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INVITED PAPER

LIGHT SCATTERING IN HEAVY FERMION COMPOUNDS

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We present results of Raman spectroscopy and magnetic susceptibility measurements in the actinide compounds UO_2 , UPt_3 and UBe_{13} . The differences in the degree of localization of the 5f-electrons in the insulating UO_2 and in the metallic, heavy fermion compounds UPt_3 and UBe_{13} are discussed. In both types of compounds we find evidence of localized electronic excitations, which yield a good fit of the magnetic susceptibility data. The spin relaxation rates of UPt_3 and UBe_{13} at $q \approx 0$ are determined experimentally from the quasielastic light scattering.

Since the discovery of the heavy fermion superconductor CeCu_2Si_2 in 1979 [1], strong attention in solid state physics has been focused on heavy fermion compounds like UPt_3 , UBe_{13} and CeCu_6 . The discovery of bulk superconductivity in UPt_3 , together with a $T^3 \ln(T/T_{\text{sf}})$ term in the low temperature specific heat, where T_{sf} is a spin fluctuation temperature, led to speculations about the important role of spin fluctuations for mediating a non-BCS electron pairing in the heavy fermion superconductors.

All heavy fermion (HF) compounds are characterized by a similarly high value of the electronic specific heat γ , which has become the standard criterion for the classification of HF systems [2]. In a Fermi liquid model, values for γ of about 100 times that of an ordinary metal indicate a very high density of states at the Fermi energy E_{F} . This can be related to narrow f bands due to a hybridization of d and f states. One then describes these highly correlated electrons and the corresponding many body effects by attributing a rather high effective mass to the f electrons. As a consequence of the large radial extent of the 5f wave function and a possible direct f–f overlap, the tendency towards delocalization, i.e. band for-

mation, is much more pronounced in 5f-compounds than in 4f-compounds. For example, one finds well defined crystalline-electric-field (CEF) levels in Kondo-type 4f-compounds, such as CeCu_2Si_3 [3] and CeB_6 [4]. Neutron scattering and Raman spectroscopy have been versatile and complementary tools in the investigation of the localized 4f ground state of CeB_6 and other 4f-compounds [4–6]. On the other hand, not much information is available about CEF levels in metallic actinide compounds.

In this paper we present light scattering results of the actinide compounds UO_2 , UPt_3 and UBe_{13} . We find evidence of localized 5f states in insulating UO_2 and at least partially localized 5f-electron character in the HF compound UPt_3 . Moreover, we review investigations of spin fluctuations in the HF compounds UPt_3 and UBe_{13} by means of light scattering, with the emphasis on the spin relaxation rate Γ_s at $q \approx 0$. This allows to test predictions of the Fermi liquid theory that $T_s \approx v_{\text{F}}^* q$, where v_{F}^* is the Fermi velocity of the heavy particles.

For 5f-electrons, the degree of localization is intermediate between 3d- and 4f-electrons. In insulating actinide compounds, the 5f-electrons are generally more localized than in metallic ones. This dependence of the localization on the chemical binding of the ion in the lattice is in contrast to the situation of the 4f-electrons.

In the past UO_2 has already been subject to light scattering and neutron scattering investigations [7,8]. IR reflectivity measurements [9] showed

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Fig. 1. Raman spectrum of UO_2 and ThO_2 at 300 K. The ThO_2 spectrum was scaled in such a way that the T_{2g} phonons in both compounds have the same intensity.

sharp structures at some hundred meV due to 5f excitations. Neutron scattering could resolve 5f-CEF excitations in UO_2 [8]. On the other hand, from magnetic susceptibility measurements a magnetic moment of $3.2\mu_B$ was determined which deviates considerably from the value $2.83\mu_B$ calculated for a Γ_5 CEF ground state [10].

Fig. 1 shows the Raman spectrum of UO_2 at 300 K. In addition to the T_{2g} phonon at 450 cm^{-1} , one observes excitations at 1150 and 1500 cm^{-1} , which are interpreted as due to CEF transitions. As there is no scattering in this region for ThO_2 and as the CEF ground state is known from several other investigations to be Γ_5 [11,12], we have assigned the observed peaks to $\Gamma_5 \rightarrow \Gamma_3$ and $\Gamma_5 \rightarrow \Gamma_4$ transitions, respectively. The symmetry assignment of the CEF levels is based on that of Pr^{3+} in CaF_2 which has also a $J = 4$ ground state [12]. Although the Russell-Saunders coupling scheme becomes increasingly inappropriate for higher lying excited states of heavy elements, a CEF level scheme of UO_2 derived on the basis of the Lea-Leask-Wolf scheme for $J = 4$ [13] can explain the observed temperature dependence of the magnetic susceptibility measured up to 400 K.

The fit of the experimental data is shown in fig. 2. Especially we want to emphasize that the kink near 200 K is well reproduced. The experimentally determined magnetic moment of $\mu_{\text{eff}} = 2.71\mu_B$ is in good agreement with the value $2.83\mu_B$ of the Γ_5 ground state of the $\text{U}^{4+}(5f^2)^3\text{H}_4$ configuration. Therefore, our findings support the conclusion that the 5f levels in UO_2 are localized and split by a crystalline electric field.

The situation is different in actinide metals, especially in compounds like UPt_3 with narrow f bands and correlation effects [2]. Fig. 3 shows the Raman spectrum of UPt_3 at 5 K under 5309 Å laser excitation up to 5000 cm^{-1} frequency shift [14]. We observe strong inelastic scattering between about 1000 and 3000 cm^{-1} which is also observed for 4762 Å excitation and is thus not due to luminescence. This is in agreement with recent observations of inelastic scattering intensities between 1000 and 3000 cm^{-1} in a UPt_3 single crystal under 5145 Å laser excitation [15]. We attribute the inelastic scattering to 5f excitations similar to those observed in UO_2 . However, in UPt_3 these inelastic excitations are very broad compared to UO_2 , in agreement with the expectation of a stronger tendency towards delocalization in metallic actinide compounds. The origin of these excitations can be either due to intraionic multiplet levels or to CEF-splittings. The splittings of the electronic ground state of this order of magnitude can explain the temperature dependence of the magnetic susceptibility. In fig. 4 we

Fig. 2. Inverse magnetic susceptibility of UO_2 between 30 and 400 K. The solid line is a fit based on the Lea-Leask-Wolf CEF scheme described in the text.

Fig. 3. Raman spectrum of UPt₃ at 5 K.

show the magnetic susceptibility of UPt₃ between 10 and 1050 K [16]. A splitting of roughly 2000 cm⁻¹ or 3000 K could describe the bending of the experimental 1/χ curve at about 750 K. This value of 3000 K should be understood as a rough estimate of an average 5f-splitting. The magnetic moments of the states in this two-level model are fit parameters.

Spin fluctuations in the HF compounds UPt₃ and UBe₁₃ have been observed by neutron scattering [17–19] for the momentum transfer $q > 1 \text{ Å}^{-1}$. As Raman scattering is a true $q \approx 0$ method, our aim was to test the linear q dependence of the spin relaxation rate Γ_s , predicted by non-interacting Fermi liquid theory. Fig. 5 shows the quasielastic

Fig. 4. Inverse magnetic susceptibility of UPt₃ between 10 and 1050 K [16]. Fig. 5. Raman spectrum of UBe₁₃ from ref. [20] for perpendicular polarization of incident and scattered light. The hatched area indicates the quasielastic scattering due to spin fluctuations with the relaxation rate Γ_s .

scattering intensity of UBe₁₃ under 5145 Å laser excitation at 350 and 40 K [20]. By the perpendicular orientation of the incident and scattered electric field vectors $E_i \perp E_s$ this scattering is identified as magnetic in origin. The scattering intensity $I(\omega)$ has been fitted by

$$I(\omega) \sim (1 + n(\omega)) \hbar \omega \frac{\Gamma_s/2}{(\Gamma_s/2)^2 + (\hbar \omega)^2}, \quad (1)$$

where $n(\omega)$ is the Bose factor; the Lorentzian is the Fourier transform of $\exp(-\Gamma_s t)$ used to describe fluctuating uncorrelated 5f spins with a spin relaxation rate Γ_s . The fit based on eq. (1) is shown by the hatched area in fig. 5. One obtains an approximately temperature independent spin relaxation rate $\Gamma_s(q \approx 0) = (110 \pm 10) \text{ cm}^{-1}$ ($= 13.6 \text{ meV}$). This result together with that from neutron scattering for $q = 2 \text{ Å}^{-1}$ [19] is shown in fig. 6. The q independence of Γ_s is evidence for the localized nature of the spin fluctuations. A similar q independence of Γ_s is found for UPt₃ [21,22]. A slight dependence of the spin relaxation rate, contrary to Fermi liquid predictions, has been observed [23] in CeCu₆ and theoretically explained in ref. [24].

Fig. 6. Spin relaxation rate $\Gamma_s/2$ of UBe_{13} as a function of momentum transfer. Raman data at 40 K and neutron data at 10 K from ref. [19].

The above results for UBe_{13} , UPt_3 and CeCu_6 are in disagreement with the simple non-interacting Fermi liquid approach. The observation of a finite-frequency zone-center contribution of the spin fluctuations reflects the fact that the spin or the magnetization is not conserved due to the strong spin-orbit coupling [24].

Conclusions

In addition to previous neutron measurements of CEF levels in the cerium-based HF compound CeCu_2Si_2 we could find first evidence of localized electronic excitations in uranium-based HF compounds which are not accessible by neutron scattering. This evidence is further supported by fits of the magnetic susceptibility data. The observation of the spin relaxation rate at $q \approx 0$, which complements neutron data at large q , has revealed that the simple non-interacting Fermi liquid theory is not applicable to HF compounds. Theoretical work in this direction has been undertaken recently [24]. The advantage of applying light scattering to HF compounds has been shown to lie in the $q \approx 0$ momentum transfer and the measurable high energy losses.

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