Novel Magnetoacoustic Effects in Heavy-Fermion Systems in High Magnetic Fields

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We observed a strong softening of longitudinal acoustic modes in UPt_3 and $CeRu_2Si_2$ in high magnetic fields where the magnetic susceptibility exhibits an anomaly. The two phenomena can be correlated with a large electronic Grüneisen parameter. Some shear modes exhibit oscillations in the same high-field and low-temperature range.

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The anomalous electronic properties and the superconductivity of heavy-fermion metals are not yet understood and still of great current interest.[1] Probing of the heavy-fermion state in a different way than with conventional experiments is therefore of considerable value. In this Letter we present a high-magnetic-field (up to 23 T) investigation of sound velocity and ultrasonic attenuation for two heavy-fermion materials: UPt_3 and $CeRu_2Si_2$. These compounds show a positive curvature in the magnetization curves M versus B up to an inflection point leading to maxima of $\chi_m = \partial M/\partial B$ at $B_c \sim 20$ and 8 T, respectively. This maximum occurs for B applied in the hexagonal plane for UPt_3 and along the tetragonal c axis for $CeRu_2Si_2$[2,3] (see Table I). Since large variations of the characteristic parameters occur with applied field and pressure (Franse and co-workers[2,4] for UPt_3 and Flouquet and co-workers[3,5] for $CeRu_2Si_2$) a huge field dependence can also be expected for the sound velocity.

In agreement with these considerations, we will show here the existence of a soft longitudinal-acoustic mode which is intimately related to the magnetic susceptibility. The coupling constant appears to be the electronic Grüneisen parameter which is also responsible for different electron-phonon effects.[6,7] The other striking phenomenon is the discovery of oscillatory behavior of sound velocity and attenuation in some shear modes. This may suggest field-induced instabilities.

The experiments were performed with the field along the easy axis giving rise to the maxima of χ_m. Table I summarizes some physical parameters of the two compounds. The similarity in the magnitudes of the linear temperature coefficient γ of the specific heat and consequently in the characteristic fluctuation temperature T^* ($T^* \sim \gamma^{-1}$) should be noticed. The difference in B_c may reflect that one in the effective moment μ so that $\mu(B_c)B_c \sim kT^*$.

The magnetic field dependence of the longitudinal modes (c_{11} for UPt_3 and c_{33} for $CeRu_2Si_2$) is shown in Figs. 1(a) and 1(b) at various temperatures. The common feature is a very strong softening of the two longitudinal modes: 9% for c_{11} in UPt_3 at 20.35 T and 28% for c_{33} in $CeRu_2Si_2$ at 8.07 T for the lowest temperatures (two UPt_3 samples from different sources gave a similar size of the anomaly). In the case of $CeRu_2Si_2$ where extensive magnetization curves $M(T,B)$ exist,[5] the temperature dependence of $\Delta c_{33}(T,B_c)$ is proportional to $\chi_m(T,B_c)$.

TABLE I. Physical properties of the materials.

Material	UPt_3	$CeRu_2Si_2$
Crystal structure	Hexagonal	Tetragonal
Specific-heat γ values (mJ/mol K^2)	420	380
Fluctuation temperature T^*	25 K	24 K
Magnetic easy direction	a,b plane	c axis
Critical field B_c for χ_m anomaly	20.35 T	8.07 T
Soft longitudinal-acoustic mode	c_{11}	c_{33}
Oscillatory shear mode	c_{66}	c_{44}

One can relate the softening of these longitudinal modes and the change in magnetic susceptibility by using scaling and thermodynamic arguments at low temperatures:

$$\Delta c_L = -\Omega_B^2 B^2 \chi_m. \quad (1)$$

Here $\Omega_B = -\partial \ln B_c / \partial \varepsilon_v$, a magnetic Grüneisen parameter, describes the coupling of the lattice to the electronic system. B_c is the critical field (Table I) and ε_v the volume strain. For Eq. (1) one takes the scaled magnetization $M(B/B_c)$ and neglects terms of $\partial^2 B_c / \partial \varepsilon_v^2$ and the small difference between adiabatic and isothermal elastic constants in magnetic fields.[7] This approach is valid for $T \ll T^*$.[8]

In UPt_3 at $T = 4.2$ K experimentally $\Delta c_{11}/c_{11} = -3\%$ [Fig. 1(a)], and with χ_m from Ref. 2 at B_c we get $\Omega_B = 60$ which is exactly the same value as obtained for the thermal Grüneisen parameter $\Omega_T = -\partial \ln T^* / \partial \varepsilon_v$ determined from thermal expansion and elastic constants.[6,7] We made the estimate at 4 K because high-field susceptibility data are not available yet for lower temperatures. Another relation between c_L and the volume magnetostriction coefficient[8] κ can be tested for

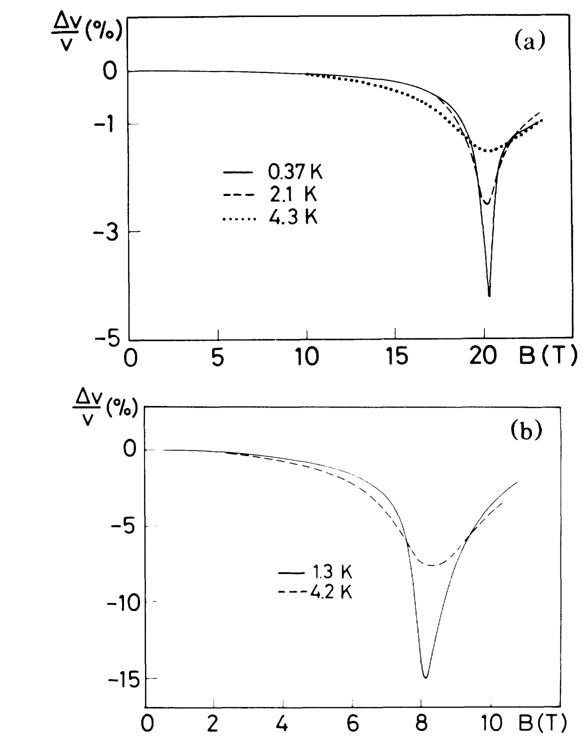

FIG. 1. (a) Relative velocity change in percent for c_{11} mode in UPt_3 at 0.37 K (full line), 2.1 K (dashed line), and 4.3 K (dotted line) as a function of magnetic field applied along b axis. (b) Relative velocity change in percent for c_{33} mode in $CeRu_2Si_2$ at 1.3 K (full line) and 4.2 K (dashed line) as a function of magnetic field applied along c axis.

UPt_3 up to 8 T at 4 K where measurements for κ exist.[9] Again $\Omega_B = 60$ is obtained.

The same analysis for $CeRu_2Si_2$ for $T = 1.3$ K gives, with $\Delta c_{33}/c_{33} = -0.27$ and with χ_m from Refs. 3 and 5, $\Omega_B = 129$. This value compares favorably with $\Omega_T = 120$ from an analysis of the temperature dependence of the elastic constant c_{33} at low temperatures.[10] A more detailed analysis using $\chi_m(B)$ from Ref. 5 and $\Omega_B = 129$ gives a very good fit for $\Delta c_{33}(B)$ or $\Delta v/v$ at $T = 1.3$ K of Fig. 1(b). In addition, the pressure dependence of the electrical resistivity and magnetic susceptibility give again Ω_T of the order of 130.[5,11] A more detailed analysis of the link between $c_L(B,T)$, $\chi_m(B,T)$, and $B_c(P)$ will be published later.[10]

The observed equality between Ω_B and Ω_T emphasizes that the ratio B_c/T^* is pressure independent. This was proved experimentally in the case of $CeRu_2Si_2$.[5] Theories of the almost-localized Fermi liquid do not give the equality of Ω_B and Ω_T.[12] Nevertheless they can describe a "metamagneticlike transition" as observed for polarized He^3.[13] Strong antiferromagnetic correlations have been observed in neutron-scattering experiments for UPt_3,[14] $CeCu_6$,[15] and $CeRu_2Si_2$.[16] The fluctuation temperature T^* and the critical field B_c of the metamagneticlike transition play the equivalent roles of the ordering temperature and of the critical field, respectively, in magnetically ordered systems,[5] although no phase transition occurs in heavy-fermion systems. We then suggest that the soft longitudinal-acoustic mode observed and discussed here is coupled to the soft order-parameter mode of the heavy-fermion system.

Another interesting feature related to the soft-mode behavior is the oscillatory phenomenon observed for some shear modes: c_{66} in UPt_3 and c_{44} in $CeRu_2Si_2$. Note that c_{66} is a shear mode with propagation and polarization in the hexagonal plane, where the susceptibility anomaly also occurs. Figure 2 gives typical examples for this effect in the two cases. We observed these oscillations both as sound velocity changes, as shown in the figure, and in sound attenuation (not shown here). The oscillations disappear for higher temperatures [Fig. 2(a)], they show a weak frequency dependence if any [Fig. 2(b)], and they depend on previous history (they can disappear and reappear after heating up the sample). The c_{44} mode in UPt_3 does not exhibit any such oscillations.

Mode interferences can be excluded as an explanation since the oscillations appear not only for UPt_3, c_{66} (where possible interferences with the c_{44} mode due to misalignment can be excluded because this effect would be too small), but also for c_{44} in $CeRu_2Si_2$ where the c_{44} mode is isotropic in the a-b plane. Mode interferences with spurious longitudinal modes can also be excluded from the frequency dependence, the delay times, and the absence of oscillations in the c_{44} mode in UPt_3. For UPt_3 the oscillations appear to be symmetric with

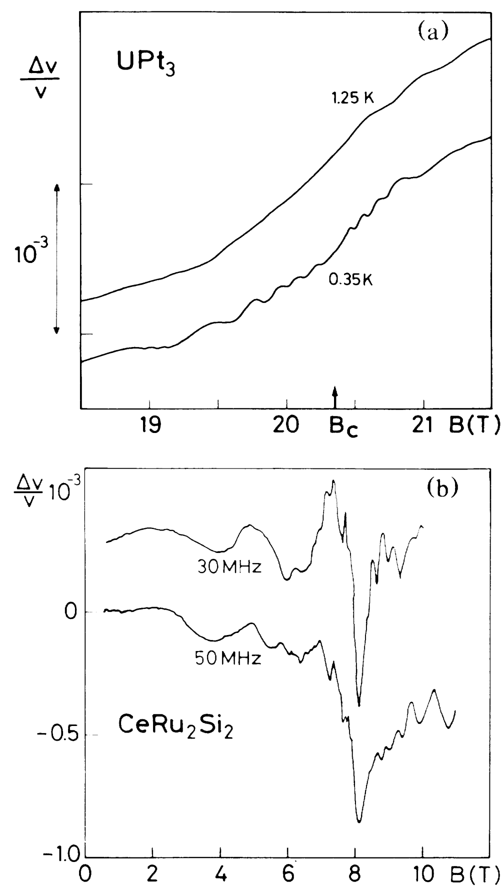

FIG. 2 (a) Field dependence of c_{66} mode at 30 MHz for two different temperatures. Plotted is the relative velocity change in units of 10^{-3}. B and q applied along b axis. (b) Field dependence of c_{44} mode at 1.2 K for two different frequencies (30 MHz, 50 MHz). Plotted is the relative velocity change in units of 10^{-3}. B and q applied along c axis.

respect to B_c, in contrast to $CeRu_2Si_2$ [Figs. 2(a) and 2(b)].

We tried to analyze the oscillations in different ways, e.g., different power laws in $|B-B_c|$. But the most appealing approach was to plot the extrema as de Haas–van Alphen oscillations, i.e., to plot n vs $1/B_n$, B_n being the field at which the nth extremum occurs. We find piecewise approximate linear dependence between n and $1/B_n$ with different slopes in different field regions, for $B>B_c$. No reasonable field dependence is evident for $B<B_c$. The slopes for $B>B_c$ would correspond to de Haas–van Alphen frequencies (α cross-sectional area) of the order of 1 kT in UPt_3 and 0.2 kT in $CeRu_2Si_2$.

Recently de Haas–van Alphen oscillations in UPt_3 were observed at very low temperatures (<100 mK) in fields up to 12 T.[17] High effective masses for large cross-sectional areas were determined. Note the difference in temperature and field region between our experiment and the one of Ref. 17. While the de Haas–van Alphen results indicate a stable Fermi surface, our oscillations above 18 T might indicate more complicated behavior as mentioned above.

We conjecture that these oscillations may be related to the vicinity of magnetic instabilities. These oscillations have not been found yet in other physical quantities (for $CeRu_2Si_2$) such as magnetization,[5] specific heat,[18] and magnetoresistance.[5] Note that the oscillations shown in Fig. 2 have small amplitudes of $\Delta v/v \sim 10^{-4}$ and would correspond to rather small cross-sectional areas of the Fermi surface. The oscillations as discussed above might suggest the existence of field-induced spin-density waves (SDW). The complex oscillations point to a Fermi surface successively altered by different regions of stable SDW.

So far field-induced SDW have been discussed only for quasi one-dimensional organic conductors.[19] The calculated phase diagram exhibits stable SDW regions separated by first-order transitions. In addition the appearance of SDW has been observed at zero magnetic field by alloying of UPt_3 with Au, Th,[20] or Pd.[21]

Other elastic modes exhibit somewhat less spectacular features.[10] For example, c_{33} in UPt_3 and $CeCu_6$ and $(c_{11}-c_{12})/2$ in $CePb_3$ show slow oscillations (one to two periods up to 20 T) in the temperature region 0.4–4 K. Correspondingly magnetization oscillations for M_z with the same period have also been observed for UPt_3 at 4 K.[22]

In summary, we have observed strong softening of longitudinal-acoustic modes for these substances in high magnetic fields and we have given a useful phenomenological description of these results in terms of Grüneisen parameter coupling. A microscopic theoretical description is still lacking. In addition, we found velocity and amplitude oscillations for some shear modes where symmetry allows a coupling to magnetic fluctuation in these systems. We conjecture that they probe field-induced electronic instabilities. Further experiments have to probe this conjecture.

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