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THE PRIMARY ROLE OF IONIC POLARIZABILITY IN EXOTIC SUPERCONDUCTIVITY

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The ionic polarizability of organic metals and of high- T_c cuprates is very high while the density of conduction electrons is low. As a result, the ionic polarizability screens out the electronic polarizability for states very close to the Fermi surface. The cut-off is determined by the dispersion of the ionic polarizability. The electron-phonon matrix element is very large for the ionically screened states, and is the cut-off for the electronically screened states. Therefore, the McMillan coupling constant λ_{ph} is very large at very low energies, and has a low energy cut-off. This model accounts for both normal and superconducting state anomalies in organic metals, as well as cuprates.

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1. Introduction

High superconducting transition temperatures can, in principle, be obtained with large coupling constants; in the conventional BCS-Gorkov-Nambu-Eliashberg theory [1], the coupling constant is the McMillan parameter λ_{ph} given by $I^2 n(E_F)/M\omega_{ph}^2$.

When the 1-dimensional organic metals were discovered in the early 1970's, we pointed out that a large λ_{ph} in these compounds will induce a Peierls transition to an insulating CDW state, and therefore λ_{ph} has to be reduced to observe superconductivity [2]. An interchain coupling helps to overcome this instability somewhat, but even if the CDW with a wave vector $Q = (2k_F, 0, 0)$ is suppressed, nesting with a wave vector $Q = (2k_F, \pi/a, 0)$ can still be effective [2].

Thus, in the quest to obtain organic superconductors with a high transition temperature, we face two challenges: First, to obtain a large λ_{ph} and second, to avoid instabilities.

Since λ_{ph} depends on 4 parameters, we must consider each of them. Decreasing ω_{ph} increases λ_{ph} , but does not increase T_c *per se* (for a strong coupling); $T_c = 0.18\omega_{ph}\sqrt{\lambda_{ph}/(1+2.6\mu^*)}$, where $\mu^* = \mu/[1 + \mu \ln(E_F/\omega_{ph})]$ [3]. Thus

$$T_c = 0.18I\sqrt{n(E_F)/M(1+2.6\mu^*)} \quad (1)$$

and ω_{ph} drops out, except for its effect on μ^* .

Thus, decreasing ω_{ph} does not help and only causes instabilities, and to overcome them a *high* ω_{ph} is necessary [2]. However, a small M is favorable for a high T_c , which is the situation in the cuprates, where the small oxygen mass may be responsible for the high T_c . In the organics, M is small for internal phonon modes, such as the C–C stretching, and these may perhaps account for the high T_c of the fullerenes [4], but in the charge-transfer complexes, the very strong pressure dependence of T_c [5] seems to indicate that this is not the case.

Since the BCS theory was formulated in the 1950's, it is commonly known that a large $n(E_F)$ is favorable for a high T_c . Specifically, in some A-15's the exponential BCS behavior was reported [6], and the very large $n(E_F)$ was found to be due to Van-Hove singularities, which are particularly strong because of the quasi-1D nature of the band structure [7]. However, the 30 year long quest to increase T_c by increasing $n(E_F)$ seems to have been saturated at $T_c = 23$ K of Nb_3Ge [8].

This leaves the electron–phonon matrix element I . Already in 1964, Marvin Cohen suggested that I is particularly large in SrTiO_3 , because of the absence of electronic screening, and as a result T_c should be abnormally high. This prediction was substantiated experimentally [9]. This work was followed by the IBM Zürich group. Bednorz and Muller found that I is particularly large in the perovskites. This was found empirically, from the large Jahn–Teller effect of the copper ion, and also on theoretical grounds it was claimed that the reduced electronic screening is the cause of the large matrix element [10].

The theory of Marvin Cohen applies to semiconductors; in the present work, and some other publications, we attempt to extend this theory to metals.

2. The Bardeen scenario

In 1937, Bardeen found that the electron–phonon matrix element I is reduced from the rigid-ion value I_{RI} (employed previously by Bloch) by screening the conduction electrons [11]. Bardeen's result can be expressed approximately as:

$$I \approx \frac{I_{\text{RI}}}{1 + (\kappa_{\text{TF}}/k_{\text{F}})^2}, \quad (2)$$

where κ_{TF} is the Thomas–Fermi screening parameter, given by

$$\kappa_{\text{TF}}^2 = \frac{4\pi e^2}{\varepsilon_{\text{background}}} n(E_{\text{F}}). \quad (3)$$

In simple metals, $\kappa_{\text{TF}}^2 = 0.66r_s k_{\text{F}}^2$ [12]. In 3D, r_s is given by $r_s = 1.92/(k_{\text{F}}a_0)$, where a_0 is the Bohr radius, $a_0 = \hbar^2 \varepsilon_{\text{background}}/m^* e^2$. Thus, this simple expression for κ_{TF} takes into account the ionic polarization, which is incorporated in the background of the dielectric constant. In typical metals, $r_s \approx 3$, thus I is reduced by about a factor of 3, and λ_{ph} by about a factor of 10, from the rigid-ion value.

Thus, if the screening is reduced, λ_{ph} will increase significantly. A low $n(E_{\text{F}})$ is favorable for this scenario, since $I^2 \propto [n(E_{\text{F}})]^{-2}$, thus $I^2 n(E_{\text{F}}) \propto 1/n(E_{\text{F}})$. Moreover, a large ionic polarizability is particularly favorable for a large λ_{ph} , since it decreases κ_{TF} , and consequently increases I , without decreasing $n(E_{\text{F}})$. We call this recipe for increasing λ_{ph} the “Bardeen scenario”.

Uemura found empirically [13] that $T_c \propto \lambda_L^{-2}$ for exotic superconductors. λ_L is the London penetration depth, and $\lambda_L^{-2} = (4\pi ne^2/\hbar k_F c^2)v_F$, $v_F \approx (n/\hbar k_F) \times [n(E_F)]^{-1}$. Thus, T_c is *inversely* proportional to $n(E_F)$, in line with the Bardeen scenario, and contrary to the “common knowledge”.

3. A self-consistent treatment of the ionic and electronic polarizabilities

In the well-known Bardeen–Pines formalism, the ionic polarizability is decreased by the dielectric function of the conduction electrons: $\epsilon_{ion}^{dressed} - 1 = (\epsilon_{ion}^{bare} - 1)/\epsilon_{el}$ [14]. This formalism applies to metals, where: $\epsilon_{el} \gg \epsilon_{ion}$. Consequently, the backwards-effect of the ionic polarizability on the electronic one can be ignored. In organic metals and cuprates, we deal with a situation where $\epsilon_{ion}^{bare} \approx 20$ (organics [15]) to 50 (cuprates, [16]). The bare electronic dielectric function is given by: $\epsilon_{el}^{bare} = 1 + (\kappa_{TF}^{bare})^2/q^2$, where $(\kappa_{TF}^{bare})^2 = 0.66r_s k_F^2$, with $a_0 = \hbar^2 \epsilon_\infty / m^* e^2$. For near-neighbors at a distance $\approx a$, $q \approx a^{-1}$; if $k_F \approx \pi/a$, $\epsilon_{el}^{bare} \approx 1 + 0.66r_s \pi^2 \approx 20$. For “simple” metals, ϵ_{ion}^{bare} is much smaller, and is usually less than 5 [17]. Thus for “simple” metals, $\epsilon_{el}^{bare} \gg \epsilon_{ion}^{bare}$. But for “exotic” metals, $\epsilon_{el}^{bare} < \epsilon_{ion}^{bare}$, and therefore the “metallic” approximation that ignores the screening of the electronic polarizability by the ionic one, no longer applies, and we must treat the two screening processes on equal footing:

$$\begin{aligned} \epsilon_{ion}^{dressed} - 1 &= (\epsilon_{ion}^{bare} - 1)/\epsilon_{el}^{dressed}, \\ \epsilon_{el}^{dressed} - 1 &= (\epsilon_{el}^{bare} - 1)/\epsilon_{ion}^{dressed}. \end{aligned} \tag{4}$$

We describe ϵ_{ion}^{bare} by the Lyddane–Sachs–Teller theory [17]

$$\epsilon_{ion}^{bare}(\omega) = \epsilon_\infty + (\epsilon_0 - \epsilon_\infty)\omega_0^2/(\omega_0^2 - \omega^2 + i\omega\gamma),$$

where γ is a damping term. (We apply here a conventional 3D theory. We do not believe that the effects that we consider *require* low-dimensional systems. In low-dimensional systems, the *details* of our theory will, however, have to be modified.)

Substituting ϵ_{el}^{bare} , ϵ_{ion}^{bare} in (4) and solving for $\epsilon_{el}^{dressed}$, $\epsilon_{ion}^{dressed}$ we obtain the behavior illustrated in Fig. 1 (this calculation was done in collaboration with J. Gersten).

For $\omega \ll \omega_0$ the polarizability is predominantly ionic with $\epsilon_{el}^{dressed} = 1 + (\kappa_{TF}^{dressed})^2/q^2$

$$(\kappa_{TF}^{dressed})^2 = \frac{\epsilon_\infty}{\epsilon_{ion}^{dressed}(\omega)} (\kappa_{TF}^{bare})^2. \tag{5}$$

We also show $\kappa_{TF}^{dressed}$ as a function of energy (Fig. 1b). (We set the electronic energy equal to ω .) The very small dressed Thomas–Fermi screening is the same as that obtained for semiconductors by Cohen [9] and Das Sarma [18]. For $\omega \gg \omega_0$, the polarizability is predominantly electronic, with $\kappa_{TF}^{dressed} \approx \kappa_{TF}^{bare}$. In Fig. 1c we illustrate the electron–phonon matrix element I as a function of the electronic energy.

These results apply for large values of q appropriate for distances of a lattice constant or smaller distances. For long distances (small q), the electronic polarizability exceeds the ionic one, in spite of the small value of $\kappa_{TF}^{dressed}$.

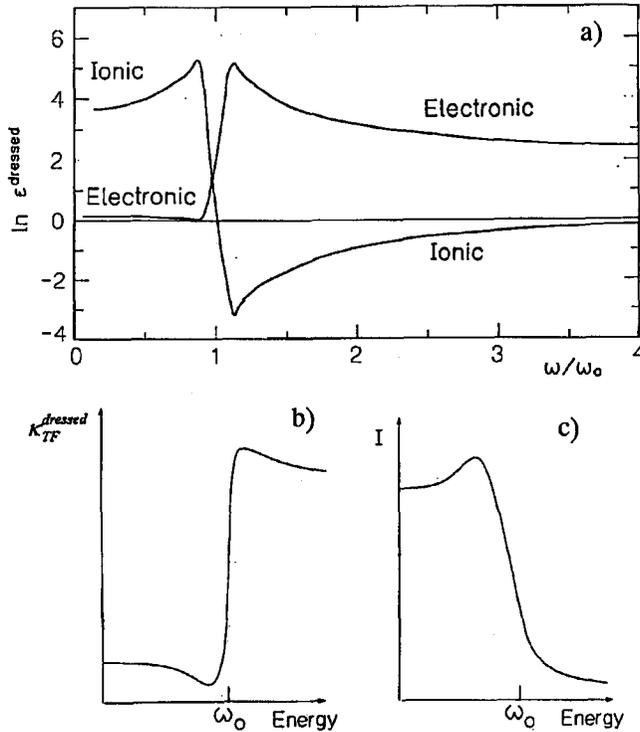


Fig. 1. Crossover from ionic screening at the FS to electronic screening some distance away. (a) The ionic and electronic dressed dielectric constants as a function of frequency. (b) The dressed Thomas-Fermi screening parameter as a function of the electronic energy. (c) The electron-phonon matrix element as a function of the electronic energy.

The cut-off frequency ω_0 can be determined experimentally, and is about 15–20 meV in the cuprates [19] and about 4 meV in the organic superconductors [15]. This is the frequency of the low optical phonons, such as the modes dominated by the barium (or yttrium) ions. These modes have a low frequency because of the high Ba mass and the softness of the Ba–O ionic bonds. In the organics, the low-frequency optical phonons are predominantly librins.

In contrast, the phonons involving the copper–oxygen stretching modes, possess a high frequency $\omega_{\text{ph}} \approx 30\text{--}70$ meV, because of the low oxygen mass and rigidity of the Cu–O covalent bond.

Thus, we expect the large, highly dispersive ionic polarizability to give rise to a very large value of λ_{ph} for electronic states close to the Fermi surface ($\xi(k) < \omega_0$), with a *small* cut-off of order ω_0 . This behavior applies to the coupling with *both* low-frequency and high-frequency phonons.

We thus describe the interaction between the conduction electrons, mediated

by the high-frequency phonons, by the following interaction function:

$$D(k, k', \omega - \omega') = g^2(k, k') \frac{2\omega_{\text{ph}}}{(\omega - i\delta - \omega')^2 - \omega_{\text{ph}}^2}, \quad (6)$$

where $g(k, k')$ is a cut-off when $\xi(k), \xi(k')$ are of order ω_0 . This interaction is seen to possess a strong, explicit dependence on $\xi(k)$, i.e. the component of k perpendicular to the Fermi surface. This strong $|k|$ -dependence is a new feature, not present in the original Eliashberg equations [20].

Equations (4) imply a competition between the electronic and ionic polarizabilities, in which even slightly larger one suppresses the other. Thus the transition from the normal, metallic situation to the anomalous situation of ionic screening, as the bare dielectric constant exceeds about 20, is rather abrupt.

4. Experiments indicating a large λ_{ph} with a small cut-off

The normal state, as well as superconducting state properties of organic metals have been a mystery for more than 20 years. We suggest that they can be accounted for by the transition from ionic screening to electronic screening at a low energy (of order 1–4 meV).

Specifically, the resistivity follows a $\rho \propto T^2$ law above about 30 K, with a freeze-out around 20 K (Fig. 2a, [21]). We account for this behavior as follows. Above the cut-off, the electron–electron interaction is not screened by the ions, therefore it is very strong and the electron–electron scattering rate $1/\tau_{\text{ee}}$ is very large (Fig. 2b). Consequently the mean-free-path $l = v\tau_{\text{ee}}$ is very small (Fig. 2c), and the electronic conduction is due to the states with energy smaller than the cut-off. Thus, at temperatures higher than the cut-off there is an extra factor of T in the expression for the resistivity, and the resistivity due to elastic scattering is proportional to T (instead of being temperature independent, as it usually is) [22], and the resistivity due to scattering by *phonons* is proportional to T^2 (instead of being proportional to T , as it usually is).

A striking feature of the resistivity of ET (ET = BEDT-TTF, bis(ethylenedithio)-tetrathiafulvalene) is the large resistance ratio — a few thousand, up to more than 10^4 for very good samples [21].

In the superconducting state, the cut-off manifests itself as an anomaly in the tunneling I/V curve (Fig. 3a). The energy of the cut-off is considerably smaller than the value of the superconducting gap (at the Fermi surface), therefore the anomaly is called a “zero bias anomaly” (ZBA). In $\alpha_T(\text{BEDT-TTF})_2\text{I}_3$, this anomaly is observed at about 0.5 meV (Fig. 3a, [23]). Since this energy is two orders less than E_F , the k -states responsible for it are within 1% of k_F ; therefore the uncertainty principle requires a junction with dimensions of about a hundred lattice constants (or more) to observe this structure. Indeed, this structure is not observed with an STM, where the dimensions of the contact are small [24].

The superconducting gap at 5 meV is extremely large, $2\Delta(0)/T_c \approx 14$, i.e. 4 times the BCS weak-coupling value. This large gap suggests a very large value of λ_{ph} .

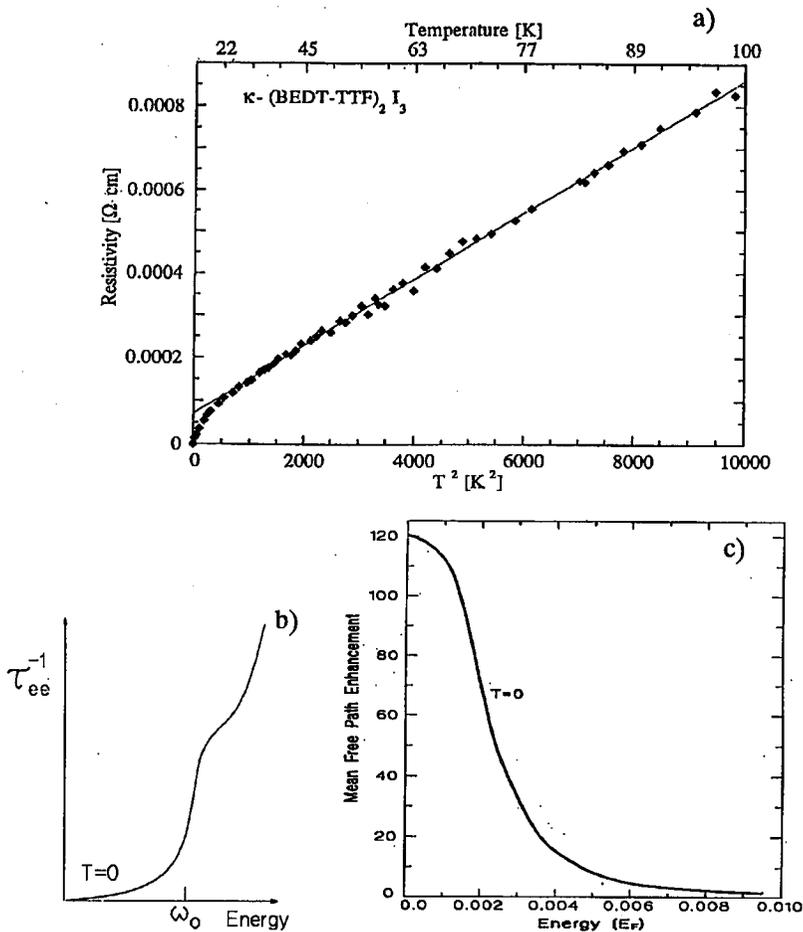


Fig. 2. The role of the small cut-off in giving rise to the $\rho \propto T^2$ law. (a) In-plane resistivity of ET as a function of T^2 [21]. (b) Calculated electron–electron scattering rate as a function of the electronic energy. (c) Transport mean-free-path as a function of the electronic energy.

The structure at 12 meV is probably a McMillan–Rowell structure, showing that the phonons responsible for the pairing possess an energy of about $\omega_{\text{ph}} \approx 7$ meV.

Neutron diffraction shows strong phonon lines at about 2, 4, and 8 meV at temperatures below T_c . At higher temperatures these lines are smeared out (Fig. 3b, [25]). This is an indication of an extremely strong electron–phonon coupling of these modes.

Raman scattering shows phonon lines at 3.5 and 5 meV at temperatures

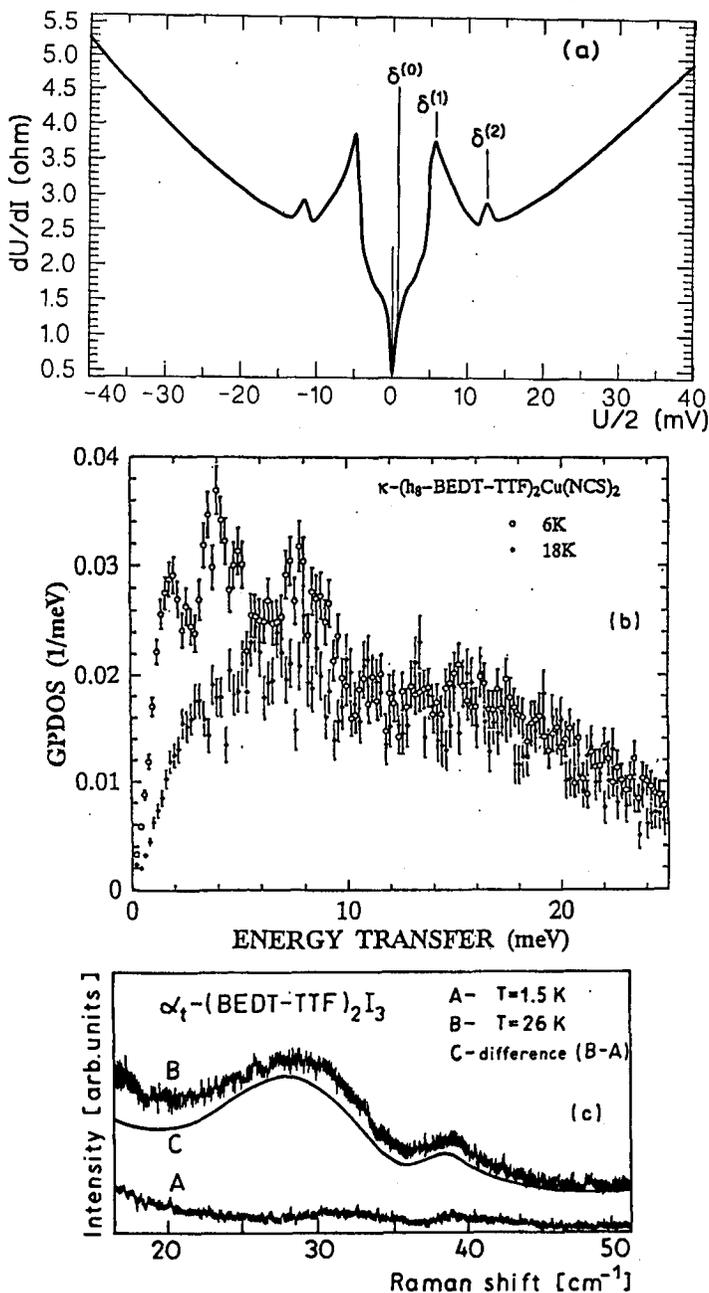


Fig. 3. Experimental manifestation of the role of very low-frequency phonons (librons) in the superconducting process. (a) Point-contact-spectroscopy data for ET [23]. (b) Neutron diffraction data for ET, by Toyota et al. [25]. (c) Raman scattering in ET, by Pokhodnia et al. [26].

above T_c , but below T_c these lines are not seen (Fig. 3c, [26]). By Le Chatelier's principle, since superconductivity has such a drastic effect on these lines, these low-energy optical phonons (librons) must have a strong effect on the superconductivity. However, we do not know yet the exact nature of this effect. A specific suggestion was made by Gherghetta and Nambu [27].

In the cuprates, a ZBA was reported by Gurvitch et al. [28]; however, it is not yet clear whether it is a genuine spectroscopic feature. Anomalies in Raman spectra, and neutron diffraction, are also seen [29], but they are not as strong and "dramatic" as in ET. One suggestion is that in the cuprates the electrons interact with many phonon modes, therefore the coupling with each mode is not very strong [30]. We suggest here an alternative explanation. In order to observe these low-energy phenomena, the elastic mean-free-path l must be very long; otherwise, these structures are smeared out. Because of the large resistance ratio of ET, l/ξ_0 , where ξ_0 is the Pippard coherence length, is very large. We show values of l/ξ_0 for YBCO, the Bechgaard salt TMTSF₂PF₆, and ET (κ -(BEDT-TTF)₂I₃) as

	YBCO	TMTSF ₂ PF ₆	BEDT-TTF
l/ξ_0	6-8	2-5	30-100.

It is seen that while YBCO can be considered to be a "clean" superconductor, ET is "superclean". Thus the inherent behavior at energies low compared with the gap $\Delta(0)$ shows up in a dramatic way.

5. Solution of the Eliashberg equations for an interaction with a very large λ_{ph} and a small cut-off

The small cut-off introduces a strong explicit $|k|$ -dependence into the electron-phonon interaction. Therefore, the Eliashberg equations must be modified to accommodate such an interaction. This requires us to introduce in addition to the well-known [1] renormalization function Z , which describes the frequency (ω) dependence of the self-energy Σ , also a renormalization function X which describes the energy ($\xi(k)$) dependence of the self-energy Σ . Because of the very strong coupling, Z and X depend on the superconducting gap Δ and we must solve for these 3 functions self-consistently. This generalization of the Eliashberg equations is described in detail in Ref. [31].

The small cut-off is found to reduce the effect of the strong Coulomb interaction. The well-known μ^* has to be replaced by

$$\mu^{**} = \frac{\mu}{1 + \mu \ln[(E_F/\omega_{ph})(E_F/\omega_0)]}, \quad (7)$$

where ω_0 is the cut-off. μ^{**} is much smaller than μ^* , and consequently the Coulomb interaction has only a very small effect on T_c [31].

When $\omega_0 < \omega_{ph}$, the cut-off replaces ω_{ph} as the prefactor in the expression for T_c :

$$T_c \approx 0.18\omega_0 \sqrt{\lambda_{ph}/(1 + 2.6\mu^{**})}. \quad (8)$$

Thus the isotope effect (coming from the isotope effect of ω_{ph}) is strongly reduced. The broadening and pulling of the high-frequency phonons (ω_{ph}) is also strongly

reduced by the small cut-off ω_0 [31]. Also, the resistivity due to the high-frequency phonons is strongly reduced by the cut-off. The cut-off thus suppresses the *real* electron-phonon processes strongly, while the *virtual* processes are not strongly suppressed. Since superconductivity is caused by a phonon-mediated interaction due to a virtual process, it is not reduced by the cut-off significantly.

An order-of-magnitude estimate of the maximum transition temperature for "conventional" superconductors ($\kappa_{\text{TF}}/k_{\text{F}} > 1$), based on formula (1) and the Bardeen formula (2), yields

$$(T_{\text{c}})_{\text{max}} \approx 0.25 E_{\text{F}} \sqrt{pm^*/M}, \quad (9a)$$

p is the degeneracy (5 for a d -band, as in Nb; 1 for $p\sigma$ orbitals in cuprates); m^* is the electronic effective mass; M is the atomic, or molecular, reduced mass. For "exotic" superconductors ($\kappa_{\text{TF}}/k_{\text{F}} < 1$), we obtain [32]

$$(T_{\text{c}})_{\text{max}} \approx 0.25 E_{\text{F}} \sqrt{pm^*/M} (r_{\text{s}}/2), \quad (9b)$$

where r_{s} is the average distance between electrons in units of the Bohr radius. In 3 dimensions, $r_{\text{s}} = 1.92e^2/[\hbar\epsilon_{\infty}v_{\text{F}}(\text{band})]$. In exotic superconductors, which are near the Mott transition, r_{s} is large (about 8–10), and as a result $(T_{\text{c}})_{\text{max}}$ is abnormally large.

6. Conclusion

Exotic superconductors, such as organics and cuprates, possess a highly dispersive background, due to the high ionic polarizability: $\epsilon_0/\epsilon_{\infty} > 10$. The role of this ionic polarizability has apparently not received adequate attention.

We find that this ionic polarizability dominates both the normal and superconducting state properties. For electronic states very close to the Fermi surface, the screening is ionic rather than electronic. Consequently, the electron-phonon coupling is extremely strong. For states somewhat removed from the FS, the screening is electronic and electron-electron Coulomb interactions are very strong. The crossover is determined by the dispersion of the ionic polarizability.

Normal state properties can be described by the Boltzmann transport equation, with a mean-free-path l_k with an extremely strong $|k|$ -dependence; near the FS it is very long (particularly at low temperatures) and it drops to nearly zero at the crossover energy.

Superconducting properties follow from a solution of the Eliashberg equations, which are generalized by introducing a strongly $|k|$ -dependent electron-phonon matrix element.

This theory accounts for the anomalous properties of both organic metals and cuprates.

The large resistance ratio of the organics provides a "superclean" situation, where the elastic mean-free-path is *two* orders larger than the Pippard coherence length. This makes it possible to determine the low-energy behavior in an accurate and reliable way, and in this way elucidate the microscopic origin of high temperature superconductivity.

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