Transverse ultrasound revisited: A directional probe of the A phase of UPt_3

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We have measured the transverse ultrasonic attenuation $\alpha_{q\epsilon}$ in a well-characterized single crystal of UPt_3 for propagation direction $\mathbf{q}$ in the basal plane and polarization ϵ in and out of the plane. Unlike previous measurements, the sample was of sufficient purity to exhibit three well-defined phases as a function of temperature and magnetic field. We uncover a functional dependence for the attenuation in the (zero-field) high-temperature (A) phase which is distinctly different than that in the low-temperature (B) phase. Our data provide new directional information on the quasiparticle momentum distribution in this largely unstudied phase of UPt_3 and direct evidence that the two zero-field phases possess different order parameters. [S0163-1829(96)03633-8]

Following the discovery of multiple superconducting phases in UPt_3[1] numerous theoretical investigations have attempted to predict the gap structure in each of the three phases. These calculations have been very successful in predicting measured phase diagrams obtained under magnetic field, hydrostatic pressure, and uniaxial stress. Indeed, the theoretical H-T-P phase diagram agrees with that observed under every experiment to date.[2,3] While many experiments agree as to the location of the phase lines, it is notable that there are very few direct measures of the *gap structure* within each phase. Here, ''direct'' means an experimental probe that couples to the quasiparticle density in such a way that the quasiparticle momentum distribution may be mapped. Potential examples include thermal conductivity,[4] ultrasound attenuation,[5] muon spin resonance,[6] and point contact spectroscopy,[7] each of which has yielded insights into the gap structure of the low-temperature (B) phase. Studies of the high temperature (A) phase have been very limited, however, largely due to the very small temperature range (about 60 mK) over which the A phase exists. Thus thermal conductivity data, which have strongly constrained parameters for the B phase, are insufficiently sensitive to the growth of the gap near T_c to similarly address the properties of the A phase. Point contact spectroscopy, while an elegant probe of the gap structure, is similarly limited since the exact temperature of the A-B phase transition was not known for the samples used. Considering the small temperature region over which the A phase may be studied, it is essential to perform experiments on independently characterized samples using an experimental probe which is sensitive to the onset of superconductivity. The data presented here satisfy these criteria and constitute the most definitive evidence for a different gap structure in the A and B phases.

Transverse ultrasonic attenuation is one of the most powerful tools for investigating the geometric properties of the superconducting gap. The strengths of transverse sound attenuation $\alpha_{q\epsilon}(T)$ lie in its strong dependence on the magnitude of the gap and also on its highly directional nature involving two independent vectors, the propagation direction $\mathbf{q}$ and the polarization ϵ. Ultrasonic attenuation changes very sharply on entering the superconducting state. The relative sensitivity of sound attenuation as compared to, for example, the thermal conductivity κ may be motivated by considering the energy dependence of the integrand of the appropriate BCS quantities:

$$\kappa \sim \int_{\Delta(T)}^{\infty} E^2 \frac{\partial f}{\partial E} dE, \quad \alpha \sim \int_{\Delta(T)}^{\infty} \frac{\partial f}{\partial E} dE. \tag{1}$$

Both α and κ depend on the number of quasiparticles per unit energy and thus on the derivative of the Fermi function, $\partial f/\partial E$. However, the expression for κ has an energy prefactor E^2 because thermal conductivity depends on energy transport. Since $\partial f/\partial E$ is peaked at $E=0$, as the gap $\Delta(T)$ becomes nonzero the energies contributing the most to the α integral are excluded and the attenuation drops sharply (as $\alpha(T)=2f[\Delta(T)]$). The integrand of κ, however, is zero at $E=0$ and thus the thermal conductivity is a much weaker function of Δ near T_c.

Transverse sound attenuation is also very sensitive to gap anisotropy. Indeed, it was transverse attenuation data that first provided definitive evidence for a highly anisotropic gap in UPt_3.[5] These measurements found a linear dependence of the attenuation for α_{ab} ($\epsilon||\hat{b}$ and $\mathbf{q}||\hat{a}$) while $\alpha_{ac}\sim T^3$. Thus the measured attenuation was not only decidedly non-BCS [$\alpha_{ij}\sim\exp(-\Delta/T)$ for all i,j at low T] but also manifestly anisotropic. In the ''dirty limit'' (see below) appropriate to all experiments to date, the analysis of Kadanoff and Falko,[8] later extended to the resonant impurity case by several groups,[9] gives for the attenuation (outside of any gapless regime near $T=0$)

$$\alpha_{q\epsilon} \propto \int_0^{\infty} d\omega \frac{\partial f(\omega)}{\partial \omega} \frac{1}{\mathrm{Im}(\Sigma_0)} \left\langle p_\epsilon^2 p_q^2 \frac{\sqrt{\omega^2-\Delta(p)^2}}{\omega} \right\rangle_p, \tag{2}$$

where $\Sigma_0(\omega,T)$ is the electronic self-energy (independent of polarization or propagation direction), $\Delta(p)$ is the gap, p_ϵ

and p_q are the projections of momentum p along the appropriate directions on the Fermi surface, and $\langle\rangle_p$ is an average over the Fermi surface restricted so that $\omega^2-\Delta(p)^2>0$. The function Σ_0 includes the effects of resonant impurity scattering[10] which modify the energy-dependent electronic mean free path. The point is that the experimental parameters q and ϵ act solely as weights in the Fermi surface average of a function of Δ_p. Therefore, in principle, the transverse attenuation allows one to directly map out the k space location of nodes. As an example, we consider the effect on α_{ac} of a line node in the hexagonal basal plane of UPt_3 [e.g., $\Delta(k_c=0)=0$]. The product of momenta $p_q p_\epsilon$ in Eq. (3) is then equal to $p_a p_c$, which vanishes in the basal plane. Thus the contribution of a line node to the attenuation α_{ac} is much reduced as compared to its contribution to α_{ab}. That this is in qualitative agreement with the results of Ref. 5 constitutes strong evidence that there is a basal plane line node in the B phase of UPt_3. The *relative* sizes of the α_{ij} for various model gaps may all be qualitatively explained in like manner. However, a caveat is essential: The functional form of $\alpha(T)$ may depend strongly on the form of Δ_p far from the node, even at relatively low temperatures. Thus only a careful, self-consistent calculation can be confidently compared with experimental data. Detailed calculations[9] based on Eq. (1) found the aforementioned experimental results[5] to be in qualitative agreement with a gap having a line node in the basal plane. These data, however, do not bear on the question of the gap structures of the multiple phases in UPt_3 insofar as the sample used predated the growth of high-purity, low-strain crystals exhibiting the now familiar phase diagram. We have performed transverse sound attenuation studies on a single crystal of UPt_3 which is known to support three superconducting phases in the temperature-field plane. Most notably, our data are unique in providing an incisive directional probe of the gap structure in the high-T–low-field A phase.

The sample used was a single-crystal rectangular prism approximately $1\times1\times2.7$ mm^3 in size. The sound was propagated along the long dimension which corresponds to the crystallographic a axis. Ultrasound was generated using 30 MHz lithium niobate transducers. The attenuation was measured using a conventional pulse-echo interferometer in a transmission geometry at frequencies ranging from 90 to 290 MHz. The frequency dependence of the attenuation was as expected for a metal, a point which will be considered in more detail below. The sample is the same one used by Adenwalla *et al.*[11] in their careful study of the phase boundaries using longitudinal sound velocity measurements. We therefore know to high precision the location of the phase boundaries *a priori*, an important advantage when studying the functional form of the attenuation over the small temperature range covered by the A phase. The T_c's obtained from our data are in excellent agreement with those obtained in Ref. 11: $T_c^+=495$ mK and $T_c^-=435$ mK.

The measured attenuations for polarization in and transverse to the hexagonal basal plane are shown in Figs. 1 and 2, respectively. The normal state attenuation is a decreasing function of temperature, which is a consequence of the fact that the electrical conductivity also falls with increasing temperature. [For a Fermi liquid in the dirty limit,[13] the attenuation is proportional to the electronic mean free path which,

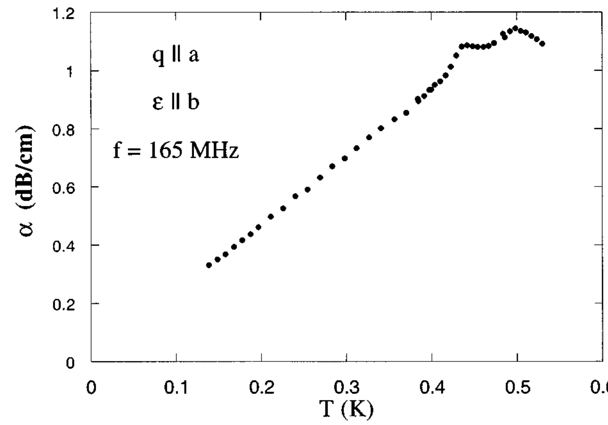

FIG. 1. Transverse ultrasonic attenuation α_{ab} for propagation along $\hat{a}$ and polarization along $\hat{b}$. The low-temperature attenuation shows no curvature down to 130 mK.

if we ignore, e.g., mass anisotropy, implies that $\alpha(T)\propto\sigma(T)$.] The experimental technique determines $\alpha(T)$ up to an additive constant corresponding to sound loss in cables, bonding layers, and so forth. We fix this parameter by demanding that $\alpha(T=0)=0$. It is important to realize that there is no thermodynamic reason for this condition to hold. Indeed, some pertinent theories predict a nonzero attenuation at zero temperature. Therefore this experiment (or any similar work of which we are aware) cannot directly address the issue of a residual attenuation at $T=0$ reflecting a finite density of zero-energy excitations. (This question is addressed by recent thermal conductivity results.[4]) Independent of any theoretical interpretation, it is evident that our B phase results (i.e., $T<435$ mK) are in agreement with those of Ref. 5: $\alpha_{ab}\sim T$ and $\alpha_{ac}\sim T^n$ with $n\sim3$. The exact power law dependence of the attenuation should not be taken too seriously, particularly for α_{ac}. We also note that the absolute magnitudes of our data are in general agreement with Ref. 5 after appropriate frequency normalization, as discussed below.

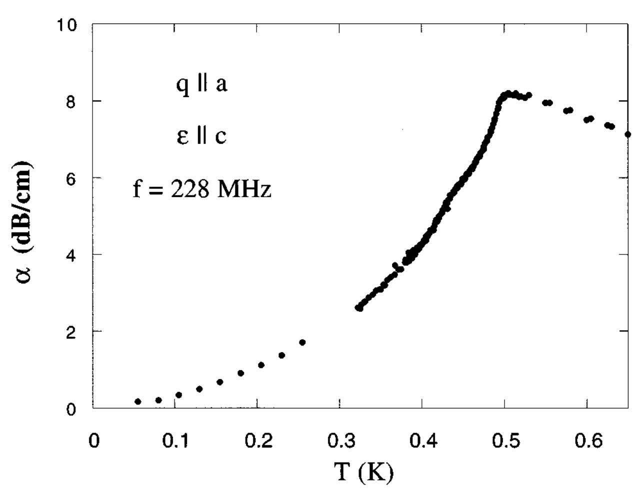

FIG. 2. Transverse ultrasonic attenuation α_{ac} for propagation along $\hat{a}$ and polarization along $\hat{c}$. The attenuation drops much more quickly than α_{ab}, reflecting the insensitivity of α_{ac} to quasiparticles in the basal plane.

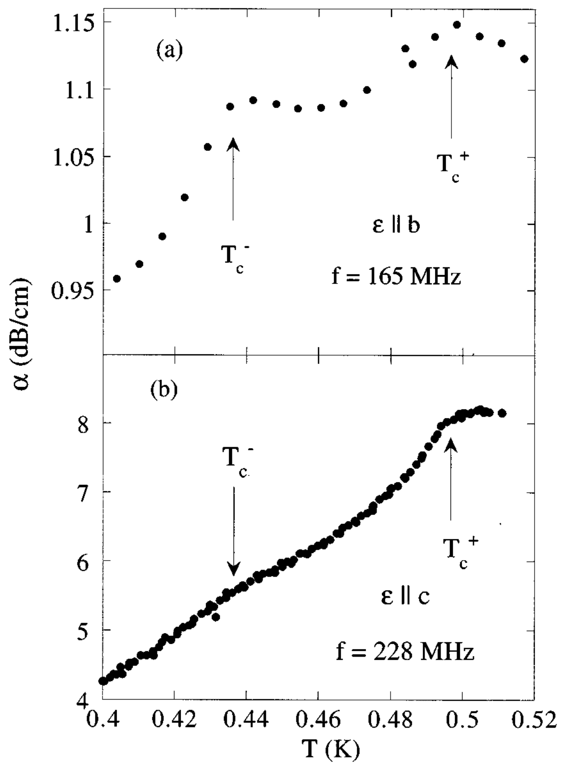

FIG. 3. (a) Detail of α_{ab} in the A phase. After a drop at T_c, the attenuation reaches a plateau before dropping sharply on entering the B phase. This behavior reflects a larger quasiparticle density in the A than in the B phase. The arrows designate T_c^- and T_c^+ as determined from longitudinal sound velocity data Ref. 10. (b) Detail of α_{ac} in the A phase. The onset of the B phase is evident as a small "bump" near $T_c^- = 435$ mK.

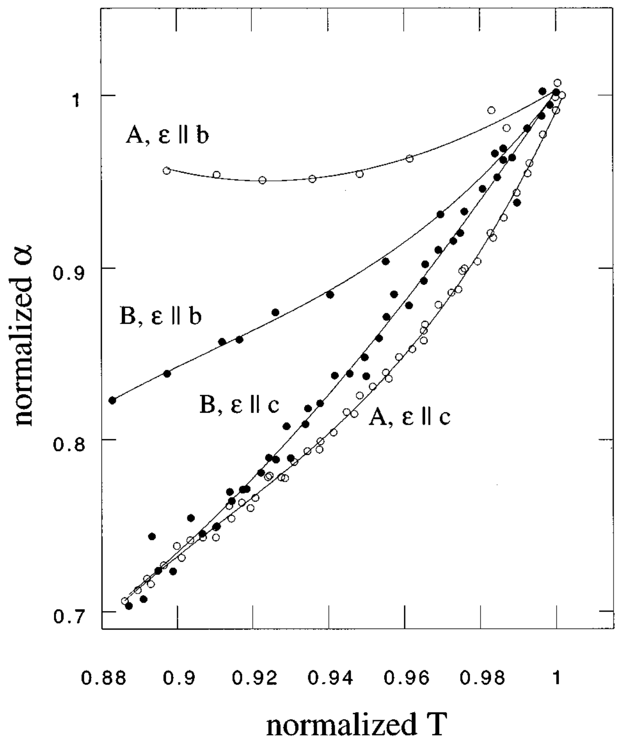

FIG. 4. Transverse attenuation data for both polarizations normalized to the value of the attenuation at T_c^+ and T_c^- as a function of $T/T_c^{+,-}$ This choice of normalization allows us to compare the attenuation in the A and B phases over the same (reduced) temperature range. The A phase shows an enhanced in-plane attenuation vs the B phase while the out-of-plane polarization data are roughly equal in the two phases. The lines are guides to the eye.

The main results of this work are the sharp differences observed in the attenuation of the A phase (500 mK $>$ T $>$ 435 mK) as compared to the B phase. As may be seen in Fig. 3, α_{ab} drops quickly with decreasing T before becoming roughly constant, while α_{ac} has a "bump" seemingly superimposed on the sharply falling attenuation observed in the B phase. Interestingly, the data of Fig. 3 show that $\alpha(T)$ attains its low-temperature asymptotic functional form for phase B within a small range $\Delta T \leqslant 25$ mK below T_c^- Making an analogous assumption about the normal-state–A phase transition, it is tempting to assume that the roughly constant $\alpha(T)$ seen in Fig. 3 may be used to predict the gap structure of the A phase without worrying about the small temperature range available. Quite aside from any other objections, however, such an assumption neglects the effect of superconductivity on the quasiparticle-quasiparticle scattering in UPt_3. Above T_c, quasiparticle-quasiparticle interactions lead to a conductivity $\sigma(T) = 1/(\rho_0 + AT^2)$, where the elastic and inelastic contributions are the same order of magnitude at T_c. On entering the superconducting state, the quasiparticle density falls, leading to a decrease in the inelastic term, presumably within a fairly small temperature interval below T_c. The corresponding increase in the quasiparticle mean free path would then lead to an increase in α with decreasing T. Quantitative interpretations of the A- phase data assuming a specific gap must carefully consider this effect, particularly since the structure and evolution of the gap at high temperatures affect the quasiparticle density and therefore the magnitude of the inelastic scattering. We emphasize, however, that while changes in inelastic scattering due to superconductivity might affect the *size* of the observed anisotropy, this anisotropy still derives from that of the gap and thus reflects the structure of the order parameter.

Qualitatively, our data allow us to immediately conclude that more quasiparticles exist in the A phase than would be present if the B phase extended up to the same temperature. Furthermore, it appears that these extra excitations preferentially scatter sound when the polarization is in the basal plane. To see this we plot, in Fig. 4, the data of Figs. 1 and 2 normalized to the attenuation at either T_c^+ or T_c^- as a function of temperature normalized to the appropriate critical temperature. This allows us to compare, say, the B phase attenuation (for either polarization) with the attenuation in the A phase over the same reduced temperature range. It is evident that α_{ab} is much enhanced in the A phase as compared to the B phase. The data for the c-axis polarization α_{ac}, however, are roughly equal in the two phases. The latter result implies that there is either a serendipitous combination of inelastic scattering effects and quasiparticle contributions far from the gap that lead to the similarity of the A and B phase results or that the gap structures in the two cases possess the same nodal weight in the a-c plane. [According to Eq. (3), this plane contributes most strongly to the attenuation for $\epsilon||c$ and $q||a$.] The latter explanation is puzzling in the context of popular theories[3] (such as the E_{1g} scenario mentioned in the discussion of the B-phase results) that predict a line node perpendicular to the basal plane in the A phase, with the orientation of the nodal plane determined by the direction of the antiferromagnetic order parameter. The latter is known to form many small domains in the sample along the three a^* (k-space) directions. Therefore, regions of the sample would possess a line node which is not perpendicular to either $\epsilon||c$ or $q||a$. According to Eq. (3), such nodes would contribute more quasiparticles with favorable momenta than the c-axis point nodes that are postulated to exist in the B phase, leading to an enhanced attenuation in the A phase. Such speculations, of course, emphasize the need for a complete theoretical treatment.

The data of Fig. 3 contain precise information on the momentum space distributions of quasiparticles and, thus, on the gap structure. As mentioned above, such detailed information can only be extracted by comparison with complete calculations analogous to the published B phase computations taking proper account of inelastic scattering. In the absence of pertinent theoretical predictions, we conclude with some strictly qualitative observations on sample dependence. The effect of anisotropy in the quasiparticle spectrum on the attenuation critically depends on the measurement frequency and the purity of the sample. In the clean limit, defined as $ql \gg 1$, where l is the electronic mean free path and q the phonon wave vector, energy-momentum conservation implies that only quasiparticles with momenta very nearly perpendicular to q may contribute to the attenuation. Though somewhat analogous to the effect of the $p_\epsilon p_q$ term in Eq. (1), the clean limit attenuation is affected by a much smaller region of the Fermi surface. In this limit, therefore, attenuation is a very effective probe of the anisotropy of both the Fermi surface and the gap in a superconductor. In fact, attenuation measurements in this regime were one of the most effective early techniques used in studies of the anisotropic gaps found in conventional superconductors.[12] We have measured the frequency dependence of the normal state attenuation in our sample and find approximate ω^2 scaling, as predicted for the hydrodynamic regime. Deviations from perfect scaling[13] indicate, however, that ql may be as large as 0.2 at the top of our frequency range (about 300 MHz). We may therefore roughly estimate the quasiparticle mean free path in the normal state as $l \approx 300$ nm. This is in accordance with estimates of the mean free path from, e.g., de Haas–van Alphen experiments.[14] Cleaner samples and/or higher frequencies may thus make the clean regime accessible in the near future.

In conclusion, we have measured the attenuation of transverse ultrasound in the A and B phases of UPt_3. Our B-phase data are in agreement with published results.[5] The A-phase data are qualitatively different and display a significant directionally dependent enhanced density of quasiparticles as compared to the B phase. In combination with resonant impurity scattering calculations which take into account electron-electron interactions, these data should provide some of the first constraints on the symmetry of the A phase.

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