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Volume effect at the metamagnetic transition in CeRu_2Si_2

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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 $B \approx B^*$. 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 heat³⁻⁵ ($\gamma \sim 350$ mJ/molK²), 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 measurements⁷ 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 (α_{\parallel}) and perpendicular (α_{\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 heat³ and magnetic susceptibility⁶ 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

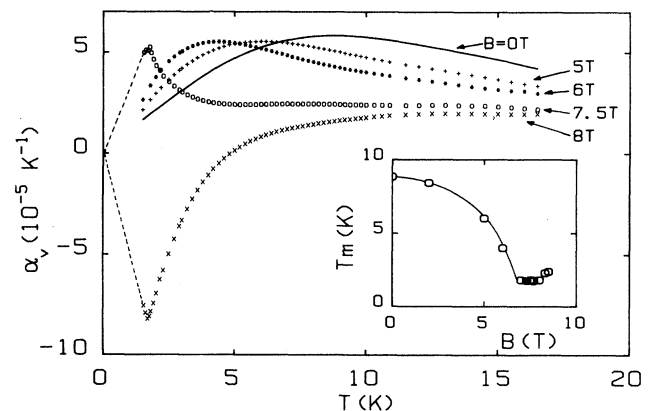


FIG. 1. Coefficient of volume expansion of CeRu_2Si_2 for magnetic fields as indicated. The inset shows the field dependence of the extremal temperature T_m .

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-temperature 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 $a^{\max} = 45.0 \times 10^{-6} \text{ K}^{-2}$ at 7.30 T and $a^{\min} = -50.8 \times 10^{-6} \text{ K}^{-2}$ 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 $(\alpha_v/T)_{T \rightarrow 0}$ 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 as¹⁰

$$S(T, B, P) = k_B S \left(\frac{T}{T_S(P)}, \frac{B}{B_S(P)} \right), \quad (1)$$

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

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

$$\Omega_B = -\frac{1}{B_S} \left(\frac{\partial B_S}{\partial P} \right) = \kappa \Gamma_B,$$

where κ is the isothermal compressibility ($\kappa = 0.95 \text{ Mbar}^{-1}$),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

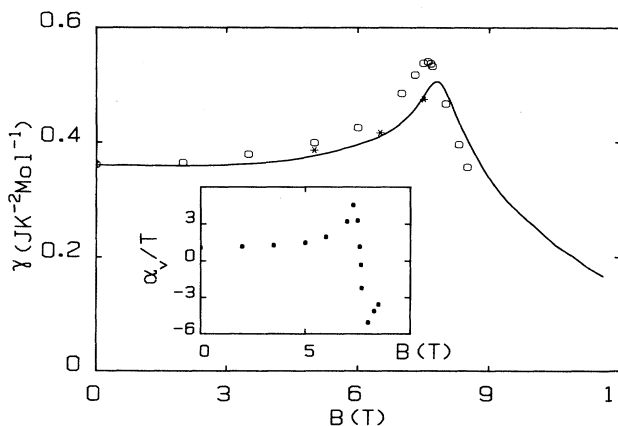


FIG. 2. Field dependence of the coefficient γ of the linear term in the specific heat: (O) γ_{SA} calculated using scaling ansatz [Eq. (3)]; (*) as measured (Ref. 4), and γ_M calculated using Eq. (4). The inset shows the linear extrapolation of α_v : $(\alpha_v/T)_{T \rightarrow 0}$ (in units of $10^{-5}/\text{K}^2$) 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 $\text{CeRu}_2\text{-Si}_2$ we have $\Omega_T \approx \Omega_B \approx 150 \text{ Mbar}^{-1} = \Omega$. From Eqs. (1) and (2) it follows that

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

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 $\text{Ce}_{0.95}\text{La}_{0.05}\text{Ru}_2\text{Si}_2$ B^* has dropped to ~ 5 T. Specific-heat measurements⁴ 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 $1.3 \text{ K} < T < 2.0 \text{ K} < T_m(B^*)$. Assuming a T^2 low-temperature limit of the magnetization (at fixed fields), from thermodynamics it follows that

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

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-lattice⁴ 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 $C = 0.46 \text{ emu K/mol}$ and a weak θ value ($\theta \leq 0.2 \text{ K}$). As already suggested by the deep minimum of T_m ($\approx 2 \text{ K}$) at B^* , the low-temperature regime has not been truly attained yet for $B = B^*$. Figure 3 shows the field dependence of the ratio $\chi(1.3 \text{ K})/\gamma_M$. 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

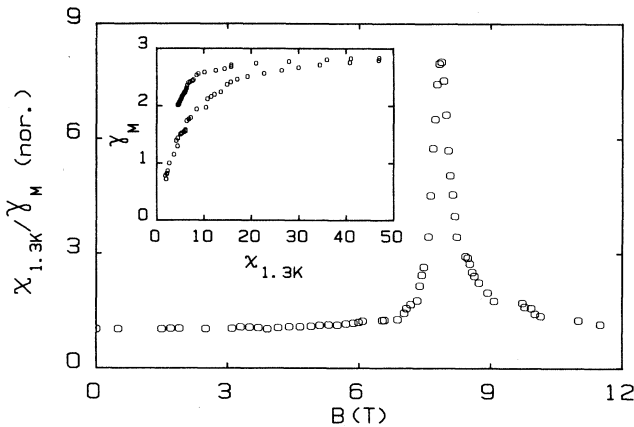


FIG. 3. Field dependence of the ratio $\chi(1.3\text{ K})/\gamma_M$ normalized to 1 at $B=0$. The inset shows γ_M vs $\chi(1.3\text{ K})$ in arbitrary units.

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 magnetism¹³ 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 en-

ergy 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:

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

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 :

$$T_K(x) = T_{0e}^{-x}, \quad (6)$$

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 $M \approx 0.5$, 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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