Title

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Journal

Solid State Communications, 62(6)

ISSN

0038-1098

Authors

Brenten, H
Zirngiebl, E
Wire, MS
et al.

Publication Date

1987-05-01

DOI

10.1016/0038-1098(87)91039-8

Solid State Communications, Vol.62,No.6, pp.387-390, 1987.
Printed in Great Britain.

0038-1098/87 $3.00 + .00
Pergamon Journals Ltd.

RAMAN SCATTERING FROM SPIN FLUCTUATIONS AND PHONONS IN THE HEAVY-FERMION SUPERCONDUCTOR UPt_3

H. Brenten, E. Zirngiebl[*], M.S. Wire, S. Blumenröder, G. Pofahl
and G. Güntherodt
II. Physikalisches Institut[+], Universität zu Köln, 5000 Köln 41,
Federal Republic of Germany

Z. Fisk
Los Alamos National Laboratory, Los Alamos, N.M. 87545, USA

(Received 22 January 1987 by B. Mühlschlegel)

Quasielastic scattering from spin fluctuations has been observed in UPt_3 by Raman spectroscopy. The experiments for wave vectors $q \approx 0$ show a nearly temperature independent linewidth for $5\ K < T < 300\ K$. Complementary to neutron scattering results this establishes the q independence of the spin relaxation rate, indicating the localized nature of the spin fluctuations. A Raman-active phonon near 79 cm^{-1} (10 meV) shows a drastic increase in linewidth with decreasing temperature, demonstrating strong electron-phonon coupling .

There has been considerable interest recently in the unusual properties of heavy fermion materials[1] such as UPt_3[1,2], which is a superconductor and exhibits spin fluctuations. The latter have been inferred from specific heat[2], magnetic susceptibility[3], and resistivity measurements[4]. From neutron scattering measurements[5] on polycrystals of UPt_3 one has deduced an energy scale of the spin fluctuations of about 9 meV (100K) independent of momentum transfers $q \geq 1$ Å^{-1}. Fermi liquid theory predicts this energy scale to be determined by $v_F \cdot q$, where v_F is the Fermi velocity. An important question is whether this linear q dependence can be observed in $q \approx 0$ Raman scattering. Moreover, since it has been suggested that the electron pairing in heavy fermion superconductors may not be due to the usual BCS-type electron-phonon interaction[6], any phonon spectroscopy is highly desirable.

In UPt_3 we have observed by means of Raman spectroscopy (a true q = 0 probe) quasielastic scattering from spin fluctuations and a temperature dependent, anomalous broadening of the linewidth of the A_{1g} (Γ_1^+) phonon. The result for the spin fluctuations complements neutron scattering investigations and establishes the q independence of the spin relaxation rate. In analogy to findings in A15 superconductors[7] we observe an increasing broadening of the phonon mode with decreasing temperature, reflecting strong electron-phonon coupling.

UPt_3 crystallizes in the hexagonal Ni_3Sn-structure (space group D^4_{6h}, $P6_3/mmc$). The sample was made by arc melting of the pure elements on a water-cooled copper hearth in a zirconium-gettered argon atmosphere. Raman measurements have been carried out between 5 and 300 K, using 5309 Å Kr^+-laser excitation with 150 mW incident power. The Raman spectra were taken on a fractured surface of the polycrystalline sample where the laser beam was focussed onto a (001) cleavage plane of a single crystal grain.

Fig. 1 shows Raman spectra of UPt_3 at 300, 77 and 5 K, obtained in backscattering geometry using perpendicular polarizations of incident and scattered light. This choice of polarizations reduces (100 % in the case of ideal polarizations) the fraction of light scattered elastically due to surface imperfections and strongly indicates the quasielastic scattering extending beyond 200 cm^{-1} in Fig. 1 to be of magnetic origin[8]. The elastically scattered light has been accounted for by a Lorentzian fit to the laser line and shows a cut-off near ± 30 cm^{-1}. Moreover, elastic scattering should show no temperature dependence, i.e. no increasing Stokes/anti-Stokes asymmetry with decreasing temperature as seen in Fig. 1. Finally, the residual Stokes scattering intensity at 5 K cannot be due to second order phonon difference processes since these processes die out very rapidly with decreasing temperature[8]. The quasielastic scattering intensity in Fig. 1 (given by the hatched area) is interpreted as due to spin fluctuations as will be described below.

According to Ref. 8 the scattering cross section for scattering by fluctuating magnetiza-

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[*] Present address: Los Alamos National Laboratory, Los Alamos, N.M. 87545, USA

[+] Work supported by Deutsche Forschungsgemeinschaft, SFB 125

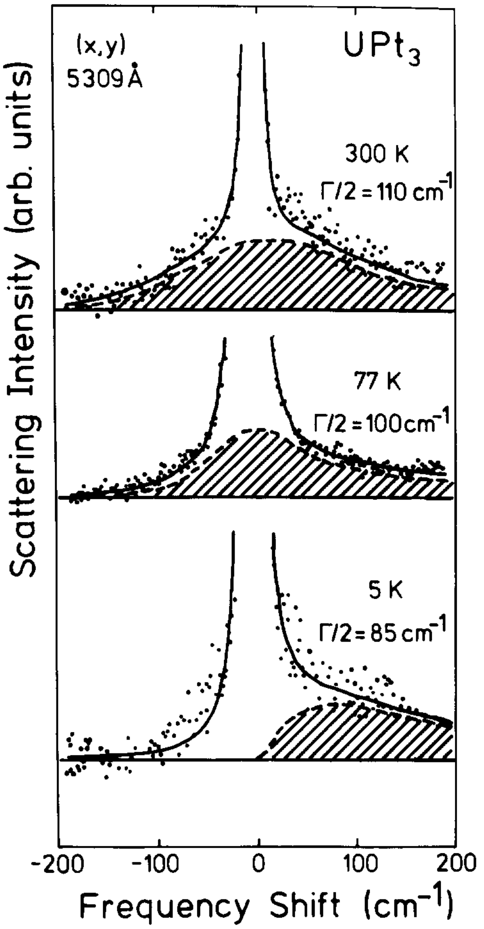

Fig 1:

Temperature dependence of the Raman spectra of UPt_3 for perpendicular polarizations of incident (5309 Å) and scattered light. The hatched area shows the contribution due to spin fluctuations with half width at half maximum, $\Gamma/2$, determined from the fit using Eq. (4).

tion components can be expressed for Stokes scattering as

$$\frac{d^2\sigma}{d\Omega d\omega_S} \sim \sum_{i,j} (\epsilon_S \times \epsilon_I)^i (\epsilon_S \times \epsilon_I)^j \times \langle M^i M^j \rangle \qquad (1)$$

where ϵ_S, ϵ_I are the polarization vectors of incident and scattered photons, M^i, M^j are the cartesian components (i,j = x,y,z) of the sample magnetization M. Using the fluctuation-dissipation theorem[9] yields

$$\frac{d^2\sigma}{d\Omega d\omega_S} \sim \sum_{i,j} (\epsilon_S \times \epsilon_I)^i (\epsilon_S \times \epsilon_I)^j \times \frac{\chi''_{ij}(q,\hbar\omega)}{1 - \exp(-\hbar\omega/kT)} \qquad (2)$$

where χ''_{ij} is the imaginary part of the susceptibility. If we are dealing with uncorrelated spins, i.e. which relax exponentially in time with a q-independent relaxation rate Γ, it should be possible to separate χ''_{ij} into q and ω dependent factors. We use a Kramers-Kronig relation and assume

$$\chi''(q,\omega) = \pi\omega\chi'(q,0)\, P(\omega) \qquad (3)$$

where $P(\omega)$ and χ' denote the spectral function and the real part of the susceptibility, respectively.

We now assume a Lorentzian for $P(\omega)$, since we expect the 5f spins to fluctuate statistically. Thus we find the magnetic scattering intensity $I(\omega)$ as a function of the frequency shift ω to be given by

$$I(\omega) = A * \frac{\hbar\omega}{1 - \exp(-\hbar\omega/kT)} * \frac{\Gamma}{(\omega^2 + (\Gamma/2)^2)} + S(\omega) \qquad (4)$$

where A denotes the magnetic scattering intensity and $S(\omega)$ is the surface roughness scattering contribution (Lorentzian line shape), which is superimposed on the magnetic excitations. We have fit our data to Eq. (4) and subtracted $S(\omega)$ to yield the pure spin fluctuation contribution to the spectra (shown by the hatched area in Fig. 1). From the fit, the half width $\Gamma/2$ at half maximum of the quasi-elastic intensity is found to decrease from 110 cm^{-1} (13.6 meV) at 300 K to 85 cm^{-1}(10.5 meV) at 5 K.

These Raman measurements, complementing the neutron scattering results for large q ($>$ 1 $Å^{-1}$), establish the q independence of the spin relaxation rate within the large error bars of the neutron data. Hence we conclude that the spin fluctuations are localized in space as expected for well separated U atoms. Coherence and Fermi liquid effects, which lead to a q dependence of the spin relaxation rate for $q \to 0$, can be ruled out for temperatures down to 5 K as a consequence of our results. In addition to our findings for UPt_3 we have reanalyzed magnetic Raman scattering data of UBe_{13}, reported[10] recently for 40 K and 350 K. We were able to fit these data to the form of Eq. (4) and the result is shown in Fig. 2 by the hatched area. For both temperatures $\Gamma/2$ is found to be 110 ($\pm$20) cm^{-1} (13.6 meV), in good agreement with neutron scattering results (13 $\pm$ 2 meV)[11].

In addition to the magnetic Raman scattering in UPt_3, we were able to investigate one of the five Raman-active phonons. Fig. 3 shows unpolarized Raman spectra of UPt_3 at 300 K, 77 K, and 5 K. The phonon mode at 79 cm^{-1} appears only for parallel polarizations of the incident and scattered light and has A_{1g} symmetry. The linewidth of the phonon peak increases with decreasing temperature from 10 cm^{-1} at 300 K to 40

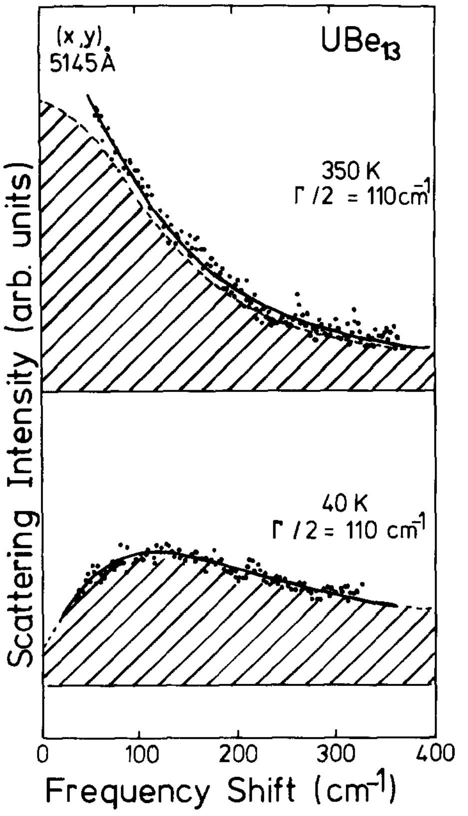

Fig 2:

Raman spectra of UBe_{13} from Ref. 10, obtained with perpendicular polarizations of incident (5145 Å) and scattered light. The hatched area shows the contribution due to spin fluctuations with halfwidth at half maximum, $\Gamma/2$, determined from the fit using Eq. (4).

cm^{-1} at 5 K. The weak excitation near 150 cm^{-1} appearing in the spectrum for 77 K is attributed to a scattering process of second order.

We ascribe the phenomenon of increasing width of the optical phonon in UPt_3 to strong electron-phonon coupling, in analogy to observations in superconducting A15 compounds[7]. Assuming that this discrete phonon interacts with a continuum of electronic excitations leads to a phonon linewidth, which is proportional to the square of the electron-phonon coupling constant[7]. Hence we conclude an increase in the electron-phonon coupling constant with decreasing temperature. The anomalous behavior observed for the phonon mode of A_{1g} symmetry is consistent with the dominant deformation potential type electron-phonon coupling concluded from elastic anomalies[12]. This most likely explains the strong Raman scattering intensities of the A_{1g} phonon compared to the other unobservable Raman-active modes (E_{1g}, 3 E_{2g}).

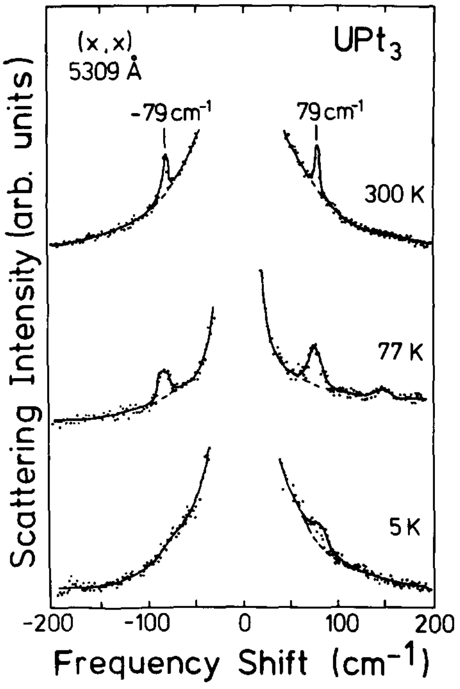

Fig 3:

Raman spectra of UPt_3 for parallel polarizations of incident (5309 Å) and scattered light. The scattering intensity of the A_{1g} phonon is separated from the background by the dashed lines.

Further evidence for strong electron-phonon coupling is given by the occurence of the two phonon-process near 150 cm^{-1} for intermediate temperatures between 300 K and 5 K, i.e. for 77 K. On the one hand the electron-phonon coupling increases with decreasing temperature, but on the other hand the phonon population decreases drastically according to the Boltzmann factor[8].

In conclusion, we would like to emphasize that spin fluctuations of heavy fermion U-compounds can be seen by Raman scattering. This is indicative of large magneto-optical coupling already observed for non-heavy fermion U-compounds[13] and perhaps characteristic for any U-compound. This may be due to the large spin-orbit coupling in U itself. Moreover the $q \approx 0$ measurements of the spin fluctuations in UPt_3 and UBe_{13} show the same characteristic energy scale as the neutron data for $q > 1$ Å^{-1}. The absence of any Fermi liquid behavior of the spin fluctuation rate remains an openquestion. Any proposed theory for heavy fermions must be able to account for this disagreement. Furthermore our measurements give evidence for a strong electron-phonon coupling, which has to be taken into acccount by any theory explaining the unusual superconductivity. A coupling of the observed A_{1g} phonon of UPt_3 to the electronic system seems even more probable because its frequency is of the same order as the spin fluctuation rate. The $q = 0$ A_{1g} phonon coincides with a flat optical phonon branch near 10 meV (80 cm^{-1}) measured for $q > 1$ $Å^{-1}$ (Ref. 5) and establishes the existence of an Einstein mode in UPt_3. The coincidence of an Einstein mode with the spin relaxtion rate is a striking similarity between UPt_3 and UBe_{13}[11,14].

Acknowledgements:
We would like to thank E. Holland-Moritz, R. Mock, and B. Hillebrands for stimulating discussions. The research in Los Alamos was supported by the U.S. Department of Energy.

References

1) G.R. Stewart, Rev. Mod. Phys., **56**, 755, (1984).
2) G.R. Stewart, Z. Fisk, J.O. Willis, and J.L. Smith, Phys. Rev. Lett. **52**, 679 (1984); A. de Visser, J.J.M. Franse, A. Menovsky, and T.T.M. Palstra, Physica **127B**, 442 (1984).
3) P.H. Frings, J.J.M. Franse, F.R. de Boer, and A. Menovsky, J. Magn. Magn. Mat. **31-34**, 240 (1983).
4) A. de Visser, J.J.M. Franse, and A. Menovsky, J. Magn. Magn. Mat. **43-47**, 43 (1984).
5) G. Aeppli, E. Bucher, and G. Shirane , Phys. Rev. **B32**, 7597 (1985).
6) P.W. Anderson, Phys. Rev. **30**, 1549 (1984); C.M. Varma, in _Theory of Heavy Fermions and Valence Fluctuations_, edited by T. Kasuya and T. Saso (Springer-Verlag, Berlin, 1985), p. 227; P.A. Lee, T.M. Rice, J.W. Serene, L.S. Sham, and J.W. Wilkins, Comments Cond. Mat. Phys. B12, 99 (1986).
7) H. Wipf, M.V. Klein, B.S. Chandrasekhar, T.H. Geballe, and J.H. Wernick, Phys. Rev. Lett. **41**, 1752 (1978).
8) W. Hayes and R. Loudon, _Scattering of Light by Crystals_, (Wiley ,New York ,1978), p. 244.
9) R.M. White, _Theory of Magnetism_, (McGraw Hill, New York, 1970), p. 213.
10) S.L. Cooper, R.T. Demers, M.V. Klein, Z. Fisk, and J.L. Smith, Physica **135B**, 49 (1985).
11) A.I. Goldman, I.M. Shapiro, G. Shirane, J.L. Smith and Z. Fisk, Phys. Rev. **B33**, 1627 (1986).
12) M. Yoshizawa, B. Lüthi, T. Suzuki, B. Renker, A. de Visser, P. Frings, and J.J.M. Franse J. Magn. Magn. Mat. **52**, 413,(1985).
13) J. Schoenes, in _Handbook on the Physics and Chemistry of the Actinides_, edited by A.J. Freeman and G.H. Lander (North Holland, Amsterdam, 1984), p. 341.
14) B. Renker, F. Gompf, W. Reichardt, H. Rietschel, and J.B. Suck, Phys. Rev. **B32**, 1859 (1985).