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# Anisotropic (triplet) superconductivity in $UPt_3$ (invited)

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Ultrasound experiments were performed on single crystals of the heavy Fermion superconductor $UPt_3$. From the ultrasound attenuation below $T_c$, a quasiparticle density of states linear in energy is deduced, which we interpret as due to an anisotropic polar-like (triplet) superconducting state. This is further supported by the agreement of the only fitting parameter $\Delta_0/kT_c$ with the theoretical value for polar $p$-wave pairing. The attenuation in the normal state gives the electron-phonon interaction strength and the product of the deformation potential times the effective mass is found to be the same as for ordinary "light" Fermion systems. The observed variations of the sound velocity are discussed.

## I. INTRODUCTION

In a small class of Ce- and U-based compounds the low-temperature properties are those of a heavy Fermi liquid which condenses in a superconducting state.[1-3] Both this ground state and the pairing interaction are quite possibly of a nonconventional type. Whereas one of us[4] and Anderson[5] suggested $S = 1$ pairing in these compounds, Razafimandimby, Fulde, and Keller[6] and Tachiki and Maekawa[7] described the heavy Fermion system with the Anderson lattice model and found a singlet superconducting state to be stable under certain conditions. In this paper we present the results of ultrasonic experiments on $UPt_3$ below 1 K. The temperature dependence of the ultrasound attenuation in the superconducting state reveals information on the quasiparticle density of states, and from the magnitude of the attenuation in the normal state, the product of the effective mass times the electron-phonon coupling strength will be deduced.

## II. SAMPLE PREPARATION

$UPt_3$ was synthesized in an argon arc furnace, using highly purified argon. The starting materials were nominally 99.98% pure uranium and 99.99% pure platinum. The melt was subsequently cast into an ingot of about 6 mm diameter and 12 cm length, and transferred into a UHV float-zone apparatus with oil-free vacuum, built by Leybold Heraeus. Float zoning is possible due to the very congruent melting behavior of this compound. Special care, however, was necessary to achieve a very stable narrow zone, due to its high density and moderate surface tension. After a few passes, low angle grain boundaries develop along the crystal axis, leading to nearly single-crystal behavior. Single crystals can be obtained by necking and simultaneously rotating the lower part at 30 rpm and speeds of up to 0.5 mm/min.

For the ultrasound experiments, the sample was cut and mechanically polished to optical flatness. Transducers of $LiNbO_3$ produced longitudinally polarized sound that propagated close to the hexagonal $c$ direction of the sample. During the ultrasound experiments, the ac susceptibility was monitored with coils wound around the sample. The transition temperature determined this way is 460 mK with a width of $\sim 25$ mK. The attenuation in the frequency range from 50 to 600 MHz was measured with a pulse spectrometer of conventional design.

## III. ATTENUATION BELOW $T_c$

Ultrasound attenuation measurements are very useful to study superconductivity and, historically, they confirmed, together with NMR experiments, microscopic aspects of the BCS theory. For singlet pairing, the longitudinal sound attenuation in the superconducting state $\alpha_s$, normalized to the normal state attenuation $\alpha_n$, is simply twice the Fermi function of the temperature-dependent energy gap $\Delta(T)$. Accordingly, $\alpha_s/\alpha_n$ drops abruptly at $T_c$ and decreases exponentially as the temperature is reduced well below $T_c$. The results on $UPt_3$ are in sharp contrast to this classical behavior (Fig. 1). Most characteristically, $\alpha_s/\alpha_n$ increases proportionally to $T^2$, rather than exponentially. In addition, $\alpha_s/\alpha_n$ does not change abruptly at $T_c$; not even a change in slope is noticed at $T_c$.

The solid line in Fig. 1, labeled "triplet," is a fit to the data of an absorption curve based on the assumption that superconducting pairing in $UPt_3$ leads to a polar-like anisotropic state. In such a state, the gap $\Delta(\mathbf{k})$ vanishes along one or more *lines* on the Fermi surface. Quite generally, the resulting density of quasiparticle states $n(E)$ is linear in energy for $E \ll \Delta$ (Ref. 8). The ultrasound attenuation by quasiparticle scattering involves the product of initial and final density of states, and is thus proportional to $(T/\Delta)^2$ for $T \ll \Delta$.

Other unconventional superconducting states may have either $\Delta(\mathbf{k})$ finite everywhere on the Fermi surface (analogous to the Balian–Werthamer state in $^3$He) or $\Delta(\mathbf{k})$ vanishing at *points* on the Fermi surface (analogous to the Anderson–Brinkman–Morel or "axial" state in $^3$He). In the former case $\alpha_s/\alpha_n$ at $T \ll \Delta$ is the same as in the singlet case, whereas $\alpha_s/\alpha_n$ in an "axial-like" state varies at $T^4$, due to the $E^2$ dependence of $n(E)$. Based on these criteria well below $T_c$ the $UPt_3$ data are clearly consistent with a polar-type anisotropic state.

The "triplet" curve in Fig. 1 is calculated from the full

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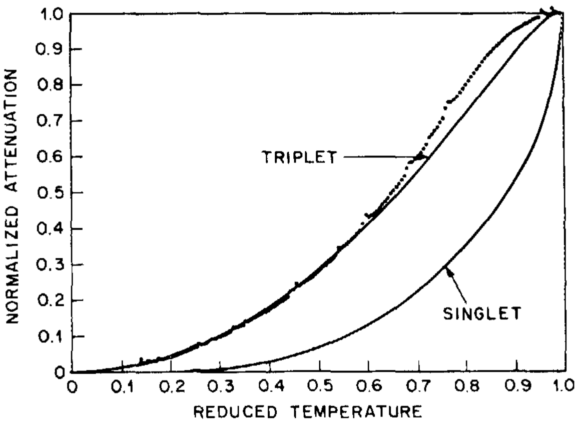

FIG. 1. Normalized ultrasound attenuation as function of the reduced temperature. The curve labeled "triplet" is a fit to the data of the attenuation expected for a polar-like anisotropic superconductor. The only adjustable parameter is $\Delta_0/kT_c$, and its value of 2.6 agrees with the theoretical one (2.46, Ref. 8).

expression for $n(E)$ and a variation of $\Delta$ with $T$ according to BCS. The best agreement with experiments is found for $\Delta\,(T=0) = \sim 2.6\,T_c$. We have become aware of recent theoretical studies of the universal constant $\Delta_0/kT_c$ for various anisotropic superconductors.[8] For a polar $p$-wave pairing this constant is 2.462 and the closeness of our experimentally determined value further supports the interpretation given above. The remaining small discrepancies in Fig. 1 may have several reasons, which are discussed in Ref. 8. In short, they are (i) finite width of the transition; (ii) details of the polar-like state may be different from the assumption $\Delta\,(\mathbf{k}) = \Delta \hat{k}_z$; and (iii) collective mode absorption.

We would like to point out that $\alpha_s/\alpha_n$ obeys a $T^2$ law over a wide temperature range $(0.1 \leqslant T/T_c \leqslant 0.6)$ which in turn indicates $n(E) \sim E$ up to $E \sim 0.6\Delta$. Obviously, this puts very stringent requirements on any interpretation other than in terms of a polar-like state. In particular, traditional "pair-breaking" mechanisms, of whatever origin the perturbing potential may be, do not result in $n(E) \sim E$ up to $E \sim 0.6\Delta$. Therefore, the temperature dependence of $\alpha_s/\alpha_n$ in the pair-breaking situation[9] is drastically different from the results in Fig. 1.

The linear density of states in a polar-like state leads at $T \ll T_c$ to a specific heat $\sim T^2$, an electronic heat conductivity $\sim T^2$, and a nuclear spin relaxation rate $\sim T^3$. Recent NMR[10] and thermal conductivity[11] measurements on $\mathrm{UBe}_{13}$ found power laws typical for the polar-like state, whereas specific heat data favored an axial-type state.[12]

## IV. ATTENUATION IN THE NORMAL STATE

The normal state attenuation is of interest because its magnitude depends on the electron-phonon interaction strength, a quantity so far unknown for heavy Fermion systems. The standard theory of phonon-electron scattering[13–15] describes the perturbation of the electron's potential due to lattice vibrations in the form of a deformation potential $E_1$

$$E\,(\mathbf{k},\delta) = E_0 + \hbar^2\mathbf{k}^2/(2m^*) + E_1\nabla\delta, \tag{1}$$

where $m^*$ is the band effective mass and $\delta$ the deformation. Taking into account the finite electron mean free path $l_e$, the attentuation coefficient $\alpha_n$ for longitudinal sound becomes

$$\alpha_n = [(m^*E_1^2)\nu/2\rho v^2\hbar^3]\,ql_e, \tag{2}$$

for $ql_e < 1$. Here $\nu$ is the sound frequency, $v$ the sound velocity, $\rho$ the density, and $q(=2\pi\nu/v)$ the phonon wave number. In the limit of small $ql_e$ the attenuation is proportional to the mean free path ($\sim$ conductivity) and to the square of the frequency $\nu$. This is observed in $\mathrm{UPt}_3$. The electrical resistivity close to $T_c$ varies as $\rho_0 + AT^2$ (Refs. 16 and 17), resulting in a $T^2$ dependence of $\alpha_n$. In Fig. 2 $\alpha(T)$ is plotted versus $T^2$ to show that both $\alpha_n$ and $\alpha_s$ vary as $T^2$. This plot allows a simple determination of $\alpha_n$ at $T > 0$ and $\alpha_s/\alpha_n(T)$. The attenuation was measured at four frequencies between 52 MHz and 508 MHz, and the $\nu^2$ dependence of the electronic contribution to $\alpha(T=0)$ is shown in Fig. 3. In order to evaluate the microscopic quantity $m^*E_1$, in (2), the mean free path $l_s$ has to be estimated. We assume $l_e$ to be 200–800 Å from existing resistivity data,[16,17] but keep in mind the simplifying assumptions necessary to calculate $l_e$. Since we do not know $m^*$ or $E_1$ independently, only the product $m^*E_1$ can be determined from the attenuation. The value for $m^*E_1/m_0$, with $m_0$ being the free electron mass, is 6–12 eV (800 Å $> l_e >$ 200 Å). This is a rather unexpected result when compared to ordinary metals like In or Sn, where $m^*E_1/m_0$ ranges from 4–7 eV (Refs. 14 and 18). Given the uncertainty in $l_e$, we consider the 6–12 eV for $\mathrm{UPt}_3$ not significantly different from the "light" Fermion systems. One concludes that the mass enhancement in the heavy Fermion systems, known from specific heat experiments to be $\sim 200$ for $\mathrm{UPt}_3$ (Refs. 3 and 19), either is not probed in the ultrasound experiments, or it is compensated by a corresponding reduction of the deformation potential. For a general discussion of similar effects see Ref. 20.

## V. SOUND VELOCITY

The longitudinal sound velocity $v$ in the $c$ direction decreases with increasing temperature up to 17 K, where it passes through a minimum and increases then up to at least 20 K. The total reduction of $v$ between 0 K and 17 K is 1300 ppm. The minimum in $v$ occurs close to the basal-plane sus-

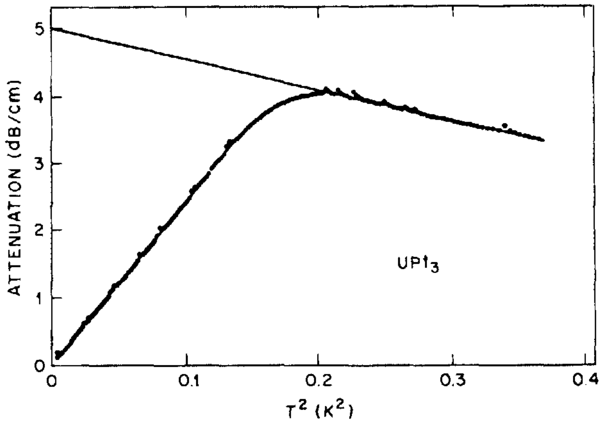

FIG. 2. Ultrasound attenuation as function of $T^2$. This plot emphasizes the $T^2$ dependence of $\alpha$ for $T \ll T_c$.

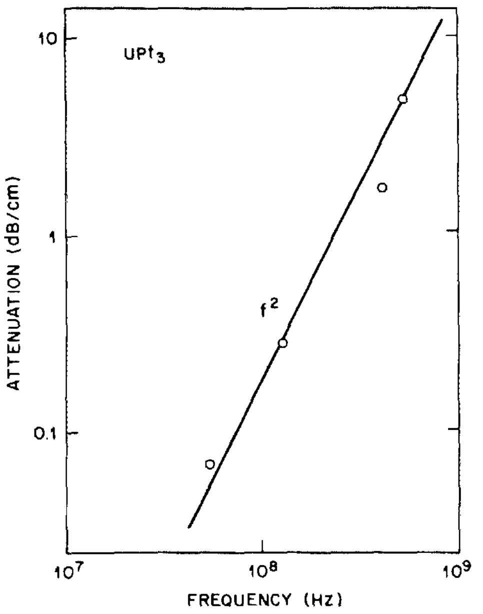

FIG. 3. Frequency dependence of the $T \to 0$ normal state attenuation.

ceptibility maximum at 17.5 K in this sample. The sound velocity in the basal plane, however, monotonously decreases with temperature up to 20 K. The coefficient of thermal expansion is positive in the basal plane and negative ($T < 45$ K) in the $c$ direction, and exhibits maxima between 10 and 15 K (Ref. 21). An interpretation of all these anisotropies, including also the susceptibility,[19] possibly involves low-lying crystal-field split multiplet levels, similar to the ones found in UPt$_3$ (Ref. 22).

The most prominent feature in Fig. 4 is the rapid decrease of $v$ when the sample is cooled below $T_c$. When the smooth background is subtracted, as shown in the insert to Fig. 4, $\Delta v/v$ amounts to 13 ppm. Such a change in $v$ is a consequence of the superconducting transition and standard thermodynamic derivation gives a change of the bulk modulus at $T_c$

$$\frac{\Delta B}{B} = B\left[\frac{H_c}{4\pi}\frac{\partial^2 H_c}{\partial p^2} + \frac{1}{4\pi}\left(\frac{\partial H_c}{\partial p}\right)^2\right]. \tag{3}$$

Here $\partial H_c/\partial p$ is the pressure derivative of the thermodynamic critical field $H_c$. Close to $T_c$, $\partial^2 H_c/\partial p^2$ is usually neglected. A different version of Eq. (3), where $\Delta B/B$ is expressed in terms of uniaxial stress dependencies of $H_c$, would be appropriate to compare $\Delta v/v (= \frac{1}{2}\Delta C_{33}/C_{33})$ with other experiments.[18] But because only the "hydrostatic" pressure dependence of $H_c$ can be inferred from existing measurements, the following numerical analysis will demonstrate the general agreement among the various data. The upper critical field slope[17,23] and $T_c$ have been measured under pressure, and $d \ln H'_{c2}/dp$ and $d \ln T_c/dp$ were found to be the same ($-0.028 \pm 0.001$ kbar$^{-1}$). Combining $H_{c1}$ and $H_{c2}$ measurements,[24] the thermodynamic critical field $H_c$ is estimated to be 500–600 Oe, and $dH_c/dp = 15.5 \pm 2$ Oe/kbar. The bulk modulus of UPt$_3$ is unknown, but comparing longitudinal sound velocities with the ones of Pt metal, we assume $B = \sim 3$ Mbar. Equation (3) then gives $\Delta B/B = 54 \pm 14$ ppm. Since $B$ involves more elastic constants besides $C_{33}$, the 26 ppm contribution of $\Delta C_{33}/C_{33}$ to $\Delta B/B$ appears very reasonable.

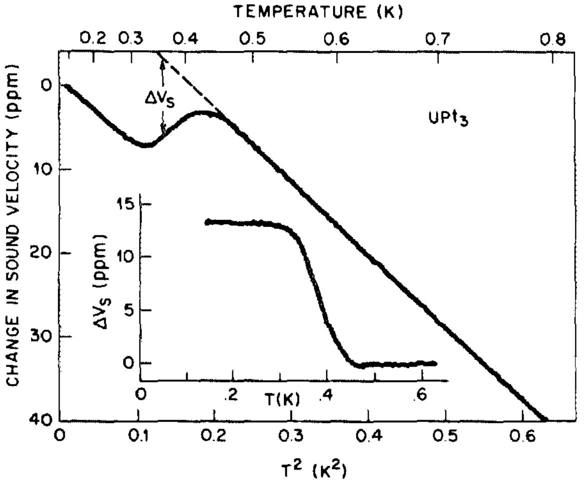

FIG. 4. Temperature dependence of the sound velocity change. The insert shows the change associated with the superconducting transition.

## VI. CONCLUSION

The results of our ultrasound experiments on UPt$_3$ strongly suggest that this heavy Fermion system condenses into an anisotropic (triplet) superconducting state. The observed $T^2$ dependence of the attenuation below $T_c$ is typical for a polar-like state in which the gap vanishes along one or more lines on the Fermi surface. Also, the value of $\Delta_0/kT_c$ supports this interpretation. The normal state attenuation, particularly its magnitude, is the same as in normal metals. This means that the product of the effective mass times the deformation potential (electron-phonon coupling) is similar in "heavy" and in "light" Fermion systems.

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