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Phonon effects at the metamagnetic transition in $CeRu_2Si_2$

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We present experiments and model calculations concerning the metamagnetic transition in $CeRu_2Si_2$. Our sound velocity, sound attenuation (down to 60 mK), and ac magnetic susceptibility measurements (down to 20 mK) all show strong anomalies near the critical magnetic field of this transition. We can account for this behavior using scaling arguments and a more microscopic model based on the Kondo volume collapse.

INTRODUCTION

The normal state of heavy-fermion systems has been studied using many different experimental methods. An interesting question is how this state can be broken up by applying a pressure or a magnetic field. In the latter case the breaking up of the heavy-fermion ground state manifests itself as a metamagnetic transition, meaning a peak in the magnetic susceptibility. This has been observed in $CeRu_2Si_2$ at $B_c=7.8$ T, in UPt_3 at $B_c=21$ T, and URu_2Si_2 at $B_c=36$ T. For a review on $CeRu_2Si_2$, see Ref. 1.

The metamagnetic transition in $CeRu_2Si_2$ has been further characterized by magnetization,[2] magnetoresistivity,[2] magnetostriction, thermal-expansion,[3] and sound-wave experiments.[4] Here we show that the susceptibility peak is very large but finite down to 20 mK. This magnetic anomaly is accompanied by a very large longitudinal elastic constant softening of up to 50% at 60 mK.

The metamagnetic transition is due to an at least partial breakup of the heavy-fermion ground state. Mechanisms proposing an explanation include crystal electric-field level crossing in an effective internal field, magnetic interactions giving rise to renormalized $M(B)$ curves, or the so-called Kondo volume collapse, which is based on the large volume dependence of the Kondo temperature T_K due to large Grüneisen parameters. Expanding the volume of the unit cell, e.g., by applying a magnetic field, causes a narrowing of the quasiparticle band at the Fermi energy. For $B>B_c$ the heavy-fermion ground state is going to be destroyed, leading to either naked $4f$ moments or a mixed-valence system.

The Kondo volume-collapse mechanism as developed in Ref. 5 offers a qualitively correct picture of the metamagnetic transition in $CeRu_2Si_2$ as far as magnetostriction, magnetization, and magnetic susceptibility are concerned, i.e., it predicts a B_c characterized by a peak in the susceptibility and magnetostriction coefficient. However, although the author realizes that at the same time the lattice must be near a mechanical instability, in his version of the model there is no direct connection between the susceptibility peak and the softening of the elastic constant. In this paper we propose an extension of the Kondo volume-collapse model which accounts for both the susceptibility peak and the strong sound-wave softening at the metamagnetic transition. We start presenting our experimental results on magnetic susceptibility and sound-wave propagation at very low temperatures. We discuss these results at first from the viewpoint of a scaling approach[6] and afterwards within the framework of the modified Kondo volume-collapse model.

EXPERIMENTS ON $CeRu_2Si_2$

Results on sound-velocity changes and ultrasonic attenuation at $T=1.3$ and 4.2 K have been given before Ref. 4. In Fig. 1 we show elastic constant results for the longitudinal c_{11} (sound-wave vector **q**||**a** axis) and c_{33} (**q**||**c** axis) modes as a function of magnetic field **B** applied parallel to the tetragonal c axis and perpendicular to it. It is found that the strong sound-wave softening for both modes occurs only for **B**||**c**. These results, together with

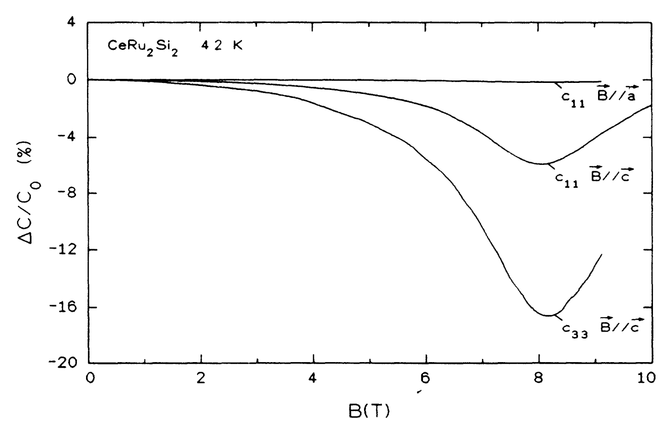

FIG. 1. Elastic constant change $\Delta c/c_0$ as a function of magnetic field B for the longitudinal modes c_{11} (with **B**||**c** and **B**||**a**) and c_{33} (with **B**||**c**) at 4.2 K.

the weak-field dependence of the transverse modes (which are volume conserving), are a beautiful demonstration of the Grüneisen parameter coupling.[6] The Grüneisen parameter coupling is a special case of the deformation potential coupling describing the strong volume dependence of the characteristic energy scales of the heavy-fermion state. Departing from a scaling ansatz for the appropriate thermodynamic potential as a function of T/T_K and B/B_c, one can derive the following results for the change of the longitudinal elastic constants c_{11} and c_{33} (noted collectively with Δc_L), see Refs. 6 and 7:

$$\Delta c_L = -\Omega_T^2 k_B T_K \qquad (1a)$$

total effect going from $T=T_K$ to $T=0$ in zero field.

$$\Delta c_L = -\Omega_B^2 B^2 \chi_m(B) \qquad (1b)$$

in an external magnetic field B.

$\Omega_B = -(1/B_c)(\delta B_c/\delta\epsilon_L)$, $\Omega_T = -(1/T_K)(\delta T_K/\delta\epsilon_L)$ are the Grüneisen parameters, ϵ_L is the longitudinal elastic strain (ϵ_{xx} and ϵ_{zz} for c_{11} and c_{33}, respectively), T_K is the Kondo temperature and $\chi_m = \delta M/\delta B$, ($M$ is the magnetization) is the magnetic susceptibility. In Eq. (1a) we have substituted $k_B T_K$ for the internal energy U. In Refs. 6 and 7 it is shown that $U = \int C dT$ (where C is the specific heat) is a better approximation. However, for the sake of comparison with our equations below we use Eq. (1a) in its present form.

In principle Ω_B and Ω_T are different, but theoretically it has been shown[8] that for heavy-fermion systems $\Omega_B = \Omega_T$. Also experimentally the two have the same numerical value, and for the purpose of this paper we consider them equal. To account for the observed strong anisotropy, we do use different uniaxial Grüneisen parameters Ω_a and Ω_c for the c_{11} and c_{33}, modes, respectively. As starting value c_0 of an elastic constant we take its value without Kondo contributions, in practice the value at $T \simeq 25$ K and $B=0$ for $CeRu_2Si_2$. Within this formalism one can very effectively describe and interrelate not only elastic constants and magnetic susceptibility but also thermal expansion, specific heat and magnetostriction.[3,7]

In Fig. 2 we show magnetic susceptibility and elastic constant of $CeRu_2Si_2$ at 160 mK versus external magnetic field B. In all cases B is parallel to the c axis, which is the configuration giving the largest effects for susceptibility and for sound-wave softening as well. Figure 3 gives results for the peak height of the magnetic susceptibility at $B=B_c$ and the corresponding minimum elastic constants c_{11} and c_{33} also at $B=B_c$ as a function of temperature. It is seen that all curves flatten towards lower temperature, indicating that neither the susceptibility χ_m nor the softening of c_{11} or c_{33} increase indefinitely. But note that the total amount of softening for c_{33} from $B=0$ to B_c reaches 50% at 60 mK! Having observed such a pronounced metamagnetic transition and this strong sound-wave softening one has to look into the possibility of at least a precursor of a phase transition. Nevertheless Eq. (1) can account for this result very nicely with only slightly temperature-dependent Grüneisen parameters $\Omega_a \simeq 110$, $\Omega_c \simeq 140$ down to the lowest temperatures measured. This means that a transition to, e.g., a fluctuating valence state (which should have a much smaller Grüneisen parameter) is not likely.

The temperature dependence of the extrema of χ_m as well as of c_{11} and c_{33} at B_c can be phenomenologically fitted below 10 K to a temperature dependence proportional to $\log_{10}(T+2\text{ K}/10\text{ K})$. The 10 K in the denominator indicates the temperature limit above which the metamagnetic transition becomes very weak. The physical meaning of the 2 K is unclear at the moment: From absolute sound-velocity measurements we noticed that 2

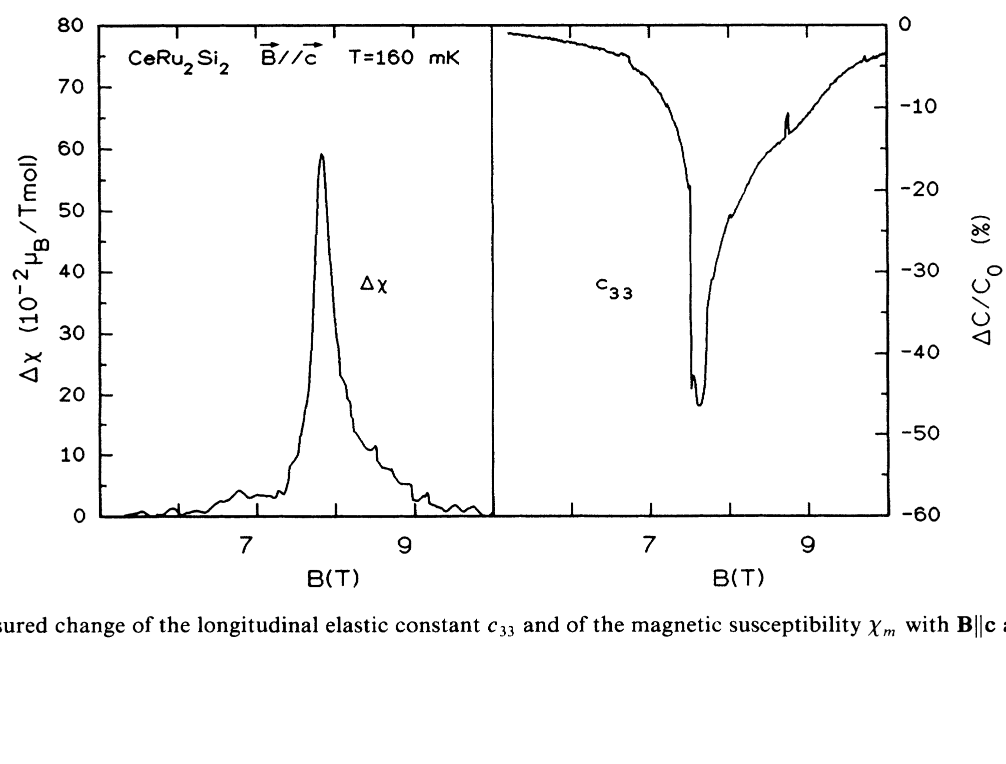

FIG. 2. Measured change of the longitudinal elastic constant c_{33} and of the magnetic susceptibility χ_m with $\mathbf{B}\|\mathbf{c}$ at $T=160$ mK vs B.

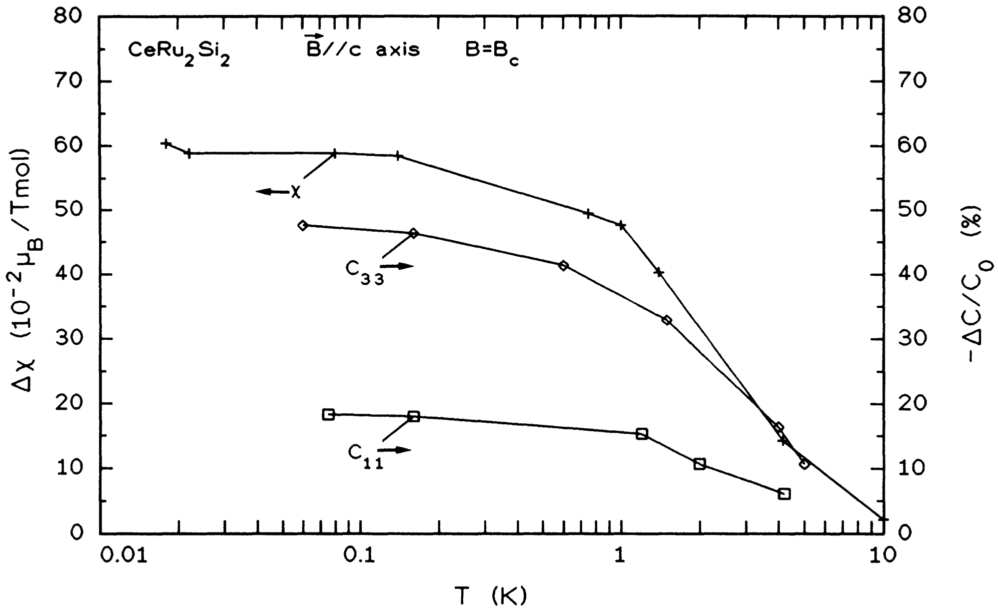

FIG. 3. Temperature dependence of the peak height of χ_m and the maximum softening of c_{33} and c_{11} at B_c and $\mathbf{B}\|\mathbf{c}$.

K is the temperature below which the softening of c_{33} near B_c becomes so strong that c_{33} crosses the slower transverse mode c_{44}. This causes additional structure in the field dependences (near B_c) of both c_{33} and c_{44} (not shown). The c_{44} anomaly has been discussed more fully elsewhere.[7,9] However, the crossing of c_{33} and c_{44} does not seem to be the direct reason for the flattening of c_{33} towards lower temperatures. This must be concluded from the similar flattening behavior of c_{11} (see Fig. 3), c_{11} being far from crossing any mode at any field value.

Thermal expansion measurements[10] in fields exhibit an extremum in the thermal expansion coefficient as a function of temperature and a sign reversal of this extremum at B_c. This effect is difficult to relate to our measurements and we do not have any explanation yet. For the three samples we used for our experiments we observed the same flattening displayed in Fig. 3.

Additionally to the sound-wave softening around B_c one observes a pronounced peak in the ultrasonic attenuation, which can be described with a Landau-Khalatnikov mechanism.[4] This enables one to obtain a relaxation time τ, which seems to be a spin-fluctuation relaxation time as it is proportional to the magnetic susceptibility and very slow (of the order of 10^{-9} sec) compared to usual electronic relaxation rates.

THEORETICAL CONSIDERATIONS

Our results for the metamagnetic transition in $CeRu_2Si_2$ indicate that the softening of the longitudinal modes and the susceptibility peak near B_c are closely intertwined and therefore a theory for this transition should account for both of them. In the following we will discuss the proposed mechanisms for the metamagnetic transition, and we will show that it is possible to construct a Kondo volume-collapse model, which fulfills the above-mentioned condition.

The crossing of the crystal-field levels in an internal field can be excluded because the smallest crystal-field splitting is of the order of 200 K.[7] The internal field necessary to make a crossing at $B_c = 8$ T had to be of the order of 50 T or more, enough to induce a ferromagnetic phase transition.

Another mechanism based on magnetic interactions[11] cannot be checked very easily because the coupling constants are not known. In addition all the experiments involving strong volume dependences cannot be explained with this mechanism without further assumptions.

Recently a model for the metamagnetic transition in heavy-fermion compounds based on the so-called Kondo volume-collapse mechanism[12] was proposed.[5] The author shows that the relevant part of the effective f-electron energy at zero temperature can be written as

$$E_f = -[(k_B T_K)^2 + (\mu_B B_{\text{eff}})^2]^{1/2} , \tag{2}$$

where k_B is the Boltzmann constant, T_K is the Kondo temperature, μ_B is the Bohr magneton and $B_{\text{eff}} = B + JM/\mu_B^2$. M is the magnetization, J is a measure for the strength of a postulated ferromagnetic interaction which is indispensable in the version of Ref. 5 in order to produce an appreciable susceptibility peak. The g factor of the electrons we assume to have the value 2. The volume dependence of E_f comes mainly from the volume dependence of T_K, which can be expressed as $T_K = T_0 e^{-\Omega\epsilon}$, since $\Omega = -\delta \ln(T_K)/\delta\epsilon$.

This form for E_f describes correctly the screening of the magnetic moments in the Kondo state: The magnetization of $M = -\delta E_f/\delta B$ is zero for $T = 0$ and $B = 0$, and regains its full value of $1\mu_B$ per f atom only at high fields $(\mu_B B > k_B T_K)$. The effect of finite temperatures is not included, but raising the temperature should likewise undo the quenching of the magnetic moments. However, this form of E_f alone does not give any pronounced peak in the magnetic susceptibility nor in the softening of the elastic constants.

To remedy this, one has to consider the equilibrium of forces inside the crystal. In Ref. 5 this is done by using the simple algebraic relations between the first derivatives of E_f with respect to ϵ and B. Assuming an elastic energy, which contains only linear and quadratic terms in ϵ, the author of Ref. 5 reaches the conclusion that the elastic constant which figures in his equilibrium equation is the background elastic constant c_0, independent of deformation or magnetic field. However, taking the strong softening of the elastic constant at face value, one can very well argue that higher (>2) order terms in the elastic energy must be taken into account. These higher-order contributions, as well as the linear one which is responsible for the Kondo volume collapse originate from the $T_0 e^{-\Omega\epsilon}$ term in E_f.

To circumvent this problem, we approach the effect from the other side: First we calculate the elastic constants, and then we use Hookes Law to evaluate the forces.

The change of the elastic constant due to E_f is $\Delta c_L = \delta^2 E_f / \delta^2 \epsilon_L$. The second derivative of E_f with respect to the effective field B_{eff} we call the effective susceptibility: $\chi_{\text{eff}} = \delta M / \delta B_{\text{eff}}$. Comparing first and second differential derivatives of Eq. (2) with respect to ϵ and B_{eff} one can express the change of the longitudinal elastic constants due to E_f as

$$\Delta c_L = -\Omega^2[(k_B/\mu_B)^2 T_K^2 + 2B_{\text{eff}}^2]\chi_{\text{eff}} . \quad (3)$$

The total elastic constant is again $c_L = c_0 + \Delta c_L$. For $B=0$, Eq. (3) reproduces the result of Eq. (1a). The field-dependent part is very similar to Eq. (1b), which altogether is quite satisfying.

The first derivative of E_f with respect to the deformation, $\delta E_f/\delta\epsilon$, represents the internal force in the crystal responsible for the large thermal expansion and magnetostriction of $CeRu_2Si_2$. This force is balanced by the elastic forces of the lattice: $-\delta E_f/\delta\epsilon_L = \int c_L d\epsilon_L$. We have to use the integral form of Hookes law, because $c_L = c_0 + \Delta c_L$ depends on ϵ. Using this formula with Δc_L given either by Eq. (1a) or Eq. (3) we can calculate the total effect in the thermal expansion between $T=0$ K and $T=T_K$ in zero field as $c_0\epsilon_0 = \Omega k_B T_0$. Since for the moment we are interested in the field-induced reversal of this Kondo volume collapse, we take ϵ_0 as our new origin and write the equilibrium condition as follows:

$$-\frac{\delta E_f}{\delta \epsilon_L} = c_0(\epsilon_0 - \epsilon) + \int_{\epsilon_0}^{\epsilon} (\Delta c_L - \Delta c_{L(B=0)}) d\epsilon \quad (4)$$

with Δc_L given by Eq. (3).

Using the equilibrium condition one gets equations for the magnetization M and external magnetic field B:

$$M = \mu_B \left[1 - e^{2\Omega\epsilon} \left[\frac{p}{k_B T_0 \Omega} \right]^2 \right]^{1/2} , \quad (5)$$

$$B = M \frac{k_B T_0}{\mu_B^2} \left[\frac{e^{-\Omega\epsilon}}{[1-(M/\mu_B)^2]^{1/2}} - j \right] , \quad (6)$$

where p is the right-hand side of the equilibrium condition Eq. (4),

$$p = c_0(\epsilon_0 - \epsilon) + \int_{\epsilon_0}^{\epsilon} [\Delta c_L - \Delta c_L(B=0)] d\epsilon$$

and $j = J/k_B T_0$ is the renormalized ferromagnetic exchange constant. Equation (6) incorporates the relation between the externally applied field B and the effective field B_{eff} introduced below Eq. (2), $B = B_{\text{eff}} - JM/\mu_B^2$.

If one puts $p = c_0(\epsilon_0 - \epsilon)$, Eqs. (5) and (6) above are equivalent to Eqs. (1) and (2) of Ref. 5. A numerical solution for M and ϵ as functions of B from these equations gives the salient features of the effect, as shown in Ref. 5: a step in the magnetization at B_c and maxima in magnetic susceptibility and magnetostriction. However, some of the parameters that are needed to fit experimental values of the magnetization and of B_c are quite different from accepted values for $CeRu_2Si_2$, notably $T_K = 50$ K instead of $\simeq 20$ K, $\Omega_c = 500$ instead of $\simeq 140$ and a relatively high j of 0.7. The main objection as seen from our side, however, is that the elastic constant is assumed constant.

Now we remove this constraint by taking Eq. (3) to calculate c_L. Using Eqs. (3)–(6) one can simultaneously calculate M, χ_m, ϵ, and c_L as a function of B. In Fig. 4 we give results of such a calculation for $\chi_m(B)$ and $c_{33}(B)$. The parameters used in this calculation are $T_0 = T_K(\epsilon=0) = 20$ K, $\Omega_c = 150$, $j = 0.38$, and c_0 (for the c_{33} mode) $= 1.2 \times 10^{12}$ erg/cm^3. These values are much closer to generally accepted ones for $CeRu_2Si_2$. It should be noted that so far neither ferromagnetic short-range nor long-range ($j=1$) correlations have been found from inelastic neutron scattering experiments in this compound.[13] The model as shown in Fig. 4 now produces both a maximum in the susceptibility and a minimum in the elastic constant of the right magnitude. Moreover, we obtain a B_c of 7.8 T as observed with more realistic

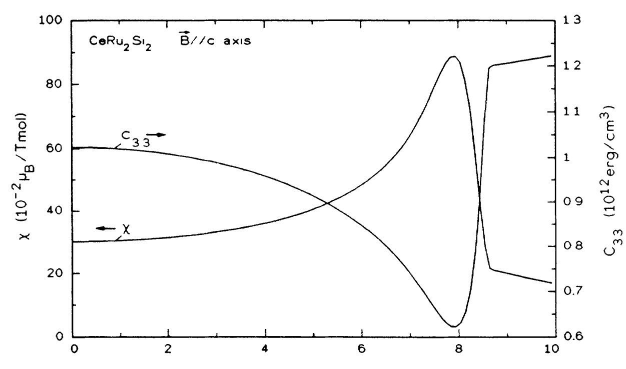

FIG. 4. Elastic constant c_{33} and magnetic susceptibility χ_m for $T=0$ K as a function of B (with $\mathbf{B}\|\mathbf{c}$) calculated from Eqs. (3)–(6). $T_0 = T_K(\epsilon=0) = 20$ K, $\Omega_c = 150$, and $j = 0.38$.

values for T_K, Ω, and j. Even with $j=0$ (all other parameters unchanged), one gets strong softening of the elastic constant and a peak in the susceptibility at a B_c of 11 T. This means that in our model the driving force for the metamagnetic instability is the large Grüneisen parameter coupling leading to the strong sound-wave softening. An experimental feature not well reproduced by the model is the width of the susceptibility and elastic constant anomalies (see Figs. 2 and 4). To improve this, one probably needs an expression for the electronic free energy which goes beyond the mean-field result of Eq. (2).

In conclusion, we have presented data for the magnetic susceptibility χ_m and of the longitudinal elastic constants c_{11} and c_{33} for fields up to 10 T and temperatures down to 20 mK. We have shown that both quantities exhibit large anomalies at the metamagnetic transition $B_c \simeq 8$ T. We can describe these results satisfactorily with a scaling Ansatz, and in more detail with a Kondo volume-collapse model.

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