

# UC Irvine

## UC Irvine Previously Published Works

### Title

Light scattering in heavy fermion compounds

### Permalink

<https://escholarship.org/uc/item/7f44s6j5>

### Journal

Journal of Magnetism and Magnetic Materials, 76(C)

### ISSN

0304-8853

### Authors

Blumenröder, S  
Brenten, H  
Zirngiebl, E  
[et al.](#)

### Publication Date

1988-12-01

### DOI

10.1016/0304-8853(88)90413-1

### Copyright Information

This work is made available under the terms of a Creative Commons Attribution License, available at <https://creativecommons.org/licenses/by/4.0/>

Peer reviewed

INVITED PAPER

LIGHT SCATTERING IN HEAVY FERMION COMPOUNDS

S. BLUMENRÖDER, H. BRENTEN<sup>1</sup>, E. ZIRNGIEBL<sup>2</sup>, R. MOCK<sup>3</sup>, G. GÜNTHERODT,  
J.D. THOMPSON<sup>4</sup>, Z. FISK<sup>4</sup> and J. NAEGELE<sup>5</sup>

*II. Physikalisches Institut, RWTH Aachen, 5100 Aachen, Fed. Rep. Germany*

We present results of Raman spectroscopy and magnetic susceptibility measurements in the actinide compounds  $\text{UO}_2$ ,  $\text{UPt}_3$  and  $\text{UBe}_{13}$ . The differences in the degree of localization of the 5f-electrons in the insulating  $\text{UO}_2$  and in the metallic, heavy fermion compounds  $\text{UPt}_3$  and  $\text{UBe}_{13}$  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  $\text{UPt}_3$  and  $\text{UBe}_{13}$  at  $q \approx 0$  are determined experimentally from the quasielastic light scattering.

Since the discovery of the heavy fermion superconductor  $\text{CeCu}_2\text{Si}_2$  in 1979 [1], strong attention in solid state physics has been focused on heavy fermion compounds like  $\text{UPt}_3$ ,  $\text{UBe}_{13}$  and  $\text{CeCu}_6$ . The discovery of bulk superconductivity in  $\text{UPt}_3$ , together with a  $T^3 \ln(T/T_{sf})$  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  $\gamma$ , which has become the standard criterion for the classification of HF systems [2]. In a Fermi liquid model, values for  $\gamma$  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  $\text{CeCu}_2\text{Si}_3$  [3] and  $\text{CeB}_6$  [4]. Neutron scattering and Raman spectroscopy have been versatile and complementary tools in the investigation of the localized 4f ground state of  $\text{CeB}_6$  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  $\text{UO}_2$ ,  $\text{UPt}_3$  and  $\text{UBe}_{13}$ . We find evidence of localized 5f states in insulating  $\text{UO}_2$  and at least partially localized 5f-electron character in the HF compound  $\text{UPt}_3$ . Moreover, we review investigations of spin fluctuations in the HF compounds  $\text{UPt}_3$  and  $\text{UBe}_{13}$  by means of light scattering, with the emphasis on the spin relaxation rate  $\Gamma_s$  at  $q \approx 0$ . This allows to test predictions of the Fermi liquid theory that  $T_s \approx v_F^* q$ , 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  $\text{UO}_2$  has already been subject to light scattering and neutron scattering investigations [7,8]. IR reflectivity measurements [9] showed

<sup>1</sup> Physikalisches Institut, Technische Universität Clausthal, Leibnizstrasse 4, 3392 Clausthal-Zellerfeld, FRG.

<sup>2</sup> Bayer AG, ZF-TPE6, 5090 Leverkusen, FRG.

<sup>3</sup> Siemens AG, ZT-SFE AMF 42, 8000 München 83, FRG.

<sup>4</sup> Los Alamos National Laboratory, Los Alamos, NM 87545, USA.

<sup>5</sup> Kernforschungszentrum Karlsruhe, Karlsruhe, FRG.

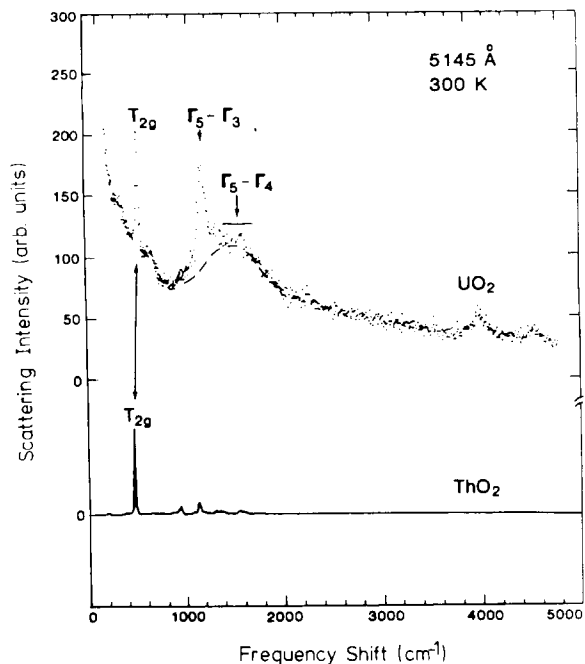


Fig. 1. Raman spectrum of  $\text{UO}_2$  and  $\text{ThO}_2$  at 300 K. The  $\text{ThO}_2$  spectrum was scaled in such a way that the  $T_{2g}$  phonons in both compounds have the same intensity.

sharp structures at some hundred meV due to 5f excitations. Neutron scattering could resolve 5f-CEF excitations in  $\text{UO}_2$  [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  $\Gamma_5$  CEF ground state [10].

Fig. 1 shows the Raman spectrum of  $\text{UO}_2$  at 300 K. In addition to the  $T_{2g}$  phonon at  $450\text{ cm}^{-1}$ , one observes excitations at 1150 and  $1500\text{ cm}^{-1}$ , which are interpreted as due to CEF transitions. As there is no scattering in this region for  $\text{ThO}_2$  and as the CEF ground state is known from several other investigations to be  $\Gamma_5$  [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  $\text{Pr}^{3+}$  in  $\text{CaF}_2$  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  $\text{UO}_2$  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  $\Gamma_5$  ground state of the  $\text{U}^{4+}(5f^2)^3H_4$  configuration. Therefore, our findings support the conclusion that the 5f levels in  $\text{UO}_2$  are localized and split by a crystalline electric field.

The situation is different in actinide metals, especially in compounds like  $\text{UPt}_3$  with narrow f bands and correlation effects [2]. Fig. 3 shows the Raman spectrum of  $\text{UPt}_3$  at 5 K under  $5309\text{ \AA}$  laser excitation up to  $5000\text{ cm}^{-1}$  frequency shift [14]. We observe strong inelastic scattering between about 1000 and  $3000\text{ cm}^{-1}$  which is also observed for  $4762\text{ \AA}$  excitation and is thus not due to luminescence. This is in agreement with recent observations of inelastic scattering intensities between 1000 and  $3000\text{ cm}^{-1}$  in a  $\text{UPt}_3$  single crystal under  $5145\text{ \AA}$  laser excitation [15]. We attribute the inelastic scattering to 5f excitations similar to those observed in  $\text{UO}_2$ . However, in  $\text{UPt}_3$  these inelastic excitations are very broad compared to  $\text{UO}_2$ , 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

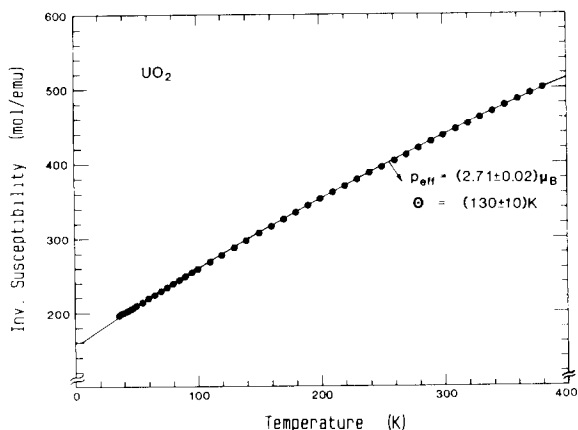
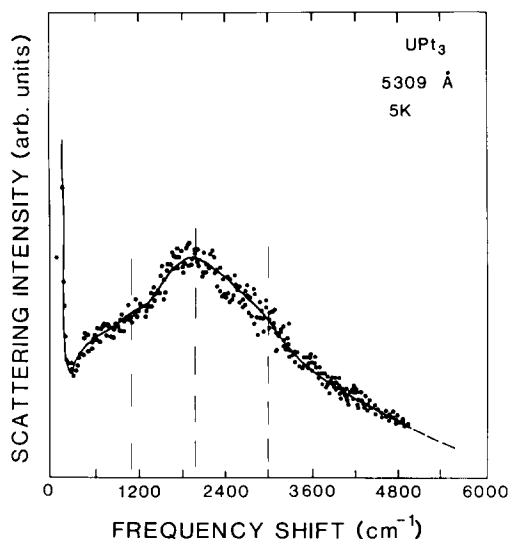


Fig. 2. Inverse magnetic susceptibility of  $\text{UO}_2$  between 30 and 400 K. The solid line is a fit based on the Lea-Leask-Wolf CEF scheme described in the text.

Fig. 3. Raman spectrum of  $\text{UPt}_3$  at 5 K.

show the magnetic susceptibility of  $\text{UPt}_3$  between 10 and 1050 K [16]. A splitting of roughly  $2000 \text{ cm}^{-1}$  or 3000 K could describe the bending of the experimental  $1/\chi$  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  $\text{UPt}_3$  and  $\text{UBe}_{13}$  have been observed by neutron scattering [17–19] for the momentum transfer  $q > 1 \text{ \AA}^{-1}$ . As Raman scattering is a true  $q \approx 0$  method, our aim was to test the linear  $q$  dependence of the spin relaxation rate  $\Gamma_s$ , predicted by non-interacting Fermi liquid theory. Fig. 5 shows the quasielastic

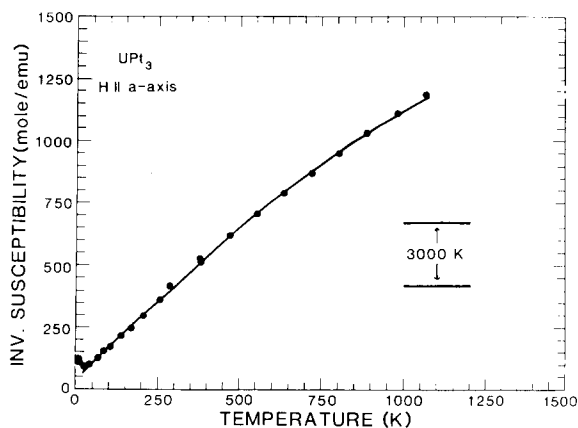
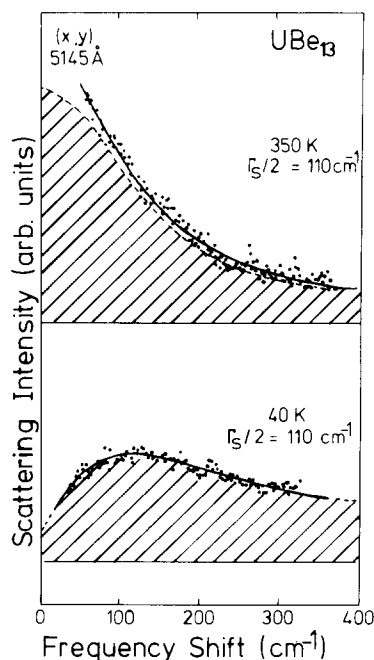
Fig. 4. Inverse magnetic susceptibility of  $\text{UPt}_3$  between 10 and 1050 K [16].

Fig. 5. Raman spectrum of  $\text{UBe}_{13}$  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  $\Gamma_s$ .

scattering intensity of  $\text{UBe}_{13}$  under  $5145 \text{ \AA}$  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  $\Gamma_s$ . 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  $q = 2 \text{ \AA}^{-1}$  [19] is shown in fig. 6. The  $q$  independence of  $\Gamma_s$  is evidence for the localized nature of the spin fluctuations. A similar  $q$  independence of  $\Gamma_s$  is found for  $\text{UPt}_3$  [21,22]. A slight dependence of the spin relaxation rate, contrary to Fermi liquid predictions, has been observed [23] in  $\text{CeCu}_6$  and theoretically explained in ref. [24].

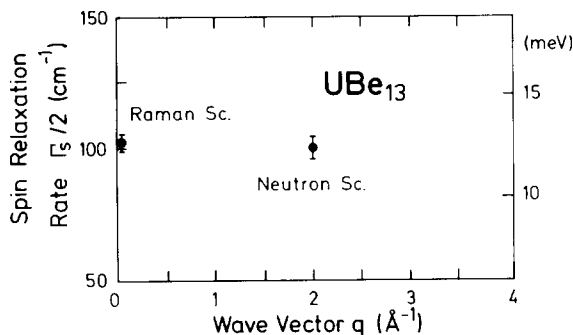


Fig. 6. Spin relaxation rate  $\Gamma_s/2$  of  $\text{UBe}_{13}$  as a function of momentum transfer. Raman data at 40 K and neutron data at 10 K from ref. [19].

The above results for  $\text{UBe}_{13}$ ,  $\text{UPt}_3$  and  $\text{CeCu}_6$  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  $\text{CeCu}_2\text{Si}_2$  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.

### References

[1] F. Steglich, J. Aarts, C.D. Bredl, W. Lieke, D. Mesched, W. Franz and J. Schäfer, Phys. Rev. Lett. 43 (1979) 1892.

- [2] G.R. Stewart, Rev. Mod. Phys. 56 (1984) 755.  
 [3] S. Horn, E. Holland-Moritz, M. Loewenhaupt, F. Steglich, H. Scheuer, A. Benoit and J. Flouquet, Phys. Rev. B 23 (1981) 3171.  
 [4] E. Zirngiebl, B. Hillebrands, S. Blumenröder, G. Güntherodt, M. Loewenhaupt, J.M. Carpenter, K. Winzer and Z. Fisk, Phys. Rev. B 30 (1984) 4052.  
 [5] J.T. Hougen and S. Singh, Phys. Rev. Lett. 10 (1963) 406.  
 [6] J.H. Koningstein and O.S. Mortensen, in: The Raman Effect, ed. A. Anderson (Marcel Dekker, New York, 1973) p. 519.  
 [7] P.J. Cowell, L.A. Rahn and C.T. Walker in: Light Scattering in Solids, ed. Balkanski, J. Schoenes, J. Chem. Soc. Faraday Trans. 2 83 (1987) 1205.  
 [8] S. Kern, C.-K. Loong and G.H. Lander, Phys. Rev. B 32 (1985) 3051.  
 [9] J. Schoenes, Phys. Rep. 63 (1980) 301.  
 [10] S. Nasu, Japan. J. Appl. Phys. 5 (1966) 1001.  
 [11] H.U. Rahman and W.A. Runciman, J. Phys. Chem. Solids 27 (1966) 1833.  
 [12] M.J. Weber and R.W. Bierig, Phys. Rev. 134 (1964) A1492.  
 [13] K.R. Lea, M.J.M. Leask and W.P. Wolf, J. Phys. Chem. Solids 23 (1962) 1381.  
 [14] S. Blumenröder, H. Brenten, G. Güntherodt, E. Zirngiebl and Z. Fisk, Bull. Am. Phys. Soc. 32 (1987) 719.  
 [15] S.L. Cooper, M.W. Klein, Z. Fisk and J.L. Smith, Phys. Rev. B 37 (1988) 2251.  
 [16] P.H. Frings, J.J.M. Franse, F.R. de Boer and A. Menovsky, J. Magn. Magn. Mat. 31-34 (1983) 240.  
 [17] A.I. Goldman, S.M. Shapiro, G. Shirane, J.L. Smith and Z. Fisk, Phys. Rev. B 33 (1986) 1627.  
 [18] H.A. Mook, B.D. Gaulin, G. Aeppli, Z. Fisk and J.L. Smith, Bull. Am. Phys. Soc. 32 (1987) 594.  
 [19] G. Aeppli, E. Bucher, G. Shirane, J.L. Smith and Z. Fisk, Phys. Rev. B 32 (1985) 7579.  
 [20] S.L. Cooper, R.T. Demers, M.V. Klein, Z. Fisk and J.L. Smith, Physica B 135 (1985) 49.  
 [21] H. Brenten, E. Zirngiebl, M.S. Wire, S. Blumenröder, G. Pofahl, G. Güntherodt and Z. Fisk, Solid State Commun. 62 (1987) 387.  
 [22] G. Güntherodt, E. Zirngiebl, R. Mock, S. Blumenröder and H. Brenten, Proc. Int. Conf. on Magnetism, Paris ICM '88) to be published.  
 [23] G. Aeppli, H. Yoshizawa, Y. Enodh, E. Bucher, J. Hufnagl, Y. Ōnuki and T. Komatsubara, Phys. Rev. Lett. 57 (1986) 122.  
 [24] A. Auerbach, J.H. Kim, K. Levin and M.R. Norman, Phys. Rev. Lett. 60 (1988) 623.