [24] find that the Hall constant changes from being strongly temperature dependent at high temperature to being very weakly temperature dependent at low temperature. For CeAl₃, which has a similar T_F to CeCu₆, this change occurs at about 300 mK.

The thermopower of Amato et al. [1] is linear above 500 mK, but the slope changes in the region of 300 mK to a much steeper slope at lower temperatures. These Hall and thermopower results

Fig. 2. Hall constant for the same CeCu₆ crystal as in fig. 1, for a field of 1 T in the direction of largest susceptibility, the orthorhombic *c*. The solid line is the high temperature $\chi\rho_m$ fit for incoherent skew scattering. The Hall constant deviates entirely from the incoherent behavior in the neighborhood of $T_F \approx 3$ K where the resistivity is dropping rapidly and C/T is rising rapidly. The change in behavior near 400 mK indicates structure in the quasiparticle band on this scale. Figure from ref. [21].

indicate that there is structure in the quasiparticle band on the scale of 300 mK.

In order to observe the true asymptotic behavior of the Fermi liquid, we measured the Hall effect down to 12 mK. Great care was taken to insure good thermal equilibrium. The sample was a bar taken from the larger bar of figs. 1 and 2. The new data are shown in figs. 3 and 4. The extremum at 400 mK is seen as before. Below that, R_H is approximately linear in T with a negative slope and negative intercept (fig. 3). The data may be fit just as well to a $T^{1.25}$ law and a fit to $T^{1.5}$ is only slightly worse. The remarkable new result is the change in behavior below 25 mK. This deviation from the linear T law observed above 25 mK is seen best in the log T plot, fig. 4. In this plot the solid line is the linear T fit from fig. 3. The dashed line is the extrapolation of the linear T fit. The range from 12 to 25 mK is too small to determine the power law. The slope, however, has clearly changed sign. The $T=0$ intercept is still negative. This result reveals that there is structure in the quasiparticle band even on this fine scale.

The recent work of Sato et al. [25] on single crystal CeCu₆ gives support to the view that there are changes in behavior at temperatures much

Fig. 3. Hall constant below 400 mK for a single crystalline bar taken from the sample of figs. 1 and 2. The current and magnetic field are along b and c , respectively, as in fig. 2. The Hall voltage is linear in field in the 0–1 T range studied. Between 30 and 300 mK, the Hall constant follows a linear T dependence with negative slope and intercept.

Fig. 4. The same Hall data as in fig. 3, but on a log T scale. The solid line is the linear T fit of fig. 3. The change in behavior below 25 mK is an indication that there is structure in the renormalized quasiparticle band on a scale much smaller than $T_F \approx 3$ K.

smaller than T_F . They have measured the resistivity and fit it to $\rho = \rho_0 + A(T)T^2$ and the thermoelectric ratio $G = S/LT$, where S is the thermopower and L is the Lorenz number. They find that between 1 K and 30 mK, both A and G change by large amounts. Only below 30 mK are they temperature independent. Coleridge [26] has measured the magnetoresistance up to 10 T at various temperatures of some of the high quality single crystals that were used for the de Haas–van Alphen measurements of Reinders et al. [27]. Coleridge finds both positive and negative contributions to the magnetoresistance for the current along the b axis and the magnetic field along c . He associates the negative part which is dominant down to about 500 mK to incoherent Kondo scattering. Below 30 mK he finds only a positive linear magnetoresistance which he associates with the coherent state. The transition to this state is smooth.

Another way to view the magnetoresistance is to measure ρ vs. T^2 for various magnetic fields as done by Amato et al. [1]. They find that the slope A decreases monotonically with H , in fact $A(H)^{1/2}$ scales roughly with $\gamma(H)$ and $\chi(H)$ showing their common origin in T_F . The intercept ρ_0 first increases (1.5 T) and then decreases (4.6 T) with

field. The region of T^2 behavior increases from about 100 mK at 0 T, to about 150 mK at 1.5 T. This result confirms the necessity for $T \ll T_F$ to observe the T^2 behavior. An interesting conclusion from this field scaling is that T_F and m^* are strongly field dependent [1]. This is in apparent contrast to the de Haas–van Alphen results of Reinders et al. [27] who found a field independent m^* . A possible solution to this contradiction is that it is the heaviest masses which dominate the specific heat and fields of order 5–7 suppress the increase in C/T below 2 K [1]. Since the measurement [27] of the field independent mass was for a mass of $m^*/m = 6$, in fields between 6 and 13 T, the apparent contradiction can be understood.

A possible objection to our claim that we have seen the effect of coherence, is that our findings could be the result of some unseen underlying uninteresting magnetic transitions unrelated to the heavy fermion renormalized band. Mignot et al. [28] have studied the low temperature metamagnetic transition in CeRu₂Si₂ which occurs at 8 T. Regnault et al. [29] have seen this transition in neutron diffraction as an increase in ferromagnetic fluctuations and a decrease in antiferromagnetic fluctuations. They also see a similar transition in CeCu₆ at 2 T. Reinders et al. [27] see structure in their de Haas–van Alphen data at 2 T. Since the Hall data is taken at 1 T and below, this metamagnetic transition probably has no bearing on our result. Furthermore, the metamagnetic transition field in CeRu₂Si₂ has been shown by Mignot et al. to scale under pressure with both χ and the A coefficient of the T^2 resistivity. The metamagnetism, therefore, has its origin in the renormalized band. If it does have a bearing on the Hall effect, it is traceable back to the renormalized band.

Recent reviews of heavy fermion systems have been given by Lee et al. [30], Fulde et al. [31] and Fisk et al. [32]. In the introduction of Fulde et al., there is a very clear description of the different energy scales. There is one scale which we have called T_F and is the scale of the quasiparticle bandwidth. In addition, one expects a number of peaks in the density of states due to the structure in the band. The origin of this structure is the periodicity, just as in a normal 3d transition metal. The widths of the peaks, T_p , vary but will be smaller than T_F . This point of view, which is the

same as Martin's, seems to fit the experiments well, since we find changes in behavior at several hundreds of mK and again at several tens of mK.

There is some confusion in the use of the word coherence because it is used to describe two different but related effects. Firstly, there is the formation of the narrow band of quasiparticles in the temperature region T_F . This is due to coherence between wave functions on different sites [30–35]. In a periodic system, as the coherence builds and C/T increases, the resistivity decreases strongly. The formation of the narrow band, however, does not depend greatly on periodicity as demonstrated by the fact that many substituted systems have enormous electronic specific heats but do not show the low resistivity associated with coherence. In these disordered systems, Bloch waves are strongly scattered but there still exists a non-periodic coherence. The second use of the term coherence is to describe the origin of the structure in the quasiparticle band. In a periodic system this structure should be much sharper than in a disordered system, just as in the case of common unrenormalized bands.

Coherence has been discussed by a number of theorists in addition to Martin [5]. Structure in the quasiparticle bands can be seen in the renormalized band structure calculations of Fulde et al. [31] for CeCu₂Si₂ and by Newns and Read [34] for CeSn₃. Brandow [36] has argued that there is but one temperature scale, T_F , and no separate coherence temperature. However, he notes that for $T \ll T_F$ the physical properties will be dominated by the particular quasiparticle band structure. This point of view is similar to that initially proposed by Martin, and discussed by Fulde et al. Coleman [37] finds one temperature scale for the Kondo lattice. This one scale is sufficient for the specific heat and the resistivity. As observed experimentally, the calculated resistivity drops rapidly for $T \approx T_F$ and for $T \ll T_F$ it follows AT^2 with $A \approx 1/T_F^2$. This point of view is similar to Brandow's. In another study, Koyama and Tachiki [38] have found for the Anderson lattice, a sharp peak in the quasiparticle density of states at E_F , but no partial gap.

In other studies, coherence pseudogaps have been explicitly found. Grewe [39] has calculated densities of states for Kondo impurities and the Kondo lattice. He finds in both cases that as the

temperature drops below T_K , a resonance occurs in the neighborhood of the Fermi energy, E_F . However, in the lattice case, there is a partial gap just above E_F due to coherence which is about $T_K/3$ wide for his parameters.

Kaga et al. [40] have found that for a Kondo impurity there is a peak in the density of states of width T_K at E_F . For the Kondo lattice they find a peak T_K wide, located T_K below E_F and a pseudogap at E_F . Furthermore, the coherent structure forms before the single site Kondo resonance is fully formed. They define a coherence temperature $T_0 \sim 0.1T_K$ as the temperature at which the coherent lattice density of states is fully formed. Here T_K is the Kondo temperature for the impurity. The transition to the coherent state is smooth.

Lacroix [41] has found that the Kondo resonance of width T_K forms without structure for $T \approx T_K$. Then for $T < T_c$ the coherence temperature, a partial gap occurs in the resonance. The coherence temperature is $T_c = T_K(T_K/D)^{1/2}$, where D is the bandwidth of the conduction band. A similar point of view was taken by Moschalkov [42] at this conference. He suggested that the transition could be either continuous or abrupt.

A rather different direction has been taken by Doniach [43] who first considered the Kondo necklace and the competition between the Kondo quenching of local moments and magnetic order. He has shown that the peak in C/T below T_F observed in CeAl₃ and CeRu₂Si₂ may be due to the proximity of a spin density wave instability. Associated with this SDW phase is a second temperature scale.

All heavy fermion theories have a quasiparticle band at E_F to explain the specific heat data. Most theories also find that for the lattice case the band has more structure than for the impurity case. In most cases it appears to be somewhat a matter of semantics whether this added structure is associated with a new temperature scale. There is a distinction between the formation of a quasiparticle band with structure and a phase transition of incoherent Kondo states into a coherent Kondo lattice.

Conclusion

The extremely narrow renormalized quasiparticle bandwidth of a heavy fermion system has a

characteristic temperature, T_F , which is measured by the electronic specific heat and is about 3 K for CeCu₆. This bandwidth is not particularly sensitive to periodicity or disorder. In purely periodic systems, the resistivity is very large above T_F , but becomes very small with a T^2 dependence for $T \ll T_F$. No abrupt changes in the resistivity are observed. Our Hall results, however, show sharp features near 400 mK and near 25 mK, which we believe are caused by structure in the quasiparticle band due to the periodicity of the lattice. Thermopower results of others also show changes in behavior on these scales. We cannot determine whether any of these features indicate a phase transition. These observations strongly support the notion that coherence in a Kondo lattice system produces effects on a scale much smaller than T_F .

We thank J.M. Rigotty for technical assistance and J. Flouquet for valuable discussions. Work at Los Alamos was done under the auspices of the US Department of Energy.

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