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Absence of acoustic signature of the quadrupolar Kondo effect in $U_{0.2}Y_{0.8}Pd_3$

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Elastic-constant data for $U_{0.2}Y_{0.8}Pd_3$ are presented between 1.5 and 100 K. The absence of any strong anomaly in their temperature dependence is strong evidence against a quadrupolar Kondo effect interpretation of the non-Fermi-liquid behavior of this compound.

Recent measurements of the specific heat, magnetic susceptibility, thermal expansion, and electrical resistivity on the cubic material $U_xY_{1-x}Pd_3$ ($x \approx 0.2$) have given indications of non-Fermi-liquid behavior.[1,2] These results were interpreted in terms of a two-channel Kondo effect (specifically a quadrupolar Kondo effect)[1] based on the multichannel Kondo model[3] or, in another approach, as a non-Fermi-liquid with a $T=0$ K phase transition.[2] In the first case a Kondo temperature $T_K=42$ K was identified for $x=0.2$. More recent specific-heat measurements on $U_{0.2}Y_{0.8}Pd_3$ revealed deviations from the behavior predicted by the quadrupolar Kondo effect at very low temperatures.[4] We show here that the absence of any observable anomalies in the temperature dependence of elastic constants in this compound makes the quadrupolar Kondo effect an unlikely cause for the cited phenomena. We concentrate on the temperature region where the screening of the quadrupoles should manifest itself, i.e., from $T > T_K$ (unscreened) to $T \ll T_K$ (screened).

Elastic-constant measurements are particularly well suited to study, via the magnetoelastic interaction, the quadrupolar response of the $4f$ or $5f$ ions. The elastic constants, as the strain susceptibility, measure the diagonal (Curie terms) and off-diagonal (Van Vleck terms) quadrupolar matrix elements, analogously to the magnetic susceptibility which samples the magnetic dipole matrix elements.[5] This is in contrast to the thermal expansion, which, in cubic symmetry, couples only to higher-order multipole components of these ions.[5]

With a Γ_3 quadrupolar-active ground state of the $U^{4+}(5f^2)$ ions and a $T_K=42$ K (Ref. 1) one would expect typical quadrupolar effects for $T \geq T_K$. Especially, symmetry elastic constants c_Γ should exhibit a strong temperature dependence according to[5,6]

$$c_\Gamma = c_\Gamma^0 - N\frac{g_\Gamma{}^2\chi_\Gamma^s}{1-g'\chi_\Gamma^s} . \tag{1}$$

Here c_Γ^0 is the background elastic constant, N the number of magnetic ions per unit volume, χ_Γ^s the corresponding strain susceptibility, g_Γ the magnetoelastic coupling constant, and g' the $\mathbf{q}=0$ interaction between quadrupoles in the Hamiltonian

$$H = -\sum_i g_\Gamma \varepsilon_\Gamma O_{\Gamma_i} - \sum_{ij} G_{ij} O_{\Gamma_i} O_{\Gamma_j} , \tag{2}$$

where ε_Γ denotes the symmetry strain, O_Γ is the corresponding quadrupolar operator, and $g' = \sum_j G_{ij}$. For a Γ_3 ground state the single-ion susceptibility χ_Γ^s diverges as $T \to 0$ K.[5] Typical manifestations of such elastic-constant behavior have been observed for rare-earth pnictides,[5,7] rare-earth vanadates,[5] and the U-compounds UO_2 (Ref. 8) and UPd_3.[9]

The latter case, UPd_3, is especially interesting because transitions, between well-defined crystal-field levels of the $5f$ ground multiplet, have been observed by inelastic neutron scattering[10] and the temperature dependence of the elastic constants can be rather well interpreted using Eq. (1).[9] UPd_3 crystallizes in the double hexagonal close-packed structure, in which the U^{4+} ions occupy quasicubic and hexagonal sites. Below $T \approx 150$ K, the temperature dependence of the elastic constants is essentially due to the crystal-field splitting at the quasicubic sites with a Γ_1 singlet ground state.[11] Therefore the strain susceptibility for these sites was inserted in Eq. (1), in order to describe the thermal evolution of two elastic constants (c_{11} and c_{44}), as shown in Fig. 1. One observes strong softening effects for these modes which are due to the magnetoelastic coupling of the sound waves to the $5f$-electronic charge distribution of the U^{4+} ions at these cubic sites. For example, the c_{44} mode exhibits an overall softening of more than 5% from the maximum at 100 K down to the quadrupolar transition temperature $T_Q=6.8$ K. This result establishes the sizable coupling constants $g^2N/c_0=0.6$ K and $g'=-6$ K for the c_{44} mode. Recent elastic neutron-scattering experiments revealed an antiferroquadrupolar ordering with predominantly Γ_5 symmetry related to local ε_{xz} strains,[12] in accordance with our analysis.

In Fig. 2 we show the temperature dependence of longitudinal and transverse modes measured on the same polycrystalline $U_{0.2}Y_{0.8}Pd_3$ sample that was previously used for the specific-heat measurements reported in Ref. 4. Since no single crystals are available for this material we show the only experimentally accessible modes, i.e., c_L and c_T.

In $U_{0.2}Y_{0.8}Pd_3$, the U^{4+} has the same near environ-

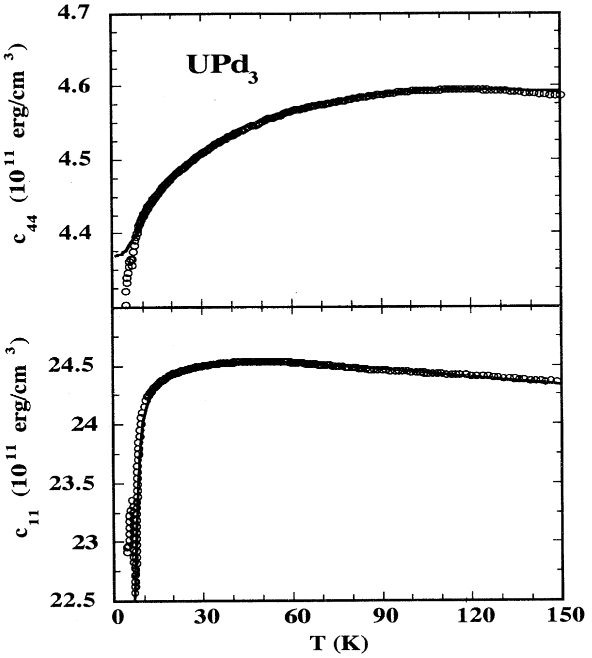

FIG. 1. Temperature dependence of the c_{44} and c_{11} elastic modes for UPd$_3$. The full lines are fits using strain susceptibilities as discussed in the text.

ment as the quasicubic sites in UPd$_3$: it has the same coordination number and its first neighbors are located at nearly exactly the same positions. The latter point is attested by the comparison of the lattice parameters, corresponding to a in hexagonal symmetry, of 5.767 Å for U$_{0.2}$Y$_{0.8}$Pd$_3$ (Ref. 1) versus 5.765 Å for UPd$_3$,[13] at room temperature. Therefore, we may expect similar crystal-field-scheme and magnetoelastic couplings as in UPd$_3$. However, while for UPd$_3$ the quasicubic sites ground state is a Γ_1 singlet with a Γ_3 doublet 18 K above, recent inelastic neutron-scattering measurements on U$_{0.2}$Y$_{0.8}$Pd$_3$ have been interpreted as resulting from a Γ_3 ground state with Γ_5 and Γ_4 excited levels.[14] This is very surprising in view of the same valence state for the U ions and the similar site symmetry.

Nevertheless, for such a Γ_3 ground state, we would expect very strong softening effects for $T \geq T_K$ and a flattening of the temperature dependence for $T < T_K$ for both c_L and c_T due to the Kondo screening of the quadrupole moments. In the framework of the quadrupolar Kondo effect, a calculation of the quadrupolar susceptibility of Γ_3 symmetry results in a logarithmic divergence below T_K for $T \rightarrow 0$ K.[15] In any case, one expects a substantial temperature dependence of the symmetry elastic constants for temperatures around T_K.

It is seen, however, that no sign of an anomalous temperature dependence is apparent in Fig. 2, over the whole temperature range 1.5 K $< T < 100$ K, for both the c_T and c_L modes. In addition the magnetic-field dependence of these modes is smaller than $(c - c_0)/c_0 < 2.10^{-4}$ at 1.5 K and fields up to 10 T.

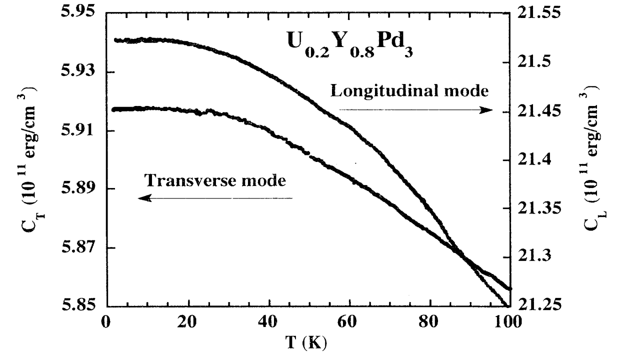

FIG. 2. Longitudinal, c_L, and transverse, c_T, elastic constants as a function of temperature for polycrystalline U$_{0.2}$Y$_{0.8}$Pd$_3$.

From these results we may conclude that the temperature dependence of c_T and c_L of polycrystalline U$_{0.2}$Y$_{0.8}$Pd$_3$ give no indication that quadrupolar effects play a significant role for the temperature-dependent elastic properties of this material. As an effect of the dilution of U with Y, the softenings should be reduced for U$_{0.2}$Y$_{0.8}$Pd$_3$ with respect to UPd$_3$. On the other hand, due to the breaking of bonds (ij), the strong quadrupolar interaction constant g', which is negative, should be strongly reduced in U$_{0.2}$Y$_{0.8}$Pd$_3$ and, consequently, the softening effects enhanced. In addition, the hypothetical Γ_3 ground state of U$_{0.2}$Y$_{0.8}$Pd$_3$ would further amplify this enhancement.

Doubts about the relevance of a quadrupolar Kondo effect for interpreting the low-temperature properties of U$_{0.2}$Y$_{0.8}$Pd$_3$ were already initiated by the results of measurements of the temperature dependence of the electrical resistivity $\rho(T)$. The two-channel Kondo model, of which the quadrupolar Kondo effect invoking a Γ_3 ground state would be a physical realization, predicts $\rho(T) \sim T^{1/2}$ well below T_K,[16] while various experimental investigations[1,2,4] revealed consistently that $\rho(T)$ varies rather linearly as $T \rightarrow 0$ K.

The results of our measurements add to the experimental indications that the quadrupolar Kondo-effect model may not be the real cause of the non-Fermi-liquid behavior of U$_{0.2}$Y$_{0.8}$Pd$_3$. An alternative view of U$_{0.2}$Y$_{0.8}$Pd$_3$ was discussed in Ref. 2. No Γ_3 ground state is invoked and the anomalous non-Fermi-liquid properties have been discussed using scaling laws and assuming a $T = 0$ K phase transition. In view of our results, this explanation is still viable. Finally it should be mentioned that Grüneisen parameter effects on the elastic constants as observed for heavy electron systems[17] are not noticeable for U$_{0.2}$Y$_{0.8}$Pd$_3$.

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