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Permalink
https://escholarship.org/uc/item/6fm1z07m

Journal
Physical Review Letters, 55(12)

ISSN
0031-9007

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Publication Date
1985-09-16

DOI
10.1103/physrevlett.55.1319

λ-Shaped Ultrasound-Attenuation Peak in Superconducting (U,Th)Be_{13}

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(Received 28 May 1985)

We report measurements of the ultrasound attenuation and the changes in the sound velocity of the heavy-fermion superconductor (U,Th)Be_{13}. This material has shown evidence of two transitions, a bulk superconducting transition at T_{c1} and a lower one at T_{c2}. Near T_{c2} we find a λ-shaped attenuation peak two orders of magnitude larger than the total contribution from particle-hole scattering, and a dip in the c_{11} elastic constant. These results are consistent with those expected near a magnetic transition. A state of coexisting anisotropic superconductivity and (antiferro-) magnetic order is suggested below T_{c2}.

PACS numbers: 74.30.Gn, 74.70.Rv

The observation of superconductivity in a class of cerium[1] and uranium[2-4] compounds with very large quasiparticle effective masses has aroused considerable interest. This is in large part due to the possibility, both suggested on theoretical grounds[5,6] and also indicated by experiments, that the pairing interaction is not of the conventional type and leads to an anisotropic superconducting state.[7-9] Further interest was generated when it was discovered by specific-heat measurements[10] that one of these compounds, UBe_{13}, when doped with a small amount of thorium (1%–6%) exhibits a second transition (T_{c2}) which is *below* its bulk superconducting transition (T_{c1}). It was speculated that this second transition might be analogous to the $A \rightarrow B$ transition in ^3He. This would provide further evidence that the pairing is not of an S-wave variety because for S-wave pairing there is only one order parameter.

In this paper we report measurements of the ultrasound attenuation and changes in sound velocity as a function of temperature for different frequencies and magnetic fields. At the lower transition T_{c2} we find a very large λ-shaped attenuation peak accompanied by a minimum in the sound velocity. An absorption maximum of the observed magnitude and shape is typically associated with magnetic ordering, but not with superconductivity.

The measurements were made on a large single crystal ($2\times4\times6$ mm^3) of $U_{1-x}Th_xBe_{13}$ with nominally 1.75% Th. The sample was cut and polished to optical flatness. The transducers were $LiNbO_3$, overtone mode with a 40-MHz fundamental. Longitudinal sound (50–250 MHz) was propagated parallel to a [100] axis of the crystal, and the ac susceptibility of the crystal was simultaneously monitored.

Shown in Fig. 1(a) is the ultrasonic attenuation at 136 MHz as a function of temperature as measured through T_{c1} and T_{c2}. The change in sound velocity is shown in Fig. 1(b), and the ac susceptibility in Fig. 1(c). Concerning Fig. 1 there are several points to be noted. (a) The magnitude of the attenuation peak at T_{c2} is of order 0.2 dB/μsec, (b) the attenuation peak is only associated with T_{c2}—the attenuation shows no evidence of T_{c1}, (c) the sound velocity varies most rapidly at T_{c1} and shows a pronounced minimum at T_{c2}, and (d) both the attenuation and sound velocity have a λ-type character at T_{c2}.

In the following discussion we will show that the present results (i) are fully consistent with a superconducting transition at T_{c1} and (ii) indicate a transition at T_{c2} which has the same strain-coupling characteristics as are usually found at magnetic transitions. In the superconducting state the ultrasound attentuation decreases because the electron-phonon scattering vanishes at $T \rightarrow 0$. This change equals the normal-state attenuation α_n, and it is too small to be measured in the present experiment. The magnitude of α_n in (U,Th)Be_{13} can be estimated in two different ways. One possibility is to assume the same microscopic electron-phonon coupling strength as in UPt_{13}[11] and apply the proper scaling according to the relevant parameters (resistivity, density, and sound velocity).[12] The second way is by comparison with the directly measured attenuation in pure UBe_{13}.[13] Both estimates are within $\sim 20\%$ and give a value for α_n of 10^{-3} dB/cm at 100 MHz. Thus, given a resolution of

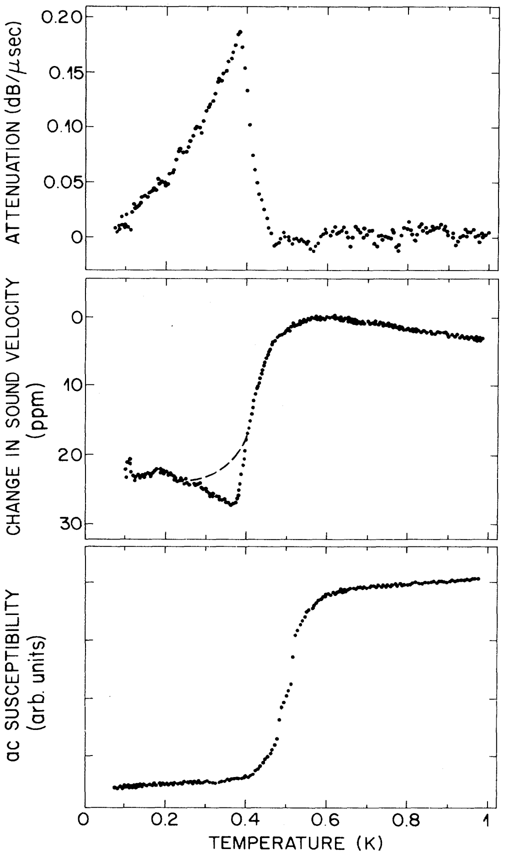

FIG. 1. Temperature dependence of (a) the ultrasound attenuation, (b) the change in sound velocity, and (c) the ac susceptibility in (U,Th)Be$_{13}$. The broken line is a symmetric continuation of the sound-velocity change at higher temperatures. This represents Δv_s associated with the (broadened) superconducting transition with the midpoint T_{c1} at ~ 425 mK. The dip in v_s and the attenuation maximum indicate the "lower" transition at $T_{c2} = 360$ mK. The nominal Th concentration is 1.75%.

$\sim 10^{-2}$ dB/cm in the present experiment, we are not able to see the reduction of attenuation in the superconducting state. An interesting point concerning the magnitude of α_n is worth mentioning here. In the heavy-fermion compounds investigated so far, the size of α_n implies that the product of the effective mass by the coupling strength has a normal value, i.e., similar to ordinary metals like In or Sn. A renormalization of the coupling strength to cancel the effect of mass enhancement in ultrasonic attenuation arises if the quasiparticle self-energy is predominantly frequency dependent.[14]

Although the superconducting transition at T_{c1} is not reflected in the attenuation data, it can be readily identified in the temperature dependence of the sound velocity v. Basic thermodynamic analysis of the superconducting phase transition links the change of elastic properties at T_c to the stress dependence of the critical field. The bulk modulus, e.g., changes typically by 10^{-5}. In UPt$_3$, the only heavy-fermion superconductor for which sufficient experimental data are available, the measured sound-velocity change at T_c is well within the expected range.[12] In Fig. 1(b) the variation of v below 1 K is shown for (U,Th)Be$_{13}$. The change at T_{c1} is not steplike, but rather S shaped, indicating a smearing of T_c by about 120 mK. The dashed line is a symmetry continuation of the curve at higher temperatures, from which the midpoint of T_c is determined to be 420–430 mK. It is not surprising that this true bulk T_c, as seen by the propagating sound, is somewhat lower than indicated by the ac susceptibility [Fig. 1(c)]. On cooling into the superconducting state, v decreases by 27 ppm, corresponding to a decrease of c_{11} by 54 ppm.

Let us now turn to the lower transition at T_{c2} where an attenuation peak about 200 times larger than the expected total ultrasonic attenuation due to scattering from conduction electrons is observed. Furthermore, the rounding of the peak is $\lesssim 10$ mK, which is about 10 times narrower than the superconducting transition at T_{c1} as seen, for instance, in the specific heat or in the change of sound velocity. These facts seem to us to rule out a superconducting transition at T_{c2}. The magnitude and the shape of the attenuation peak at T_{c2}, as well as the dip in the sound velocity, are reminiscent of the anomalies associated with magnetic ordering.[15] This statement can be made somewhat more quantitative. Near T_c magnets have an attenuation which varies as

$$\alpha_{\pm} = A_{\pm}[(T-T_c)/T_c]^{-\theta_{\pm}},$$

where + and − refer to $T > T_c$ and $T < T_c$. Although the values for these parameters vary from system to system, it is generally found that $\theta_+ = \theta_-$ and $A_+ < A_-$. An appropriate plot of the data on (U,Th)Be$_{13}$ indicates that $\theta_+ = \theta_- \cong 0.2$ and $A_+ < A_-$. Although the data are of insufficient quality that these exponents should be taken too seriously, this analysis suggests that the transition at T_{c2} is very similar in character to a magnetic transition. In the literature, the values of the exponent θ at known antiferromagnetic transitions range from ~ 0.15 in MnF$_2$ to 1.0–1.6 in the rare-earth metals.

Shown in Fig. 2 is the amplitude of the attenuation peak as a function of frequency. The data are consistent with a linear frequency dependence. The attenuation in the hydrodynamic regime, where the frequency of the sound wave is much less than the characteristic spin-fluctuation frequency, is proportional to the square of the frequency. A linear frequency dependence can arise if one is well inside the critical regime, which is unlikely in our experiment, or if be-

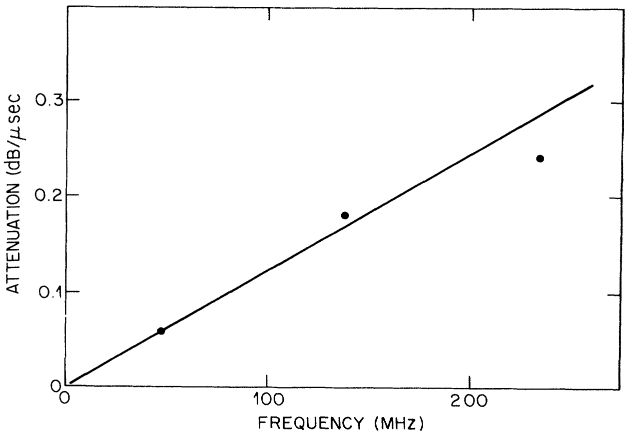

FIG. 2. Frequency dependence of the peak attenuation at $T \simeq 370$ mK.

cause of magnetic anisotropy or impurities the dominant spectral weight of fluctuations near the transition is at finite frequencies.

We have measured the amplitude of the peak as a function of magnetic field. Up to fields of 20 kG we find a small (approximately linear) depression of the peak amplitude with field. This is also consistent with our hypothesis of an antiferromagnetic transition.

Although the ultrasonic attenuation peak near T_{c2} is two orders of magnitude larger than the total contribution due to quasiparticle scattering, it is much less than that observed at the magnetic transition of, for example, the rare-earth metals where near the transition it varies on the scale of 10 dB/cm.[15] The ordered moment in $(\mathrm{U,Th})\mathrm{Be}_{13}$ must therefore be quite small. This is more directly seen from the comparison of the specific heat "jump" ΔC at T_{c2} in $(\mathrm{U,Th})\mathrm{Be}_{13}$ with those of known antiferromagnets with low T_{N}. Within mean-field theory we note that for small ordered moments $\Delta C \sim M_0$ and we get $(0.02\text{–}0.05)\mu_{\mathrm{B}}$ as a rough estimate for M_0. This explains why the observed shift in the Be resonance below T_{c2} is so small (~ 10 G).[8] No such shift is observed between T_{c1} and T_{c2} which is consistent with our hypothesis that the transitions at T_{c1} and T_{c2} are of different nature. No anomaly in the Be nuclear relaxation rate T_1^{-1} near T_{c2} has been observed. Ultrasonic attenuation couples directly to the divergent long-wavelength fluctuations while NMR integrates over all wave vectors directly. For this reason T_1^{-1} is less divergent than the ultrasonic attenuation coefficient. When we use the relationship between the ultrasonic attenuation exponent and the NMR exponent given by Kawasaki[16] and note that the former is about 0.2, no feature in T_1^{-1} is expected.

We have no good explanation for the physics of the proposed coexisting phase of antiferromagnetism and (extrapolating from UBe_{13}) anisotropic superconductivity. Some points are worth making, however. All the heavy-fermion materials appear to have a predilection to an antiferromagnetic state and some like $\mathrm{U_2Zn_{17}}$ and $\mathrm{UCd_{11}}$ achieve it in the pure state.[17,18] The heavy-fermion entropy is undoubtedly associated with spin fluctuations of what at high temperatures were local moments. Yet there is no evidence of any significant magnetic-susceptibility enhancement over the specific-heat enhancement, which suggests that most of the magnetic fluctuations are at large wave vectors. It is possible that perturbations, whose precise origin is unknown to us, push these fluctuations over the critical value to the antiferromagnetic state. With respect to the coexisting phase there are two possibilities. One is that because of metallurgical reasons Th makes a second long-range ordered phase in UBe_{13}, which has magnetic order alone. Despite a careful analysis of the lattice parameters of single crystals and arc-melted samples, we could not detect any deviation from a systematic, linear variation of a_0 as function of Th substitution. This, together with the absence of any extra diffraction line, strongly suggests phase purity in $(\mathrm{U,Th})\mathrm{Be}_{13}$. The other is that Th goes in uniformly and the uniform phase has both transitions. Within this category a relatively simple possibility is that, because of random Th defects, anisotropic superconductivity occurs in a gapless state over significant portions of the Fermi surface. Leftover portions of the Fermi surface can acquire a gap through an antiferromagnetic transition further reducing the free energy.

In conclusion we have measured the ultrasonic attenuation and sound velocity through the two transition temperatures in $(\mathrm{U,Th})\mathrm{Be}_{13}$. At the lower transition we see a large attenuation peak and a sound velocity minimum. Both the shape and the magnitude are typical for antiferromagnetic transitions. We suggest the lower transition in $(\mathrm{U,Th})\mathrm{Be}_{13}$ to be a magnetic transition within the (anisotropic) superconducting state.

We would like to acknowledge helpful discussions with E. Abrahams, E. I. Blount, and B. Lüthi. The work at Los Alamos was performed under the auspices of the U. S. Department of Energy.

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