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Volume effect at the metamagnetic transition in $CeRu_2Si_2$

A. Lacerda, A. de Visser,* L. Puech, P. Lejay, P. Haen, and J. Flouquet
Centre de Recherches sur les Très Basses Températures,
Centre National de la Recherche Scientifique, Boîte Postale 166 X, 38042 Grenoble-CEDEX, France

J. Voiron and F. J. Okhawa†
Laboratoire Louis-Néel, Centre National de la Recherche Scientifique, Boîte Postale 166 X, 38042 Grenoble-CEDEX, France
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Thermal-expansion (α) measurements performed in magnetic fields down to 1.3 K on the heavy-fermion compound $CeRu_2Si_2$ show that the temperature at which the extremum in α occurs has a deep minimum at the metamagnetic transition ($B^* = 7.8$ T). In agreement with scaling theory, the change of sign in α occurs for $B \simeq B^*$. A Kondo collapse model with ferromagnetic molecular fields partially explains the results. However, ferromagnetic fluctuations may play the major role in the field enhancement of the effective mass at B^*.

The understanding of the competition between the local fluctuations and the intersite coupling in heavy-fermion compounds is still an open problem.[1] An elegant way to try to elucidate this problem experimentally is to change the external variables such as pressure (P) or magnetic field (B). A sufficiently high pressure results in a situation where the $4f$ (or $5f$) electron recovers its full degeneracy and the intersite coupling is drastically reduced,[2] while a strong magnetic field will break up the intersite correlations. In the high-field limit, a narrow band of renormalized quasiparticles arises from the local fluctuations. In this spirit, extensive experimental studies have been performed on $CeRu_2Si_2$. This tetragonal compound can be characterized by a large coefficient of the linear term in the electronic specific heat[3-5] ($\gamma \sim 350$ mJ/mol K^2), an extraordinary large electronic Grüneisen parameter[6,7] ($\Gamma_{el} \sim 150$), i.e., a large pressure variation of the electronic parameters, and a metamagneticlike transition at a magnetic field $B^* \sim 7.8$ T applied along the tetragonal axis.[3,8] Inelastic neutron-scattering experiments[1,9] have shown unambiguously a collapse for $B > B^*$ of the antiferromagnetic correlations, which develop below 70 K in zero field. In magnetoresistivity experiments[8(a)] the metamagneticlike transition can be observed up to about the same temperature.

Motivated by this exemplary system we have performed thermal-expansion (α) measurements in applied magnetic fields, since α is extremely sensitive to the pressure and field dependence of the entropy, and probes as well the proximity of a pressure or magnetic instability. Our previous thermal-expansion measurements[7] in zero field revealed a huge broad low-temperature anomaly, centered at $T_m = 9$ K, that was attributed to the presence of magnetic correlations, in particular a competing interaction between the single-site Kondo effect and intersite coupling. An analysis in terms of a simple resonance-level model yielded a Kondo-like temperature $T_K = 19$ K. In this Rapid Communication we present measurements of the coefficients of linear thermal expansion, $\alpha = (1/L) \times (dL/dT)$, of a single-crystalline sample of $CeRu_2Si_2$, in strong magnetic fields ($B < 8.5$ T) applied along the tetragonal (c) axis. In order to obtain the volume expansion (α_v), α has been measured along ($\alpha_\parallel$) and perpendicular ($\alpha_\perp$) to the tetragonal axis, as $\alpha_v = \alpha_\parallel + 2\alpha_\perp$. Data were gathered using a sensitive three-terminal capacitance method. The sample was mounted in a parallel-plate capacitance cell, machined out of oxygen-free high-conductivity copper. Experimental details and sample preparation have been described elsewhere.[7]

For all applied fields we observed that $\alpha_\parallel \sim 3\alpha_\perp$. Here we will leave the anisotropy in the thermal-expansion coefficient out of consideration and concentrate in the following analysis on the volume effect. An overall picture of $\alpha_v(T)$ in magnetic fields of 0, 5, 6, 7.5, and 8 T is presented in Fig. 1. In zero field a broad anomaly is centered at $T_m = 9$ K. Since the low-temperature extrema in the specific heat[3] and magnetic susceptibility[6] are found at 11 and 10 K, respectively, these anomalies are likely attributed to the same physical phenomenon. In a magnetic field the anomaly in α_v rapidly shifts towards lower temperatures where it becomes very sharp and, surprisingly, changes sign near the metamagnetic transition. The field

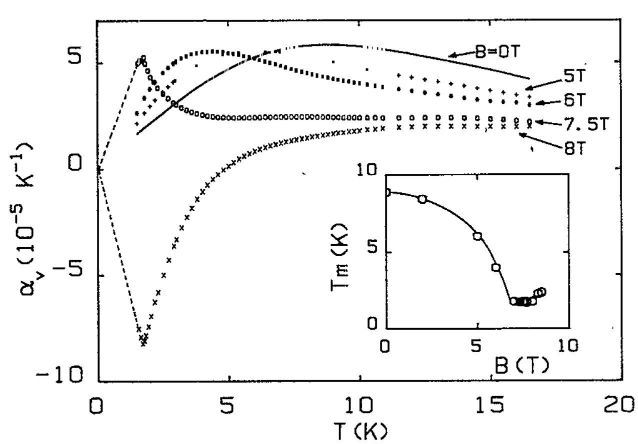

FIG. 1. Coefficient of volume expansion of $CeRu_2Si_2$ for magnetic fields as indicated. The inset shows the field dependence of the extremal temperature T_m.

dependence of T_m is shown in the inset of Fig. 1. Note that for $B>B^*$, T_m rises. $T_m(B)$ seems to mimic qualitatively the boundary of a crossover phase diagram, separating a highly correlated antiferromagnetic low-temperature phase and a polarized paramagnetic phase.

By using thermodynamic relations we can couple the linear low-temperatures terms in the thermal expansion, $\alpha_v=aT$, and the γ term of molar specific heat, $C_P=\gamma T$, to $a=-V_0^{-1}\partial\gamma/\partial P$. Here V_0 is the molar volume. In the vicinity of the instability, i.e., near the critical field B^*, α_v/T rapidly changes sign, implying a large variation of the pressure dependence of γ. Assuming $\alpha_v=aT$ below $T=1.3$ K, we obtain the extreme values $a^{\max}=45.0\times10^{-6}$ K^{-2} at 7.30 T and $a^{\min}=-50.8\times10^{-6}$ K^{-2} at 8.00 T (inset of Fig. 2).

We will use a scaling ansatz (SA), extensively verified in earlier work,[6,8(b),10] to relate the slope $(\alpha_v/T)_{T\to0}$ analytically to the field dependence of γ at zero pressure. Assuming that only one single temperature and field scale with pressure (and volume) exists, the entropy with SA can be written as[10]

$$S(T,B,P)=k_B s\left[\frac{T}{T_S(P)},\frac{B}{B_S(P)}\right], \tag{1}$$

where k_B is the Boltzmann constant. In this approach the relative pressure dependences of T_S and B_S are given by

$$\Omega_T=-\frac{1}{T_S}\left(\frac{\partial T_S}{\partial P}\right)=\kappa\Gamma_T,$$
$$\Omega_B=-\frac{1}{B_S}\left(\frac{\partial B_S}{\partial P}\right)=\kappa\Gamma_B, \tag{2}$$

where κ is the isothermal compressibility ($\kappa=0.95$ Mbar^{-1}),[11] $\Gamma_T=-\partial\ln T_S/\partial\ln V$ and $\Gamma_B=-\partial\ln B_S/\partial\ln V$ are the usual thermal and magnetic Grüneisen parameters. Ω_T has been determined from a combination of specific-heat and thermal-expansion measurements,[7]

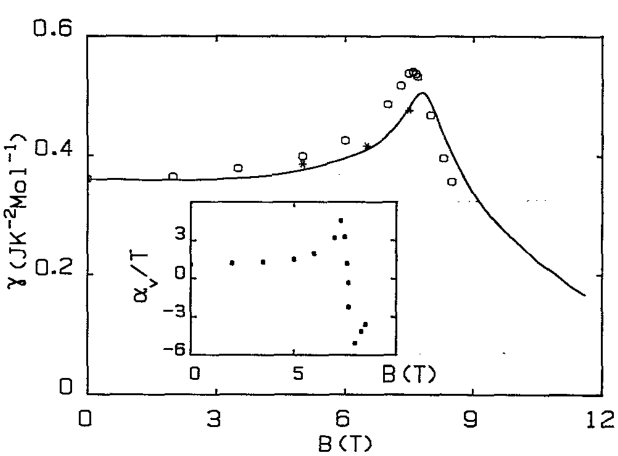

FIG. 2. Field dependence of the coefficient γ of the linear term in the specific heat: (○) γ_{SA} calculated using scaling ansatz [Eq. (3)]; (*) as measured (Ref. 4), and γ_M calculated using Eq. (4). The inset shows the linear extrapolation of α_v: $(\alpha_v/T)_{T\to0}$ (in units of 10^{-5}/K^2) as a function of magnetic fields.

while Ω_B has been determined from magnetostriction data,[7,8(b)] field dependence of the sound velocity,[12] or pressure dependence of the magnetization.[6] For $CeRu_2Si_2$ we have $\Omega_T\simeq\Omega_B\simeq150$ Mbar$^{-1}=\Omega$. From Eqs. (1) and (2) it follows that

$$a=\frac{\Omega}{V_0}\left(\gamma+B\frac{\partial\gamma}{\partial B}\right). \tag{3}$$

Upon resolving the differential equation the field dependence of γ_{SA} is shown in Fig. 2. γ_{SA} is predicted to increase by as much as 50% at B^*. In Fig. 2 we also compare γ_{SA} with the experimental determination γ_C obtained from specific-heat measurements in the field.[4] The remaining discrepancy between the calculated and measured γ values may be due to the difficulties in extrapolating α_v and C_p in the low-temperature limit. From resistivity measurements in applied magnetic fields[8(a)] a 30% increase of γ at B^* has been conjectured, assuming a scaling via the coefficient of the term linear in temperature. Chemical alloying has led to systems in which both sides of B^* can be studied more easily. For instance, in $Ce_{0.95}La_{0.05}Ru_2Si_2$ B^* has dropped to ~5 T. Specific-heat measurements[4] on this compound show a 30% increase of γ at B^*.

Another way to derive the relative field dependence of $\gamma(B)$ is to perform low-temperature magnetization measurements. Such complementary magnetization studies have been performed (on the same single crystal as used for the data in Fig. 1) in a Bitter magnet ($B<15$ T) at the Service National des Champs Intenses-Grenoble, France for 1.3 K $<T<2.0$ K $<T_m(B^*)$. Assuming a T^2 low-temperature limit of the magnetization (at fixed fields), from thermodynamics it follows that

$$\frac{\partial M}{\partial T^2}=\frac{1}{2}\frac{\partial\gamma}{\partial B}, \tag{4}$$

and thus a new independent determination γ_M of $\gamma(B)$ is obtained. In the high-field limit (15 T) γ_M has dropped to 130 mJ/mol K^2. Below B^*, the agreement between γ_{SA} and γ_M is excellent. Within the different experimental accuracies (2%), the positions of the maxima in γ_{SA} and γ_M coincide. The difference between γ_{SA} and γ_M can be reduced by choosing slightly different Grüneisen parameters Γ_B and Γ_T. Very low-temperature experiments are now needed to discuss quantitatively the insufficiency of a SA description. Furthermore, the difference and sharpness in γ for $B\sim B^*$ between alloys and the perfect-lattice[4] data will necessitate improvement of the quality of the material.

In contrast to the weak enhancement of γ, a huge field variation of the differential susceptibility χ occurs at B^*; $\chi(T)$ at B^* follows a Curie-Weiss law with a Curie constant $C=0.46$ emu K/mol and a weak θ value ($\theta\lesssim0.2$ K). As already suggested by the deep minimum of $T_m(\simeq2$ K) at B^*, the low-temperature regime has not been truly attained yet for $B=B^*$. Figure 3 shows the field dependence of the ratio $\chi(1.3$ K$)/\gamma_M$. In a rigid-band model the system is described by a local density of states at the Fermi level, and the ratio χ/γ cannot increase with B. Here this ratio attains a sharp maximum at B^*. Furthermore, it is asymmetric with respect to B^* (inset in Fig. 3). The

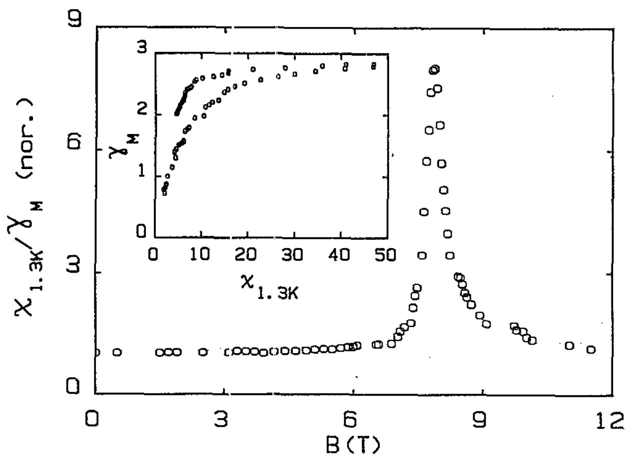

FIG. 3. Field dependence of the ratio χ (1.3 K)/γ_M normalized to 1 at $B=0$. The inset shows γ_M vs χ (1.3 K) in arbitrary units.

maximum is associated with a collapse of the antiferromagnetic correlations and the entrance into a regime where only local fluctuations play a role. As the field further increases above B^*, the Zeeman effect will become dominant. Static magnetic measurements show a large increase of the uniform susceptibility at B^*. The mass enhancement at B^* arises probably from the optimum ferromagnetic instability at this field. This corresponds to the narrowness of the dynamical susceptibility for vanishing wave vector (q) at $B=B^*$. The surprising result is that, despite a large variation in χ/γ, these thermodynamic quantities can be described in a first approximation by only one scaling parameter. The basic reason for this might be that the local fluctuations and intersite coupling have equal weight at $B=0$. The local fluctuations may prevent the collapse of the linewidth of the dynamical susceptibility at $B=B^*$ and thus govern at first approximation the T and B responses (i.e., $\Omega_T \sim \Omega_B$). In the theory of itinerant magnetism[13] the collapse of the energy linewidth for $q \rightarrow 0$ is the driving mechanism for the divergence of the mass enhancement at the ferromagnetic instability.

A first attempt to model the data can be made by introducing the effect of ferromagnetic coupling in the Kondo-lattice collapse model.[14] At $T=0$ K the ground-state energy per unit cell is the sum of a f-electron contribution $E_f(V)$ plus a lattice contribution $E_{\text{lat}}(V)$. In the presence of a ferromagnetic molecular field $E_f(V)$ is replaced by an effective energy:

$$E_f^{\text{eff}} \sim -[T_K^2 + (\tfrac{1}{2} g\mu_B H_{\text{eff}})^2]^{1/2}, \qquad (5)$$

where g is the Landé factor and μ_B the Bohr magneton. The effective field (H_{eff}) is the linear combination of B and the molecular field due to the ferromagnetic exchange J. The volume equilibrium is governed by the huge volume dependence of the Kondo temperature T_K:

$$T_K(x) = T_0 e^{-x}, \qquad (6)$$

where $x = \Gamma(V-V_0)/V_0$. The large Grüneisen parameter enhances the volume effect by 2 orders of magnitude. When J and T_K are comparable a pronounced maximum appears in χ at B^*. The metamagnetic transition occurs for a critical value of the magnetization $M \simeq 0.5$, whatever the strength of J is, as also has been found experimentally.[6,8(b)] However, in this simple model no universal curve of M in B/B^* is found, the enhancement of γ at B^* is only 3% and the maximum of γ coincides with B^* only for large J. This implies that the interplay of ferromagnetic exchange and Kondo effect must be treated beyond the molecular-field approximation.

In conclusion, thermal-expansion experiments provide a very powerful technique for the study of systems near a field instability (for instance, the metamagnetic transition in $CeRu_2Si_2$). In heavy-fermion $CeRu_2Si_2$ the characteristic temperature (T_m) decreases drastically for $B \rightarrow B^*$ and can be extrapolated linearly to zero at $B=B^*$. This suggests a magnetic instability for $B=B^*$ at 0 K. The linear field dependence of T_m near B^* may reflect the importance of quantum fluctuations. The relatively weak mass enhancement at B^* when compared to the large increase in the differential susceptibility, i.e., the large value of the ratio χ/γ at B^*, shows that a large part of the mass enhancement is due to local fluctuations.

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*Also at Natuurkundig Laboratorium, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands.

†Permanent address: Physics Department, Hokkaido University, Sapporo 060, Japan.

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