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Model for the orientation, magnetic field, and temperature dependence of the specific heat of $CeCu_6$

A. S. Edelstein
Naval Research Laboratory, Washington, DC 20375-5000
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The results of a model calculation of the orientation, magnetic field, and temperature dependence of the specific heat C of $CeCu_6$ are found to be in good agreement with single-crystal data of Amato *et al.* The model incorporates both Kondo and crystal-field effects. It is suggested that the low-temperature Wilson's ratio $C/T\chi$, where χ is the susceptibility, may not change in an applied field H and that both C/T and χ at low temperatures as a function of H may be proportional to the many-body density of states at the energy μH.

Heavy-fermion systems[1,2] have a very large term in their low-temperature specific heat which is linear in the temperature, i.e., of the form γT. They were given this name since such a large linear term can be viewed as resulting from electronic excitations into a narrow, flat band at the Fermi surface. Electrons in such a band would have a heavy mass. The considerable interest heavy fermions have received is in part due to their unusual superconducting properties.[2,3]

The fact that the properties of heavy-fermion systems are strongly temperature dependent probably indicates that there is a small energy that characterizes these systems. Some of the properties of heavy-fermion systems can be understood by treating them as concentrated Kondo systems.[2,4] This view provides a simple explanation of both the large linear term in the specific heat γT and the small energy scale. The latter in this case is the Kondo energy, kT_K, where T_K is the Kondo temperature.

That these systems are characterized by a small energy motivated investigations of whether the specific heat could be modified by the application of a magnetic field. It was found[5-8] that the specific heat of two heavy-fermions systems, $CeAl_3$ and $CeCu_6$, is strongly field dependent while that of UPt_3 and UBe_{13} is not.[9-11]

The somewhat larger, crystal-field energies are also important. A crystalline electric field has the effect of decreasing the multiplicity at low temperatures and it usually introduces anisotropy. A form of the resonant level model[12] (RLM), which included crystal-field anisotropy, was previously[5] applied to $CeAl_3$. The model's predictions[5] are in reasonable agreement with the observed magnetization and the temperature and magnetic field dependence of the specific heat of $CeAl_3$. Unfortunately, since no single-crystal data exists, it was impossible to test the model's predictions about the anisotropy of both the magnetization and the field dependence the specific heat. To make a comparison with experiment it was necessary to assume that the experimental, polycrystalline sample had no preferred orientation and to average the model's predictions over all crystal orientations.

In the present work, a form of the RLM will be developed for treating $CeCu_6$. This system was chosen because of the availability of single-crystal data.[6,8,13] The specific heat and magnetization calculated from the model are in reasonable agreement with the single-crystal data.[6,13] Further, in qualitative agreement with experimental results[7] on polycrystalline samples, we find that the application of a magnetic field suppresses the γT term in the specific heat.

The following is a discussion of the model. Above 50 K the inverse susceptibility of both $CeAl_3$ and $CeCu_6$ is proportional to $T-\theta$. The θ values for both compounds are approximately equal to -50 K. These nonzero θ values probably arise because of crystalline electric field splittings. The crystal-field splittings[14] of $CeAl_3$ are 60 and 89 K. Because the θ values of the two compounds are similar, it is likely that they have comparable crystal-field splittings. Thus one can estimate that the crystal-field splittings of $CeCu_6$ are of order 50 K. Though this estimate is insufficient for performing a complete calculation[15] of the zero-field specific heat $C(T,0)$, we can calculate $C(T,0)$ for $T \ll 50$ K if we make two assumptions. We shall assume that the crystal-field ground state is a doublet and that $C(T,0)$ can be calculated from the RLM by treating $CeCu_6$ as a dilute Kondo system.

With these assumptions it follows that $C(T,0)$ is given by

$$C(T,0) = 2A\frac{\partial}{\partial T}\int_{-\infty}^{\infty} \varepsilon N(\varepsilon) f(\varepsilon/kT)\, d\varepsilon \ , \qquad (1)$$

where A is Avogadro's number, f is the Fermi function, and $N(\varepsilon) = \Delta/2\pi(\varepsilon^2+\Delta^2)$.

Now we consider the field dependence. By a thermodynamic Maxwell relation the specific heat $C(T,\mathbf{H})$ in a magnetic field $\mathbf{H}$ is given by

$$C(T,\mathbf{H}) = C(T,0) + T\frac{\partial^2}{\partial T^2}\int_0^{\mathbf{H}} \mathbf{M}(T,\mathbf{H}')\cdot d\mathbf{H}' \ . \qquad (2)$$

(Here in contrast to Ref. 5, $\mathbf{M}$ is the magnetization per mole.) Thus, to calculate $C(T,\mathbf{H})$, even at low temperatures, one must include the crystal-field anisotropy of the magnetization. Unfortunately, little is known about the crystal fields of $CeCu_6$. Despite this, some general statements follow just from the fact that the magnetism in $CeCu_6$ is associated with the $4f^1$ electronic configuration of Ce. For example, in the absence of magnetic ordering and many-body effects, there must be at least one crystal direction in which the low-temperature susceptibility

diverges as $1/T$. For $CeAl_3$, which is hexagonal, this direction is the c axis. For $CeCu_6$, which is orthorhombic, the susceptibility comes closest to following a $1/T$ temperature dependence in the [001] direction. Because of this, we will assume that the [001] direction is the "easy" axis, i.e., the direction in which the susceptibility would have had a $1/T$ temperature dependence in the absence of many-body effects or magnetic ordering. Presumably, since the system does not magnetically order, the absence of a $1/T$ temperature dependence of χ in the [001] direction can be attributed to many-body effects. It is known, for example, that the Kondo effect removes the $1/T$ temperature dependence of the susceptibility in dilute alloy Kondo systems.

For $CeAl_3$ the crystal-field splittings are larger than the Kondo energy.[5] Using the above estimate of 50 K for the crystal-field splittings of $CeCu_6$ and the fact that T_K of $CeCu_6$ is approximately equal[16] to 5 K, it appears that the crystal-field splittings in $CeCu_6$ are also much larger than the Kondo energy in $CeCu_6$. We shall assume that this is the case. We consider the effect of the crystal-field splittings, the larger energies, first. For kT less than the crystal-field splittings, there is a condensation into the crystal-field ground state. This condensation is the likely cause for the fact that the susceptibility in the [100] and [010] directions is smaller and less temperature dependent than it is in the [001] direction. Suppose a magnetic field is applied in the [100] or [010] direction. Since the crystal field has already removed most of the temperature dependence of the susceptibility, one might expect, to lowest order, that the susceptibility is unaffected by Kondo scattering. For this reason, we assume that we can neglect the Kondo scattering if **H** is applied in either the [100] or [010] direction. As discussed above, the susceptibility data indicate that [001] is the easy axis, i.e., the direction in which the susceptibility would diverge as $1/T$ in the absence of many-body effects. For **H** in the [001] direction, we assume that the Kondo scattering plays a dominant role and that we can apply the RLM to calculate the magnetization.

Since the magnetization in the [100] and [010] directions has a weak temperature dependence, it follows from Eq. (2) that the specific heat should be approximately independent of **H** in these directions. This result is in agreement with experiment.[6]

Now let us apply the model to calculate $C(T,\mathbf{H})$ and the magnetization in the [001] direction, M_{001}. For **H** in the [001] direction the RLM prediction is

$$M_{001}=A\mu\sum_{\sigma=\pm 1}\int_{-\infty}^{\infty}\sigma N(\varepsilon+\sigma\mu H)f(\varepsilon/kT)d\varepsilon\ . \quad (3)$$

Using Eqs. (1)–(3), one can compute both the magnetization and the field dependence of the specific heat for **H** in the [001] direction. In order to fit the zero-field specific heat we have taken $\Delta=5.4$ K. In order to fit the field dependence of C, we have taken $\mu=1.5\mu_B$. These values for Δ and μ are in approximate agreement with the experimental values of Walter, Wohlleben, and Fisk[16] of $T_K=4.6$ K and $\mu=1.6\mu_B$. The computed values of $C(T,H)$ for **H** in the [001] direction are compared in Fig. 1 with the experimental values of Amato *et al.*[6] One sees

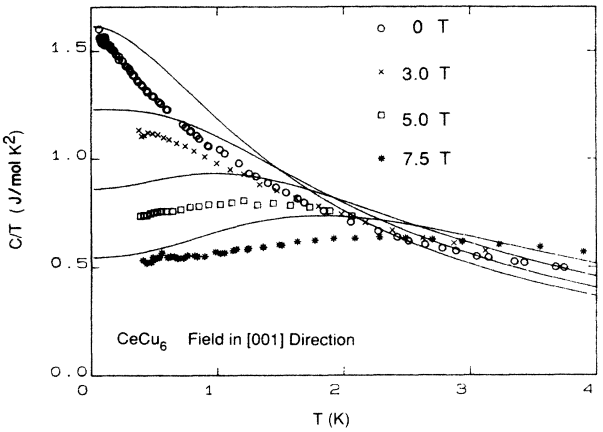

FIG. 1. Plot of C/T for $CeCu_6$ vs T for several fields in teslas in the [001] direction. The solid curves are computed from the model. The experimental data are from Ref. 6.

that the model provides a reasonably good fit to the data. The model does not provide as good a fit to the less precise data of Satoh *et al.*[8]

One of the features found in both the experimental and theoretical work on $CeAl_3$ was that $C(T,H)$ decreased approximately linearly as a function of H. This result was a little surprising since the model predicted that $C(T,\mathbf{H})$ should decrease as H^2 for **H** along the c axis. Presumably the difference in the field dependence is due to the averaging over orientations. Amato *et al.*[6] observed that the initial decrease in C/T of $CeCu_6$ for **H** in the [001] direction is quadratic in H. Figure 2 shows a comparison between the model's values and the experimental values[6] of C/T for **H** in the [001] direction as a function of H^2 at $T=0.39$ K. One sees that the calculated values are in good agreement with the experimental values.

Figure 3 shows a comparison of the experimental[13] and model's values of M_{001} at $T=1.3$ K. One sees that the model fails to fit the magnitude of M_{001}. If, however, one multiplies the model's values of M_{001} by a scale factor of 1.81, then the scaled values agree with the data for $H<10$ T.

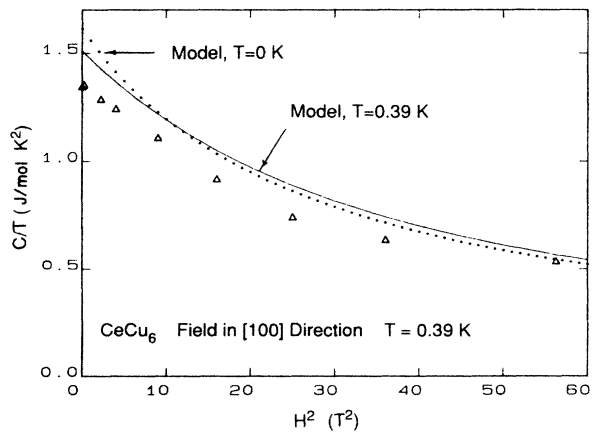

FIG. 2. Plot of C/T for $CeCu_6$ vs H^2 for **H** in the [001] direction and $T=0.39$ K. The curves are computed from the model. The experimental data are from Ref. 6.

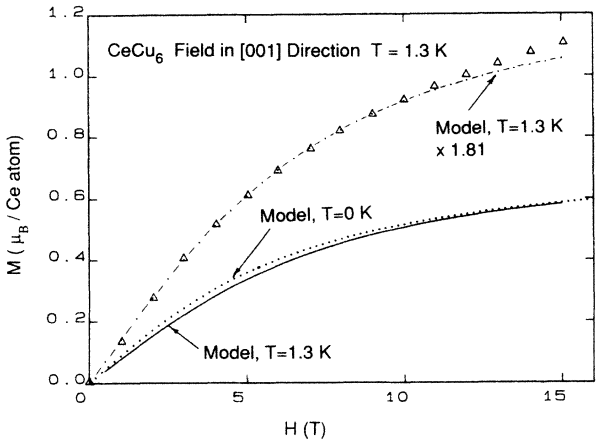

FIG. 3. Plot of the magnetization M of $CeCu_6$ vs H for **H** in the [001] direction and $T=1.3$ K. The curves are computed from the model. The top calculated curve has been multiplied by a scale factor of 1.81. The experimental data are from Ref. 13.

The model provides an excellent fit to most features of the data for $C(T,\mathbf{H})$ from Ref. 6 and the field dependence of M_{001}. For $CeAl_3$, the model fitted[5] $C(T,H)$ and both the field dependence and the magnitude of the magnetization up to 10 T. The parameters Δ and μ in the model for $CeCu_6$ are nearly equal to the values obtained from experiment. Satoh *et al.*[8] have applied two other forms of the RLM to interpret $C(T,H)$ of $CeCu_6$. They did not discuss crystal-field effects in their models nor have they incorporated the thermodynamic relationship [Eq. (2)]. In their first model, they assumed that the magnetic field broadens the resonance. This first model does not predict the observed peak in C/T observed by Amato *et al.*[6] for $H \geq 5$ T. Their second model, which involves band shifting, does predict a peak in C/T but does not predict other aspects of their data.

It is not surprising that the present model is not quantitatively correct. Here, and in Ref. 5, the Lorentzian was multiplied by the factor obtained in a calculation[17] of the zero-field susceptibility of dilute alloy Kondo systems. This normalization, however, leads to the result that $M \rightarrow \mu/2$ in the limit that $H \rightarrow \infty$. If one attempts to correct the high-field value of M by increasing the density of states by a factor of 2, then the fit to the specific-heat data is not very good. One knows[12] that the low-temperature, zero-field value for $C/T\chi$, where χ is susceptibility, calculated by this model [Eq. (6) below] is a factor of 2 smaller than that given by Wilson's[18] renormalization-group calculation. In part these deficiencies are due to the fact that vertex corrections[19] are not included in the RLM.

Despite these difficulties, the qualitative agreement between the model's predictions and experiment for **M** and $C(T,\mathbf{H})$ of $CuCu_6$ and $CeAl_3$ makes it worthwhile to discuss other qualitative features of the model. Consider using a general density-of-states function $N(\varepsilon)$ in Eqs. (1) and (3). By taking the $T \rightarrow 0$ limits of Eqs. (1)–(3), it can be shown that C/T and the susceptibility $\chi_{001} \equiv \partial M_{001}/\partial H$ are given by

$$C(T,H)/T \rightarrow \pi^2 A k^2 [N(+\mu H)+N(-\mu H)]/3 \ , \quad (4)$$

$$\chi_{001} \rightarrow A\mu^2 [N(+\mu H)+N(-\mu H)] \ . \quad (5)$$

Thus, in the low-temperature limit

$$C/T\chi_{001} \rightarrow \pi^2 k^2/3\mu^2 \quad (6)$$

independent of the magnitude of H. This suggests that, where Kondo scattering is dominant, the limit $T \rightarrow 0$ of $C/T\chi$, i.e., the Wilson ratio, may be independent of field and that both C/T and χ as a function of H may be proportional to the many-body density of states at the energy μH.

For the special case of the Lorentzian density of states we have employed in our calculation, the low-temperature limits of $C(T,H)/T$ and M_{001} are given by

$$C/T \rightarrow (\pi A k^2 \Delta/3)/[(\mu H)^2+\Delta^2] \ , \quad (7)$$

$$M_{001} \rightarrow (\mu A/\pi)\tan^{-1}(\mu H/\Delta) \ . \quad (8)$$

These low-temperature limits are shown in Figs. 2 and 3. One sees that the finite-temperature results are not very different from these low-temperature limits.

Note added in proof. Recent measurements [W. Joss, J. M. van Ruitenbeek, G. W. Crabtree, J. L. Tholence, A. P. J. van Deursen, and Z. Fisk, Phys. Rev. Lett. **59**, 1609 (1987)] show that the cyclotron mass of the Kondo system CeB_6 is strongly reduced by the application of a magnetic field. Since the cyclotron mass is related to γ, the model presented here may also be useful in interpreting the CeB_6 data.

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