High temperature superconductivity due to a long-range electron-phonon interaction and super-light bipolarons in cuprates.

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Strong electron-phonon interaction in cuprates and other high-temperature superconductors has gathered support over the last decade in a large number of experiments. Here I introduce the "Froehlich-Coulomb" multi-polaron model of high-temperature superconductivity, which includes strong on-site repulsive correlations and the long-range Coulomb and electron-phonon (e-ph) interactions. The bipolaronic extension of the BCS theory to the strong-coupling regime with a long-range *unscreened* electron-phonon interaction naturally explains the critical parameters, isotope effects, pseudo-gaps, thermomagnetic transport, and checkerboard modulations of the tunnelling density of states in cuprates.

1. The Froehlich-Coulomb model

A significant fraction of research in the field of high-temperature superconductivity[1] suggests that the interaction in novel superconductors is essentially repulsive and unretarted, but it provides high Tc without any phonons. A motivation for this concept can be found in the earlier work by Kohn and Luttinger[2] who showed that the Cooper pairing of repulsive fermions is possible. However, the same work showed
that Tc of repulsive fermions is extremely low, well below the mK scale. Nevertheless, the BCS and BCS-like theories (including the Kohn-Luttinger consideration) heavily rely on the Fermi-liquid model of the *normal* state. This model fails in cuprates, so that there are no obvious *a priory* reasons to discard the dogma, if the normal state is not the Fermi-liquid. Strong onside repulsive correlations (Hubbard U) are indeed essential in the cuprates, which are doped charge-transfer insulators.

However, independent of any experimental evidence the Hubbard U (or t-J) models share an inherent difficulty in determining the order. While some groups claimed that they describe high-Tc superconductivity at finite doping, other authors could not find any superconducting instability without an additional (i.e. e-ph) interaction. Therefore it has been concluded that models of this kind are highly conflicting and confuse the issue by exaggerating the magnetism rather than clarifying it [3] . There is also another serious problem with the Hubbard-U approach to high temperature superconductivity in the cuprates. The characteristic (magnetic) interaction, which might be responsible for the pairing in the Hubbard model, is the spin exchange interaction, $J=4t^2 / U$, of the order of 0.1 eV. It turns out much smaller than the (inter-site) Coulomb repulsion and the unscreened long-range (Froehlich) e-ph interaction each of the order of 1 eV, routinely neglected within the approach. There is virtually no screening of e-ph interactions with c-axis polarized optical phonons in cuprates because the upper limit for the out-of-plane plasmon frequency (< 200 cm^{-1}) is well below the characteristic phonon frequency, ω= 400 - 1000 cm^{-1}. Because of the poor screening, the magnetic interaction remains small compared with the Froehlich interaction at any doping. Hence, any realistic approach to superconductivity in cuprates should treat the Coulomb and unscreened e-ph interactions on an equal footing.

We have developed a so-called "Froehlich-Coulomb" model [4] to deal with the strong Coulomb and long-range e-ph interactions in cuprates and other doped oxides. The model Hamiltonian explicitly includes the long-range electron-phonon and Coulomb interactions as well as the kinetic and deformation energies. The implicitly present large Hubbard U term prohibits double occupancy and removes the need to distinguish fermionic spins. Introducing spinless fermionic, $c_{\mathbf{n}}$ and phononic, $d_{\mathbf{m}}$ operators the Hamiltonian of the model is written as

$$H = \Sigma_{\mathbf{n},\mathbf{n}'} \, [\, t(\mathbf{n}-\mathbf{n}')\, c_{\mathbf{n}}^{+} c_{\mathbf{n}'} + V_c\,(\mathbf{n}-\mathbf{n}')\, c_{\mathbf{n}}^{+}\, c_{\mathbf{n}'}^{+}\, c_{\mathbf{n}'}\, c_{\mathbf{n}}] - \omega \Sigma_{\mathbf{n},\mathbf{m}}\, g(\mathbf{m}-\mathbf{n})\, \mathbf{e}\mathbf{u}_{\mathbf{m}-\mathbf{n}}\, c_{\mathbf{n}}^{+} c_{\mathbf{n}} (d_{\mathbf{m}}^{+} + d_{\mathbf{m}}) + \omega \Sigma_{\mathbf{m}} (d_{\mathbf{m}}^{+} d_{\mathbf{m}} + 1/2),$$

where $\mathbf{e}$ is the polarization vector of the vibration coordinate, $\mathbf{u}_{\mathbf{m}-\mathbf{n}} = (\mathbf{m}-\mathbf{n})/|\mathbf{m}-\mathbf{n}|$ is the unit vector in the direction from electron $\mathbf{n}$ to the ion $\mathbf{m}$, $g(\mathbf{m}-\mathbf{n})$ is a dimensionless e-ph coupling function, and $V_c\,(\mathbf{n}-\mathbf{n}')$ is the intersite Coulomb repulsion. This Hamiltonian can be solved analytically in the extreme case of the strong e-ph interaction with the e-ph dimensionless coupling constant $\lambda = E_p/zt > 1$ using $1/\lambda$ multipolaron expansion technique[4] . $E_p = \omega \Sigma_{\mathbf{n},\mathbf{m}} [g(\mathbf{m}-\mathbf{n})\, \mathbf{e}\mathbf{u}_{\mathbf{m}-\mathbf{n}}]^2$ is the polaron level shift about 1 eV and zt is the half-bandwidth in a rigid lattice.

The model shows a reach phase diagram depending on the ratio of the inter-site Coulomb repulsion V_c and the polaron (Franc-Condon) level shift E_p[5] . The ground state is a *polaronic* Fermi liquid at large Coulomb repulsions, a *bipolaronic* high-temperature superconductor at intermediate Coulomb repulsions, and a charge-segregated insulator at weak repulsion. The model predicts *superlight* bipolarons with a remarkably high superconducting critical temperature. It describes many other properties of the cuprates[4], in particular the isotope effects, normal state transport and

real-space modulations of the single-particle density of states (DOS) as discussed below.

2. Band-structure isotope effect

The isotope substitution, where an ion mass M is varied without any change of electronic configurations, is a powerful tool in testing the origin of electron correlations in solids. In particular, a finite value of the isotope exponent α=-dlnTc/dln M proved that the superconducting phase transition at T=Tc is driven by the electron-phonon interaction in conventional low-temperature superconductors. Advances in the fabrication of isotope substituted samples made it possible to measure a sizable isotope effect also in many high-temperature superconductors. This led many authors to a general conclusion that phonons are relevant for high Tc. Essential features of the isotope effect on Tc, in particular its large values in low Tc cuprates, an overall trend to decrease as Tc increases, and a small or even negative α in some high Tc cuprates, were understood in the framework of the bipolaron theory of high-temperature superconductivity[6] .

The most compelling evidence for (bi)polaronic carries in novel superconductors was provided by the discovery of a substantial isotope effect on the (super)carrier mass [7]. The effect was observed by measuring the magnetic field penetration depth of isotope-substituted copper oxides. The carrier density is unchanged with the isotope substitution of O^{16} for O^{18} , so that the isotope effect on the penetration depth measures directly the isotope effect on the carrier mass m*. A carrier mass isotope exponent α_m=-dlnm*/dln M was observed, as predicted by the bipolaron theory[6] . In ordinary metals, where the Migdal adiabatic approximation is believed to be valid, α_m=0 is expected. However, when the e-ph interaction is sufficiently strong, the effective mass m* depends on M as m* = m exp (γ E_p/ ω). Here m is the band mass in the absence of the electron-phonon interaction, and γ is a numerical constant less than 1 that depends on the radius of the electron-phonon interaction. In the expression for m* only the phonon frequency depends on the ion mass. Thus there is a *large isotope effect on the carrier mass* in (bi)polaronic conductors, α_m = (1/2)ln(m*/m), in contrast with the zero isotope effect in ordinary metals.

Recent high resolution angle resolved photoemission spectroscopy (ARPES)[8] provided another compelling evidence for a strong e-ph interaction in the cuprates. It revealed a fine phonon structure in the electron self-energy, and a complicated
isotope effect on the electron spectral function in Bi2212.

These and many other experimental and theoretical observations point towards unusual e-ph interactions in complex oxides, which remain to be quantitatively addressed.

We have performed quantum Monte Carlo (QMC) simulations [9] of a single-polaron band dispersion with any-range e-ph interaction in the most relevant intermediate region of the coupling strength, λ ω1, and of the adiabatic ratio, ω/t ω1, where any analytical or even semi-analytical approximation (i.e. dynamic mean-field approach in finite dimensions) is unreliable. Unlike the strong-coupling limit [6] the band-structure isotope effect depends on the wave vector in the intermediate region of parameters. It also depends on the radius of the e-ph interaction. If we define a band-structure isotope exponent as

$$\alpha_b(\mathbf{k}) = -d \ln E(\mathbf{k})/d \ln M,$$

it does not depend on the wave-vector **k** in the extreme strong-coupling limit, λ >>1 because e-ph interactions do not change the band topology in this limit , and α_b is the same as α_m. Notwithstanding QMC results[9] show that the isotope exponent becomes a nontrivial function of the wave vector in the intermediate-coupling regime, because e-ph interactions substantially modify the band topology in this regime especially for a short-range e-ph interaction. The isotope exponents $\alpha_b(\mathbf{k})$ are presented in Fig.1,2 for the small Holstein polaron (SHP) with the short-range interaction, Fig.1, and for the small Froehlich polaron (SFP) with the long-range interaction, Fig.2. The polaron spectra are calculated for two phonon frequencies, ω = 0.70 t and ω =0.66 t , whose difference corresponds to a substitution of O^{16} for O^{18} in cuprates. There is a significant change in the dispersion law (topology) of SHP, Fig.1, which is less significant for SFP, Fig.2, rather than a simple band-narrowing. The strongest dispersion of $\alpha_b(\mathbf{k})$ is observed for SHP. Importantly, the isotope effect is suppressed near the band edge.

Using the 1/λ one can calculate the total electron spectral density [4] and predict the isotope effect also on the incoherent background due to phonon dressing of carriers [9]. While our prediction is qualitatively robust it is difficult to quantify the ARPES isotope effect in the intermediate region of parameters. The role of electronic correlations should be also addressed in connection with ARPES. While the results shown in Figs.1,2 describe band-structure isotope effects in slightly-doped conventional and Mott-Hubbard insulators with a few carriers, their spectral properties could be significantly modified by the polaron-polaron interactions , in particular by the bipolaron formation at finite doping. On the experimental side, a separation of the coherent and incoherent parts in ARPES remains rather controversial.

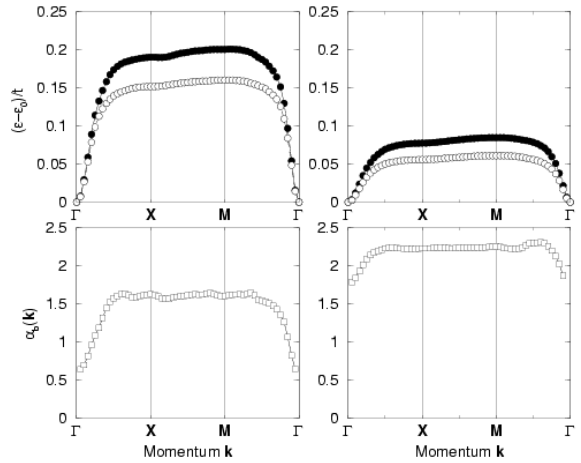

Fig.1. SHP polaron band dispersion and isotope effect along different directions of the Brillouin zone for two phonon frequencies and λ=1.1 (left panels) and λ=1.2 (right panels).

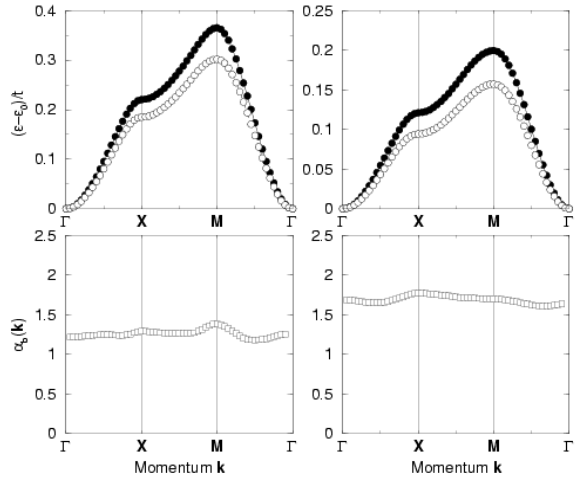

Fig.2. SFP polaron band dispersion and isotope effect along different directions of the Brillouin zone for two phonon frequencies and λ=2.5 (left panels) and λ=3.0 (right panels).

3. Pseudogaps and thermomagnetic transport

There is much evidence for the crossover regime at T* and normal state charge and spin gaps in the cuprates [4]. Within the Froehlich-Coulomb model these energy gaps could be understood as being half of the binding energy, Δ_p and the singlet-triplet exchange energy of preformed bipolarons, respectively. Notwithstanding more "direct" evidence for the existence of a charge 2e Bose liquid in the normal state of cuprates is highly desirable. Mott and Alexandrov[10] discussed the thermal conductivity, the contribution from the carriers given by the Wiedemann-Franz ratio depends strongly on the elementary charge as and should be significantly suppressed in the case of e*=2e compared with the Fermi-liquid contribution. As a result, the Lorenz number, L differs significantly from the Sommerfeld value $L_e = \pi^2 / 3$ of the standard Fermi-liquid theory, if carriers are bipolarons. Ref. [10] predicted a rather low Lorenz number $L = 6L_e /(4\pi^2) \omega 0.15 L_e$ due to the double charge of carriers, and also due to their nearly classical distribution function above Tc.

Unfortunately, the extraction of the electron thermal conductivity has proven difficult since both the electron term and the phonon term are comparable to each other in the cuprates. Only recently a new way to determine the Lorenz number has been realized by Zhang et al. [11] based on the thermal Hall conductivity. The thermal Hall effect allowed for an efficient way to separate the phonon heat current even when it is dominant. As a result, the Lorenz number has been directly measured in YBCO because transverse thermal and electrical conductivities involve only the electrons. Remarkably, the measured value of L just above Tc was found about the same as predicted by the bipolaron model. The experimental L showed a strong temperature dependence, which violates the Wiedemann-Franz law. Based on the Froehlich-Coulomb model and the conventional Boltzmann kinetics we [12] explained the Lorenz number in the cuprates. We have demonstrated that the Wiedemann-Franz law breaks down because of the interference of polaron and bipolaron contributions to the heat transport. When thermally excited polarons and also triplet pairs are included, our model explains the violation of the Wiedemann-Franz law in the cuprates and the Lorenz number as seen in the experiment, Fig.3.

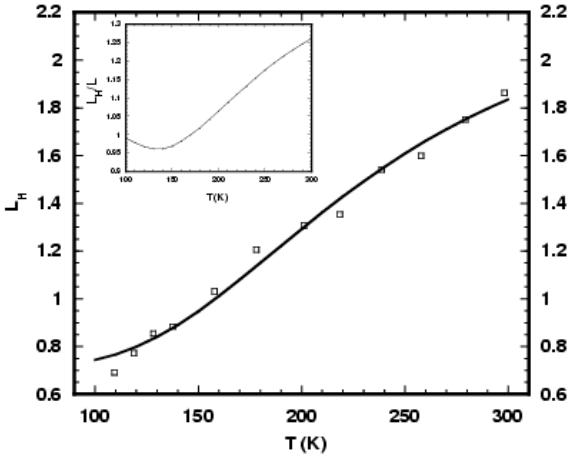

Fig.3. The Hall Lorenz number of the bipolaron model [12] fits the experiment in YBa2Cu3O6.95 [11]. Charge and spin pseudogaps are taken as 675 K and 150 K, respectively, and the ratio of the polaron and bipolaron Hall angles is 0.36. The inset gives the ratio of Hall Lorenz number to Lorenz number in the model.

4. Checkerboard spatial modulations of the tunnelling DOS.

We have proposed a simple phenomenological model [13] explaining a difference between the normal and superconducting gaps in cuprates. The main assumption, supported by a parameter-free estimate of the Fermi energy is that the attractive potential is large compared with the polaron Fermi energy, so that the ground state is the Bose-Einstein condensate of tightly bound real-space pairs. Here I argue that if pairs are Bose-condensed with nonzero center-of-mass momenta, the tunnelling density of states (DOS) should be modulated in real space revealing the checkerboard patterns [14] as observed [15] .

Real-space pairs might have an unconventional symmetry due to a specific symmetry of the pairing potential as in the case of the Cooper pairs, but in any case the ground state and DOS are homogeneous, if pairs are condensed with **K**=0. However, if a pair band dispersion has its minima at finite **K** in the center-of-mass Brillouin zone, the Bose condensate is inhomogeneous. In particular, the center-of-mass bipolaron energy bands could have their minima at the Brillouin zone boundaries at $\mathbf{K} = (\pi, 0)$ and three other equivalent momenta. These four states are degenerate, so that the condensate wave function in the real (Wannier) space, is their superposition. Then solving the Bogoliubov equations describing the interaction of single polarons with the pair Bose condensate[14] one obtains the checkerboard DOS similar to those observed in a few cuprates[15] .

Our 'kinetic' interpretation of charge modulations in cuprates, originally proposed [14] before STM results[15] became available, is consistent with the inelastic neutron scattering, where incommensurate inelastic peaks were observed only in the superconducting state [16] . The vanishing at Tc of the incommensurate peaks is inconsistent with any other heterogeneity picture, where a characteristic distance needs to be observed in the normal state as well.

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