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ELECTRON–PHONON COUPLING IN URANIUM COMPOUNDS

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We present results of elastic measurements for UN, UPd_3, UBe_{13} and UPt_3. Exchange striction effects are seen for UN and crystal field anomalies for UPd_3. Polycrystalline UBe_{13} exhibits an anomalously low Poisson ratio. UPt_3 shows low temperature anomalies in various elastic constants and strong anharmonic effects for the c_{44} mode.

1. Introduction

We present results of elastic constant measurements of various uranium compounds and discuss the various types of electron–phonon coupling. In addition inelastic neutron scattering measurements together with elastic constant results give evidence of a strong dispersion for transverse phonons in UPt_3. We discuss exchange striction effects in UN, magnetoelastic effects in UPd_3 and deformation potential coupling in UPt_3 and UBe_{13}.

2. Exchange striction effects in UN

Elastic constants have been measured before [1] in UN. Our own measurements shown in fig. 1 agree within a few percent in the absolute value of the elastic constants, but show distinct differences in the paramagnetic region and in the vicinity of the antiferromagnetic phase transition of $T_N = 53$ K. We observe also a pronounced attenuation for the $c_{11} - c_{12}$ mode for $T \approx 180$ K, but we did not find precursor phenomena. In the vicinity of T_N all elastic modes exhibited steplike discontinuities which are reminiscent of structural phase transitions [2] and may be due to domain-wall stress effects or due to a strain–order parameter coupling of

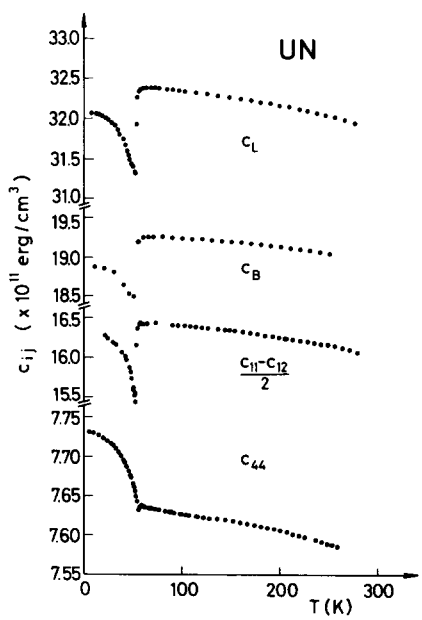

Fig. 1. Elastic constants as a function of temperature for UN. (mass density $\rho = 14.3$ g/cm^3)

the form $E_{int} = d\epsilon_\Gamma \eta^2$ where ϵ_Γ is the strain of symmetry Γ and η is the order parameter. In the ordered region all the elastic modes exhibit a stiffening with decreasing temperature contrary to the older experiments. This behaviour could be due to exchange striction [1,3] but again domain wall stress effects could also play an important role. No effect due to crystal fields or deformation potential coupling is noticeable in the whole temperature region.

3. Crystal field effects and structural transition in UPd_3

In UPd_3 inelastic neutron scattering experiments [4] revealed various crystal electric field transitions due to $5f^2(^3H_4)$. In addition specific heat, magnetic susceptibility and electrical resistance measurements [5] gave indications of two consecutive phase transitions at $T \approx 6$ and 7 K, respectively. The nature of these transitions is probably of structural origin. In fig. 2 we show elastic constants for the c_{44}- and $(c_{11} - c_{12})/2$-mode for UPd_3. Whereas the relative measurement of these modes is very accurate as seen from the smooth temperature dependence, the absolute value of the elastic constants quoted is only accurate within 9% for the c_{44}-mode and 20% for the $(c_{11} - c_{12})/2$-mode because of limited crystal quality and size. The results in fig. 2 show a strong softening of the two modes: 11% for the c_{44} and even 24% for the $(c_{11} - c_{12})/2$-mode. This softening is strong evidence for the structural nature of the low temperature phase transitions. In the insert of fig. 2 strong anomalies for both modes are noticeable at 5 and 7 K, respectively.

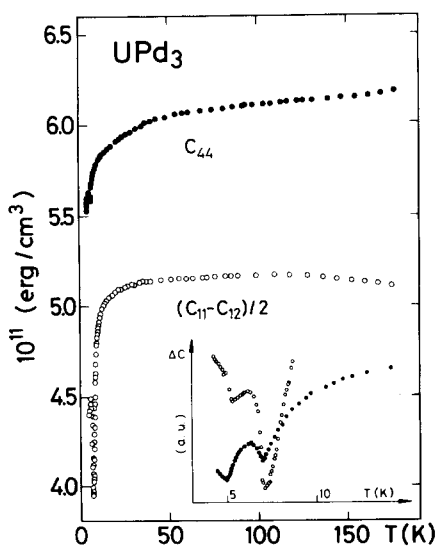

Fig. 2. Elastic constants c_{44} and $(c_{11} - c_{12})/2$ of UPd_3. Insert the same modes at low temperatures (mass density $\rho = 11.6$ g/cm^3).

A quantitative interpretation of these results is difficult because in UPd_3 with the $TiNi_3$-type structure (double hexagonal) one has two nonequivalent U-sites, whose ions exhibit different crystal field splittings. Therefore one has too many arbitrary parameters to make a meaningful analysis and one has to wait for more detailed information. Nevertheless the strong softening of the $(c_{11} - c_{12})/2 = c_{66}$ mode indicates that the upper transition is of induced quadrupolar type involving magnetoelastic coupling to $J_x J_y$ quadrupole operators and strong effective quadrupole–quadrupole coupling, leading to a structural transition from hexagonal to orthorhombic or monoclinic [6].

Application of a magnetic field, B, has a strong effect on the c_{66} mode. If B is applied perpendicular to the c-axis we have a sharp velocity increase at 8.5 T and 7.6 K, at 4 T and 6 K and at 5.5 T and 4 K. These effects are indications of the phase transition line as a function of field and temperature.

4. Heavy fermion system UBe_{13}

Recently the superconductivity in UBe_{13} [7] was identified as due to heavy fermions [8]. Narrow 5f bands with high density of states exist at temperatures $T \leqslant 10$ K ($\gamma = 0.2$ J/mol K^2). Therefore strong deformation potential effects can be expected. In fig. 3 we show transverse and longitudinal sound velocities for a polycrystalline sample of UBe_{13} in the temperature region 1–150 K. Both longitudinal and transverse velocities show a normal temperature dependence down to 10 K and a slight anomaly below. These anomalies are much smaller than in the other heavy fermion compound UPt_3 discussed below, indicating that the deformation potential coupling is relatively small [9]. However the Poisson ratio $\nu = -\epsilon_\perp/\epsilon_\parallel = \frac{1}{2}(((v_L/v_T)^2 - 2)/(v_L/v_T)^2 - 1))$ is anomalously small though not negative. For the temperature range 4–300 K, $\nu = 0.05$. The implication of a small ν for narrow bands is discussed elsewhere [9]. With the mass density $\rho = 4.37$ g/cm^3 we can estimate for the bulk modulus $c_B = \rho(v_L^2 - \frac{4}{3}v_T^2) = 16.1 \times 10^{11}$ erg/cm^3 at $T = 200$ K. This value is not small. Together with $\nu > 0$ and small deformation potential effects it means that in UBe_{13} anomalous

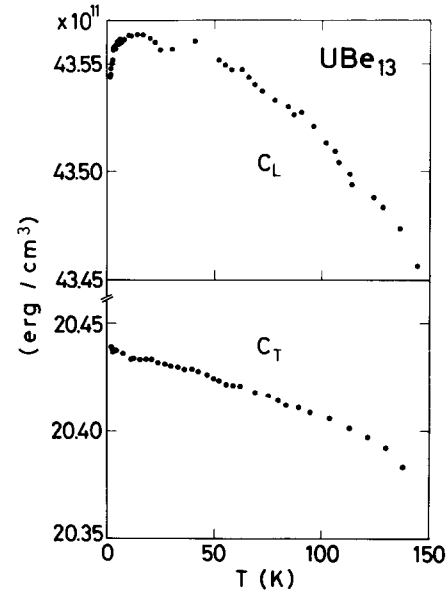

Fig. 3. Temperature dependence of longitudinal and transverse elastic constants for polycrystalline UBe_{13} (mass density $\rho = 4.37$ g/cm^3).

elastic effects are much less pronounced than for UPt_3 probably because of the great number of Be atoms per UBe_{13} molecule.

5. Deformation potential effects in the heavy fermion system UPt_3

One of the most interesting uranium compounds, UPt_3 ($MgCd_3$type structure), which has a high electronic γ-value (0.42 J/mol K^2 at 1 K) and a transition to a superconducting state [10,11] exhibits also strong electron–phonon effects. In fig. 4 the temperature dependence of some of the elastic constants (c_{33}, c_{44}, c_{66}, c_{11}) are shown as a function of temperature. The absolute values at room temperature are in reasonable agreement with a recent determination [12]. From the data in fig. 4, low temperature anomalies of 0.03–1.1% are clearly seen for all modes for $T < 50$ K, and the strongest anomaly is observed for c_{11}. At first sight these anomalies have some resemblance to crystal field anomalies observed in dhcp Pr [13,14], although the CEF effects in Pr are more pronounced. However, the comparison of the thermal expansion curves for UPt_3 [12] and Pr [15] clearly show that the low temperature anomalies in UPt_3 is due to a strain coupling to itinerant electrons. Therefore a deformation type electron–phonon coupling is responsible for the low temperature anomalies in UPt_3. The small anomalies for c_{66} (0.1%) and c_{44} (0.03%) compared to c_{33} (0.3%) and c_{11}(1.1%) indicate that the deformation potential coupling constant $d(\Gamma_5, c_{66})$ and $d(\Gamma_6, c_{44})$ are rather smaller than

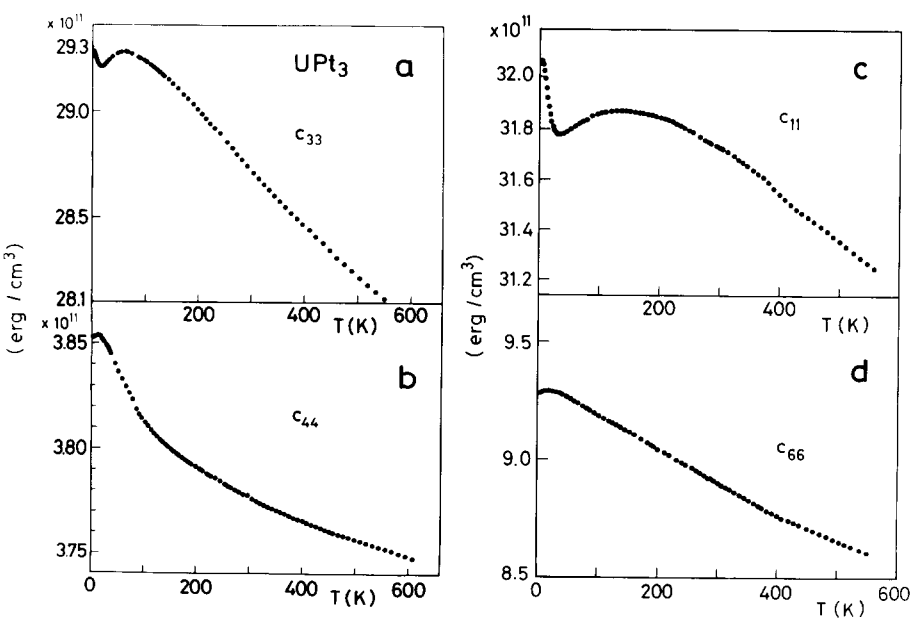

Fig. 4. Temperature dependence of the elastic constants c_{33}, c_{44}, c_{11}, c_{66} for UPt_3 from 1 to 600 K (mass density $\rho = 19.4$ g/cm^3).

$d(\Gamma_1, c_B)$. For the longitudinal mode we can make estimates using the Grüneisen parameter [9,16] $\Omega = -(1/E)(\partial \bar{E}/\partial \epsilon_v) = c_B \beta / C$, where $\bar{E}$ is a typical 5f-band width, β the volume thermal expansion and C the electronic specific heat. Taking values [10,12] for C, β, c_B at 1 K we get $\Omega = 60$. A recent band structure calculation [17] gives a high density of states at the Fermi energy with a width of ≈ 0.5 eV. In order to account for the measured heavy fermion density of states at low temperatures, one has to assume additional structure developing in the density of states, similar to the coherent Kondo lattice state [18]. With a bandwidth of $T_{sf} \approx 20$ K we get $d(\Gamma_1) \approx 1200$ K, a typical value also observed for $CeAl_3$, $CeCu_2Si_2$ etc. [9]. With these values we get a semiquantitative description of the low temperature anomalies in c_{11}, c_{33} and β. It is difficult to calculate $c_{ij}(T)$ and $\beta(T)$ quantitatively [9], because γ and the density of states are temperature dependent for $T \leqslant T_k$ for heavy fermion systems [10,18]. $d(\Gamma_1)$ provides also the appropriate electron–phonon coupling for the ultrasonic attenuation in superconducting UPt_3 [19]. With $m^*/m_0 = 180$ [10] one gets $(m^*/m_0)\, d(\Gamma_1) = 18$ eV, a value which accounts for the electronic contribution to the normal state ultrasonic attenuation [9].

In addition to these low temperature anomalies in the elastic constants we observe other effects as well. As seen in fig. 4 the high temperature dependence of c_{44} is rather anomalous. Whereas c_{33} has a linear T dependence for $T > 300$ K, the T dependence of c_{44} is slower than linear. This seems to suggest strong anharmonic behaviour. We also observe strong difference between zero and first sound for the transverse acoustic mode along the c-axis but not for the other modes. Fig. 5 shows a detail of phonon dispersion curves which have been measured at 300 and 10 K. In order to investigate the anomalous behaviour of c_{44} more precisely the corresponding TA phonon branches were determined with improved horizontal and vertical collimations. No pronounced temperature dependence shows up in the present neutron results when the sample is cooled down to 10 K. As seen from fig. 5 the ultrasonic c_{44} velocity is about 9% smaller than the high frequency phonon velocity determined by inelastic neutron scattering. Such a large change in c_{44} is unusual [20]. It is not yet clear whether these anomalies are directly related to the electron–phonon coupling for the narrow 5f bands. The Poisson ratios for UPt_3 range from 0.1–0.3 and do not reflect this coupling in any dramatic way [9]. Further details will be published elsewhere.

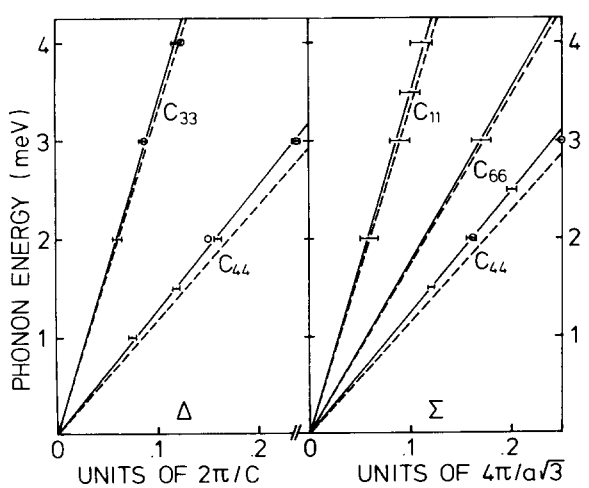

Fig. 5. Slopes of acoustic phonon branches for 300 K. Phonon frequencies measured at 10 K are shown by circles. The dashed lines are calculated from the values of the elastic constants at 300 K.

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