Polarized light scattering study of UPt$_3$

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We have performed polarized Raman scattering experiments on single-crystal samples of UPt$_3$ 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 UPt$_3$ 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. Evidence for spin fluctuations in UPt$_3$ 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, and more recently Raman scattering, 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 UPt$_3$, in the form of a dramatic broadening with decreasing temperature in the phonon identified as the $A_{1g}$ breathing mode. This behavior was cited as evidence for a strong deformation-potential coupling of this phonon to the electronic states of UPt$_3$.

In this paper, we report on a polarized-light-scattering investigation of single-crystalline UPt$_3$, 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 UPt$_3$.

Our polarized Raman scattering study was conducted on oriented single-crystal samples of UPt$_3$, 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.

UPt$_3$ crystallizes in the hexagonal Ni$_3$Sn structure with space group $D_{6h}^5$ (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 UPt$_3$, 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 UPt$_3$, 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](image)

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

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

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

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., $b \approx a$).

Our symmetry assignment also contradicts that made in an earlier Raman scattering study\textsuperscript{3} which ascribed the $A_{1g}$ symmetry to the phonon near 79 cm\textsuperscript{-1}. As shown in Fig. 1, the appearance of the 79 cm\textsuperscript{-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\textsuperscript{-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\textsuperscript{-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\textsuperscript{-1}), and is found to be confined between ±15 cm\textsuperscript{-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 UP\textsubscript{T3} space group, $A_{2g}$, characteristic of magnetic scattering. The observed quasielastic scattering can be nicely fit to the power spectrum

$$S(\omega) = \frac{\omega \Gamma}{(\frac{1}{2} \Gamma)^2 + \omega^2},$$

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.\textsuperscript{3} The quasielastic half widths extracted from this fit are found to have a slight temperature dependence, which ranges from a value of 140 cm\textsuperscript{-1} at 340 K, to 90 cm\textsuperscript{-1} at 5 K. The latter half width is consistent with values reported near 5 K by neutron\textsuperscript{2} and Raman\textsuperscript{3} scattering. The comparable linewidths noted between the $q = 0$ Raman studies and the neutron scattering results at much higher $q$ suggest that the spin-fluctuation relaxation is $q$ independent down to 5 K. However, neutron scattering studies\textsuperscript{6} have observed a $q$ dependence in the spin fluctuation intensity, i.e., the static susceptibility $\chi(q)$, which provides evidence that the spin fluctuations do in fact have antiferromagnetic correlations in UP\textsubscript{T3}.

The observation of $q = 0$ spin fluctuations is particularly noteworthy inasmuch as noninteracting Fermi-liquid theory sets the energy scale of the imaginary part of the magnetic susceptibility at $\nu_F q -(\nu_F/p_F)q^2$ (where $\nu_F$ and $p_F$ are the Fermi velocity and momentum, respectively). Therefore, finite spin-fluctuation strength at $q = 0$ illustrates the absence of simple Fermi-liquid behavior in UP\textsubscript{T3}. A zone-center spin-fluctuation contribution is al-

<table>
<thead>
<tr>
<th>Phonon</th>
<th>Frequency (cm\textsuperscript{-1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_{2g}$</td>
<td>78</td>
</tr>
<tr>
<td>$E_{1g}$</td>
<td>86</td>
</tr>
<tr>
<td>$A_{1g}$</td>
<td>150</td>
</tr>
<tr>
<td>$E_{2g}$</td>
<td>158</td>
</tr>
</tbody>
</table>

Table II. Observed phonon frequencies in UP\textsubscript{T3} at 330 K.
followed 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, as well as with crystal-field excitations reported by Raman scattering.

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 $E_2$) should have a small deformation-potential coupling to the electronic system. This point is further illustrated by comparing the $E_2$ 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. If confirmed, this fact could be consistent with the low-symmetry determined for this mode, which is expected to hinder a strong modulation of the electronic configuration.

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5. A very small linear term, representing contributions to the 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.