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RAMAN SCATTERING STUDY OF UBe_{13}

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Raman scattering experiments on single-crystal UBe_{13} have been performed at temperatures $2 \leq T \leq 300$ K. We observe Raman-active optical phonons consistent, in number and symmetry, with those allowed by the space and site group symmetries previously determined for UBe_{13} (space group O_h^6-Fm3c). Certain phonon modes are seen to have strongly anomalous behaviour with decreasing temperature.

We have, in addition, observed inelastic scattering from magnetic excitations, which we identify as spin fluctuations. The temperature dependence of these excitations is discussed.

1. Introduction

It was recently discovered [1] that the exotic material UBe_{13} manifests such seemingly incongruous features as, for example, a Kondo-like electrical resistivity at higher temperatures, but a superconducting ground state. In order to gain insight into such anomalous properties, it is important to investigate the various excitations prevalent in this system, as well as to study their temperature dependence.

An important technique, in this regard, is that of polarized Raman scattering, which allows one to identify the symmetry and energy of certain excitations in the system, and consequently identify the excitations themselves. In this paper, we will discuss the preliminary results of such an investigation of UBe_{13}.

2. Experimental

We conducted our experiments on single-crystal samples of UBe_{13}, using either the 4880 or the 5145 Å line of an argon laser as an excitation source. A rotating polaroid filter is used to select out a specific polarization of the scattered light, which is then dispersed and detected using a triple-grating monochromator and a photomultiplier tube detector. To permit low temperature studies, our system includes a liquid helium cryostat, capable of maintaining temperatures as low as 2 K.

3. Factor group analysis

From the previously observed O_h^6 crystal structure of UBe_{13} [2], we determined, using a factor group analysis, that 22 of the 81 optical phonons are Raman-active. These include four modes each of E_g and T_{2g} symmetry and two of A_{1g} symmetry (in agreement with the earlier analysis of ref. 3). All of these modes involve only those Be atoms which lie on the faces of a cube surrounding the U atom (Be^{II}(96)). The U(8) and Be^{I}(8) atoms, sitting at much higher symmetry positions in the crystal, have no Raman-activity. Using a backscattering geometry and isolating specific polarizations of the incident and collected light allow us to

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couple different to components of the Raman tensor, and thereby identify these phonons in our spectra. Table I presents the allowed light-scattering symmetries, for the group O_h^6, in the different geometries used. A_{1g}, E_g and T_{2g} all represent symmetric tensors, while T_{1g} is antisymmetric.

Table I

Geometry (e_i, e_s)	Allowed symmetries
(x, x)	$A_{1g} + E_g$
(x, y)	$T_{2g} + T_{1g}$
(x', x')	$A_{1g} + \frac{1}{4}E_g + T_{2g}$
(x', y')	$\frac{3}{4}E_g + T_{1g}$

$x = (1, 0, 0)$, $y = (0, 1, 0)$,
$x' = (1/\sqrt{2})(1, 1, 0)$, $y' = (1/\sqrt{2})(1, -1, 0)$.

4. Results and discussion

All of the Raman-active modes delineated by our factor group analysis are observed within an energy range of 300–650 cm^{-1}. Figs. 1 and 2 illustrate, and table II summarizes our results at room temperature. (Note: all figures have been offset. Therefore, the scales are relevant only for the peak heights relative to background.) The absence of the lowest T_{2g} mode in the 5145 Å spectra is attributable to a higher luminescence background at 5145 Å than exists in the 4880 Å spectra, resulting in a loss of signal.

Also indicated in figs. 1 and 2 is a broad "tail" of low energy scattering extending to about 330 cm^{-1}. This feature is evident only in the depolarized spectra, (x, y) and (x', y') (that is, in those geometries in which the collected polarization is perpendicular to the incident polarization), and therefore displays purely antisymmetric character, T_{1g}, as seen from table I. Excitations of such symmetry are indicative of magnetic scattering, presenting strong evidence that we are observing spin fluctuations of the 5f electrons in UBe_{13}.

Table II

Phonon	cm^{-1} (resolution: 2 cm^{-1})
T_{2g}	320
E_g	347
T_{2g}	424
E_g	432
T_{2g}	482
A_{1g}	522
E_g	539
A_{1g}	593
T_{2g}	595
E_g	637

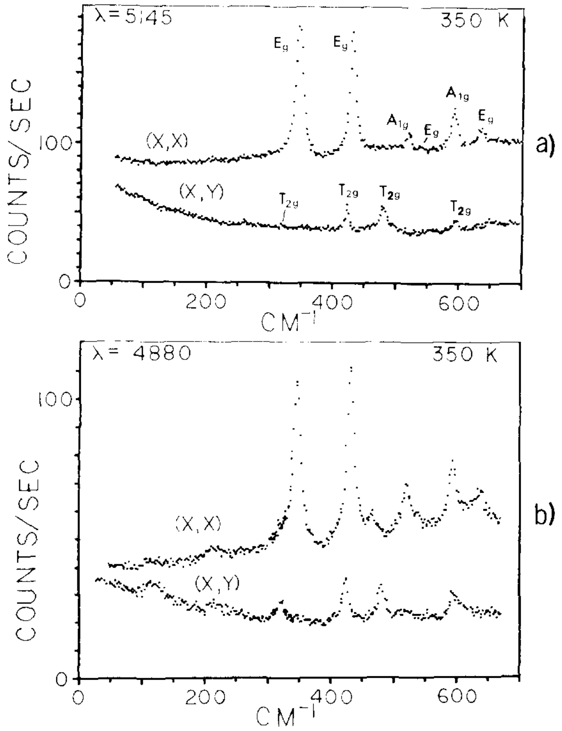

Fig. 1. $A_{1g} + E_g = (x, x)$, and $T_{2g} + T_{1g} = (x, y)$ Raman spectra for UBe_{13} using (a) 5145 Å excitation line; (b) 4880 Å excitation line. All spectra taken at 350 K. All figures have been offset.

At lower temperatures, several anomalies in the phonon spectrum develop. This point is illustrated in fig. 3. The T_{2g} modes at 320 and 424 cm^{-1} are seen to harden by about 27 and 8 cm^{-1}, respectively, soon after cooling below room temperature. This is possibly due to the thermal variation in the size of the unit cell. Additionally, all of the T_{2g} modes broaden and gain intensity with decreasing temperature. This temperature dependence is again most evident in

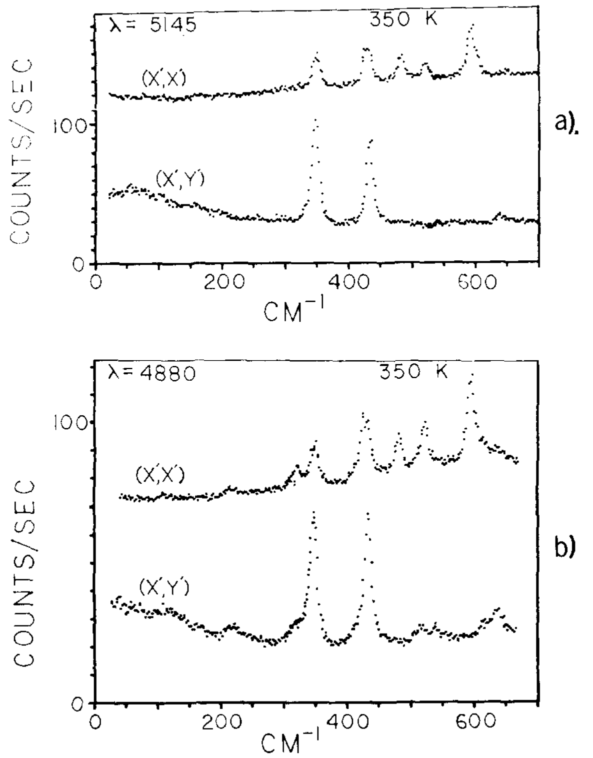

Fig. 2. $A_{1g} + \frac{1}{4}E_g + T_{2g} = (x', x')$, and $\frac{3}{4}E_g + T_{1g} = (x', y')$ Raman spectra using (a) 5145 Å excitation line; (b) 4880 Å excitation line. $T = 350$ K. All figures have been offset.

the lowest frequency T_{2g} mode. There seem to be no significant anomalies in the other Raman-active phonons, evidenced by fig. 4.

One possible explanation for the broadening of the T_{2g} peaks is that of spin defect-induced scattering. As temperature drops, the thermal fluctuations become less important than the spin fluctuations, and for spin fluctuations rates on the order of phonon frequencies, the phonons see a different spin environment each oscillatory period. Such dynamic spin defects effectively relax the kinematic constraint (i.e. $q = 0$) of the Raman scattering process, leading to a broadening of the phonons. This process may share features discussed previously for Eu_3S_4 [4]. The T_{2g} modes, which from our factor group analysis involve motion in the x, y, and z directions, would be more strongly affected (i.e. see more disorder) than would either the E_g or A_{1g} modes, which have only planar motion. Furthermore, since only phonons which oscillate on a time scale which is slower than that of the spin fluctuations will be affected, one would expect the lower frequency T_{2g} modes to be the most affected, as is observed.

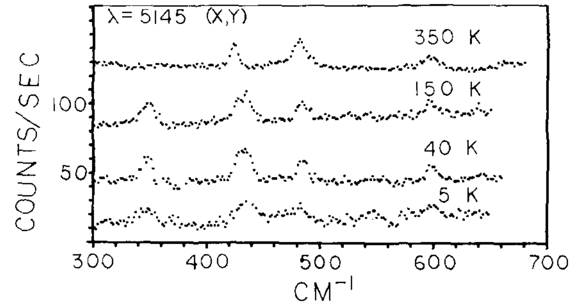

Fig. 3. Temperature dependence of the $T_{2g} + T_{1g}$ spectra between 300–700 cm^{-1}. The spectrum at 5 K was taken with 90% less laser power than in the other spectra and has been scaled (×10). All figures have been offset.

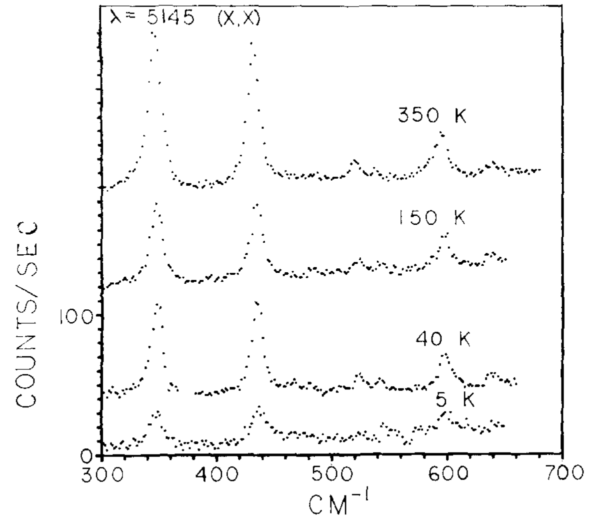

Fig. 4. Temperature dependence of the $A_{1g} + E_g$ spectra between 300–700 cm^{-1}. The 5 K spectrum was taken with 90% less power and has been scaled (×10). All figures have been offset.

An alternative possibility is that the broadened peaks represent unresolved splittings of the triply degenerate T_{2g}, due to a structural phase transition. Indeed, the possibility of such a transition has been recently proposed [5]. It should be noted, however, that such a transition should have a strong signature in the Raman scattering spectra, for any breaking of crystal symmetry would likely Raman-activate the U and/or Be^{I} atoms and give rise to new modes. As yet, no such modes have been observed. The resolution of this question must clearly await a more complete investigation.

We also observe scattering from spin fluctuations down to 2 K, the only evident temperature dependence being that due to the declining Boltzmann factor with temperature. The effect of temperature is exemplified in fig. 5. At 40 K, a broad peak is evident in the low energy spectra, centered roughly at $115\,cm^{-1}$ (14.3 meV). This result is similar to that obtained, at lower temperatures, from a recent magnetic neutron scattering study of UBe_{13} [6]. This result is rather striking, when one recalls that Raman scattering is effectively a $q = 0$ probe, in ordered materials. In a non-interacting Fermi liquid, however, one expects the peak in the magnetic structure factor to have the following q-dependence [7]:

$$P(q) = v_f q - (v_f/p_f)q^2/2\,, \quad q < 2p_f\,.$$

Therefore, in a Raman scattering experiment, one would normally expect a peak at zero frequency. That we observe otherwise may be another indication that Raman scattering is not constrained to probe only the zone center, implying

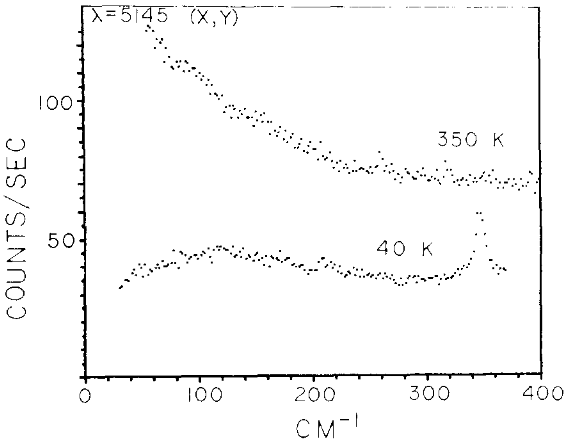

Fig. 5. Temperature dependence of the $T_{2g} + T_{1g}$ spectra between $0–400\,cm^{-1}$. All figures have been offset.

broken symmetry of some sort. It is important that neutron scattering experiments investigate fully the q-dependence of the dynamical magnetic structure factor.

5. Conclusions

Raman scattering is seen to be an important probe in any investigation into the excitations of a material. The principal observation in this study is that of light scattering from spin fluctuations in UBe_{13}. Of additional interest is our observation of phonon anomalies at low temperatures, opening the possibility that precursor effects to the novel low temperature properties in UBe_{13} *may* be manifesting themselves in the phonons. It is therefore pertinent to study the properties of the phonons, whether or not they are deemed to mediate any of the interesting behavior at low temperatures. Future work will more thoroughly consider the phonons at low temperatures.

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