UC Irvine
UC Irvine Previously Published Works

Title
Polarized light scattering study of UPt3

Permalink
https://escholarship.org/uc/item/0141z87v

Journal
Physical Review B, 37(4)

ISSN
2469-9950

Authors
Cooper, SL
Klein, MV
Fisk, Z
et al.

Publication Date
1988-02-01

DOI
10.1103/physrevb.37.2251

Peer reviewed

Polarized light scattering study of UPt₃

S. L. Cooper and M. V. Klein

*Department of Physics and Materials Research Laboratory, University of Illinois at Urbana-Champaign,
1110 W. Green Street, Urbana, Illinos 61801*

Z. Fisk and J. L. Smith

Materials Science and Technology Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545

(Received 7 July 1987)

We have performed polarized Raman scattering experiments on single-crystal samples of UPt₃ in the temperature range 5 K $\leq T \leq$ 340 K. Four of the five Raman-allowed phonons have been observed, and their symmetries identified. None of these phonons is seen to demonstrate any anomalous behavior, contrary to previous Raman scattering results. Additionally, quasielastic scattering from spin fluctuations has been observed, having the symmetry of the antisymmetric representation, A_{2g}, and displaying a slightly temperature-dependent half width at half maximum. The energy scale of this scattering is consistent with results reported earlier by both neutron scattering at $q \geq 1$ Å^{-1} and Raman scattering at $q = 0$ (where q is the momentum transfer), suggesting that the spin-fluctuation relaxation is q independent.

Since UPt₃ was recently shown to exhibit the coexistence of spin fluctuations and a superconducting transition, a great deal of effort has been devoted to its study.[1] Evidence for spin fluctuations in UPt₃ is principally suggested by a $T^3 \ln T$ term in the low-temperature specific heat, while a large specific heat jump at $T_c = 0.5$ K presages the superconducting transition. Neutron scattering,[2] and more recently Raman scattering,[3] studies have established the low-temperature energy scale (half width at half maximum, $\Gamma/2$) of the spin fluctuations to be roughly 80 cm^{-1} (10 meV), independent of momentum transfer q. This q independence seems to suggest that the spin fluctuations are not strongly correlated down to the lowest temperatures observed.

Additionally, anomalous phonon behavior has been reported in UPt₃, in the form of a dramatic broadening with decreasing temperature in the phonon identified as the A_{1g} breathing mode.[3] This behavior was cited as evidence for a strong deformation-potential coupling of this phonon to the electronic states of UPt₃.

In this paper, we report on a polarized-light-scattering investigation of single-crystalline UPt₃, in an effort to study further the magnetic and lattice excitations in this material, and in particular, to investigate the earlier report of strong electron-phonon coupling. The occurrence of strong coupling between a phonon and an electronic state demands not only that both excitations have comparable time scales, but also that the phonon has an appropriate symmetry for a strong modulation of the electronic configuration. Thus, the utility of polarized light scattering for extracting both excitation symmetry and energy information allows a careful consideration of possible strong electron-phonon coupling in UPt₃.

Our polarized Raman scattering study was conducted on oriented single-crystal samples of UPt₃, with both (0001) and (1010) exposed faces. These orientations were confirmed by Laue x-ray diffraction. Light scattering measurements were carried out using the 5145 Å line of an argon laser. The incident light was polarized along the different crystalline axes in order to isolate the excitation symmetries, and a triple stage monochromator with a cooled photomultiplier tube detector was used to disperse and collect the scattered light.

UPt₃ crystallizes in the hexagonal Ni₃Sn structure with space group D_{6h}^4 ($P6_3/mmc$), which has five Raman-active phonons. These phonons have the symmetries $A_{1g} + E_{1g} + 3E_{2g}$, and from the site symmetries of UPt₃, one can determine that the A_{1g} and E_{1g} phonons involve Pt atoms, while the E_{2g} phonons involve both Pt and U. The different symmetries can be selectively coupled to, and the symmetries of the observed excitations consequently identified by varying the scattering geometry of our light scattering experiment. Table I outlines the allowed excitation symmetries for each scattering geometry used. As the wave vector of light used in our investigation is a small fraction of the Brillouin zone, the excitations probed by light scattering are effectively at $q = 0$.

In our study, we have been able to observe and identify four of the five Raman-allowed phonons in UPt₃, all of which are shown in Fig. 1. The most conspicuous feature we observed is a very intense phonon at 150 cm^{-1}, which from the geometry used is clearly identified as the A_{1g} breathing mode of Pt atoms (top spectrum, large figure). This phonon may be associated with the optical mode ob-

TABLE I. Allowed excitation symmetries for various scattering geometries in UPt₃ [$\hat{x} = (1,0,0)$, $\hat{y} = (0,1,0.)$, $\hat{z} = (0,0,1)$, $\hat{y}' = 1/\sqrt{2}(1,-1,0)$].

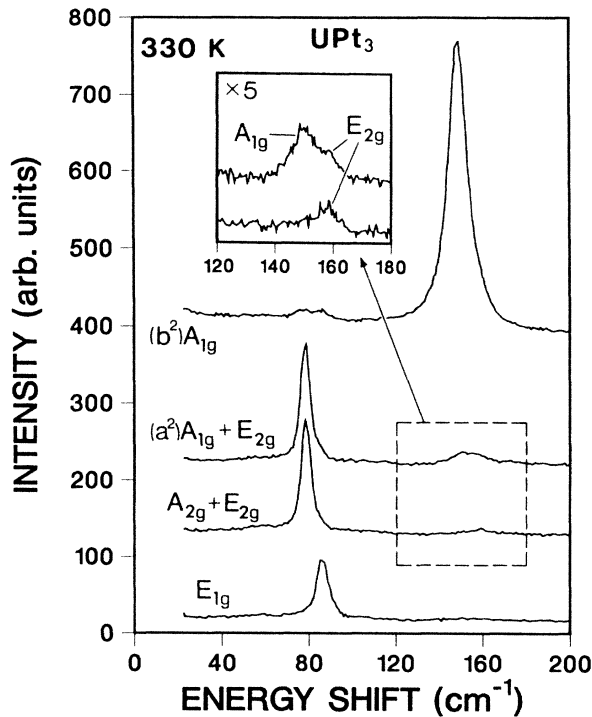| Propagation direction ($\mathbf{k}_i$)	Geometry ($\hat{e}_i, \hat{e}_s$)	Allowed symmetries
c axis	$(\hat{x}, \hat{x})$	$(a^2)A_{1g} + E_{2g}$
	$(\hat{x}, \hat{y})$	$A_{2g} + E_{2g}$
Basal plane	$(\hat{z}, \hat{z})$	$(b^2)A_{1g}$
	$(\hat{z}, \hat{y}')$	E_{1g}

FIG. 1. Phonon spectra of UPt₃ at 330 K with symmetries as shown. The inset is a 5× enlargement of the $(a^2)A_{1g}+E_{2g}$ and $A_{2g}+E_{2g}$ spectra between 120 and 180 cm^{-1}. The small feature around 80 cm^{-1} in the $(b^2)A_{1g}$ spectrum is E_{1g} and E_{2g} leakage due to imperfect alignment of $\mathbf{k}_i$ and $\hat{\mathbf{e}}_i$ along the crystalline axes. All of the spectra have been offset for clarity. Resolution: 3 cm^{-1}.

served at 20 meV in a recent optical reflectivity study.[4] The A_{1g} phonon is particularly notable in that it demonstrates a large anisotropy in its polarizability tensor, given by

$$a(A_{1g}) = \begin{bmatrix} a & 0 & 0 \\ 0 & a & 0 \\ 0 & 0 & b \end{bmatrix} .$$

The weakness of the A_{1g} mode in the $(a^2)A_{1g}+E_{2g}$ spectrum (second spectrum, large figure) compared to that in the $(b^2)A_{1g}$ spectrum (top spectrum, large figure) indicates a large polarizability of this phonon along the c axis compared to that in the basal plane (i.e., $b \gg a$).

Our symmetry assignment also contradicts that made in an earlier Raman scattering study[3] which ascribed the A_{1g} symmetry to the phonon near 79 cm^{-1}. As shown in Fig. 1, the appearance of the 79 cm^{-1} phonon in both the $A_{1g}+E_{2g}$ and $A_{2g}+E_{2g}$ spectra clearly indicates its symmetry to be E_{2g}. This symmetry analysis raises questions regarding the earlier report of strong electron-phonon coupling involving this mode, as will be discussed later. Our study shows, furthermore, that the broad feature near 150 cm^{-1} in the $A_{1g}+E_{2g}$ spectra is not two-phonon scattering, as reported in Ref. 3, but is rather a doublet of the A_{1g} phonon and one of the E_{2g} phonons (see inset, Fig. 1). Indeed, both peaks are found to demonstrate a temperature dependence consistent with one-phonon

scattering. The remaining phonon we observe is the E_{1g} mode at 86 cm^{-1}, illustrated in the bottom spectrum of Fig. 1. The results of our phonon symmetry assignments have been tabulated in Table II.

Figure 2 illustrates the temperature dependence of the $A_{2g}+E_{2g}$ spectrum, in which the two observed E_{2g} phonons, as well as broad quasielastic scattering from spin fluctuations (hatched area), are evident. One clearly observes the growing Stokes–anti-Stokes asymmetry in both the E_{2g} phonon and the spin-fluctuation scattering as the temperature is reduced. Part of the elastically scattered contribution has also been included for comparison (truncated peak at 0 cm^{-1}), and is found to be confined between ±15 cm^{-1}.

The central peak which we observe in the $A_{2g}+E_{2g}$ spectrum is absent in all other scattering geometries, which indicates that the quasielastic scattering has the symmetry of the totally antisymmetric representation of the UPt₃ space group, A_{2g}, characteristic of magnetic scattering. The observed quasielastic scattering can be nicely fit to the power spectrum

$$S(\omega) \propto [1+n(\omega)] \frac{\omega\Gamma}{(\frac{1}{2}\Gamma)^2+\omega^2} ,$$

which reflects the simple relaxational response of the spin fluctuations. In the power spectrum above, $n(\omega)$ is the Bose factor, and $\Gamma/2$ is the temperature-dependent half width at half maximum. The quasielastic contribution is illustrated at each temperature by the hatched areas in Fig. 2.[5] The quasielastic half widths extracted from this fit are found to have a slight temperature dependence, which ranges from a value of 140 cm^{-1} at 340 K, to 90 cm^{-1} at 5 K. The latter half width is consistent with values reported near 5 K by neutron[2] and Raman[3] scattering. The comparable linewidths noted between the $\mathbf{q}=0$ Raman studies and the neutron scattering results at much higher $\mathbf{q}$ suggest that the spin-fluctuation relaxation is $\mathbf{q}$ independent down to 5 K. However, neutron scattering studies[6] have observed a $\mathbf{q}$ dependence in the spin fluctuation intensity, i.e., the static susceptibility $\chi(\mathbf{q})$, which provides evidence that the spin fluctuations do in fact have antiferromagnetic correlations in UPt₃.

The observation of $\mathbf{q}=0$ spin fluctuations is particularly noteworthy inasmuch as noninteracting Fermi-liquid theory sets the energy scale of the imaginary part of the magnetic susceptibility at $v_F q - (v_F/p_F)q^2$ (where v_F and p_F are the Fermi velocity and momentum, respectively). Therefore, finite spin-fluctuation strength at $\mathbf{q}=0$ illustrates the absence of simple Fermi-liquid behavior in UPt₃. A zone-center spin-fluctuation contribution is al-

TABLE II. Observed phonon frequencies in UPt₃ at 330 K.

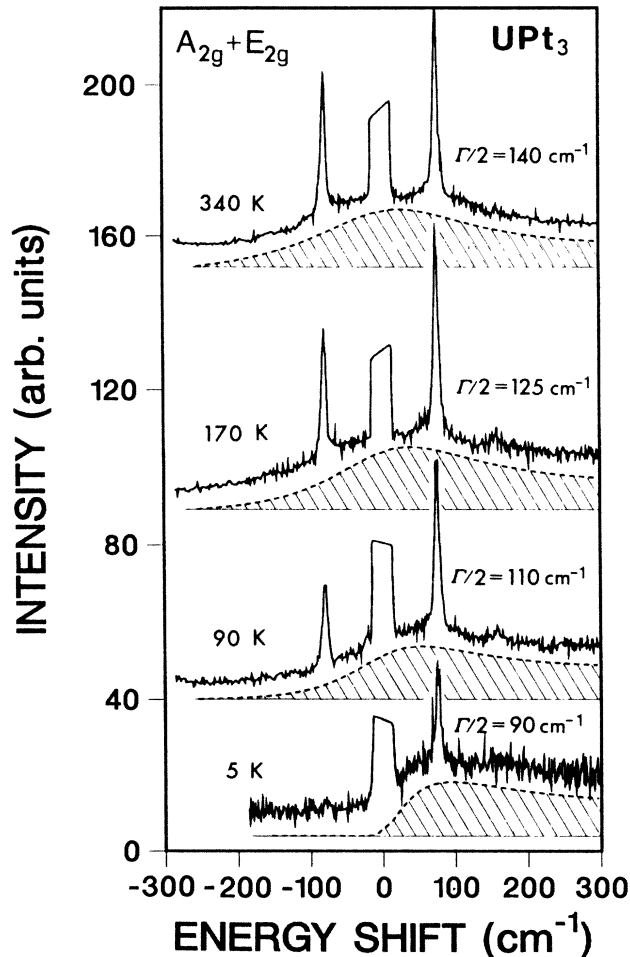| Phonon	Frequency (cm^{-1}) (Resolution: 3 cm^{-1})
E_{2g}	78
E_{1g}	86
A_{1g}	150
E_{2g}	158

FIG. 2. Temperature dependence of the $A_{2g}+E_{2g}$ spectrum. The hatched area shows the contribution to the spectrum from spin fluctuations, with the half widths obtained from the fit as shown. The dashed fit has been offset from the data for clarity.
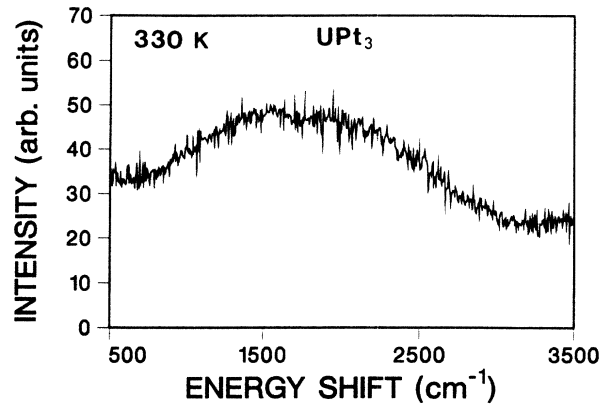

FIG. 3. Electronic scattering with mixed symmetry, attributed to crystal-field excitations. Temperature: 330 K.

tions reported by Raman scattering.[7]

Finally, another notable feature in Fig. 2 is the absence of broadening in the 79 cm^{-1} phonon linewidth as was observed in Ref. 3, which questions any suggestion of strong electron-phonon coupling involving this mode. Indeed, the lack of broadening is more consistent with one's expectation that a lower-symmetry phonon (in this case E_{2g}) should have a small deformation-potential coupling to the electronic system. This point is further illustrated by comparing the E_{2g} phonon's scattering intensity to that of the much larger A_{1g} mode at 150 cm^{-1}, reflecting the substantially greater breathing mode coupling to the electronic configuration (see Fig. 1). It should be noted, however, that no anomalous temperature dependence is observed in any of the phonons seen in our investigation.

In conclusion, we have observed quasielastic scattering from spin fluctuations in UPt$_3$, corroborating earlier reports by both neutron and Raman scattering. As in the latter study, we conclude that the quasielastic linewidth is q independent and only slightly temperature dependent. We have also been able to observe, and to identify the symmetries of, four of the five Raman-active phonons. We have been unable, however, to confirm earlier reports of strong electron-phonon coupling of the 79 cm^{-1} phonon. We find this fact to be consistent with the low-symmetry determined for this mode, which is expected to hinder a strong modulation of the electronic configuration.

lowed in heavy electron materials because the magnetization is not conserved in these systems, due to the presence of a strong spin-orbit interaction.

Despite the apparent evidence for correlations in UPt$_3$ (Ref. 6), the localized nature of the f electron levels in UPt$_3$ is also observed in our Raman-scattering study, where extremely broad inelastic scattering due to excitations between well defined crystal-field levels is observed between 1000 and 3000 cm^{-1} (see Fig. 3). This scattering demonstrates mixed symmetry and coincides with electronic scattering centered at 270 meV in an earlier optical reflectivity study,[4] as well as with crystal-field excita-

background from broad inelastic scattering, is also included in the fits displayed in Fig. 2. This term, however, makes a negligible contribution to the hatched (quasielastic) areas shown.

[1]G. R. Stewart, Rev. Mod. Phys. **56**, 755 (1984).

[2]G. Aeppli, E. Bucher, and G. Shirane, Phys. Rev. B **32**, 7597 (1985).

[3]H. Brenten, E. Zirngiebl, M. S. Wire, S. Blumenröder, G. Pofahl, G. Güntherodt, and Z. Fisk, Solid State Commun. **62**, 387 (1987).

[4]F. Marabelli, G. Travaglini, P. Wachter, and J. J. M. Franse, Solid State Commun. **59**, 381 (1986).

[5]A very small linear term, representing contributions to the

[6]G. Aeppli (unpublished).

[7]E. Zirngiebl (unpublished); see also, S. Blumenröeder, H. Brenten, G. Güntherodt, E. Zirngiebl, and Z. Fisk, Bull. Am. Phys. Soc. **32**, 719 (1987).