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ULTRASOUND STUDIES OF THE HEAVY FERMION SUPERCONDUCTORS UPt₃, UBe₁₃ AND (U,Th)Be₁₃

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Ultrasound studies are reported on the heavy fermion superconductors UPt₃, UBe₁₃ and (U,Th)Be₁₃. In UPt₃ 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 UBe₁₃ 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 UBe₁₃, we find a λ -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 ($(0.02\text{--}0.05)\mu_B$). 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)Be₁₃.

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 UPt₃, UBe₁₃ and (U,Th)Be₁₃ 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 UPt₃ are indicative of an anisotropic superconducting state with the gap vanishing along lines on the fermi surface. In UBe₁₃, the attenuation peaks sharply at T_c and is reminiscent of the attenuation in superfluid ³He with collective mode excitations. (U,Th)Be₁₃ 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 α of ultrasound by electrons can be expressed by a combination of microscopic and macroscopic quantities

$$\alpha_n = \frac{4(m^*E_1)^2\nu}{5\pi\rho u^2\hbar^2} ql,$$

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 ρ , the sound frequency ν 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 ($\approx 150 \mu\Omega \text{ cm}$ in UPt_3 , $\approx 1 \mu\Omega \text{ cm}$ in UPt_3) which requires very good single crystals and high ultrasound frequencies ($\alpha \approx \nu^2$). In UPt_3 we verified both the ν^2 and the $l(T)$ dependence of α_n . From the measured attenuation coefficient α_n , the product of m^* and E_1 can be evaluated and compared with values for other metals. In the heavy fermion compounds UPt_3 and UPt_3 the quantity $(m^*/m_0)E_1$ (m_0 – 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 ΔB which is proportional to the integral over the specific heat $c(T)$. In UPt_3 , the sound velocity varies as $u_0 - u_1 T^2$ in the normal state up to $\approx 1 \text{ K}$, in agreement with the expectations [8]. We find the value of u_1 to be larger for sound propagating in the basal plane than it is for propagation along the hexagonal axis [14].

The superconducting state

The ultrasonic attenuation by electrons decreases rapidly when a metal is cooled below the superconducting transition temperature T_c . In BCS theory the opening of the gap, the redistribution of density of states and the coherence factors lead to an abrupt decrease of α_s/α_n at T_c and an exponentially vanishing α_s/α_n for $T \rightarrow 0$. The results on heavy fermion superconductors are in sharp contrast to BCS theory and will be discussed now.

3. Results

UPt₃

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, α_s/α_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 $0.1T_c$ to $\approx 0.6T_c$.

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 $\Delta(T=0)$ is set to $2.6kT_c$. A recent theoretical study of the Δ_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 α 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.

UPt₃

The ultrasound attenuation in UPt_3 , 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 ^3He . 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 α_s/α_n 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 ≈ 27 ppm.

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

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 ($T_{c1} = 420\text{--}430\text{ mK}$) the sound velocity decreases by ≈ 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} ($ql \approx 10^{-4}$) the electronic contribution to the attenuation is only of order 10^{-3} dB/cm. No reduction of α 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 $\alpha = A_{\pm}|t|^{-\theta}$, where $t = (T - T_c)/T_c$. The exponent θ is close to 0.2 and $A_+(T > T_c)$ 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 $(0.02\text{--}0.05)\mu_B$. 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 $(\text{U,Th})\text{Be}_{13}$ points to the possibility of complicated order parameters in heavy fermion systems.

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