Low-frequency excitation in the optical properties of superconducting $CeCoIn_5$

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The far-infrared optical response of $CeCoIn_5$, a superconducting heavy fermion metal with a T_C of 2.3 K, was investigated from 5–40 cm^{-1} at temperatures from 0.5–2.5 K using a polarizing interferometer and a He^3 cryostat. A strong absorption feature is revealed at low temperatures which appears to be a gap in the density of states, reminiscent of the energy gap seen in the hidden order state in URu_2Si_2. The depth of the spectral structure decreases with increasing temperature from 0.5 to 2.5 K indicating that the characteristic temperature for this behavior is close to the superconducting T_C. A peak in the superconducting state Kramers-Kronig-derived optical conductivity occurs just above the gap at 1.5 meV.

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I. INTRODUCTION

The superconducting pairing mechanism in strongly correlated f-electron systems has intrigued researchers during the last decade. An attractive interaction between the electrons resulting from the virtual exchange of spin fluctuations was suggested to be responsible for the superconductivity rather than the one usually seen in ordinary metals which is due to the exchange of phonons.[1–3] It is believed that the superconductivity found among these materials occurs in the vicinity of a quantum-critical point (QCP) where the characteristic temperature of a magnetically ordered ground state approaches zero temperature.[1,4] An interesting example of a magnetically ordered state is the "hidden-order" state in URu_2Si_2 that occurs at 17 K, well above the superconducting transition at 1 K.[5] The optical reflectance of this material develops a sharp minimum at 17 K and Kramers-Kronig analysis shows that this is the result of a gap in the frequency-dependent conductivity.[6] Tunneling measurements show that this is a gap in the density of states.[7,8] However dc resistivity shows very little change at 17 K suggesting that only parts of the Fermi surface are gapped and the remaining regions are able to supply the carriers responsible for the superconducting transition. The initial suggestions that the state that develops at 17 K is a spin-density-wave state have been challenged recently[5] on grounds that while quantities, such as the specific-heat coefficient are large, the ordered moment is very small and at this time the 17 K state in URu_2Si_2 is not well understood. For this reason it would be interesting to find other systems where a hidden-order state might be present. We suggest that one such material might be $CeCoIn_5$ which also has an anomalously large specific-heat coefficient but no measurable bulk moment. $CeCoIn_5$ has the highest superconducting critical temperature, T_C=2.3 K, among the known heavy fermion materials at ambient pressure. In the limit as $T \to 0$ tuning parameters such as chemical doping,[9] pressure,[10] and magnetic field[11,12] yield phase transitions to a magnetic state indicating proximity to quantum-critical behavior.

As $T \to 0$, a Landau Fermi liquid exhibits a T^2 dependence of the resistivity and constant C/T and χ, where C is the specific heat and χ is the magnetic susceptibility. In contrast, the resistivity in the normal state of $CeCoIn_5$ is approximately linear in temperature while the susceptibility exhibits a modified Curie-Weiss behavior and the specific heat diverges as $T \ln T$ when a magnetic field is applied to quench the superconductivity.[11,13–17] A deviation from the Landau Fermi-liquid state occurs when a system is located near a QCP and its physical properties are affected by strong spin fluctuations. The non-Fermi-liquid behavior exhibited by $CeCoIn_5$ is interpreted to be an indication that it lies near an antiferromagnetic (AFM) region in phase-space and that a transition to the magnetically ordered state would have been observed in the absence of superconductivity at the QCP.[10,18] This is further supported by normal-state nuclear-spin-lattice relaxation-rate measurements which find rather than the linear temperature dependence expected for a Fermi liquid, a $T^{1/4}$ dependence of $1/T_1$ characteristic of when $1/T_1$ is governed by strong AFM spin fluctuations.[13]

At ambient pressure the magnetic field at which Fermi-liquid behavior is recovered, H_{QCP}, has been found to be very close to H_{C2}, the magnetic upper critical field at which superconductivity disappears,[11,12] which initially suggested that superconducting fluctuations might be responsible for the non-Fermi-liquid behavior in $CeCoIn_5$.[19] Recent work[20,21] however has shown that under pressure H_{QCP} and H_{C2} separate, from which it is concluded that the coincidence of H_{C2} and H_{QCP} at ambient pressure is accidental and that the quantum-critical fluctuations which produce the non-Fermi-liquid behavior have another origin.

Recent neutron-scattering measurements have found a sharp spin resonance which develops in the superconducting state of $CeCoIn_5$ at Ω=0.60 meV in the absence of long-ranged spin correlations, similar to that observed in the superconducting state of the cuprate superconductors $YBa_2Cu_3O_{6+x}$ and $Bi_2Sr_2CaCu_2O_8$ near 41 meV.[22] Since the physics governing the competing magnetic and superconducting ground states lies in the details of the electronic

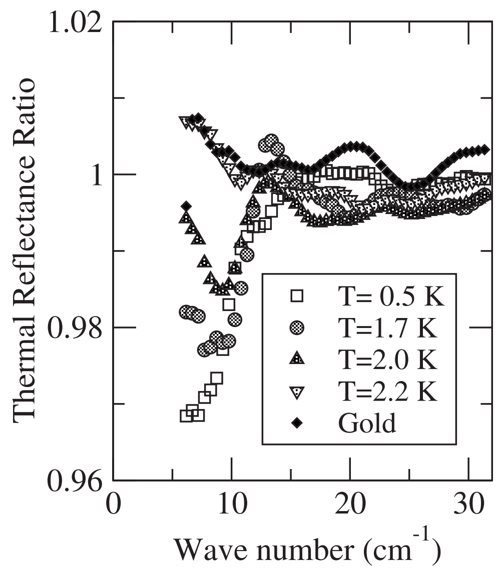

FIG. 1. Frequency dependence of the ratio of the reflectance at various temperatures to that at 2.3 K just above T_C in the normal state. The curve with solid diamonds shows the corresponding ratio of reflectances for the gold-coated sample at 0.5 K to that at 2.3 K.

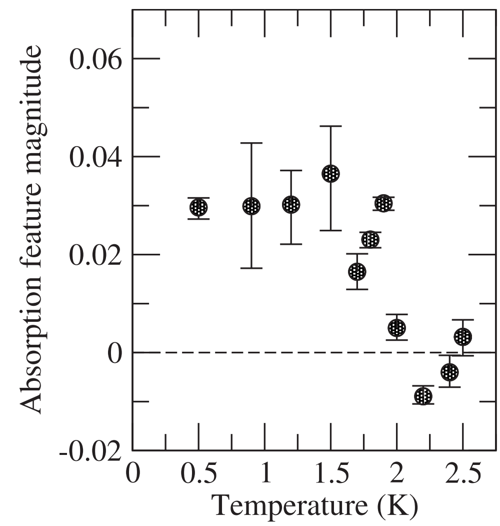

FIG. 2. Temperature dependence of the depth of the dimensionless low-frequency absorption feature observed in the thermal-reflectance ratio. (See text for definition).

structure at low energy, it is of significant interest to examine optically the spectrum of excitations of $CeCoIn_5$ at low energy and temperature.

The normal-state optical properties of $CeCoIn_5$ have previously been investigated.[23–25] In these studies it is found that with decreasing temperature the optical conductivity evolves from Drude-type behavior at room temperature to a multicomponent form at the lowest temperatures consisting of a highly renormalized narrow mode at low frequencies separated by a minimum from a higher frequency feature centered near 600 cm^{-1}. This behavior results from hybridization between the localized f electrons and conduction electrons. Intraband transitions within the lower band produce the narrow Drude-type feature while interband transitions between the hybridized bands produce the higher energy feature. Herein we present the results of optical measurements of $CeCoIn_5$ which provide spectroscopic evidence for the presence of a gap in the density of states that develops just above the superconducting transition temperature.

II. EXPERIMENT AND RESULTS

Frequency-dependent measurements of the electromagnetic response of a single crystal of $CeCoIn_5$ at various temperatures were carried out with a Martin-Pupplett-type polarizing interferometer and a ^{3}He cryostat. This combination of instruments is optimized for measurements at low energy. The ratio of the power reflected from the sample to that from a reference mirror was measured. In Fig. 1 the reflectance ratio between selected temperatures below T_C and that at 2.3 K, which is above T_C, are presented. Such ratios of the reflectance at two different temperatures are referred to as "thermal-reflectance" ratios and represent the relative change in the reflectance between two temperatures. Note that while the thermal reflectance ratio is essentially unity (indicating no change in reflectance between the two temperatures of interest) above approximately 12 cm^{-1} that at lower frequencies a strong absorption feature sets in as the temperature is lowered. The oscillations on the order of $\pm 0.5\%$ that can be seen in the data are due to incomplete cancellation of interference fringes resulting from windows, filters, beam splitters, *etc.* within the optical path. The measurements were repeated with a metallic gold film evaporated *in situ* onto the sample in order to ensure that the temperature dependence observed was intrinsic to the sample. Since the reflectance of gold has negligible temperature dependence at these low temperatures the thermal-reflectance ratio is expected to be unity. The curve indicated by solid diamonds in Fig. 1 shows the thermal-reflectance ratio of the gold-coated sample at a temperature of 0.5 K compared to that at 2.3 K. Note that as expected, within experimental uncertainty, the thermal-reflectance ratio is unity, and that the absorption feature is absent.

III. DISCUSSION AND ANALYSIS

To parameterize this absorption feature we plot in Fig. 2 an estimate of its depth as a function of temperature at all temperatures for which spectra were collected by subtracting the average value of the thermal-reflectance ratio at the lowest frequencies measured from the average value of the baseline between 20 and 30 cm^{-1}. The appearance of this feature in the spectra seems to correspond closely with the temperature of onset of the superconductivity. (Without higher temperature measurements one cannot rule out from these thermal-reflectance data, the possibility that the suppression starts above T_C. We show below that the absolute reflectance confirms the absence of the feature at 2.3 K in the normal state). In conventional superconductors one expects the zero-temperature superconducting-state reflectance, R_S, to be unity at frequencies below the superconducting gap while the normal-state reflectance, R_N, follows a monotonically decreasing Drude form with increasing frequency. Thus the thermal reflectance ratio R_S/R_N will peak at the gap with a value greater than unity. At finite temperatures below T_C the peak becomes smaller and shifts down in frequency as the superconducting gap is reduced in magnitude. Materials that have shown such behavior include the layered Ruthenate Sr_2RuO_4 (Ref. 26) and the graphenelike superconductor CaAlSi.[27] By comparison the results for $CeCoIn_5$ are quite different. The observed optical response of $CeCoIn_5$ is reminiscent of that of materials in which the excitation spectrum of itinerant charge carriers is affected by a Fermi-surface instability which gives rise to a magnetic state, such as the spin-density-wave (SDW) transition in Chromium,[28] AFM in $U\mathrm{Cu}_5$,[29] $U\mathrm{Pd}_2\mathrm{Al}_3$ and $U\mathrm{Pt}_3$ (Ref. 30) and the hidden-order transition in $U\mathrm{Ru}_2\mathrm{Si}_2$.[6] In these materials an energy gap appears at low frequencies in the optical conductivity associated with the onset of the phase transition. Its signature in the reflectance is a strong absorption feature at finite frequency, which gives rise to a pseudogap in the optical conductivity. Note that in these materials it is believed that only a fraction of the carriers are involved in the SDW-like phase transition; the remainder contributing to the low-frequency Drude behavior. If the phase transition occurs at a temperature well below the onset of coherence then the Drude peak is very narrow and the absorption feature appears essentially as a peak on a very low background conductivity (assuming electronic transitions occur at higher frequencies). This is the case for $U\mathrm{Ru}_2\mathrm{Si}_2$,[6] $U\mathrm{Pd}_2\mathrm{Al}_3$, and $U\mathrm{Pt}_3$.[30] If on the other hand the phase transition occurs at a temperature comparable to the coherence temperature as in $U\mathrm{Cu}_5$ (Ref. 29) or in a material that does not show low-temperature heavy-electron behavior such as Chromium,[28] then the gap excitation appears as a small peak or shoulder on the Drude background.

In the case of $CeCoIn_5$ the temperature at which the absorption feature appears (≈ 2 K) is well below the temperature at which the resistivity begins to fall (≈ 40 K) from its maximum value and thus behavior similar to that of $U\mathrm{Ru}_2\mathrm{Si}_2$, $U\mathrm{Pd}_2\mathrm{Al}_3$, and $U\mathrm{Pt}_3$ would be expected. That is, an absorption peak on an otherwise low background conductivity. It is thus of interest to examine the low-temperature optical conductivity which can be obtained from the absolute reflectance via Kramers-Kronig analysis. In principle the gold-coated spectrum allows one to determine the absolute reflectance.[31] Fig. 3 shows the absolute reflectance at 0.5 and 2.3 K obtained by dividing the sample spectrum by the gold-coated sample spectrum. As shown by the error bars the uncertainty is on the order of ± 0.0075. Thus structure in the data of this order of magnitude is indistinguishable from spurious noise or oscillations, which if they cause the reflectance to vary close to, and exceed unity cause difficulties for the Kramers-Kronig procedure. For this reason it was not possible to perform Kramers-Kronig analysis on the 2.3 K data. Note however that within the error bars no absorption feature is detected. Since the absolute reflectance of the 0.5 K data is several percent less than unity at low frequencies the Kramers-Kronig procedure could be carried out. At higher frequencies the reflectance data was extended using results of Singley *et al.*[23] at 10 K. The levels of the two data sets matched well. More of a challenge was the extension of the

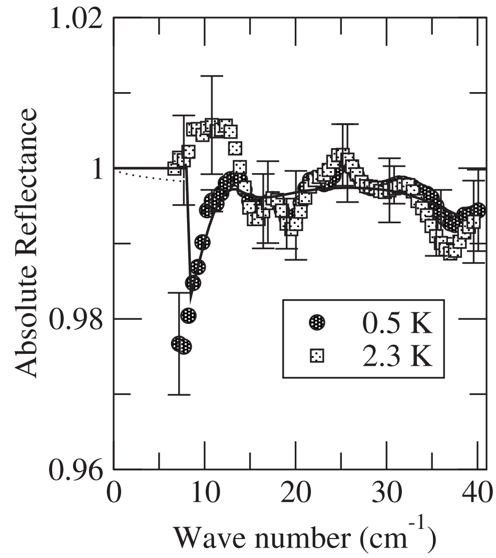

FIG. 3. Absolute reflectace at 0.5 K in the superconducting state and at 2.3 K in the normal state. At low-frequency solid and dashed curves show extensions of, respectively, $R=1$ and HR with $\rho=2$ $\mu\Omega$ cm. Also shown (solid curve) is a smoothed result through the 0.5 K data used for Kramers-Kronig analysis.

data to zero frequency. Since $CeCoIn_5$ is metallic the reflectance is expected to be unity at zero frequency. We found that extensions from $\omega=0$ to the start of the data that yielded physical values of $\sigma_1(\omega)$ were those with high reflectance (R) such as Drude or Hagen-Rubens (HR) extrapolations consistent with the dc resistivity above T_C or $R=1$ (as would be expected in a conventional superconductor below T_C) followed by a sharp decrease to the start of the measured data. Two such example extensions are shown in Fig. 3. Also shown is a smoothed curve through the data at higher frequencies which eliminates the spurious oscillatory structure discussed above. In the inset to Fig. 4 we show the Kramers-Kronig-derived optical conductivity for the smooth curve with the two low-frequency extensions shown in Fig. 3. There is little difference in the results above ≈ 8 cm^{-1}, which sets a lower limit on the reliability of $\sigma_1(\omega)$. In the main figure we show in addition to the data of the inset, the 0.5 K conductivity estimated from microwave surface impedance measurements of Ormeno *et al.* who have resolved a very narrow Drude peak with a scattering rate on the order of 0.5 cm^{-1} providing evidence for a clean-limit superconductor with a superconducting order parameter with line nodes similar to the High T_C cuprate superconductors.[32] The microwave measurements show the peak narrowing by almost two orders of magnitude *below* T_C. Ormeno *et al.* attribute the strong temperature dependence of the scattering rate below T_C to interactions with spin fluctuations. The pseudogap of magnitude $\omega \simeq 12$ cm$^{-1} \simeq 1.5$ meV, that we observe in the optical conductivity is reminiscent of that observed in $U\mathrm{Pd}_2\mathrm{Al}_3$ and $U\mathrm{Pt}_3$ attributed to magnetic correlations between localized and delocalized carriers, and that associated with the hidden order state in $U\mathrm{Ru}_2\mathrm{Si}_2$. In these materials its appearance correlates not with the onset of coherence but rather with the emergence of a low-temperature magnetic-phase transition. From this we infer that it is not

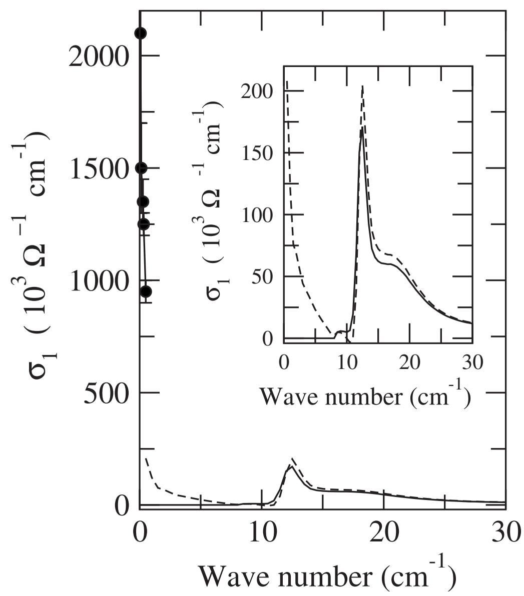

FIG. 4. Real-optical conductivity at 0.5 K. The solid circles at low frequency are from the microwave measurements of Ormeno *et al.* Curves are the data of this work derived from Kramers-Kronig analysis. The solid and dashed curves are the results for low-frequency extensions of, respectively, $R=1$ and HR with $\rho=2\ \mu\Omega$ cm. The inset shows a magnified view of the results of this work.

the superconductivity that gives rise to the observed pseudogap in $CeCoIn_5$ but rather magnetic excitations resulting from the proximity to a QCP.

IV. CONCLUSION

Our results indicate that there are carriers in $CeCoIn_5$ that are sensitive to an underlying ordered state, which may be of magnetic origin, with a characteristic temperature that lies very close to the superconducting T_C. Their presence is manifested in a low-frequency pseudogap feature of magnitude 1.5 meV that is distinct from the hybridization gap and which is similar to a feature observed in the optical response of other heavy-electron superconducting materials.

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