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POLARIZED LIGHT SCATTERING STUDIES OF HEAVY FERMION SUPERCONDUCTORS

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Polarized light scattering measurements of some heavy fermion superconductors are presented and discussed. Light scattering affords a unique opportunity to study both the f electrons, which are thought to participate in the formation of the superconducting state, and the lattice excitations, which can demonstate strong electron–phonon interactions and elastic anomalies in these compounds. These points are illustrated by presenting the results of light scattering experiments on single crystal samples of $CeCu_2Si_2$, UBe_{13}, UPt_3, and URu_2Si_2.

The use of polarized light scattering has recently proven useful for studying the complex excitation spectra of heavy fermion systems. For example, this $q = 0$ probe can provide information about crystal field excitations and spin fluctuations which is complementary to that provided by neutron scattering at higher q. Additionally, the high resolution of light scattering allows a careful study of strong electron–phonon coupling effects, which are anticipated in these compounds due to the existence of electronic energies on the order of typical phonon frequencies.

A number of interesting phonon properties have been revealed in these materials by light scattering. For example, the phonon spectra of both UPt_3 [1], and URu_2Si_2 [2] are characterized by an extremely intense A_{1g} breathing mode (at $150\,cm^{-1}$ in UPt_3, and at $430\,cm^{-1}$ in URu_2Si_2), which dwarfs the other phonons observed in these materials. The huge intensities of these modes illustrate the large breathing-type deformation potential coupling of the phonons to the electronic configuration of U, consistent with the elastic anomalies [3, 4] observed in these compounds. Furthermore, the A_{1g} phonon in URu_2Si_2 is particularly notable in that it demonstrates a strong increase in scattering intensity with decreasing temperature. This behavior is thought to reflect a strong magnetoelastic coupling of this A_{1g} mode to a crystal field level near $400\,cm^{-1}$ (50 meV) [5].

As alluded to previously, a number of interesting electronic and magnetic excitations have also

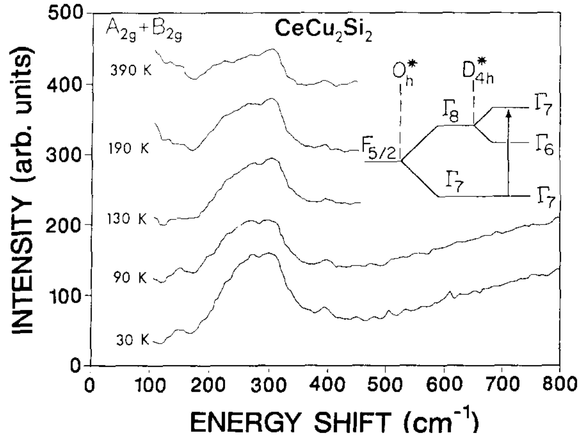

Fig. 1. Temperature dependence of the $A_{2g} + B_{2g}$ spectra of $CeCu_2Si_2$, showing crystal field excitations centered near $290\,cm^{-1}$. The inset illustrates the observed electronic transition, and the expected splitting of the $J = \frac{5}{2}$ Ce^{3+} level in subsequent cubic (O_h^*) and tetragonal (D_{4h}^*) crystal fields. The spectra have been offset for clarity.

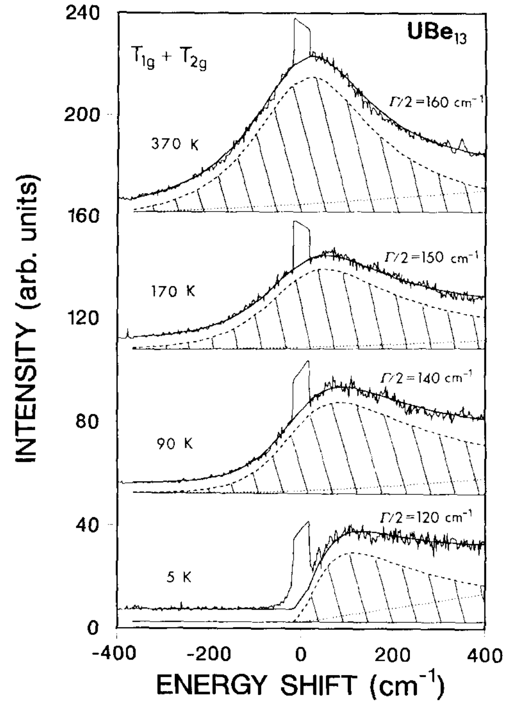

Fig. 2. Temperature dependence of the $T_{1g}+T_{2g}$ spectra of UBe_{13}, illustrating the spin fluctuation spectrum. The hatched area shows the quasielastic contribution as described in the text, the dotted line shows the linear (inelastic) contribution, and the solid line is the sum of these contributions with a small offset. The spectra have been offset.

been observed in our studies. In $CeCu_2Si_2$ [6], for example, crystal field excitations are evident, displaying a broad peak centered at 290 cm^{-1} (see fig. 1). That this peak results from electronic transitions is clear from its increasing intensity with decreasing temperature, its absence in isostructural $LaCu_2Si_2$, and its antisymmetric symmetry, A_{2g}, which is characteristic of electronic or magnetic scattering. From this symmetry assignment, we can further deduce that these excitations involve transitions between two Γ_7 levels of the tetragonally split $Ce^{3+}J=\frac{5}{2}$ multiplet (inset, fig. 1).

In UBe_{13} [7], strong quasielastic scattering from spin fluctuations is evident, displaying the symmetry of the antisymmetric representation, T_{1g} (see fig. 2). It was initially found that the spectral response of this scattering could not be fit well to a simple relaxational model

$$S(q,\omega) \propto (1+n(\omega)) \frac{\omega\Gamma}{\left(\frac{\Gamma}{2}\right)^2+\omega^2}$$

as would be expected of quasielastic scattering. However, as shown in fig. 2, by presuming a rather large linear term, $\propto (1+n(\omega))\omega$ (dotted lines), in addition to the quasielastic response (hatched area), the observed response can be fit nicely (solid line). This linear term is also observed to a lesser extent in the spectra of UPt_3 and URu_2Si_2 (see figs. 3 and 4), and is presumed to arise from very broad crystal field scattering centered at higher energies (>1000 cm^{-1}).

Quasielastic light scattering from spin fluctuations has also been observed in UPt_3 [1,8] (hatched area in fig. 3), similarly displaying the symmetry of an antisymmetric representation, A_{2g}. It should be noted that the quasielastic scattering in UBe_{13} is much greater than that

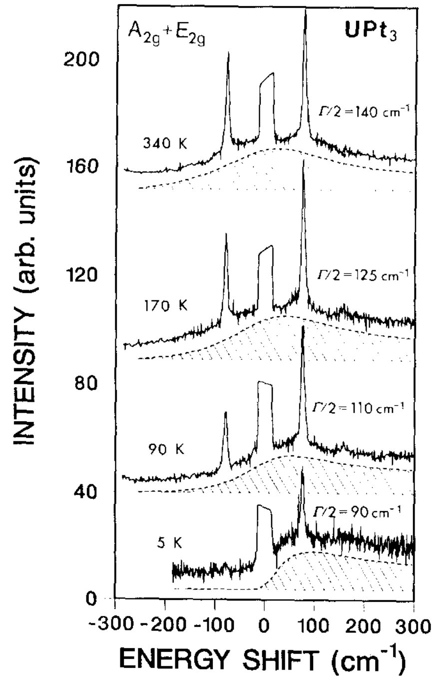

Fig. 3. Temperature dependence of the $A_{2g}+E_{2g}$ spectra of UPt_3, showing the spin fluctuation spectra. The hatched area shows the quasielastic contribution which includes a small linear (inelastic) term which may be considered negligible. The phonon observed near 80 cm^{-1} is one of the E_{2g} modes. The spectra have been offset.

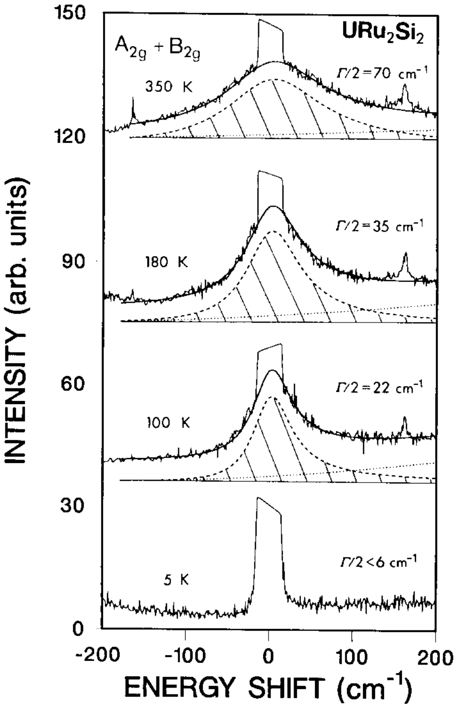

Fig. 4. Temperature dependence of the $A_{2g} + B_{2g}$ spectra of URu_2Si_2, showing the spin fluctuation spectra. The hatched area shows the quasielastic contribution, the dotted line gives the linear (inelastic) contribution, and the solid line is the sum of these contributions with a small offset. The small feature near 150 cm^{-1} is slight leakage of the B_{1g} phonon into this spectra. The spectra have been offset.

observed in either UPt_3 or URu_2Si_2 (see figs. 3 and 4). Indeed, the unexpectedly large scattering intensity of the response observed in UBe_{13} has led to the suggestion that the observed scattering possibly results from low energy, highly damped crystal field excitations [7]. However, it is likely that the large spin fluctuation scattering strength in UBe_{13} arises instead from a larger magneto-optical coupling in UBe_{13} than in UPt_3 and URu_2Si_2.

The spin fluctuation scattering we observe in URu_2Si_2 [2] (see fig. 4) is interesting in that it demonstrates a strongly temperature dependent linewidth. In fact, a detailed study of the linewidth with temperature indicates a single-ion, linear regime above 70 K, and an exchange dominated, temperature independent regime between 70 K and 23 K. Furthermore, a careful inspection of the linewidth through the magnetic transition at $T_N = 17$ K shows an abrupt decrease in the linewidth, consistent with critical slowing. Indeed, by 5 K no quasielastic scattering is observed within our experimental resolution (<6 cm^{-1}), as shown in the bottom spectrum of fig. 4. This dramatic change in the damping rate is thought to corroborate earlier specific heat evidence [9] for a gap opening transition, which removes relaxation channels along with the Fermi surface.

It is interesting to compare the spin fluctuation linewidths we observe at $\boldsymbol{q} = 0$ in these materials, with higher $\boldsymbol{q}$ neutron scattering results. For example, the $\boldsymbol{q} = 0$ spin fluctuation linewidth we see in UBe_{13} is much larger than that reported at the zone boundary by recent neutron scattering results on single crystals [10]. Such narrowing of the linewidth towards the zone boundary indicates that correlations exist with a critical $\boldsymbol{q}$ [11] near the zone boundary. A large $\boldsymbol{q}$ dependence of the spin fluctuation linewidth is also observed in URu_2Si_2 [2], although in this system the low temperature linewidths we observe at $\boldsymbol{q} = 0$ are roughly half those seen by neutron scattering at higher $\boldsymbol{q}$ [12]. This suggests longer wavelength correlations in URu_2Si_2, with a critical $\boldsymbol{q}$ closer to the zone center. Finally, in UPt_3, the spin fluctuation linewidth we observe at $\boldsymbol{q} = 0$ is comparable to that found at higher $\boldsymbol{q}$ by neutron scattering [11], indicating the absence of any $\boldsymbol{q}$ dependence in the linewidth. However, in this material, a $\boldsymbol{q}$ dependence in the spin fluctuation intensity, i.e. the static susceptibility, $\chi(\boldsymbol{q})$, has been observed by neutron scattering, suggesting the presence of antiferromagnetic correlations in UPt_3 [11].

Furthermore, the very existence of low temperature spin fluctuations in our $\boldsymbol{q} = 0$ Raman studies is notable, inasmuch as non-interacting Fermi liquid theory sets the energy scale of the imaginary part of the susceptibility at $v_F q - (v_F/2p_F)q^2$ (where v_F and p_F are the Fermi velocity and momentum, respectively). Therefore, nonzero spin fluctuation scattering at $\boldsymbol{q} = 0$ alludes to an absence of *simple* Fermi liquid effects in these materials. This finite spin fluctuation scattering at $\boldsymbol{q} = 0$ is allowed because the magnetization is not conserved in these systems due to their strong spin–orbit interactions. This issue, therefore, demands further theoretical guidance [13].

In conclusion, we have been able to observe a number of interesting excitations in the heavy fermion superconductors. Particularly interesting is the presence of finite spin fluctuation scattering at $\boldsymbol{q} = 0$, and the consequent absence of simple Fermi liquid behavior, noted for the spin fluctuation scattering in UBe_{13}, UPt_3, and URu_2Si_2. Finally, a number of novel features in the phonon spectrum have been observed, including evidence for magnetoelastic coupling to crystal field levels.

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