

## Ultrasonic Attenuation in $UPt_3$

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We report measurements of the ultrasonic absorption in  $UPt_3$  through the superconducting transition. The attenuation varies as  $T^2$  at low temperature and is inconsistent with the identification of  $UPt_3$  as a singlet superconductor. Our results can be well explained by assuming that  $UPt_3$  is an anisotropic (triplet) superconductor in a polarlike state, where the gap vanishes along a line on the Fermi surface.

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The discovery of superconductivity<sup>1</sup> in a class of materials which behave as Fermi liquids with large effective masses  $O(10^2-10^3)$  has raised important questions about the nature of the superconducting state and the interactions that are responsible for it. One of us has suggested<sup>2</sup> that conventional  $s$ -wave pairing through phonon-mediated interactions is unlikely in these materials and that  $S=1$  pairs must be involved. Anderson<sup>3</sup> has further argued this point of view while Tachiki and Maekawa<sup>4</sup> and Razafimandimby, Fulde, and Keller<sup>5</sup> favor the conventional form of superconductivity. The measured anisotropy of the critical field<sup>6</sup> in  $UPt_3$  has been used to show<sup>7</sup> the existence of a polar-type  $S=1$  state, which has a line of zeros in the gap function on the Fermi surface. Also, the nonexponential nature of the specific heat in  $UBe_{13}$  has been suggested<sup>8</sup> as some indication for a nonsinglet superconducting state for  $UBe_{13}$ . The specific heat<sup>1</sup> in  $CeCu_2Si_2$  is also nonexponential but this has been taken as an indication for gapless behavior.

In this paper we report measurements of the temperature dependence of the ultrasonic attenuation in  $UPt_3$  to temperatures well below  $T_c$ . Our results are quite inconsistent with that expected for singlet pairing and consistent with  $S=1$  pairing with the gap function having a line of zeros on the Fermi surface as suggested earlier.<sup>7</sup> We have also made high-precision measurements of the sound velocity, which we will discuss elsewhere.

The measurements were performed on a large crystal of  $UPt_3$  first synthesized in an arc furnace and then zone remelted. The sample was then cut and polished to optical flatness. The sample had a final diameter of 4 mm and a finished length of 7.12 mm. The ultrasonic transducers were attached at the ends, and ultrasound of longitudinal polarization was propagated along the long direction, which

was parallel to the hexagonal  $c$  axis. The transducers were 50-MHz overtone-mode crystals of  $LiNbO_3$  and were fastened to the sample with epoxy resin. The sample was mounted in an ac susceptibility coil so that the susceptibility and the acoustical properties could be simultaneously measured. The superconducting transition occurred at about 480 mK with a width as measured by the susceptibility of  $\sim 25$  mK.

A pulse spectrometer of conventional design was used to measure the attenuation at frequencies from 50 to 600 MHz. In Fig. 1 we show the ultrasonic attenuation and the ac susceptibility. Two points are readily apparent. At  $T_c$  the attenuation does not drop off abruptly as it would for a singlet superconductor. The data shown here (at 508 MHz) and shown later at 52 MHz are consistent with the attenuation approaching  $T_c$  with zero or a very small slope. The second and the most impor-

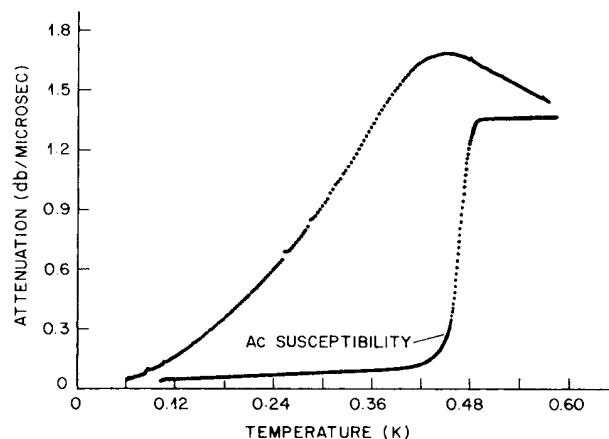


FIG. 1. The measured ultrasonic attenuation (at 507.9 MHz) and ac susceptibility for  $UPt_3$  as functions of temperature. The velocity of sound is  $3.9 \times 10^5$  cm/sec.

tant point is that at low temperatures the attenuation does not vary exponentially with temperature as is theoretically and experimentally found for singlet superconductors.

In Fig. 2 we show the attenuation plotted versus  $T^2$  for two different frequencies. We obtain a good  $T^2$  fit at low temperatures. This result is to be contrasted with  $\alpha_S \sim \exp(-\Delta/kT)$  for a singlet superconductor.<sup>9</sup> Shown in the inset in Fig. 2 is the frequency dependence of the change in electronic attenuation as measured from  $T_c$  to the extrapolated value at  $t=0$ . The solid line is proportional to  $\omega^2$ .

The  $\omega^2$  frequency dependence is observed also on the normal metallic side where the attenuation varies roughly as the conductivity.<sup>6</sup> This suggests<sup>10</sup> that we are in the limit  $ql \ll 1$ , where  $q$  is the wave vector of sound and  $l$  is the electron mean free path. On the superconducting side the attenuation can result from three mechanisms: (a) quasiparticle scattering with absorption of sound, (b) pair creation and annihilation, and (c) absorption by collective modes of the superconducting state. Except for very near  $T_c$  we expect (a) to dominate over (b). Experimentally we have found no good evidence for (c), on which we will comment later.

We will concentrate on the calculation for quasiparticle scattering. In the limit  $ql \ll 1$  but  $l/\xi \gg 1$ ,<sup>11</sup> the longitudinal sound attenuation coefficient is given by<sup>12</sup>

$$\alpha_S = \pi \lambda^2 l \sum_{\vec{k}\vec{k}'} \text{Tr}[f(E_{\vec{k}}) - f(E_{\vec{k}'})] \left[ 1 - \frac{\bar{\Delta}_{\vec{k}}^+ \bar{\Delta}_{\vec{k}'}^-}{E_{\vec{k}} E_{\vec{k}'}} \right] \delta(\omega - E_{\vec{k}} + E_{\vec{k}'}), \quad (1)$$

where  $\lambda$  is the electron-phonon matrix element,  $\bar{\Delta}_{\vec{k}}$  is the gap function,<sup>13</sup> and  $E_{\vec{k}}$  is the quasiparticle energy. This will in general yield  $\alpha_S \sim \omega^2$ , but its behavior as a function of temperature depends on the type of superconducting state.

For singlet superconductors this yields the classic result<sup>9</sup>

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

where  $\Delta(T)$  is the singlet superconducting gap; this gives an attenuation which decreases exponentially at low temperatures and clearly disagrees with our results in UPt<sub>3</sub>.

For anisotropic superconductors  $\alpha_S/\alpha_N$  will depend on the type of state realized. These may be generally grouped into three classes: (a) States in which the gap exists over the entire Fermi surface (like the Balian-Werthamer or isotropic state in <sup>3</sup>He). For these  $\alpha_S/\alpha_N$  has the temperature dependence given in Eq. (2). (b) States in which the gap vanishes at points on the Fermi surface (like the Anderson-Brinkman-Morel or axial state in <sup>3</sup>He). This has a low-energy density of states  $\sim E^2$  and can be shown to have an attenuation proportional to  $T^4$  at low temperatures, in disagreement with our

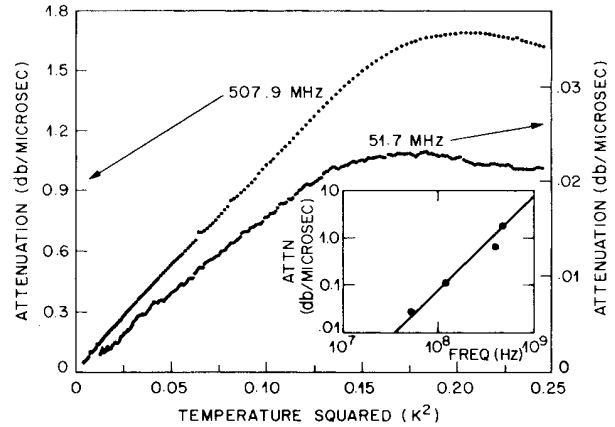


FIG. 2. Ultrasonic attenuation at two different frequencies plotted vs  $T^2$ . The inset shows the frequency dependence. The solid line has an  $\omega^2$  frequency dependence.

observations. (c) States in which the gap vanishes along a line (or lines) on the Fermi surface. For these the density of states for quasiparticles is quite generally proportional to  $E$  for  $E \ll \Delta$ . To be specific we consider the polar state of  $L=1$  symmetry,  $\Delta_{\vec{k}} \sim \Delta \hat{k}_z$ . The density of states is

$$N(E) = \begin{cases} (\pi/2)n(0)(E/\Delta), & E < \Delta, \\ n(0)(E/\Delta)\sin^{-1}(E/\Delta), & E > \Delta. \end{cases} \quad (3)$$

The normalized ultrasonic attenuation coefficient calculated for this state from Eq. (1) has the form

$$\alpha_S/\alpha_N \sim (T/\Delta)^2 \quad \text{for } T \ll \Delta. \quad (4)$$

As was shown in Fig. 2 this temperature dependence is found experimentally. To compare theory and experiment over the entire temperature range  $\alpha_S/\alpha_N$  must be evaluated numerically. This is shown in Fig. 3. For  $\Delta(T)$  we have used the BCS expression with  $\Delta(T=0) \cong 2.6T_c$  chosen to obtain the best fit. Also shown in Fig. 3 is  $\alpha_S/\alpha_N$  for a BCS singlet superconductor using  $\Delta(T=0) = 1.76T_c$ .<sup>14</sup> The data and fit shown in Fig. 3 are strong evidence that UPt<sub>3</sub> is an anisotropic (triplet)

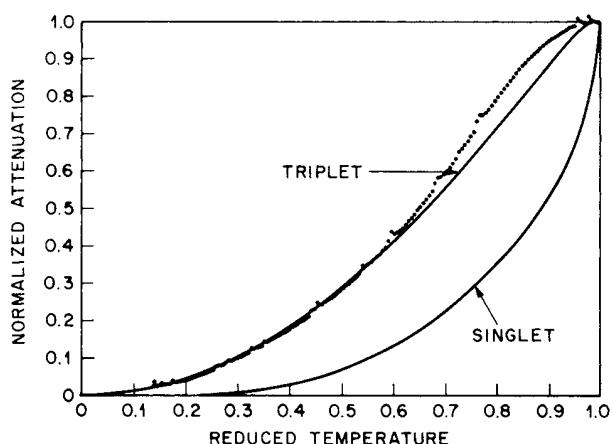


FIG. 3. The normalized attenuation vs normalized temperature for the data at 508 MHz (dots). Also shown are the theoretical results discussed in the text.

superconductor with a polarlike state.

The discrepancy near  $T_c$  can be due to a number of reasons: (i) The  $\sim 25$ -mK width of the transition when taken into account gives a theoretical behavior near  $T_c$  closer to the experimental result without affecting the low-temperature fit. (ii) The superconducting state must have lines of vanishing gap but without precisely being the state assumed. This is almost certainly so given the strong spin-orbit and crystal-field couplings. (iii) There may be strong-coupling corrections of both the Eliashberg and the Fermi-liquid type: The value of  $\Delta(0)/T_c$  of  $\sim 2.6$ , about twice the weak-coupling value, indicates this but in view of (ii) should not be considered conclusive at this point. (iv) There is absorption into collective modes. For  $T - T_c \sim \hbar\omega/k_B T$  the ultrasonic attenuation in  $^3\text{He}$  is dominated by resonant absorption by collective<sup>15</sup> modes of the orbital angular momentum variables. In materials like  $\text{UPt}_3$ , the crystal-field energies and the spin-orbit energies are considerably larger than even the effective heavy-electron bandwidth. Therefore, continuous symmetries for spin and orbit quantization axes do not exist. Propagating collective modes are therefore not expected. However, relaxational modes of the spin-orbit axes are still to be expected with significant amplitude near  $T_c$ .

In conclusion, we have measured the ultrasonic attenuation and sound velocity in  $\text{UPt}_3$ . Our results are unambiguous that superconductivity in this compound is not due to singlet pairing. We obtain good agreement with a model assuming anisotropic (triplet) pairing with a polarlike state.

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<sup>11</sup>From the resistivity, specific heat,  $T_c$ , and sound velocity one can estimate  $l \geq 1000 \text{ \AA}$ ,  $\xi \leq 200 \text{ \AA}$ , and the wavelength of the ultrasound at 500 MHz to be  $\sim 10 \mu\text{m}$ .

<sup>12</sup>This may be obtained as a modification of the relevant part of Eq. (64) of R. Balian and N. R. Werthamer, *Phys. Rev.* **131**, 1553 (1963) for the case  $ql \ll 1$ .

<sup>13</sup>See for example, P. W. Anderson and W. F. Brinkman, in *The Physics of Liquid and Solid Helium, Part II*, edited by K. H. Bennemann and J. B. Ketterson (Wiley, New York, 1978); A. J. Leggett, *Rev. Mod. Phys.* **47**, 331 (1975).

<sup>14</sup>A value of  $\Delta(0)/T_c$  of 2.6 for the singlet state would worsen the discrepancy with the experimental result.

<sup>15</sup>See for example, P. Wölfle, in *Progress in Low Temperature Physics*, edited by D. F. Brewer (North-Holland, Amsterdam, 1978), Vol. 7A.