Evidence of Electromagnetic Absorption by Collective Modes in the Heavy Fermion Superconductor UBe_{13}

J. R. Feller,[1,2] C.-C. Tsai,[1] J. B. Ketterson,[1] J. L. Smith,[3] and Bimal K. Sarma[2]

[1]*Department of Physics and Astronomy, Northwestern University, Evanston, Illinois 60208*
[2]*Department of Physics, University of Wisconsin–Milwaukee, Milwaukee, Wisconsin 53201*
[3]*Los Alamos National Laboratory, Los Alamos, New Mexico 87545*

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We present results of a microwave surface impedance study of the heavy fermion superconductor UBe_{13}. We clearly observe an absorption peak whose frequency and temperature dependence scales with the BCS gap function $\Delta(T)$. Resonant absorption into a collective mode, with energy approximately proportional to the superconducting gap, is proposed as a possible explanation. Fits to the data provide a simple relation between $\Delta(T)$ and the collective mode frequency.

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The superconducting states in heavy fermion compounds [1] such as UPt_3 and UBe_{13} exhibit behavior that differs markedly from the predictions of the theory of Bardeen, Cooper, and Schrieffer (BCS). Quantities such as the specific heat and ultrasonic attenuation, for example, display a power-law dependence on temperature instead of the usual exponential form, suggesting the existence of nodes in the superconducting energy gap. Such unconventional behavior has been explained, at least qualitatively, within the framework of Ginzburg-Landau theory, assuming a multicomponent order parameter. Much of the work on heavy fermion superconductivity, especially from a theoretical point of view, has been strongly influenced by the paradigm provided by superfluid ^{3}He. In the superfluid phases of ^{3}He, quasiparticle pairs form in states with relative orbital angular momentum quantum number $l = 1$ (p wave), as opposed to the BCS $l = 0$ (s-wave) state. Consequently, the order parameter possesses a large number of degrees of freedom. This, in turn, gives rise to multiple superfluid phases and a rich spectrum of order parameter collective modes [2,3]. The observation and classification of collective modes, notably by means of ultrasound absorption experiments, was instrumental in determining the symmetries of the order parameters corresponding to each phase. With mounting evidence that the heavy fermion superconductors might be characterized by unconventional ^{3}He-like order parameters, it was natural to wonder whether they also could support collective oscillations. A number of theoretical investigations [4–6] of unconventional charged superfluids, assuming order parameters of various symmetries, have predicted mode frequencies (measured relative to the energy gap) similar to those found in ^{3}He. High frequency (~2 GHz) longitudinal ultrasound measurements [7] of UBe_{13} revealed a sharp attenuation peak just below the superconducting transition temperature T_c. This was initially interpreted as the signature of a low-lying collective mode. It was soon realized [8], however, that the attenuation peak could more simply be explained in terms of enhanced pair-breaking and coherence effects, which are consequences of the unusually large quasiparticle effective masses. Similar features have since also been seen in UPt_3 [9] and URu_2Si_2 [10].

While very well suited to the study of ^{3}He, ultrasound is not the ideal probe of collective mode behavior in heavy fermion superconductors. One expects characteristic energies on the order of the gap function $\Delta \sim k_B T_c$, which translates to a frequency of roughly 20 GHz. Ultrasonic measurements at such frequencies are problematic. Since we are dealing with a system of charges, electromagnetic excitation of collective modes is a natural alternative. The obvious experiment is a microwave surface impedance measurement employing a resonant cavity. Low temperature microwave cavity measurements have been performed both on UPt_3 [11] and UBe_{13} [12], but no clear evidence of collective mode absorption was found. While these null results are discouraging, they may merely be an indication of overdamping attributable to high impurity concentrations.

Here we report the results of recent surface impedance measurements of a high quality UBe_{13} single crystal at temperatures down to ~80 mK. The sample is an approximately rectangular slab with dimensions $\sim 4.5 \times 3.6 \times 1.3$ mm^3. It was prepared by putting U, Be, and Al in the atomic ratio 1:15:174 into an outgassed BeO crucible and heating to 1200 °C in flowing helium gas. It was then cooled for 300 h to the freezing point of aluminum. The aluminum was removed in a concentrated NaOH solution revealing UBe_{13} single crystals with natural [100] facets of the cubic structure. The sample's surface was lightly etched in dilute sulfuric acid. For the measurements, a cylindrical, lead-plated copper cavity, mounted on a dilution refrigerator, was employed. The sample, attached to the bottom of a lead-plated copper plug, was top-loaded into the cavity through a central access hole. A frequency modulation technique was used, giving both the quality factor Q and the resonant frequency f_0 as functions of temperature. These quantities give, respectively, the surface resistance and reactance. We examined primarily the TE_{01p} modes (TE_{011}, and its overtones TE_{012}, TE_{013}, and

TE_{014} with resonant frequencies at 17.22, 19.58, 23.00, and 27.09 GHz, respectively). In an ideal cylindrical cavity, TE_{01p} is degenerate with TM_{11p}, but this degeneracy is lifted by perturbations in the top plate (e.g., introduction of the antennas), so that mode interference is not an issue. At the top of the cavity, where the sample was situated, the field distributions of all four modes are approximately the same. Modes with $p > 4$ could not reliably be measured due, perhaps, to inefficient coupling or heavy losses in the coaxial lines.

The temperature dependence of the surface resistance for each measurement frequency is presented in Fig. 1. Each curve has been normalized to unity above $T_c = 0.905$ K, and zero at low temperatures. This is consistent with the Mattis-Bardeen theory [13], assuming the London limit. In principle, given the field distribution over the sample's surface, as well as the unloaded (empty) cavity Q, the absolute surface resistance can be calculated. Such calculations, however, lead to inconsistent results for the four modes examined. The cause of the inconsistency is likely uncertainty in the positioning of the sample (or the bare plug, in the case of the empty cavity measurement) on which the field distribution and cavity Q sensitively depend. Fortunately, the actual normalization procedure is not critical, since we are here primarily concerned with the frequency and temperature dependence of well-defined anomalies. Superimposed on the usual monotonic responses are prominent peaks whose positions T^* depend strongly on the measurement frequency. At the lowest frequency (17.22 GHz), no anomalies are seen before background subtraction. Above T_c, the impedance is flat, implying a very weak temperature dependence of the normal state resistivity. Each resistance peak is accompanied by an S-shaped anomaly in the reactance (Fig. 1, inset), its zero crossing coinciding with the peak maximum. This is precisely the behavior one expects at a resonance crossing. Because of high noise levels and normalization difficulties, the reactance data yield little quantitative information, and so are not referred to in the following analysis.

To more accurately analyze the positions and shapes of the resistance anomalies, the monotonic backgrounds were subtracted. This was accomplished by first fitting the 17.22 GHz data with the normalized surface resistance predicted by the Mattis-Bardeen theory [13]. The only adjustable parameter was the ratio $c \equiv \Delta(0)/k_B T_c$, where $\Delta(0)$ is the value of the gap function at $T = 0$. Corresponding curves (dotted lines in Fig. 1) were then generated for the higher-frequency modes using the same value of c. The physical validity of this fitting procedure will be addressed below. For the moment, it is merely used to provide smooth reference curves for the background subtractions. The positions of the 19.58 and 23.00 GHz peak maxima can be read directly from the subtracted curves (Fig. 2); they occur at reduced temperatures $t^* = T^*/T_c = 0.79$ and 0.69, respectively. At the highest frequency, 27.09 GHz, two anomalies are observed: a peak centered at $t = 0.74$, and a broad plateau at low temperatures. Upon subtraction of the 17.22 GHz data, a broad feature below T_c appears. It is nearly obscured by noise, but a prejudiced observer might interpret it as a washed-out peak at $t \sim 0.85$.

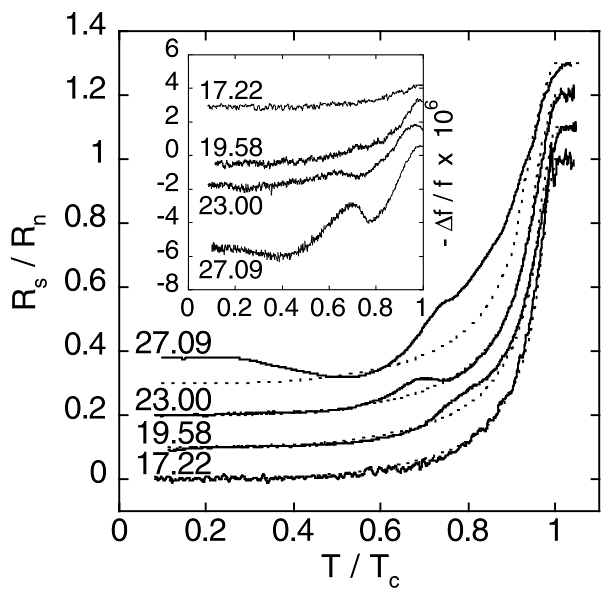

FIG. 1. Surface resistance of UBe_{13} normalized to unity at T_c and zero at $T = 0$. Successive curves have been shifted upward for clarity. Each curve is labeled with the measurement frequency in GHz. The dotted lines are the Mattis-Bardeen fits used for background subtraction. Inset: Corresponding resonant frequency shift (unnormalized reactance) for each mode.

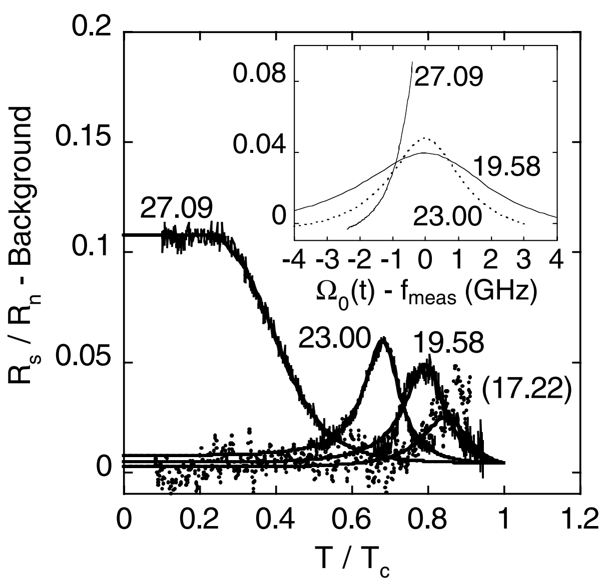

FIG. 2. Normalized surface resistance anomalies with monotonic backgrounds subtracted. The solid lines are Lorentzian curves with a temperature-dependent resonant frequency proportional to the BCS gap function. Inset: Lorentzian fits plotted versus frequency. Curves are labeled with the measurement frequency in GHz.

Our interpretation is that the resistance peaks appearing at 19.58 and 23.00 GHz, along with the 27.09 GHz plateau, represent absorption by a single temperature-dependent resonant mode with frequency $\Omega_0(t)$. In this picture, the second resistance peak seen at 27.09 GHz is presumably indicative of a separate, higher frequency mode. At lower measurement frequencies, the resonance crossing occurs at higher temperatures, implying that $d\Omega_0/dt < 0$. The development of the plateau further implies that $d\Omega_0/dt \to 0$ as $t \to 0$. This temperature dependence is similar to that displayed by the BCS gap function $\Delta(t)$. If we make the assumption that

$$h\Omega_0(t) \approx b\Delta(t), \tag{1}$$

where b is a dimensionless constant, then one would expect to see an absorption peak at a temperature t^* given by $hf = b\Delta(t^*)$, or

$$\frac{hf}{k_B T_c} = bc\,\delta(t^*), \tag{2}$$

where $\delta(t)$ is the gap function normalized to unity at $t = 0$, which is calculated from the usual BCS integral equation, and c is the (unknown) scaling factor, defined above, that gives the value of Δ at absolute zero.

Each peak was fitted with a Lorentzian line shape defined by a temperature-dependent center frequency given by (1) and an effective relaxation time τ, which is inversely proportional to the peak width (in the frequency domain). Best fits are plotted with the 19.58, 23.00, and 27.09 GHz subtracted data in Fig. 2. The inset shows the same curves in the frequency domain, where they are symmetric about the point $f_{\mathrm{meas}} = \Omega_0(t^*)$. Each fit gives an independent value of the parameter bc, or, equivalently, the mode frequency at $t = 0$: $\Omega_0(0) = bck_BT_c/h$. The results [$bc = 1.442, 1.441, 1.417$, or $\Omega_0(0) = 27.18, 27.17$, 26.7 GHz from the 19.58, 23.00, and 27.09 GHz fits, respectively] are summarized in Fig. 3. The errors in the frequency measurements, ~20 kHz, judging from the noise level, are negligible on the scale of the plot. Thermometry errors (temperature was measured using a calibrated germanium resistor) are assumed small relative to the fitting errors. The horizontal error bars in Fig. 3 are conservative estimates of the uncertainties in the crossing temperatures t^*. These errors are somewhat difficult to quantify since they depend on how the resistance curves are normalized. This is especially true in the case of the 27.09 GHz data where a well-defined zero is lacking. The position of the 27.09 GHz peak depends on whether or not $\Omega_0(t)$ is crossed at that frequency. The best fit suggests that there is no crossing ($\Omega_0 < f_{\mathrm{meas}}$ at all temperatures). If Ω_0 is crossed, there should be a peak. However, it is conceivable that a small peak might be obscured by noise. To estimate the error in $\Omega_0(0)$, fits were attempted at a range of fixed values of $\Omega_0(0)$. We conclude that $\Omega_0(0)$ could possibly lie between 26 and 27.3 GHz. The large 27.09 GHz error bar in Fig. 3 is a consequence of the flatness of the assumed $\Omega_0(t)$ at low temperatures.

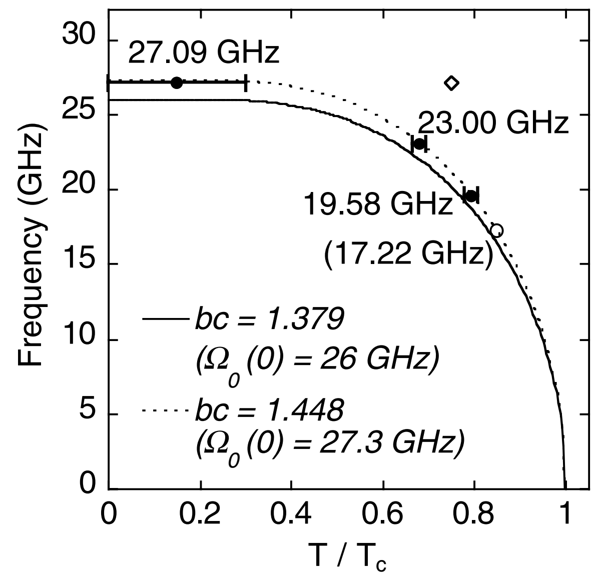

FIG. 3. Measurement frequency versus crossing temperature t^*. Solid circles represent observed surface resistance peaks. The open circle is the predicted position of the peak at 17.22 GHz, for which there is no clear evidence in the data. The curves are proportional to the BCS gap function and represent the approximate upper and lower bounds on the proposed collective mode frequency. The single diamond represents a second peak observed at 27.09 GHz.

We propose that the observed anomalies are signatures of resonant absorption by an order parameter collective mode whose temperature dependence is approximately that of the order parameter itself, as expressed in Eq. (1). A number of such modes exist in superfluid ^{3}He. The collective mode spectrum of a superconducting crystal is expected to be more limited, due to crystal field splitting. However, calculations assuming a p-wave state [4] and d-wave E_{1g} state with an order parameter of the form $k_x \pm ik_y$ [5,6] yield an optical mode, analogous to the clapping mode found in ^{3}He-A, with energy given by (1) with $b \sim 1.2$. Comparison of this with the value of the product bc determined above gives $c \sim 1.2$. Higher values of c constrain the mode to lower frequencies relative to the energy gap. Lack of an independently determined value for this parameter hampers further analysis. The BCS theory gives $c = 1.764$, but for unconventional superconductors we should not be surprised to find significant deviations. Analysis of early UBe_{13} specific heat data, assuming an Anderson-Brinkman-Morel state and including strong-coupling corrections, yielded a value between 1.65 and 1.9 [14]. A naive application of Mattis-Bardeen to the 17.22 GHz resistance curve, as described above, results in $c = 1.45$. This, however, is somewhat questionable considering that gap anisotropy has not been taken into account. Incorporating effects due to point nodes in an average way, we find that c is increased to nearly the BCS value. In addition, it is known that impurities can act as pair breakers, significantly affecting the electromagnetic response [15]. A full analysis unfortunately introduces other unknown parameters. Traditionally, ultrasound has provided an effective probe of the energy gap. The attenuation of longitudinal ultrasound in UBe_{13} [7] exhibits a clear T^2 dependence at low temperatures. Such behavior is typically taken as evidence for the existence of point nodes in the energy gap. A fit to the published data using a modification of the simple BCS expression for the attenuation coefficient [16], assuming an anisotropic energy gap of the form $\Delta(T,\theta) = \Delta_0(T)\sin\theta$, reproduces the T^2 dependence and gives $c \sim 1.2$. It has, however, been demonstrated [8] that ultrasound measurements of unconventional superconductors, when approached in such a straightforward way, can be very misleading; it is not clear that one can even differentiate between nodal structures (e.g., between point nodes and line nodes). Tunneling spectroscopy data might be less equivocal. Recent spectroscopy measurements [17] of UBe_{13} suggest a much larger value of c, placing on it a *lower* limit of 3.35. This would imply a rather low-lying collective mode with energy $h\Omega_0 < 0.43\Delta$.

It is expected that an order parameter collective oscillation will be damped both by quasiparticle excitations at the gap nodes and by the influence of impurities and other defects. The theory is not well developed at this time, but an overall decrease in damping effectiveness as $T \rightarrow 0$ has been predicted [18]. This is qualitatively consistent with our observations. The width of the observed absorption peak can be taken as a measure of the damping strength. The Lorentzian fits give an effective relaxation time τ for each measurement frequency, or, more correctly, for each crossing temperature t^*. It was assumed that $\tau = \tau(t)$ is approximately constant over the width of each peak. A physical interpretation of τ requires knowledge of the actual damping mechanisms, but it should depend strongly on the defect concentration and therefore on the normal state carrier mean-free time. The relaxation time is found to decrease dramatically as the measurement frequency f_0 is decreased (i.e., as T^* approaches T_c): $\tau = 313 \times 10^{-12}$, 197×10^{-12}, and 81×10^{-12} s at $f_0 = 27.09$, 23.00, and 19.58 GHz, respectively. The fitting errors here are quite large because the value of τ depends sensitively on the normalization procedure. As with the values of t^* and $\Omega_0(0)$, the error is especially severe in the case of the 27.09 GHz fits, where τ varies between $\sim 200 \times 10^{-12}$ and $\sim 500 \times 10^{-12}$ s, while for the other two resistance peaks the error is $\pm \sim 5010^{-12}$ s. For $f_0 = 17.22$ GHz, we have estimated a time ($\tau \sim 10 \times 10^{-12}$ s) that would render the resistive peak practically unobservable. The 17.22 GHz curve in Fig. 2 was drawn using this value of τ and $bc = 1.44$. It is included to demonstrate that the resulting peak could plausibly be hidden in the noise.

In summary, we have performed microwave surface impedance measurements of the heavy fermion superconductor UBe_{13}. Clearly seen is a resistance peak whose frequency and temperature dependence scales approximately as the BCS gap function. We interpret this as evidence of electromagnetic power absorption by an order parameter collective mode. This interpretation is consistent with existing theory. Three independent curve fits give the mode frequency at $T = 0$: $\Omega_0(0) \sim 27$ GHz. Further measurements in this vein on very pure samples of UBe_{13} as well as other heavy fermion superconductors could serve to illuminate the still undetermined symmetries of the order parameters in these materials.

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