# Far-Infrared Properties of $URu_2Si_2$

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Measurements of the far-infrared reflectance of $URu_2Si_2$ between 10 and 720 $cm^{-1}$ have been made at temperatures from 2 to 90 K. Above the coherence temperature the optical conductivity increases monotonically with increasing frequency and at lower temperatures the development of the narrow mode responsible for the high dc conductivity is clearly observed. In the antiferromagnetic state an energy gap with a size between 46 and 65 $cm^{-1}$ is observed whose shape is reminiscent of the energy gap observed in the spin-density-wave state of Cr.

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$URu_2Si_2$ has attracted interest as the first heavy-fermion metal shown to exhibit both superconductivity and magnetic ordering. Specific-heat measurements indicate that the effective mass of the electrons in this material is enhanced at low temperature by a renormalization parameter, $\lambda$, of the order of 25.[1] The transport properties of $URu_2Si_2$ are also similar to other heavy-fermion metals. Above 70 K the dc resistivity decreases with increasing temperature[2] in a manner similar to that observed in metals containing isolated magnetic impurities. This similarity to the Kondo problem suggests that the high-temperature behavior of $URu_2Si_2$ might be related to the scattering of conduction electrons by isolated magnetic impurities.[3] Below 70 K lies the coherent regime which is the temperature range in which the resistivity drops rapidly with decreasing temperature.[3] Reflectance measurements are presented here both above and below the coherent temperature, $T_{co}$, the 70-K boundary between the single-impurity regime and the coherent regime. At 17.5 K there is a resistivity anomaly similar to the one at the spin-density-wave transition (SDW) in Cr (Ref. 2) and neutron scattering has confirmed that this is an antiferromagnetic transition.[4] Far-infrared measurements probe the energy gap associated with this transition. A coexisting superconducting transition near 1 K is too low in temperature to observe in the far infrared.[2]

The single crystal used in this study was grown from U, Ru, and Si which were premelted, cleaned where applicable, and weighed. They were reacted and homogenized in an inert gas in a water-cooled-hearth arc furnace. The $URu_2Si_2$ crystal was grown by the Czochralski technique in a Reed-type Triarc furnace which has been modified to give a directly water-cooled hearth and seed rod. Ti gettered argon at 100 kPa was used as the chamber atmosphere. The growth rate was 20 mm/h. The rod was rotated at 9 rpm and the hearth at 60 rpm in the same direction.

The crystal was cleaved perpendicular to the *c* axis and the reflectance of the cleavage plane was measured against a stainless-steel reference mirror. An accurate ($\pm 0.5\%$) value of the reflectance was obtained by coating the sample with Pb *in situ* in order to correct for differences between the position of the sample and reference in the optical path.[5] The reflectance was measured from 10 to 160 $cm^{-1}$ with a Martin-Puplett-type polarizing interferometer and from 110 to 720 $cm^{-1}$ with a Michelson interferometer. The reflectance measured in the region of overlap between the two interferometers agreed to within 0.5%.

Figure 1 shows the reflectance of $URu_2Si_2$ at three different temperatures. The two sharp features at 108 and 377 $cm^{-1}$ are due to phonons and the feature near 42 $cm^{-1}$ seen in the 2-K data is caused by an energy gap associated with the antiferromagnetic transition. The reflectance was analyzed by fitting the data with a model for the dielectric function and by Kramers-Kronig analysis. Above the Néel temperature ($T_N = 17.5$ K), the response of the charge carriers can be fitted to a Drude term plus an overdamped harmonic oscillator; fits to a Drude term alone are poorer. The oscillator does not have a particular physical significance, it is merely a con-

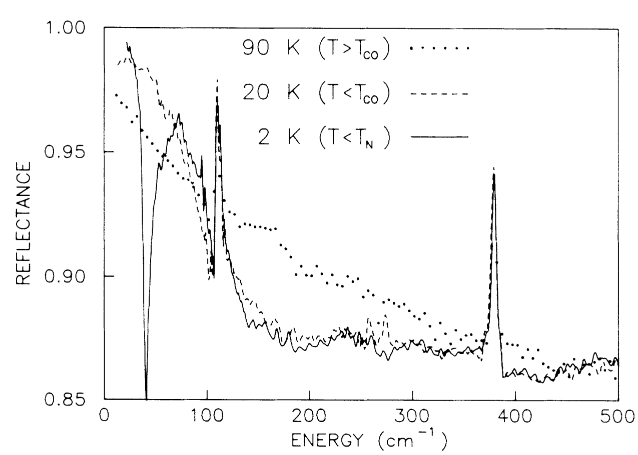


FIG. 1. The reflectance of $URu_2Si_2$ above the coherence temperature, below the coherence temperature, and below the Néel temperature.

TABLE I. Parameters used to fit the response of the charge carriers of $URu_2Si_2$. $\omega_p$ and $\Gamma$ are the plasma frequency and scattering rate of the Drude part and $\omega_0$, $\gamma$, and $\Omega_p^2$ are the position, width, and oscillator strength of a Lorentzian oscillator.

| $T$ (K) | $\omega_p$ ($cm^{-1}$) | $\Gamma$ ($cm^{-1}$) | $\omega_0$ ($cm^{-1}$) | $\gamma$ ($cm^{-1}$) | $\Omega_p^2$ ($eV^2$) |
|---|---|---|---|---|---|
| 95 | 7567 | 313 | 987 | 2770 | 8.90 |
| 40 | 4748 | 103 | 789 | 2828 | 9.36 |
| 20 | 3724 | 32 | 753 | 2552 | 9.67 |

venient way of parametrizing the data. The results of these fits, summarized in Table I, can be used to generate the extrapolations needed for evaluation of the Kramers-Kronig integral. Since the reflectance above 450 $cm^{-1}$ is independent of temperature the same overdamped oscillator is used to generate the high-frequency extrapolation for all temperatures. At temperatures above $T_N$ the Drude parameters obtained from the fits can be used to estimate the dc conductivity. That is, the plasma frequency, $\omega_p$, and scattering rate, $\Gamma$, obtained from a least-squares fit can be used to calculate dc conductivity via $4\pi\sigma_1(\omega=0)=\omega_p^2/\Gamma$. The conductivities obtained in this way are in good agreement with the dc resistivity measurements by Palstra, Menovsky, and Mydosh[2] confirming the accuracy of the measured reflectance. Thus, above $T_N$ a Drude extrapolation to zero frequency can be made at each temperature by use of parameters obtained from the fits. The low-frequency extrapolation below $T_N$ is more uncertain and use of a variety of extrapolations indicates that the Kramers-Kronig results cannot be trusted below 20 $cm^{-1}$.

Figure 2 shows the real part of the optical conductivity, $\sigma_1(\omega)$, at three temperatures above $T_N$. Beneath two sharp phonon peaks lies a smooth background conductivity due to the response of the charge carriers. Above $T_{co}$, at 90 K, $\sigma_1(\omega)$ increases with increasing frequency unlike the decreasing conductivity of a simple Drude metal. The conductivity in the coherent regime is even more striking. As the sample is cooled through the temperature range where the dc conductivity increases dramatically, the far-infrared conductivity between 40 and 300 $cm^{-1}$ decreases. The narrow mode responsible for the high dc conductivity in the coherent regime is visible at 40 and 20 K as a sharp upturn in $\sigma_1(\omega)$ at low frequencies.

To further analyze this behavior, one can use an extension of Drude theory involving a frequency-dependent scattering rate, $\Gamma(\omega)$. In order to satisfy causality, an associated frequency-dependent renormalization, $\lambda(\omega)$, is introduced whose low-frequency limit, $\lambda(\omega=0,T=0)$, is close to the specific-heat mass enhancement, $\lambda$. The Drude expression for the conductivity is then modified to[6]

$$\sigma(\omega)=\frac{1}{4\pi}\frac{\omega_{\rm eff}^2}{\Gamma_{\rm eff}-i\omega},$$

where

$$\Gamma_{\rm eff}=\frac{\Gamma(\omega)}{1+\lambda(\omega)},\quad \omega_{\rm eff}^2=\frac{\omega_p^2}{1+\lambda(\omega)}.$$

If the conductivity of $URu_2Si_2$ is analyzed in this way one obtains the $\Gamma(\omega)$ and $\lambda(\omega)$ shown in Figs. 3 and 4 with the phonon peaks removed for clarity. At 90 K $\Gamma(\omega)$ decreases with increasing frequency and the $\lambda(\omega)$ associated with this is negative and approaches zero at high frequency. At low temperature $\Gamma(\omega)$ becomes peaked in the far infrared instead of decreasing monotonically. Above 110 $cm^{-1}$ $\Gamma(\omega)$ decreases with increasing frequency as in the single-impurity regime, but below 110 $cm^{-1}$ it is strongly suppressed. Thus, 110 $cm^{-1}$

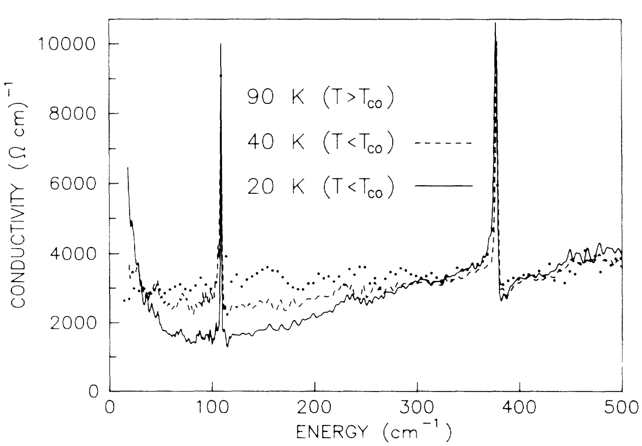


FIG. 2. The optical conductivity of $URu_2Si_2$ at three temperatures above the Néel temperature. At 90 K $\sigma_1(\omega)$ is monotonically increasing, but at 40 and 20 K a narrow mode develops at low frequency.

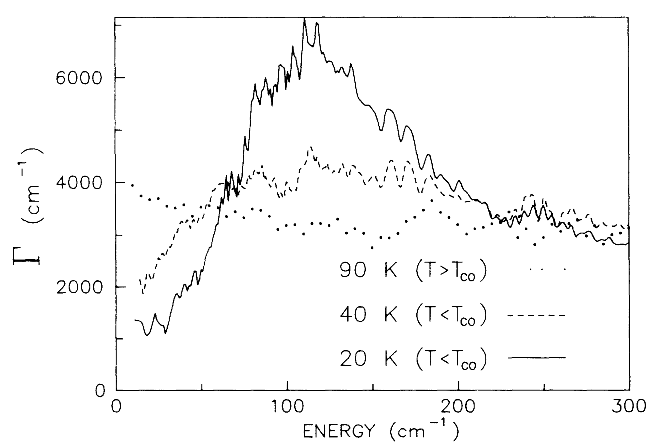


FIG. 3. The unrenormalized scattering rate, $\Gamma(\omega)$, of $URu_2Si_2$. At 90 K $\Gamma(\omega)$ is monotonically decreasing, but at 40 and 20 K it is suppressed at low frequencies and broadly peaked at 110 $cm^{-1}$.

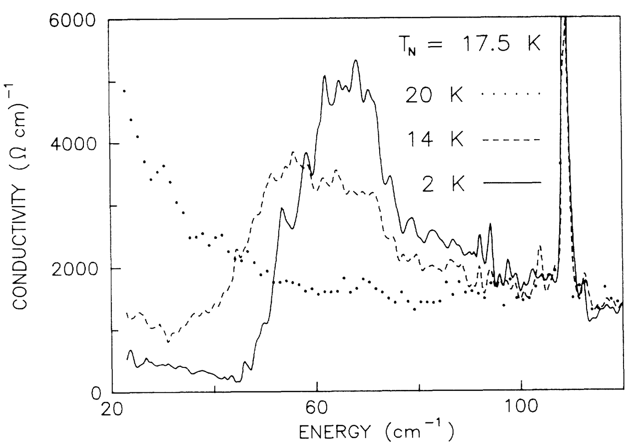


FIG. 4. The frequency-dependent renormalization, $\lambda(\omega)$, of $URu_2Si_2$. At 90 K $\lambda(\omega)$ is negative in the far infrared, but at 40 and 20 K it is large and positive at low frequency.

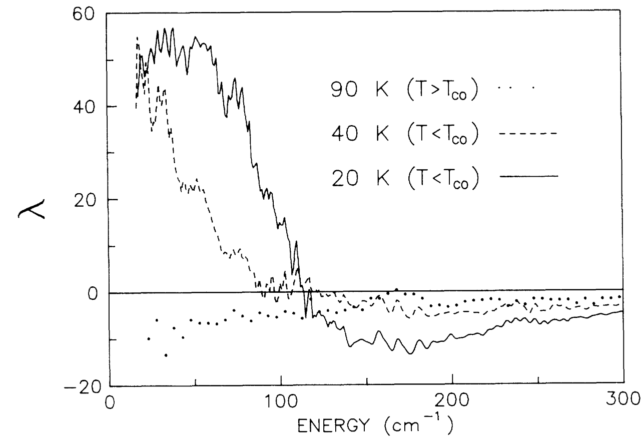


FIG. 5. The optical conductivity of $URu_2Si_2$ at low temperatures. At 20 K the mode at the origin responsible for the high dc conductivity is seen. In the antiferromagnetic state the development of a feature reminiscent of the SDW gap in Cr is observed.

might be taken as the energy which separates the coherent behavior from the single-impurity scattering and this energy is comparable to the temperature below which coherent behavior is observed (110 cm$^{-1}$ $\sim 2.2 k_B T_{co}$). In fact, $\Gamma(\omega)$ at 20 K is quite similar in shape to the temperature dependence of the dc resistivity. The peaked $\Gamma(\omega)$ observed in the coherent regime is associated with a $\lambda(\omega)$ that rises from negative values to large positive values below 110 cm$^{-1}$ and at 20 K $\lambda(\omega)$ starts to turn down again below 40 cm$^{-1}$.

There are significant similarities and differences between the results presented here and the behavior of $\Gamma(\omega)$ and $\lambda(\omega)$ reported by Webb, Sievers, and Mihalisin[7] for $CePd_3$. In both metals at low temperature, $\Gamma(\omega)$ is depressed at low frequency and rises rapidly with increasing frequency. The chief difference is that $\Gamma(\omega)$ in $CePd_3$ was found to saturate rather than decrease at higher frequencies. The difficulty here involves separating intraband and interband conductivities as an interband peak can affect the shape of $\Gamma(\omega)$ and $\lambda(\omega)$ at higher frequencies. Results presented for $UBe_{13}$ are strongly affected by a low-energy interband transition at 0.093 eV.[8] The $CePd_3$ results were obtained after removing an interband peak at 0.235 eV through fitting it to a Lorentzian. However, the width of the Lorentzian used to fit $\sigma_1(\omega)$ in $CePd_3$ was temperature dependent and one must worry that this method may not adequately separate the interband and intraband contributions. In the absence of mid-infrared measurements for $URu_2Si_2$ it is impossible to say whether or not the decreasing $\Gamma(\omega)$ is caused by an interband transition. It is, however, intriguing to note that far-infrared measurements of the dilute Kondo system, Cu:Fe, indicate a scattering rate that decreases with increasing frequency.[9] This suggests a connection between the scattering of conduction electrons by isolated magnetic ions and the high-temperature (or high-frequency) behavior of $URu_2Si_2$.

The existence, in the coherent regime, of a narrow peak at low frequency in $\sigma_1(\omega)$ has been predicted on theoretical grounds.[10] It is a Drude-like peak centered at the origin with a width given by the scattering rate of the charge carriers, but in a heavy-electron material the width is narrowed by a factor of $1+\lambda$. This mode is observed here at a relatively high temperature of 20 K and one cannot simply estimate the mass enhancement, $\lambda$, from these data because $\lambda(\omega)$ varies considerably over the width of the mode. Lower-frequency measurements are required in order to compare $\lambda(\omega)$ to the specific heat $\lambda$ because $\lambda$ loses its meaning as a mass enhancement at finite frequencies. What is clear in these results is that the narrowness of the mode comes from two sources. First, $\Gamma(\omega)$ is suppressed at low temperatures and this is responsible for the low resistivity that develops in the coherent state. This suppressed scattering rate is further reduced by the mass enhancement giving rise to a small effective scattering rate and a narrow mode.

Figure 5 shows $\sigma_1(\omega)$ at 20 K and at two temperatures in the antiferromagnetic state. At 2 K the mode at the origin is too sharp to observe in the far infrared and instead one sees an energy gap. The conductivity is very low below 46 cm$^{-1}$, rises rapidly to a peak near 65 cm$^{-1}$, and then approaches the normal-state conductivity at higher frequencies. As the temperature increases towards $T_N$ the peak broadens and shifts down in frequency. The shape of this feature is reminiscent of the gap and peak seen in the optical conductivity of Cr in the SDW state.[11] Theories exist for the optical conductivity of charge-density-wave[12] and SDW[13] systems but they assume that the scattering rate of the charge carriers is much smaller than the energy gap, and thus they produce conductivities which fall to zero at frequencies well above the energy gap.[14] Here the conductivity falls to the finite normal-state conductivity above the energy gap.

One can still make an estimate of the size of the energy gap since it should lie somewhere between the upturn in $\sigma_1(\omega)$ and the point at which it reaches its maximum value. At 2 K this puts the energy gap in the range from 46 to 65 $cm^{-1}$ and this direct measurement of the energy gap is comparable to the 88-$cm^{-1}$ gap inferred from specific-heat measurements.[1] The ratio of the gap size to the transition temperature is $2\Delta/k_B T_N = 3.6$ to 5.1 which is larger than the mean-field value of 3.5.

$URu_2Si_2$ exhibits a rich spectrum in the far infrared. The energy scales in this material are such that one can observe single-impurity scattering at high temperature and coherent behavior at low temperature. In the single-impurity regime the charge carriers have a scattering rate, $\Gamma(\omega)$, which decreases monotonically with increasing frequency; whereas, in the coherent regime $\Gamma(\omega)$ below 100 $cm^{-1}$ is suppressed, giving rise to a peaked frequency-dependent scattering rate. The development of a narrow mode in the conductivity in the coherent state is clearly observed in the far infrared. At low temperatures, the development of an energy gap associated with the antiferromagnetic transition at 17.5 K is seen. The size of this gap is between 46 and 65 $cm^{-1}$ (5.6 to 7.5 meV) and its shape is similar to that observed in the SDW state of Cr.

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