ULTRASOUND STUDIES OF THE HEAVY FERMION SUPERCONDUCTORS
UPt$_3$, UBe$_{13}$ AND (U,Th)Be$_{13}$

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Ultrasound studies are reported on the heavy fermion superconductors UPt$_3$, UBe$_{13}$ and (U,Th)Be$_{13}$. In UPt$_3$ we find the ultrasound attenuation to vary as $T^2$ well below the transition temperature. This indicates an anisotropic, polar-like superconducting state. In pure UBe$_{13}$ a pronounced attenuation peak appears at $T_c$ which suggests absorption of phonons by collective excitations of the order parameter. At the lower transition in Th-doped UBe$_{13}$, we find a $\lambda$-shaped attenuation maximum. The shape and magnitude are consistent with magnetic order within the (anisotropic) superconducting state. The ordered moment, coexisting with superconductivity is estimated to be small ($0.02-0.05\mu_B$). The variation of the sound velocity in all these compounds can be explained by (1) the large entropy associated with the heavy fermions; (2) the basic thermodynamic relationships of the superconducting transition; and (3) the magnetic order in (U,Th)Be$_{13}$.

1. Introduction

The superconducting ground state of heavy fermions is of great interest [1-3] because mounting experimental evidence [4-9], as well as theoretical suggestions [10, 11], point to non-conventional pairing. Ultrasound experiments are a powerful tool to investigate the superconducting state and we describe here the results of our studies on UPt$_3$, UBe$_{13}$ and (U,Th)Be$_{13}$ single crystals. From the magnitude of the ultrasound attenuation in the normal state the electron-phonon coupling strength can be inferred, whereas the temperature dependence of the attenuation below $T_c$ depends on the quasiparticle density of states and possibly on collective mode excitation. The data on UPt$_3$ are indicative of an anisotropic superconducting state with the gap vanishing along lines on the fermi surface. In UBe$_{13}$, the attenuation peaks sharply at $T_c$ and is reminiscent of the attenuation in superfluid $^3$He with collective mode excitations. (U,Th)Be$_{13}$ exhibits an additional transition below $T_c$ with a very strong ultrasound absorption maximum. This lower transition is consistent with magnetic order within the superconducting state.

2. Theory

The normal state

Following the deformation potential ansatz [12], the absorption $\alpha$ of ultrasound by electrons can be expressed by a combination of microscopic and macroscopic quantities

$$\alpha_n = \frac{4(m^*E_1)^2\nu}{4\pi\rho u^2k^2} ql,$$

where the deformation potential $E_1$ and the band effective mass $m^*$ are assumed to be isotropic. The above expression is valid only for $ql \ll 1(q$ - phonon wave number, $l$ - electron mean free path). The mass density $\rho$, the sound frequency $\nu$ and velocity $u$ are known quantities, and the mean free path $l$ can be estimated from resistivity data. We note here the experimental difficulties created by the rather high resistivities.

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of heavy fermion metals ($=150 \mu \Omega \text{ cm in UBe}_{13}$, $=1 \mu \Omega \text{ cm in UPt}_{3}$) which requires very good single crystals and high ultrasound frequencies ($a \approx \nu^2$). In UPt$_3$ we verified both the $\nu^2$ and the $l(T)$ dependence of $\alpha_n$. From the measured attenuation coefficient $\alpha_n$, the product of $m^*$ and $E_1$ can be evaluated and compared with values for other metals. In the heavy fermion compounds UBe$_{13}$ and UPt$_3$ the quantity $(m^*/m_0)E_1$ (free electron mass) is of order 5 eV. This value is very similar to the ones found in other metals, and certainly not larger by two orders of magnitude as one might expect from the enhancement of $m^*$. It has to be concluded, therefore, that the enhancement of $m^*$ is compensated by a reduction of the coupling strength $E_1$. This follows if the momentum dependence of the quasi-particle self-energy is negligible compared to the energy dependence, and certain Landau parameters within a Fermi liquid description are restricted by this observation [13].

Also the sound velocity is affected by the presence of the heavy fermion state. Keeping in mind the dominance of the electronic contribution to the entropy over the lattice contribution, basic thermodynamic arguments link the variation of the sound velocity to the specific heat. In particular, one gets a contribution to the bulk modulus variation $\Delta B$ which is proportional to the integral over the specific heat $c(T)$. In UPt$_3$, the sound velocity varies as $u_0 - u_0 T^2$ in the normal state up to $\approx 1$ K, in agreement with the expectations [8].

3. Results

UPt$_3$

UPt$_3$ is the first compound that we studied successfully. The attenuation in UPt$_3$ differs in two points significantly from the classical BCS behavior. First, $\alpha_n/\alpha_n$ remains unchanged at $T_c$. Second, the variation at $T \ll T_c$ is not exponential, but like $T^2$. This $T^2$ law is observed over a rather wide temperature range extending from the lower experimental limit of $0.1 T_c$ to $\approx 0.6 T_c$.

These two salient features can be well reproduced by a calculation which assumes anisotropic superconductivity with a gap function $\Delta(k)$ vanishing along one or more lines on the Fermi surface. The density of quasi-particle states for such a polar-like state varies linearly with $E$ for $E \ll \Delta$. Best agreement between theory and experiment is obtained when the only adjustable parameter $\Delta_0(T=0)$ is set to $2.6kT_c$. A recent theoretical study of the $\Delta_0/T_c$ ratio for various types of anisotropic superconductors found for polar-like $p$-wave pairing a value of 2.46 [15]. This is additional support for our conclusion that the heavy fermions in UPt$_3$ condense in a polar-like superconducting state. A more recent calculation suggests the $T^2$-attenuation to be indicative of an axial superconducting state with $\Delta(k)$ vanishing at points on the Fermi surface. It fails, however, to reproduce the smooth variation of $\alpha$ at $T_c$ [16].

The longitudinal sound velocity changes by 13 ppm at the superconducting transition, which is in the range expected from other measurements and applying standard thermodynamics.

UBe$_{13}$

The ultrasound attenuation in UBe$_{13}$, measured from 0.5 to 2.5 GHz, is very different from all known superconductors. Instead of an abrupt decrease at $T_c$ or no change at all as in UPt$_3$, a narrow attenuation peak is observed at $T_c$. The magnitude of this peak is comparable to the electronic attenuation in the normal state, and within the accuracy given by the broadening of $T_c$, it is located at $T_c$. In an applied field of 20 kOe no
shift of the maximum with respect to $T_c$ could be found. The origin of this excess attenuation is not clear at this point. We suggest as a possibility the coupling of sound to collective modes of the order parameter, in analogy to similar effects in superfluid $^3$He. It seems highly unlikely that the absorption peak is caused by ultrasound-induced pair breaking, since the phonon energy is only a few percent of the gap parameter. In conventional s-wave superconductors, an appreciable enhancement of $\alpha_s/\alpha_0$ below $T_c$ is only expected for phonon energies exceeding the gap energy, but this condition cannot be met in practice. The change in sound velocity at $T_c$ amounts to $\approx 27$ ppm.

$$\text{(U,Th)Be}_{13}$$

Partial substitution of U by Th not only depresses $T_c$, but induces also a second transition below $T_c$, which shows up in the specific heat experiments as a maximum of similar magnitude as the anomaly at $T_c$ [17]. The value of the second transition is unknown so far and the ultrasound experiments should provide useful information. At the superconducting transition ($T_{c1} = 420$–430 mK) the sound velocity decreases by $\approx 27$ ppm. (The ac susceptibility would indicate a slightly higher $T_{c1}$.) Since these experiments were done in the 50–250 MHz range, and because the mean free path is as short as in UBe$_{13}$ ($ql = 10^{-4}$) the electronic contribution to the attenuation is only of order $10^{-3}$ dB/cm. No reduction of $\alpha$ is therefore seen at $T_{c1}$. On further cooling, an attenuation peak is observed with its maximum at 377 mK. This additional attenuation is by two orders of magnitude larger than the electronic contribution. The shape can be described by $\alpha = A_+ |t|^{-\theta}$, where $t = (T - T_2)/T_c$. The exponent $\theta$ is close to 0.2 and $A_+(T > T_2)$ is smaller than $A_-(T < T_c)$. Attenuation features like this are usually associated with magnetic transitions [18]. We also find at $T_{c2}$ a small dip of 5–8 ppm in the sound velocity, which is of the right magnitude in relation to the attenuation. The ordered moment, as it can be estimated from the ultrasound attenuation or from the specific heat anomaly at $T_{c2}$, will be rather small, of order $(0.02–0.05)\mu_B$. Nuclear magnetic resonance experiments [6] found only a minute (≈10 Oe) variation in the Be spectra at a temperature considerably below $T_{c2}$, which appears consistent with a very small ordered moment. The absence of an anomaly in the nuclear relaxation rate $T_1^{-1}$ at $T_{c2}$ is also compatible with our suggestion of a magnetic order at $T_{c2}$. For phase-space reasons the divergence of $T_1^{-1}$ is much weaker than the one of the ultrasound attenuation. Given an exponent of only 0.2 for the latter, no anomaly in $T_1^{-1}$ is expected.

4. Conclusion

Ultrasound experiments have provided unique insight into the heavy fermion state and its ordered ground states. Clear evidence for the unconventional pairing is found in the $T^2$ power law of the attenuation in UPt$_3$ and the attenuation peak at $T_c$ in UBe$_{13}$. The proposed coexistence of superconductivity and magnetism in (U,Th)Be$_{13}$ points to the possibility of complicated order parameters in heavy fermion systems.

References

[1] F. Steglich, J. Aarts C.D. Bredl, W. Lieke, D. Meschede, W. Franz and H. Schäfer, Phys. Rev. Lett. 43 (1979) 1892.
[2] H.R. Ott, H. Rudigier, Z. Fisk and J.L. Smith, Phys. Rev. Lett. 50 (1983) 1595.
[3] G.R. Stewart, Z. Fisk, J.O. Willis and J.L. Smith, Phys. Rev. Lett. 52 (1984) 679.
[4] H.R. Ott, H. Rudigier, T.M. Rice, K. Ueda, Z. Fisk and J.L. Smith, Phys. Rev. Lett. 52 (1984) 1915.
[5] D.J. Bishop, C.M. Varma, B. Batlogg, E. Bucher, Z. Fisk and J.L. Smith, Phys. Rev. Lett. 53 (1984) 1009.
[6] D.E. MacLaughlin, Cheng Tien, W.G. Clark, M.D. Lan, Z. Fisk, J.L. Smith and H.R. Ott, Phys. Rev. Lett. 53 (1984) 1833.
[7] J.J.M. Franse, A. Menovsky, A. de Visser, C.D. Bredl, U. Gottwick, W. Lieke, H.M. Mayer, U. Rauchschwalbe, G. Sparn and F. Steglich, Z. Phys. B: Cond. Matter 59 (1985) 15.
[8] B. Batlogg, D.J. Bishop, E. Bucher, C.M. Varma, Z. Fisk and J.L. Smith, J. Appl. Phys. 57 (1985) 3060.
[9] D. Jaccard, J. Flouquet, Z. Fisk, J.L. Smith and H.R. Ott, to be published.
[10] C.M. Varma, in: Proceedings of the NATO Advanced Summer Institute on the Formation of Local Moments in
Metals, W. Buyers, ed. (Plenum, New York, 1984) p. 83; Bull. Am. Phys. Soc. 29 (1984) 404.

[11] P.W. Anderson, Phys. Rev. B30 (1984) 1549.
[12] See e.g. A.B. Pippard, Phil. Mag. 46 (1955) 1104.
R.W. Morse, Progress in Cryogenics, vol. 1, K. Mendelsohn, ed. (Academic Press, New York, 1959) p. 219.
M.S. Steinberg, Phys. Rev. 111 (1958) 425.
[13] C.M. Varma, J. Appl. Phys. 57 (1985) 3064.

[14] Similar observations are also reported by B. Lüthi, Proc. ICCF5 (Sendai, 1985), J. Magn. Magn. Mat. 52 (1985) 70.
[15] M. Tachiki, M. Nakahara and R. Teshima, preprint.
[16] J.P. Rodriguez, Phys. Rev. Lett. 55 (1985) 250.
[17] H.R. Ott, H. Rudigier, Z. Fisk and J.L. Smith, Phys. Rev. B31 (1985) 1651.
[18] See e.g. a review by B. Lüthi, T.J. Moran and R.J. Pollina, J. Phys. Chem. Solids 31 (1970) 1741.