Ultrasonic Determination of Different Phases in Superconducting UPt_3

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The ultrasonic attenuation of the heavy-fermion superconductor UPt_3 is investigated as a function of temperature, magnetic field strength, and frequency. It is shown that in the presence of a dc magnetic field at least one metastable and two stable phases exist in the superconducting state.

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Until recently ^3He was known as the only existing superfluid Fermi liquid in nature where the formation of the superfluid pairs is via triplet pairing. Since the discovery of superconductivity in some of the so-called "heavy-fermion" systems,[1] however, whose low-temperature behavior may also be well described[2] by Landau's Fermi-liquid approach, there is an increasing number of experimental and theoretical findings which support speculations that some of these compounds—and in particular[3,4] UPt_3—are promising candidates for anisotropic odd-parity-pairing superconductivity. One of the characteristic features of an odd-parity-pairing superfluid (or superconducting) state is the larger number of possible spin configurations than in the singlet case which give rise to the various phases in superfluid ^3He and to a phase diagram which is profoundly influenced by a magnetic field. In ^3He ultrasonic experiments[5] have substantially contributed to the identification of the different superfluid phases and to the understanding of the physical nature of the pairing mechanism. Therefore, the idea suggests itself that ultrasonic experiments should also provide valuable information about phase transitions and the pairing mechanism in superconducting heavy-fermion compounds.

It is the main purpose of this paper to present ultrasonic attenuation experiments in the superconducting state of UPt_3 and to demonstrate for the first time that—as in superfluid ^3He—the phase diagram is strongly affected by a dc magnetic field so that, in the presence of a dc magnetic field, at least one metastable and two stable superconducting phases exist in UPt_3.

The measurements where performed on a single crystal of UPt_3 of about 7 mm length and 7 mm diameter whose c axis forms an angle of about 21° with the cylinder axis. Longitudinal sound was propagated along the c axis and generated by means of a 30-MHz overtone X-cut quartz transducer. A conventional ultrasonic pulse spectrometer was used to measure the ultrasonic attenuation coefficient α, and the ac susceptibility (at 30 Hz) of the sample was monitored simultaneously. In all experiments the dc magnetic field $\mathbf{B}_0$ was oriented along the cylinder axis of the specimen, thereby giving rise to some difficulties in the calculation of the true local field because, in the sample investigated here, no common coordinate system exists in which both the magnetic susceptibility and the "demagnetization factor" are diagonal tensors. On the other hand, this orientation guarantees that $\mathbf{B}_0$ has a component perpendicular to the ultrasonic wave vector $\mathbf{q}$. The latter is a necessary condition to make sure that zero sound may couple to the three collective modes[6] (normal flapping, clapping, and superflapping) if superconductivity in UPt_3 is via Anderson-Brinkman-Morel-type odd-parity pairing. (Note that for $\mathbf{B}_0 \parallel \mathbf{q}$ only the clapping mode will be excited.)

In a recent paper[7] we have reported for zero magnetic field and longitudinal ultrasound a λ-like attenuation peak in UPt_3 just below the superconducting transition temperature T_c. After subtraction of that ultrasonic attenuation part, which—in a two-fluid model—is associated[7] with the electrical conductivity of the nonsuperconducting electrons, and may be determined by means of transverse sound,[7] the resulting peak structure is shown in Fig. 1. We would like to mention that within experimental error, the peak position does not depend on frequency. In the inset the peak height is shown for three different ultrasonic frequencies $\omega/2\pi$, where the full line represents the frequency dependence if the peak height were to follow the theoretical curve[7-9] $\alpha=\alpha_0(\omega\tau_p)^2/[1+(\omega\tau_p)^2]$ with $\alpha_0=A/\tau_p$. Here τ_p is the relaxation time of the ultrasonically distorted subsystem which gives rise to the attenuation peak, and A is a quantity which does not depend on τ_p. Although we have shown in Ref. 7 that the *total* attenuation will increase $\propto\omega^2$, it is clear from Fig. 1 that the peak itself does not follow an ω^2 law (nor an ω law as predicted[9] for a Landau-Khalatnikov mechanism) but shows a behavior which is surprisingly well described by the above equation with $\tau_p \simeq 1.14\times10^{-9}$ s. (Note that $1/\tau_p$ is given by that angular frequency for which $\alpha/\alpha_0=\frac{1}{2}$.)

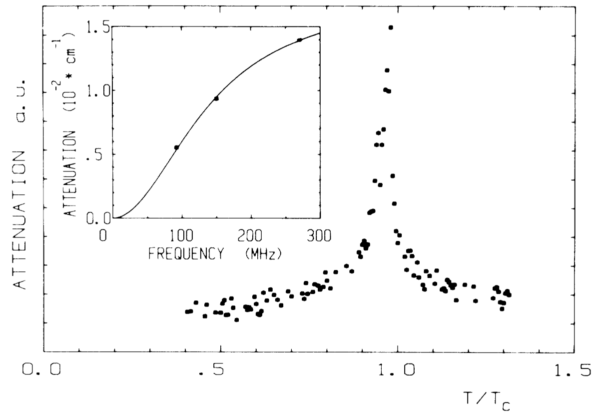

FIG. 1. Longitudinal (92 MHz) ultrasonic absorption peak after subtraction of the attenuation data $[\alpha(T)-\alpha(0.15\ \mathrm{K})]/[\alpha(0.6\ \mathrm{K})-\alpha(0.15\ \mathrm{K})]$ for transverse sound from those for longitudinal sound. The inset shows the frequency dependence of the peak height and the full line is a fit to the data (see text).

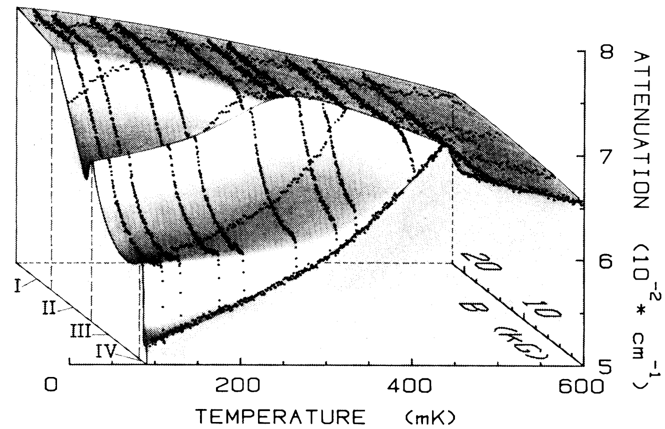

FIG. 2. Surface of ultrasonic attenuation α for a frequency of 92 MHz as a function of temperature and magnetic field strength. The shaded area at the top of the figure indicates the normal-state region.

For ultrasonic frequencies $\omega \gg 1/\tau_p$ (collisionless or zero-sound regime), i.e., $\omega/2\pi > 139$ MHz, one therefore might speculate[10] that, as in ^{3}He, excitations of order-parameter collective modes might provide a reasonable explanation for the appearance of the ultrasonic attenuation peak just below T_c. However, such an explanation is rather unlikely because in contrast to theoretical predictions[6] the peak does not vanish in the hydrodynamic regime $(\omega < 1/\tau_p)$ but even becomes more pronounced (see Ref. 7). Furthermore, a condition for the propagation of zero sound is[11] that its velocity must be greater than the Fermi velocity which seems to be unrealistic in UPt_3.[12] We therefore conclude that in the superconducting state of UPt_3 the λ-like attenuation peak is less indicative for the excitation of order-parameter collective modes than for a phase transition. As shown by the summarizing illustration in Fig. 2, and as we will discuss below, this speculation is further corroborated by the temperature and magnetic field dependence of the longitudinal ultrasonic attenuation α and ac susceptibility χ in the superconducting state.

A striking feature of Fig. 2 is that for a fixed dc magnetic field $\mathbf{B}_0$ and for $B_0 \gtrsim 0.7$ T there is a marked shift of the (temperature of the) attenuation peak relative to the superconducting transition temperature $T_c(B_0)$. For $B_0 \gtrsim 1$ T, however, and for the lowest temperatures available no attenuation peak can be detected. We note that (on the temperature scale) the field dependence of the position of the λ-like attenuation peak in UPt_3 differs essentially from that discovered in superconducting[10] UBe_{13} where *no* marked shift of the attenuation peak relative to the superconducting transition point has been reported. It is worthwhile to mention that at the superconducting transition temperature T_c of the high-temperature superconductor V_3Si, also a *pronounced peak* in the ultrasonic attenuation has been observed[13] which shows the same field dependence as the attenuation peak in UBe_{13}. Since V_3Si is believed to be a *conventional* (singlet pairing) superconductor, the attenuation peak in UBe_{13} therefore seems not to provide much evidence for nonsinglet, anisotropic superconductivity.

As a function of the magnetic field strength, the ultrasonic attenuation in UPt_3 also shows an unusual behavior. With decreasing temperature the shift of the ultrasonic peak position increases with respect to the upper critical field $B_{c2}(T)$ and the peak structure becomes more pronounced when the temperature is lowered. On the B_0 scale the peak position does not depend on frequency and the peak height shows a behavior similar to that of Fig. 1. Furthermore, we would like to emphasize that, in contrast to resistivity and susceptibility measurements, the ultrasonic attenuation (just as the specific heat) is sensitive to bulk superconductivity only. Hence, our measurements clearly reflect bulk properties. Although various theoretical models exist[4,9,14] which in part agree reasonably well with the zero-field ultrasonic attenuation data, the field dependence of the peak position has not been considered so far. Hence, it cannot be decided which model describes more adequately the physics behind the attenuation peak. Nevertheless, all models basically assume that the appearance of an attenuation peak is ultimately related to a phase transition. Furthermore, the shape of the attenuation peak is typical for a phase transition whose transition temperature (field) is characterized by the peak maximum. Thus from the peak position and the ultrasonically determined upper critical field $B_{c2}(T)$ (which agrees well with B_{c2} data[15] obtained from resistivity measurements) we may construct the phase diagram in the superconducting state of UPt_3 as presented[16] in Fig. 3. Here the roman number I refers to the normal-state phase and II, III, and IV for phases in the superconducting state.

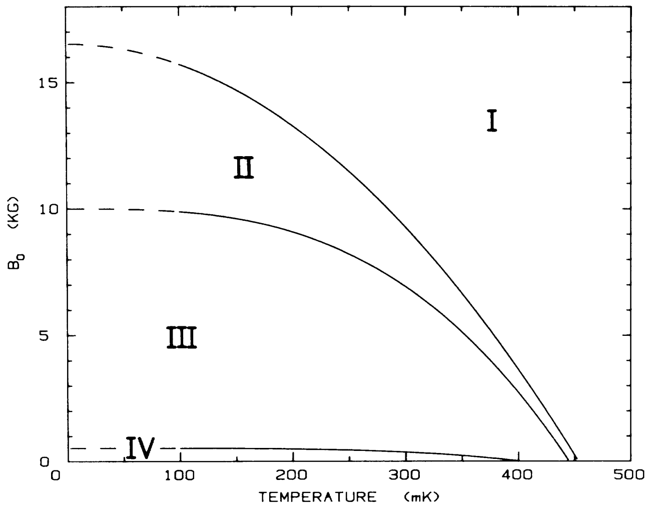

FIG. 3. Phase diagram of superconducting UPt_3 as derived from Fig. 2 by the onset of superconductivity $[B_{c2}(T)]$, the peak position, and the low-field attenuation step.

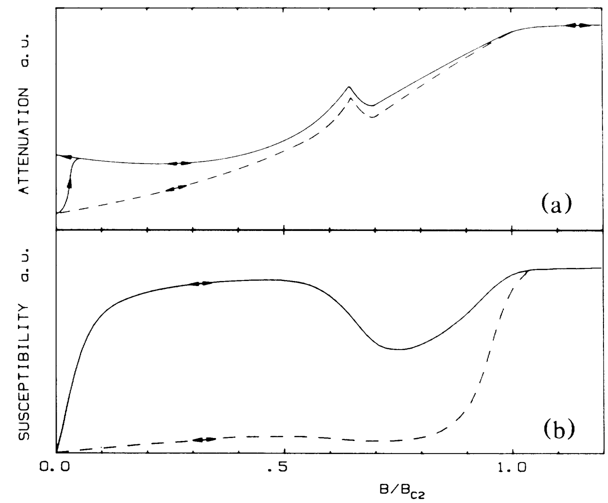

FIG. 4. (a) Ultrasonic attenuation and (b) ac susceptibility at 30 Hz in the relaxed (dashed line) and unrelaxed (full line) superconducting state at 150 mK.

Concerning the phase diagram, and in particular the "low-field phase IV," some comments are indispensable because at a fixed temperature the data of Fig. 2 were obtained by ramping of the magnetic field at a rate of 2.5 mT/s. In order to avoid flux-pinning effects, the sample was first heated above T_c and then cooled in zero field before the ramping of the magnetic field. As can be clearly seen from Fig. 4 the position and shape of the absorption peak are, within experimental error, independent of whether the measurements are performed under dynamic or steady-state conditions. Also the peak does not alter if B_0 is swept up or down. Therefore, and because the peak is also present in zero magnetic field, its origin cannot be due to flux-pinning effects and the lines between I and II and between II and III (see Fig. 3) will mark phase boundaries between stable superconducting states. For $0 < B_0 \lesssim 500$ G and for $T \lesssim 400$ mK, however, we observe for increasing field a steep increase in the ultrasonic attenuation (and ac susceptibility) which becomes less pronounced for higher temperatures and vanishes if the magnetic field is swept down. If, on the other hand, the dc magnetic field is changed in a stepwise manner and the measurements are performed under steady-state conditions, i.e., after waiting for about ten minutes at a fixed field, then the results (see Fig. 4) are reversible and neither a steplike change in the ultrasonic attenuation nor one in the ac susceptibility can be detected for $0 < B_0 \lesssim 500$ G. This "viscomagnetic" ultrasonic behavior is in some respect reminiscent of spin-glass systems and to our knowledge has not been reported for UPt_3 so far. On the other hand, hysteresis effects of the low-field magnetic susceptibility are well known[17] in superconducting UPt_3 so that flux-pinning effects cannot be ruled out as responsible for the appearance of the (low-field) *steplike change* in the ultrasonic attenuation. However, it should be noticed that such a phenomenon has not been reported as yet for any superconductor. In order to characterize this unusual state we therefore will term it the "metastable phase IV" although it does not refer to a steady state and its physical origin is not clear so far. In this context it might be interesting that in superfluid ^{3}He-*A*, fluctuations in time of the ultrasonic attenuation with a very long correlation time (20–25 s) have been observed by Lawson *et al.*[18] in the presence of an applied dc magnetic field. But in zero field no fluctuations were seen. Lawson *et al.* therefore speculated that the ultrasonic fluctuations may be related to susceptibility fluctuations. Although the same might be true for UPt_3, we actually are not in the position to decide whether or not there is any correlation between the viscomagnetic properties and long-range ferromagnetic spin fluctuations.[3] Because of the characteristic feature that the ultrasonic attenuation is sensitive to bulk properties only, one also might speculate that the relaxation phenomena indicate that superconducting currents cannot be stable in the interior of an odd-parity Anderson-Brinkman-Morel–type single-domain superconductor but only at the surface.[19] With the assumption that, as in ^{3}He-*A*, interfaces exist between two degenerate superconducting states (domains) with different orientations of the angular momentum, then, in analogy to a model proposed by Joynt, Rice, and Ueda[20] for $U_{1-x}Th_xBe_{13}$, the attenuation peak in UPt_3 would stem from the motion of domain walls and therefore would provide strong evidence for a transition into an anisotropic nonsinglet-pairing superconducting state.

Summarizing our experimental results, there is much evidence that in the superconducting state of UPt_3 at least two stable superconducting phases exist in the presence of a dc magnetic field. Furthermore, we have shown that the behavior of both the ultrasonic attenuation and ac susceptibility is by no means reminiscent of what is known for conventional superconductors. This again demonstrates that superconductivity in UPt_3 does not belong to a trivial class of superconductivity.

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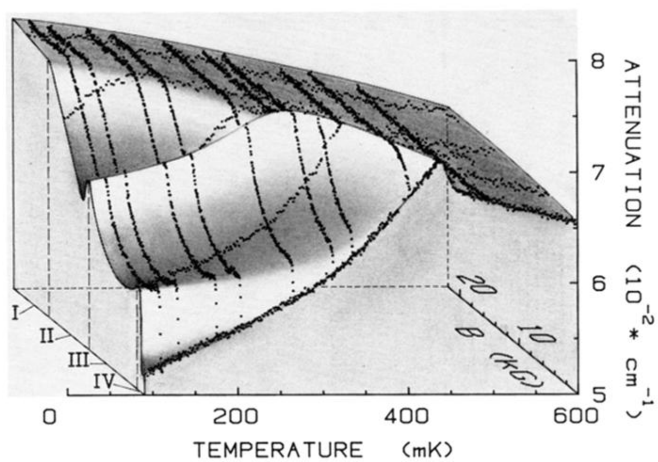

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