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Electron-phonon coupling in UBe_{13}: Absence of conventional superconductivity

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Calculation of the electron-phonon interaction strength λ_{ep} using the conventional theory for metals, which should be applicable in the normal regime, leads to the very small value of 0.035 for the heavy-fermion superconductor UBe_{13}. It is argued that the interactions responsible for the large observed mass enhancement in the heavy-fermion regime will tend to further decrease λ_{ep}, strengthening the conventional wisdom that either unconventional electron-phonon coupling or electronic interactions are responsible for pairing.

The intense interest in the heavy-fermion compounds (HFC) is due in large part to the unexpected discovery of superconductivity in the range 0.4–0.9 K in UBe_{13} (Ref. 1), UPt_3 (Ref. 2), and $CeCu_2Si_2$ (Ref. 3). With the possible exclusion of the class of HFC's to be discussed here, all known superconductivity is understood in terms of Cooper pairing of quasiparticles mediated by phonons. Moreover, in a wide variety of superconducting systems it is reasonable to model the scattering of electrons by the phonons in terms of an effective potential which moves rigidly with the vibrating atoms as well as determines the band structure of the static lattice. *We refer to this description as the conventional mechanism of superconductivity. (Unconventional mechanisms* could include unusual electron-phonon coupling involving highly correlated electrons as well as purely electronic interactions.) For conventional superconductivity the critical temperature T_c is determined primarily by λ_{ep}, the electron-phonon interaction strength. Because of the apparent strong Coulomb interactions in HFC's which are highly detrimental to conventional superconductivity, several unconventional mechanisms have been proposed to account for the observed superconductivity. Such models are largely untested to date, however, and it has not been demonstrated convincingly that conventional superconductivity is ruled out in HFC's. Indeed Oguchi and Freeman[4] have argued that superconductivity in UPt_3 is conventional. Here we report calculations of the electron-phonon interaction strength for UBe_{13} which indicate that superconductivity in UBe_{13} *cannot be due to the conventional mechanism,* as defined above.

The unusual low-temperature properties of HFC's, including extremely large heat capacities and susceptibilities, anomalous temperature-dependent resistivities, and large negative magnetoresistances, have been reviewed by Stewart.[5] The very large densities of states apparent in the thermodynamic properties are usually discussed in terms of highly enhanced effective masses $m^*/m \sim 10^2$ arising from strong Coulomb interactions between electrons. Within this picture λ_{ep} (which is of the order of unity in conventional superconductors) is considered to be negligible. Alternative phonon mechanisms to the conventional phonon-mediated interactions include the Kondo volume collapse mechanism[6–8] and the exchange-enhanced electron-phonon interaction.[9] The Kondo-like peaks in the normal-state heat capacity and susceptibility and the large negative magnetoresistance have led to suggestions[2,10–14] that supercondictivity in HFC's may arise from exchange of spin fluctuations.

The existence of a periodic lattice in HFC's leads to a coherent ground state with Bloch-like quasiparticle excitations. At present the only way of taking full account of crystal-structure effects is through band-structure calculations which, as is well known, replace the two-body Coulomb interaction between electrons with a judiciously determined mean field. The resulting theory describes several ground-state properties exactly (in principle), and in "ordinary" (non-f-electron) metals the theory provides an excellent zeroth order description of single-particle excitations. Even in f band metals, the theory has had notable success describing trends in ground-state properties[15] and Fermi surfaces.[16] The single particles described by band theory also provide the standard for estimating many-body enhancements of susceptibility and specific heat.

We have carried out linearized augmented-plane-wave (LAPW) band-structure calculations[17] on UBe_{13} in its measured crystal structure, with O_h^6 space group. The calculations utilize a general potential (i.e., no shape constraints) and the spin-orbit interaction is included[18] throughout. Referred to the uranium sphere radius R_U the augmented-plane-wave basis set is truncated at $R_U K_{max} = 8.5$, corresponding to ~ 460 LAPW's. As has been found in previously reported calculations[4,19] on the other U-based HFC UPt_3, the Fermi level E_F falls in a region of hybridized U $f_{5/2}$ states [Fig. 1(b)] and the band structure near E_F is very anisotropic.

The Fermi level is found to lie in a narrow region of relatively low density of states within the U $f_{5/2}$ bands, as

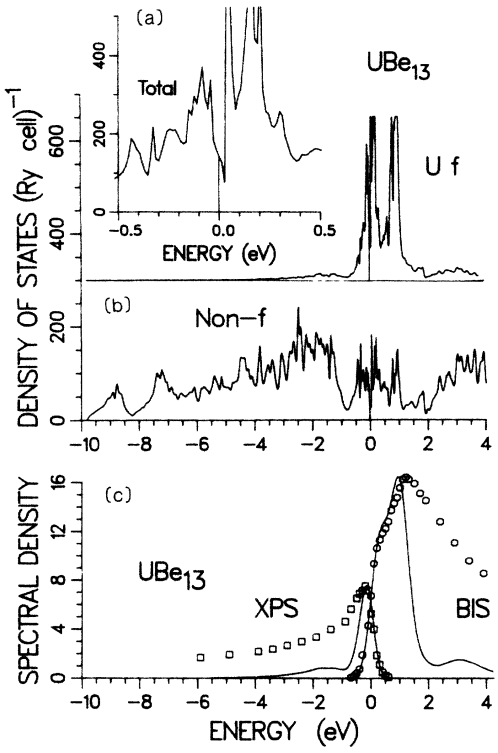

FIG. 1. (a) Total DOS of UBe_{13} near E_F. Note that $E_F=0$ falls in a pronounced minimum. (b) Uranium f (upper) and non-f (=total$-f$, lower) DOS in the range -10 to 4 eV. (c) Theoretical occupied and unoccupied spectral densities, broadened by instrumental resolution (lines), compared with XPS and BIS data (points) from Wuilloud *et al.* (Ref. 32).

shown in Fig. 1(a). As a result the density of states $N(E_F)=71$ states/Ry U atom is not as sensitive to zone sampling[20] or the precise position of E_F as in related HFC's. Using the extrapolated value of the linear specific-heat coefficient $\gamma=1.1$ J/moleU K^2, the mass enhancement $m^*/m_{\text{band}}\approx 90$ for UBe_{13}. This enhancement over the calculated band mass is the largest yet reported.

The Fermi-surface averaged electron-ion matrix elements were evaluated using the rigid muffin-tin expression of Gaspari and Györffy.[21] Quantities entering the calculation of the McMillan-Hopfield constant $\hat{\eta}$ are presented in Table I. (For compounds the quantity which characterizes the contribution to λ from a given *type* of atoms is $\hat{\eta}=L\eta$, where L is the number of atoms of that type in the unit cell.) In UBe_{13}, $\hat{\eta}_U=0.052$ eV/Å^2 is extremely small even for an f-electron atom,[22] and $\hat{\eta}_{Be}=0.118$ eV/Å^2. Although only total p, d, and f partial state densities are listed in Table I, the calculation of $\hat{\eta}_U$ includes spin-orbit corrections.[22] The small values of $\hat{\eta}_U$ and $\hat{\eta}_{Be}$ result because the U f states dominate $N(E_F)$ (making $\hat{\eta}_{Be}$ small), but they couple very poorly to the plane-wave-like conduction states, resulting in very small electron-ion coupling $\langle I^2\rangle_U=\hat{\eta}_U/N(E_F)$.

We use the approximation

$$\lambda=\frac{\hat{\eta}_U}{M_U\langle\omega^2\rangle_U}+\frac{\hat{\eta}_{Be(I)}+\hat{\eta}_{Be(II)}}{M_{Be}\langle\omega^2\rangle_{Be}}=\lambda_U+\lambda_{Be}\,, \tag{1}$$

TABLE I. Quantities entering the calculation of λ_{ep}: sphere radius R_{MT} (a.u.), Fermi energy (Ry), total $[N(E_F)]$, and partial (n_l) state densities (states/Ry spin), and the electronic stiffness $\eta_{l,l+1}$, $\hat{\eta}$, and lattice stiffness $M\langle\omega^2\rangle$ (eV/Å^2).

	UBe_{13}		
	U	Be_I	Be_{II}
R_{MT}	3.5719	1.9025	1.9025
E_F	0.762	0.762	0.762
$N(E_F)$	71.0	71.0	71.0
n_s	0.029	0.035	0.029
n_p	0.163	0.384	0.205
n_d	1.020	0.012	0.027
n_f	27.04	0.001	0.002
η_{sp}	0.0004	0.0014	0.0007
η_{pd}	0.013	0.0034	0.0038
η_{df}	0.013	0.0000	0.0000
$\hat{\eta}$	0.053	0.010	0.108
$M\langle\omega^2\rangle$	4.76	5.01	5.01
λ_{atom}	0.011	0.002	0.022
λ_{ep}	. . .	0.035	. . .

which has been highly successful in other compounds[23] whose atoms have very different masses. The data of Renker *et al.*[24] can be used to provide experimental values of the mean vibrational frequencies $\langle\omega^2\rangle_{U,Be}$ of U and Be atoms. For U, we have estimated $\langle\omega^2\rangle_U=\Omega_U^2/2$, where $\Omega_U=13$ MeV$=150$ K corresponds to the experimental (Einstein-like) peak due to the U local modes. The factor of $\frac{1}{2}$ is more appropriate for a Debye-like spectrum and in the present case will tend to give an *overestimate* of λ_U, which we find to be 0.011. For Be we use $\langle\omega^2\rangle_{Be}^{1/2}=50$ meV$=560$ K which is well below the midpoint of the Be vibrations[23] and again tends to overestimate λ_{Be}, which by our calculation is 0.024. The total electron-phonon interaction strength is $\lambda_{ep}=0.035$, which is much smaller than any reasonable effective Coulomb pseudopotential μ^* ($\mu^*\geq 0.13$ in conventional superconductors but is probably much larger in UBe_{13}). The transition temperature, normally given by[25]

$$T_c=(\omega_{\log}/1.2)\exp[-1/(\lambda-\mu^*)]\,, \tag{2}$$

where $\omega_{\log}$ is logarithmic mean frequency, vanishes for $\lambda=\lambda_{ep}<\mu^*$ because the effective interaction is repulsive. The clear conclusion is that the conventional electron-phonon interaction (EPI) cannot be responsible for superconductivity in UBe_{13}, and this conclusion is very *insensitive to any of the approximations* in the calculations. Small values of λ_{ep} have also been calculated for $CeCu_2Si_2$ (Ref. 26) and UPt_3 (Ref. 27). The present results are consistent with the very small ultrasonic attenuation[28] reported for UBe_{13}.

In light of these calculations which rule out conventional superconductivity in UBe_{13}, it is appropriate to discuss the applicability of Eliashberg theory, which provides the theortical foundation for conventional superconductivity, to heavy-fermion compounds. The most obvious obstacle is the existence of an electronic energy scale, the coherence temperature $T_0\sim 2-5$ K, which is (much) less than the

maximum phonon energy $\omega_0 \approx 900$ K. The result is the formal invalidity of Migdal's theorem[29] and therefore the Eliashberg theory. It may be noted that the same objection holds for the lattice dynamics; the adiabatic approximation is not formally valid in HFC's. Yet the phonon spectrum in UBe_{13} is indistinguishable[24] from that of the normal compound $ThBe_{13}$, and also is unaffected[24] by the onset of the heavy-fermion behavior below 5 K. Therefore it appears that the phonon spectrum may nonetheless be described by the standard adiabatic theory in spite of the lack of validity of the usual derivation. This apparently results because the onset of coherence results in only a very small rearrangement of electron spectral density.

This observation suggests the possibility that the Eliashberg theory may provide a reasonable guideline for the effect of the conventional electron-phonon interactions, if renormalization effects are taken into account. Rigorous Eliashberg theory[29] is couched in terms of Coulomb *quasiparticles* interacting with phonons. Therefore, the conventional coupling constant $\lambda = z\lambda_{ep}$, which determines T_c via Eq. (2), is λ_{ep} renormalized by the quasiparticle amplitude z. In conventional superconductors the spectral weight $1-z$ outside the quasiparticle peak is presumed to be small[30] (on the order of 0.1) and z is usually replaced by unity. In HFC's it is frequently suggested that $m^*/m_{\rm band}$ is dominated by nonphonon contributions, making $z \approx m_{\rm band}/m^* \ll 1$. Since the quasiparticle picture is valid only for excitations with $\omega < T_0$ it is inappropriate to renormalize the electron-phonon interaction which extends over an energy range ω_0 by a renormalization which is effective only for $\omega < T_0 \approx 10^{-2}\omega_0$. For energies greater than T_0 the renormalization vanishes and the remaining fraction of the spectral weight $1 - m_{\rm band}/m^* \approx 0.9 - 0.99$ may be described reasonably by the band density of states. An effective renormalization averaged over $E_F \pm \omega_0$ then is likely to be of order $1 + (T_0/\omega_0)(m^*/m_{\rm band}) \sim 2$ rather than of order $(m^*/m_{\rm band})$. Considering the very small value of λ_{ep} in UBe_{13}, the renormalization factor (giving further reduction) is unimportant in any case. We conclude that the conventional EPI is far too small to account for superconductivity in UBe_{13}. Because of the lack of understanding of the important electron-electron interactions in HFC's, the rigorous calculation of T_c remains an unsolved problem.

The large mass enhancement $z^{-1} \approx 90$ reflects strong interactions involving f electrons which are not described by the local-density approximation. The value of the band calculation is its accurate account of the dynamics of the non-f electrons and its correct description of crystal structure effects and spin-orbit coupling. It also includes conduction-f hybridization and the full fourteen-fold degeneracy of the f states. Martin and Allen[31] have emphasized that periodic f-electron systems must satisfy the Luttinger sum rule—the Fermi surface must enclose a number of states identically equal to the number of electrons in the system. This feature is satisfied by the band picture by construction. A feature less widely recognized is that, where the Landau quasiparticle (qp) picture holds $(\omega, T < T_0)$ the spectral density S satisfies

$$S_{\rm qp}(k,\omega) = -\frac{1}{\pi}\,{\rm Im}\left[\frac{z}{\omega - zE_k + i\delta}\right] = S_{\rm band}(k, z^{-1}\omega)\ . \tag{3}$$

As long as $S_{\rm band}$ is structureless within $\pm z^{-1}T_0$ of E_F, the renormalization gives little net rearrangement of spectral density. Unfortunately, instrumental resolution makes spectral-density measurements on such a fine scale difficult to realize. However, the renormalization is strong only at $\omega < T_0$ and decreases rapidly at higher energies, so the band spectral density may also closely approximate the experimental spectral density away from E_F.

In Fig. 1(c) we compare the experimental x-ray photoemission (XPS) and bremsstrahlung-isochromat-spectrum (BIS) data of Wuilloud *et al.*[32] to the U f density of states (DOS) (zone-averaged spectral density) broadened by the experimental resolution. The data have been scaled to the DOS curve so only the positions of structure and the overall bandwidth are meaningful to compare. The shoulder and peak in the BIS data at 0.4 and 1.2 eV are reproduced by the unoccupied DOS curve, although the peak in the latter occurs around 0.9 eV. Both the XPS data and the occupied DOS curve peak at -0.3 eV, reflecting the broadening of the Fermi edge. Both XPS and BIS spectra are, however, roughly twice as broad as the calculated f bands. Including intrinsic broadening would increase the theoretical widths, but some of the difference may be due to high-frequency correlations involving charge fluctuations, reminiscent of corresponding features in Ce compounds,[33] but much weaker.

To summarize, detailed calculations find the conventional electron-phonon interaction strength to be very small, at least an order of magnitude too small to account for superconductivity at 0.85 K. This result gives substantial support to the notion that some as yet undetermined unconventional mechanism is responsible for superconductivity in UBe_{13}.

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