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 demonstrate 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$_2$Si$_2$, UBe$_{13}$, UPt$_3$, and URu$_2$Si$_2$.

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$_2$Si$_2$ [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$_2$Si$_2$), 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$_2$Si$_2$ 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$_2$Si$_2$, 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$^*_f$) and tetragonal (D$^*_4h$) crystal fields. The spectra have been offset for clarity.](image-url)
maintained, and the observed response can be fit nicely (solid line). This linear term is also observed to a lesser extent in the spectra of UPt₃ and URu₂Si₂ (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 UPt₃ [1, 8] (hatched area in fig. 3), similarly displaying the symmetry of an antisymmetric representation, A₂₋. It should be noted that the quasielastic scattering in UBe₁₃ is much greater than that observed in our studies. In CeCu₂Si₂ [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 LaCu₂Si₂, and its antisymmetric symmetry, A₂₋, which is characteristic of electronic or magnetic scattering. From this symmetry assignment, we can further deduce that these excitations involve transitions between two Τ levels of the tetragonally split Ce⁵⁺J = ½ multiplet (inset, fig. 1).

In UBe₁₃ [7], strong quasielastic scattering from spin fluctuations is evident, displaying the symmetry of the antisymmetric representation, T₁₈ (see fig. 2). It was initially found that the spectral response of this scattering could not be fit well to a simple relaxational model

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

as would be expected of quasielastic scattering. However, as shown in fig. 2, by presuming a rather large linear term, \( \omega (1 + n(\omega)) \) (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 UPt₃ and URu₂Si₂ (see figs. 3 and 4), and is presumed to arise from very broad crystal field scattering centered at higher energies (>1000 cm⁻¹).
observed in either UPt$_3$ or URu$_2$Si$_2$ (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$_2$Si$_2$.

The spin fluctuation scattering we observe in URu$_2$Si$_2$ [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 $q = 0$ in these materials, with higher $q$ neutron scattering results. For example, the $q = 0$ 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 $q$ [11] near the zone boundary. A large $q$ dependence of the spin fluctuation linewidth is also observed in URu$_2$Si$_2$ [2], although in this system the low temperature linewidths we observe at $q = 0$ are roughly half those seen by neutron scattering at higher $q$ [12]. This suggests longer wavelength correlations in URu$_2$Si$_2$, with a critical $q$ closer to the zone center. Finally, in UPt$_3$, the spin fluctuation linewidth we observe at $q = 0$ is comparable to that found at higher $q$ by neutron scattering [11], indicating the absence of any $q$ dependence in the linewidth. However, in this material, a $q$ dependence in the spin fluctuation intensity, i.e. the static susceptibility, $\chi(q)$, 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 $q = 0$ 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 $q = 0$ alludes to an absence of simple Fermi liquid effects in these materials. This finite spin fluctuation scattering at $q = 0$ is allowed because the mag-
netization 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 \( q = 0 \), and the consequent absence of simple Fermi liquid behavior, noted for the spin fluctuation scattering in \( \text{UBe}_13 \), \( \text{UPt}_3 \), and \( \text{URu}_2\text{Si}_2 \). 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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