Observation of a Collective Mode in Superconducting UBe$_{13}$

Brage Golding, D. J. Bishop, B. Batlogg, and W. H. Haemmerle

AT&T Bell Laboratories, Murray Hill, New Jersey 07974

Z. Fisk and J. L. Smith

Los Alamos National Laboratories, Los Alamos, New Mexico 87545

and

H. R. Ott

Laboratorium für Festkörperphysik, Eidgenössische Technische Hochschule—Hönggerberg, 8093 Zurich, Switzerland

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Sound propagation in the heavy-fermion superconductor UBe$_{13}$ is studied at frequencies from 0.9 to 2.4 GHz and temperatures from 0.01 to 100 K. A peak in the acoustic attenuation is observed below the superconducting transition of 0.9 K, instead of the rapid drop expected for a BCS superconductor. Absorption into a collective mode of a nonsinglet, anisotropic superconductor is proposed.

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The exotic properties of the heavy-fermion superconductors$^1$ (CeCu$_2$Si$_2$, UPt$_3$, and UBe$_{13}$) have raised questions concerning the correct description of the superconducting ground state of these systems.$^{2,3}$ It is a matter of considerable interest to determine whether the pairing interaction is in an $L=1$ (or higher) state or whether the $L=0$ Bardeen-Cooper-Schrieffer (BCS) state, or some variant, is responsible. Previous experiments with UBe$_{13}$ and UPt$_3$ have suggested qualitative deviations from BCS superconductors. For example, temperature dependences of the specific heat, thermal conductivity, ultrasonic attenuation, and nuclear spin-lattice relaxation rate are unusual in that they seem to obey power laws as $T ightarrow 0$.$^{4-8}$

The present experiments on high-frequency sound propagation in UBe$_{13}$ show a $T^2$ temperature-dependent absorption coefficient at low temperature. More significantly, the general features of the absorption near $T_c$ are grossly different from the expectations of BCS theory.$^5$ Rather than a rapid decrease below $T_c$ reflecting the appearance of the superconducting gap, an enhanced absorption occurs in UBe$_{13}$ which peaks at or just below $T_c$. This feature has no precedent in any superconductor. We interpret the peak as arising from absorption into a collective mode of the superconducting condensate. It is the analog of similar collective-mode absorptions studied extensively in the triplet superfluid phases of $^3$He.$^{11,12}$ These results provide direct evidence for additional degrees of freedom of the UBe$_{13}$ superconducting order parameter and support models based on a nonsinglet ground state.

Sound propagation was studied in a flux-grown single crystal of UBe$_{13}$, $2.4 	imes 1.5$ mm$^2$, oriented along (001). Longitudinal sound from 0.9 to 2.4 GHz was generated and detected by a sputtered thin film of ZnO. The sound velocity, absorption, and low-frequency ac susceptibility were monitored simultaneously and continuously as the sample was slowly cooled or heated in a $^3$He-$^4$He dilution refrigerator. Local regions in the sample could be probed since the 0.5-mm-diam sound beam was much smaller than the crystal dimensions.

Figure 1 shows the attenuation and velocity changes from 0.1 to 1.0 K. Both the sound velocity and ac susceptibility (not shown) indicate a superconducting transition which starts just below 0.90 K with midpoint 0.86 K. A half-width of approximately 40 mK indicates a homogeneous superconductor, representative of the best heavy-fermion crystals yet produced. The discontinuous change in velocity at $T_c$ is expected from thermodynamics. The agreement between $\Delta v/v$ and the specific-heat discontinuity $\Delta C_p$ confirms that the sound wave is coupled to the heavy-mass fermions responsible for superconductivity.$^{13}$

The sound absorption shown in Fig. 1 is anomalous. On the approach to $T_c$ from above the absorption starts to increase at the onset of superconductivity as revealed by $\Delta v/v$ and the susceptibility. The absorption increases to a maximum at the transition midpoint and decreases monotonically as $T ightarrow 0$. Experiments were carried out at six frequencies between 0.98 and 2.4 GHz. In all cases the absorption maximum $T_p$ occurred at the midpoint of the velocity discontinuity. No systematic shift in $T_p$ with frequency was detectable to within $\pm$ 10 mK.

The frequency dependence of the absorption relative to the normal state is shown in Fig. 2. The upper data are the attenuation changes at the peak, $\alpha_p - \alpha_N$; the lower data represent the attenuation change at about 100 mK, $\alpha_N - \alpha_0$. Both quantities are consistent with an $\omega^n$ law over the frequency range studied and sug-
gest a hydrodynamic regime. The overall magnitude of the electronic absorption, $\alpha_N - \alpha_S$, is extremely small. It is nevertheless in agreement with the $q L_e \ll 1$ limit of the electronic-absorption coefficient$^{14}$ ($L_e$ is the electron-scattering length extracted from the conductivity)

$$\alpha \approx \frac{(m^*E_1)^2v}{2\rho\nu\hbar^3} q L_e,$$

provided $m^*E_1$ is unrenormalized. That is, the product of the effective mass $m^*$ and the deformation potential $E_1$ possesses a normal metallic value. As discussed elsewhere,$^{15}$ the sound attenuation in both UBe$_{13}$ and UPt$_3$ behaves as if $m^*/m \sim 1$. Varma$^{16}$ has shown that $m^*E_1$ for interacting fermions is unrenormalized when the energy dependence of the self-energy dominates its momentum dependence.

The existence of an ultrasonic-attenuation peak in a superconductor has not been seen previously. Instead, it is observed that the electronic attenuation decreases below $T_c$ in accordance with the predictions of the BCS theory of superconductivity.$^{9,17}$ In the low-frequency regime, $\omega \ll \Delta$, where $\Delta(T)$ is the energy gap, quasiparticle scattering of phonons yields

$$\alpha_S/\alpha_N = 2f(\Delta),$$

where $f$ is the Fermi function, leading to an exponential decrease of $\alpha$ well below $T_c$.

A recent ultrasonic study of the heavy-fermion superconductor UPt$_3$ has shown deviations from BCS theory.$^8$ For $T \lesssim \frac{1}{3} T_c$, a $T^2$ attenuation was discovered and interpreted in terms of quasiparticle scattering of phonons in an anisotropic gap possessing a line of zeros. No evidence for a peak below $T_c$ was found. The attenuation of UBe$_{13}$ below $\approx \frac{1}{2} T_c$ is inconsistent with an exponential temperature dependence but is well characterized by a $T^2$ dependence as shown in Fig. 3. Whether the asymptotic $T \to 0$ absorption can be attributed solely to quasiparticle scattering, since the additional absorption mechanism responsible for the peak is present, is unclear. Nevertheless, this result is in general agreement with power laws inferred from recent thermal-conductivity$^5$ and NMR spin-lattice relaxation measurements.$^6$

A potential contributor to enhanced attenuation below $T_c$ in a superconductor is phonon-induced pair
breaking. It occurs when the condition

$$\hbar \omega \geq 2\Delta(T)$$

(3)
is satisfied. This condition is satisfied only very close to \(T_c\) \((T_c - T \leq 2 \text{ mK})\) for the acoustic frequencies utilized here. The magnitude of the pair-breaking absorption should be no greater than \(10^{-2}(\alpha_N - \alpha_0)\), and should drop discontinuously to the BCS value when Eq. (3) is no longer satisfied.\(^1\) This behavior is clearly not observed in Fig. 1. In the presence of an anisotropic gap, pair-breaking contributions should be even less apparent since zeros of the gap allow for Eq. (3) to be satisfied for some directions in \(k\) space, even at \(T = 0\).

The most likely candidate for the large acoustic-damping peak below \(T_c\) is absorption into collective modes. For an anisotropic superconductor, the order parameter will have components corresponding to the additional degrees of freedom associated with oscillations of orbital axes. One can view these modes as excited bound pairs whose excitation energies \(\hbar \Omega_k\) lie within the gap. If a particular mode couples to density fluctuations, one can expect absorption of sound when the condition \(\hbar \omega = \Omega_k(T)\) is satisfied.\(^2\)\(^,\)\(^3\) A recent calculation\(^4\) of collective-mode energies in an \(L = 1\) polar superfluid has predicted a low-frequency orbital mode which couples to the density. The collective mode occurs at \(1.2\Delta_0\) at \(T = 0\) but, more significantly, appears to have an unusually weak temperature dependence near \(T_c\). Thus the symmetry and weak temperature-dependent energy of the mode are suggestive of our observations near \(T_c\).

In \(^3\)He, theory and experiment have shown that the collective-mode frequencies near \(T_c\) generally scale with the gap, i.e., \(\Omega_k = a \Delta(T)\), where \(a\) is a con-

cstant.\(^1\) The resonance condition yields a peak occurring at temperature \(T_p = [1 - (\hbar \omega/2a'kT_c)^2]T_c\), with \(a' \sim 1\). The present data do not provide evidence for a frequency dependence of the peak temperature. Two factors may be responsible. First, in contrast to the zero-sound propagation in \(^3\)He, the UBE\(_{13}\) experiments take place in a hydrodynamic, collision-dominated regime. One expects any sharp, resonant structure to disappear. Second, one must take into account the inhomogeneous broadening of the transition and the subsequent averaging over the transition width. Experiments at higher acoustic frequencies should resolve this issue. Inclusion of crystal-field perturbations and spin-orbit coupling may be expected to modify collective-mode energies in crystals,\(^2\)\(^,\)\(^3\) but such corrections may prove to be minimal in a cubic system such as UBE\(_{13}\). The apparent absence of an attenuation peak below \(T_c\) in hexagonal UPE\(_{13}\) may be indicative of the role of crystal-field splittings or anisotropic pairing interactions\(^5\) in crystals of lower symmetry.

We have studied the effect of magnetic fields up to \(2 \text{ T}\) on the absorption. The peak moves to lower temperature in general accord with the phase boundary, but, surprisingly, its amplitude increases by approximately \(20\%\). The velocity discontinuity is unchanged, indicating that the transition is still sharp in the relatively weak field. In the absence of a theory of the collective-mode energies in a magnetic field, two possibilities for the enhanced absorption seem reasonable. The first is that preferred alignment of the orbital axes is occurring (\(H\) is applied parallel to the sound-wave vector). Additionally, a larger ratio of \(\omega/\Delta(H)\) in finite fields should lead to increased absorption.

In conclusion, the appearance of an acoustic-attenuation peak in the superconducting state argues for a new absorption mechanism in heavy-fermion systems. The excitation of a collective mode by sound waves is the most likely explanation for the phenomenon. We believe that this observation is the most compelling evidence yet offered for non-BCS, anisotropic superconductivity.

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3C. M. Varma, Bull. Am. Phys. Soc. 29, 404 (1984), and
The relationship between the elastic constant $c_{11}$ ($=\rho v^2_L$) and the specific-heat discontinuity at $T_c$ is 
\[
\Delta c_{11}/c_{11} = 2\Delta v/\nu = (\Delta C_p/T_c)B(dT_c/dP)^2,
\]
where $B$ is the bulk modulus. We calculate $\Delta c_{11}/c_{11} = 56 \times 10^{-6}$ using values of $\Delta C_p$ from Ref. 4 and $dT_c/dP$ from J. W. Chen et al., Physica (Amsterdam) 126B, 325 (1984). This value is in excellent agreement with $\Delta c_{11}/c_{11} = 54 \times 10^{-6}$ obtained from the data in Fig. 1.