



Article

Electron Correlation and High-Temperature Superconductivity

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Abstract

Strong electron correlation plays a central role in the high-temperature superconductivity (HTSC) of cuprates. However, to date, research has focused only on its role in spin dynamics and related effects, even though it is becoming increasingly clear that spin alone may not be sufficient to create HTSC. Here, we discuss a possible role of electron correlation in the Bose–Einstein condensation (BEC) of Cooper pairs. Recently, we succeeded in observing dynamic electron correlation via inelastic X-ray scattering through results presented in real space. We discovered that electron correlations are strongly modified in the plasmon, proving that electron dynamics significantly affect electron correlation. Earlier, we found that in ^4He , the atom–atom distance in the BE condensate is 10% longer than that in the non-condensate. These results suggest the possibility that the reduction in electron-repulsion energy upon BEC is driving T_c to high values. Thus, electron correlation itself could be the origin of the HTSC phenomenon.

Keywords: electron correlation; cuprates; high-temperature superconductivity; Bose–Einstein condensation

1. Introduction

The discovery of high-temperature superconductivity (HTSC) in the cuprates [1] was a huge surprise, as it defied conventional wisdom based on the BCS theory [2–5]. As the parent compounds are antiferromagnetic Mott insulators, strong electron correlation and its effect on spins became the central theme of theoretical research. Various spin-based theories, such as the RVB theory [6,7], were proposed. At the same time, a number of reports on anomalous lattice effects [8–28] suggested that the phenomena are more complex, possibly involving phonons to form the polaronic or vibronic state [29–36]. After the initial rapid advances, progress in the field slowed down, mainly because of major theoretical and computational challenges posed by the many-body nature of strong electron correlation.

Nearly 40 years after its discovery, we may now finally be making progress toward the answer due to recent advances in computational power and numerical methods, such as the quantum Monte Carlo method and the density matrix renormalization group approach. The results are rather dramatic; it is becoming apparent that the single-band Hubbard model does not support superconductivity [37]. As the single-band Hubbard model is the minimalist model for the spin-only scenario, this negative result raises serious doubt regarding simple spin mechanisms as the sole origin of the HTSC phenomenon. At the same time, the bipolaron mechanism has been revisited, focusing on Fe-pnictides, and it has been shown that bond-stretching phonons can achieve a much higher transition temperature, T_c , than expected earlier [38]. Although the controversy still rages, it may



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be time to expand the range of arguments and to reconsider the question from different points of view. In this article, based upon our recent direct observation of dynamic electron correlation and our earlier observation of dynamic atomic correlation in superfluid ^4He , we propose that the changes in electron correlation upon Bose–Einstein condensation (BEC) may play a major role in the HTSC phenomenon.

2. Direct Observation of Dynamic Electron Correlation

Electrons interact with each other through strong Coulomb repulsion. Yet the nearly-free electron model is amazingly successful, because electron-electron scattering is suppressed due to the Pauli exclusion principle as elucidated by the Landau quasiparticle picture [39]. This led to the success of density functional theory (DFT) [40], making the accurate calculation of electronic states in solids and molecules possible, except for strongly correlated electron systems, which require higher-level models, such as the Hubbard model [41].

In contrast to these advances in theory, only a few attempts have been reported on direct experimental studies of electron correlation. They rely on X-ray diffraction from electrons [42–44]. However, X-ray scattering from electrons is inherently inelastic. In X-ray diffraction experiments, a detector records all the scattered X-ray photons without energy discrimination. Thus, it captures only the same-time (snapshot) correlation [45]. To date, no direct observation of dynamic electron correlation has been conducted. To observe dynamic electron correlation, we measured the dynamic structure factor, $S(Q, E)$, for electrons where Q and E are the momentum and energy exchanges upon scattering, via medium-resolution inelastic X-ray scattering using polycrystalline beryllium as a sample. The details of the experiment are given in ref. [46].

The $S(Q, E)$ for electron has been measured many times by various researchers. However, the results were analyzed only in terms of electronic excitations [47–53]. At present, the standard analysis in terms of excitation assumes the random-phase-approximation (RPA) decoupling of the initial and final states in the expression of the X-ray scattering cross-section [54]. If we do not apply RPA decoupling, $S(Q, E)$ is a direct Fourier-transformation of a two-time density correlation function, as shown by Van Hove [55],

$$S(Q, E) = \frac{1}{V} \iint G(\mathbf{r}, t) \exp(-i[\mathbf{Q} \cdot \mathbf{r} - t]) d\mathbf{r} dt \quad (1)$$

where $E = \hbar\omega$, and $G(\mathbf{r}, t)$ is the Van Hove function,

$$G(\mathbf{r}, t) = \frac{1}{V} \iint \langle (\mathbf{r}', t') \rho(\mathbf{r}' + \mathbf{r}, t' + t) \rangle d\mathbf{r}' dt' \quad (2)$$

This expression shows that $S(Q, E)$ contains information on dynamic two-body correlation; however, unlike the information on single-particle excitation, the signal is spread over the entire Q - E space and almost impossible to perceive.

To extract the information on correlation it is best to transform $S(Q, E)$ into the energy-resolved dynamic pair-distribution function (E -resolved DyPDF) [56],

$$g(r, E) = \frac{1}{2\pi^2 \rho r} \int S(Q, E) \sin(Qr) Q dQ \quad (3)$$

The E -resolved DyPDF shown in Figure 1 contains two contributions, one from the self-correlation that describes single-particle excitations, and the other from dynamic two-body correlations. $g(r, E) = 0$ corresponds to the average electron density, or the absence of correlation. The portion below 1 Å is due to the self-term, which is spread due to quantum uncertainty. Below $E = 20$ eV, the density correlation is suppressed below 2 Å (blue area),

and this area signifies the exchange-correlation hole. This size, $\sim 2 \text{ \AA}$, is consistent with theoretical estimates [57]. However, around the plasmon energy of 21 eV, the suppression in correlation, and thus the exchange-correlation hole, is extended up to $\sim 5 \text{ \AA}$. Within the plasmon, electrons move together more or less in parallel, with less chance of colliding with each other. This apparently enlarges the exchange-correlation hole. This result proves that electron dynamics can significantly affect electron correlation.

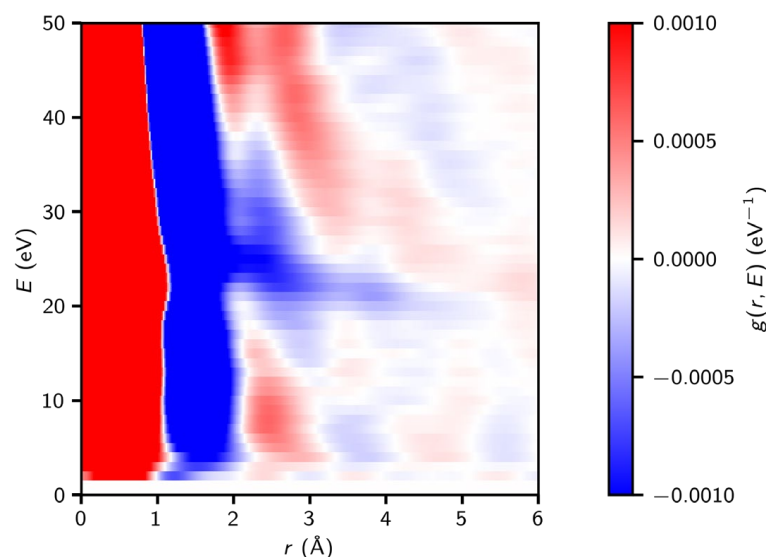


Figure 1. Electronic energy-resolved dynamic PDF of beryllium [46]. Features below 1 Å are due to self-part (single particle excitations). The negative part below 2 Å signifies the exchange-correlation hole, which is extended at the plasmon energy of 21 eV.

3. Bose–Einstein Condensation in ^4He

^4He undergoes BEC to the superfluid state. However, due to van der Waals interaction, only 7% of He atoms actually condense to the ground state [58–61], whereas the He–He distance is known to be 3.6 Å from the neutron diffraction PDF [62], which represents the He–He distance in the non-condensate. To find the actual He–He distance in the condensate, we measured the energy-resolved dynamic PDF using pulsed neutron scattering. By focusing on the energy range of the roton excitation at 0.8 meV [63] and considering the difference in $g(r, E)$ below and above the superfluid transition, we determined the PDF of the BEC atoms [64]. We found that the actual He–He distance in the condensate is 4.0 Å, longer by 10% than that in the non-condensate, as shown in Figure 2 [65]. In the condensate, all He atoms are in the ground state with zero momentum. Thus, they do not collide with each other, aside from quantum fluctuations. As a result, similarly to electrons in plasmons, they stay further apart, resulting in the longer He–He distance as observed.

Actually, it is known that the atom–atom distances in many metallic liquids become shorter as temperature increases, in contrast to thermal expansion [66,67]. This occurs because higher kinetic energy allows atoms to approach closer to each other. Even though the average density decreases with increasing temperature, the direct atom–atom distance decreases at the expense of the coordination number, which also decreases. This common behavior of liquid metal naturally explains the increased He–He distance in the BE condensate.

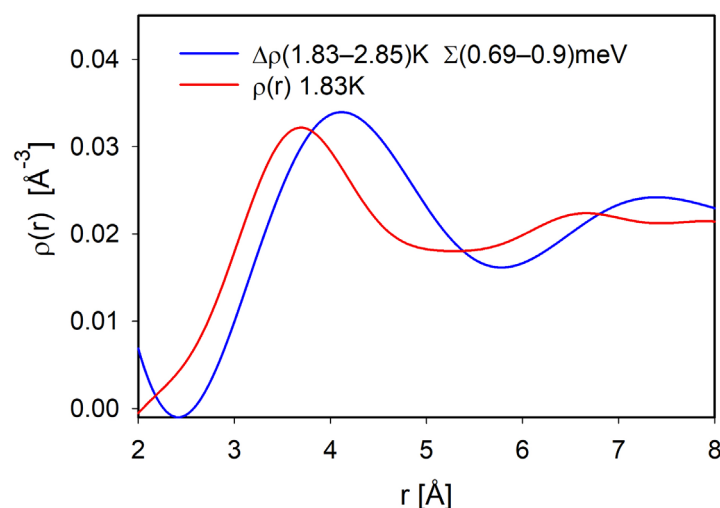


Figure 2. The total PDF (red) and the PDF of the BEC atoms (blue) [64]. See text for details.

4. Lattice Effects in the Cuprates

4.1. Lattice Effects at T_c

In regular BCS superconductors, Cooper pairs are large in size and overlap with each other. They B-E condensate in reciprocal space, and spatially they are uncorrelated. However, in the HTSC cuprates, the superconductive coherence length is short, and Cooper pairs are more localized in space, being hindered by spin correlations [68–70]. Furthermore, various experimental, computational, and theoretical results have indicated that doped holes may form polarons [29–34], or more specifically vibrons, in which many phonons form wave packets to trap electrons [35,36].

Early signs of anomalous lattice effect include the observations of dynamic structural anomaly at the superconductive transition temperature, T_c [19,20]. As shown in Figure 3, the pulsed neutron PDF peak height at 3.4 Å for $\text{Ti}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$ shows anomalous temperature dependence in the vicinity of T_c . This distance, 3.4 Å, corresponds to the separation between the apical oxygen and the in-plane oxygen. This effect is dynamic in origin. In terms of the atomic position, $S(Q, E)$ is given by [55],

$$S(Q, \omega) = \frac{1}{2\pi\hbar N} \sum_{j,j'} \int \langle e^{iQ[R_j(0) - R_{j'}(t)]} \rangle e^{i\omega t} dt \quad (4)$$

where $R_j(t)$ is the position of the j -th atom at time t , and $E = \hbar\omega$. Thus, the same-time (snapshot) PDF ($t = 0$) is obtained by Fourier-transforming the total structure function,

$$S(Q) = \int S(Q, \omega) d\omega \quad (5)$$

In diffraction experiments, however, the energy integral is performed by default by a detector placed at 2θ , with no energy discrimination. Because Q depends on E , the measured intensity,

$$S_{\text{Diff}}(Q_0) = \int S(Q(E), E) dE \quad (6)$$

where $Q_0 = Q(E = 0)$. This is slightly different from Equation (5), and the difference depends on the incident energy of the probe. Indeed, the anomalous peak in Figure 3 depends on the incident energy of neutrons, as expected for a dynamical effect [20].

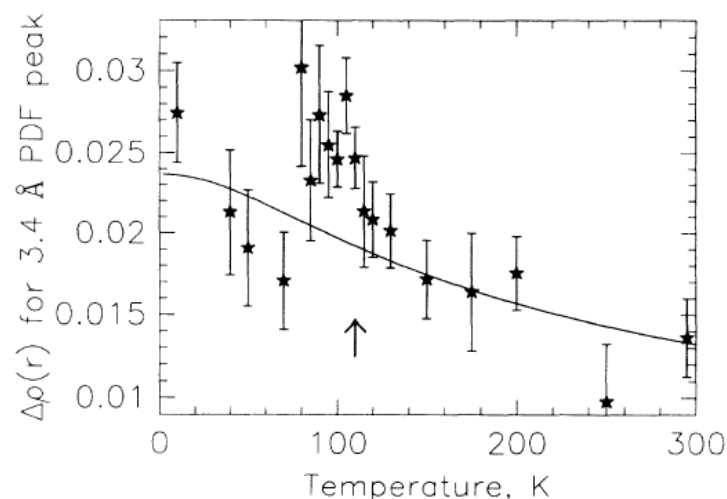


Figure 3. Temperature dependence of the pulsed neutron PDF peak height of $\text{Ti}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$ at 3.4 Å [20]. The arrow indicates T_c (≈ 110 K). The curve is the normal (Debye-Waller) temperature dependence.

At the same time, the extended X-ray absorption fine structure (EXAFS) measurements on $\text{YBa}_2\text{Cu}_3\text{O}_7$ detected a similar anomaly at T_c [21], which was interpreted in terms of HTSC-induced tunnelling of apical oxygen ion [22]. The apical oxygen has two sites, separated by 0.13 Å, and resonant hopping between the two sites with HTSC fluctuations creates the observed anomaly. Indeed, the re-evaluation of the effect in Figure 3 by separating elastic and inelastic scattering showed the two dynamic apical oxygen sites separated by 0.3 Å for $\text{Ti}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$, even though there is only one site in the time average [71]. These results demonstrate that the HTSC fluctuations couple to the apical oxygen ions, just below T_c , at which point the SC gap size becomes comparable with the oxygen tunneling frequency.

4.2. Phonon Anomalies

More direct evidence of lattice effects is found in the behavior of Cu-O bond stretching phonons. The longitudinal oxygen mode in the CuO_2 plane, the so-called half-breathing mode, shows strong mode softening, from 66 meV to 55 meV, in the Q range $h > 0.25$, as temperature decreases for $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$ (Figure 4a) [28]. However, the softening with a decrease in temperature is not homogeneous, and it occurs through a gradual spectral weight transfer from the high-energy to the low-energy branch, with the amount of transfer proportional to the HTSC order parameter, as shown in Figure 4b [28]. This demonstrates that the coupling is local, and the density of phonons involved in local coupling, soft phonons, increases as the HTSC order parameter increases. In other words, the coupling occurs in real space, forming vibrons.

Further evidence of local coupling is found in doping dependence. In $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$, as x is increased, the energies of the upper and lower branches do not change, while the spectral weight changes proportionally to x , as shown in Figure 5 [72]. These results prove that the lower branch is created because of local coupling of the half-breathing phonon modes to doped holes forming vibrons. At the optimal doping ($x = 0.95$), almost all the intensity is transferred to the lower branch. Interestingly, this coupling does not start at T_c , but instead at the pseudogap temperature, T^* . As shown in Figure 6, the intensity of the upper branch decreases with decreasing temperature below ~ 200 K for $\text{YBa}_2\text{Cu}_3\text{O}_{6.8}$ ($T_c = 80$ K and $T^* = 200$ K) and ~ 300 K for $\text{YBa}_2\text{Cu}_3\text{O}_{6.6}$ ($T_c = 60$ K and $T^* = 300$ K). The origin of the pseudogap is still controversial, but a large amount of data suggests that it is the temperature at which local Cooper pairs are formed [73–75]. The data in Figures 4–6

support this idea and suggest that the half-breathing phonons are directly involved in local pair formation. Even though the coupling between a hole and a single phonon is weak [4], through the formation of a vibron, in which many phonons collectively couple to a hole by forming an electron-phonon wave packet, this coupling becomes very strong [35].

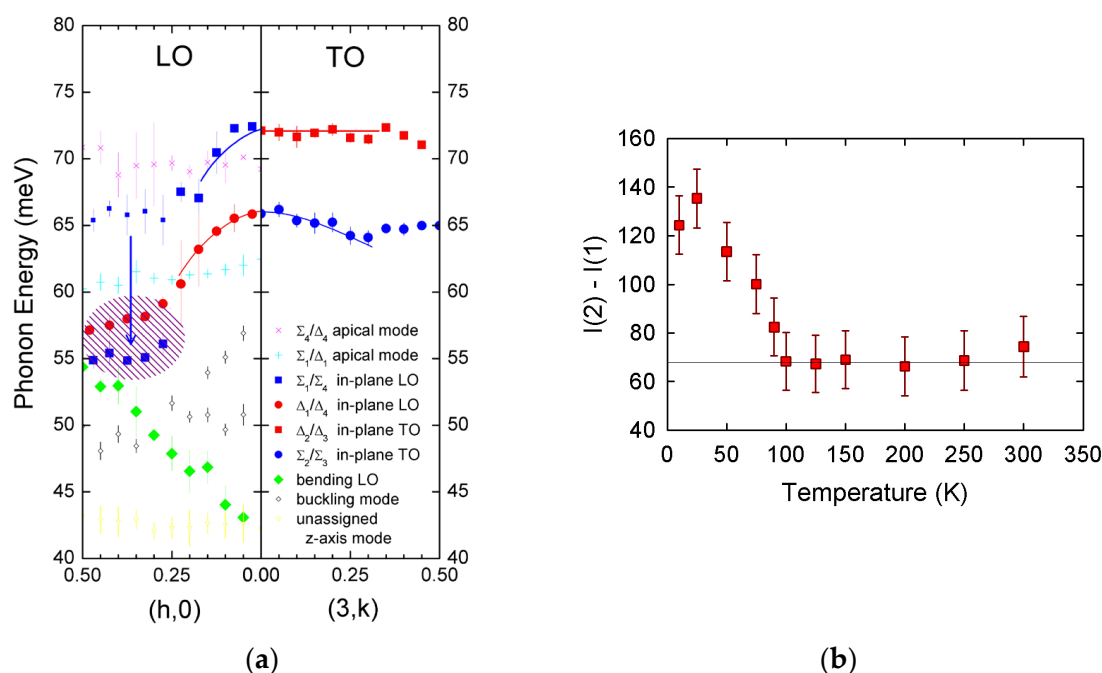


Figure 4. (a) Dispersion of the half-breathing phonon mode (blue square) of YBa₂Cu₃O_{6.95}, softening from 66 to 55 meV, indicated by an arrow, (b) The spectral weight shift from I(1): 56–68 meV to I(2): 51–55 meV at $h = 0.25$ as a function of temperature [28].

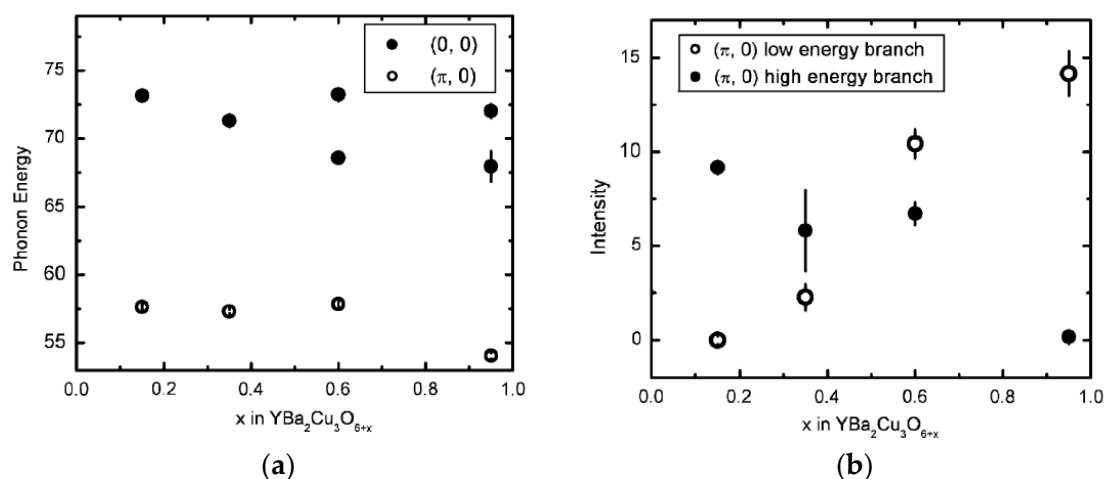


Figure 5. (a) The energies of the half-breathing phonon mode of YBa₂Cu₃O_{6+x} at (0, 0) and (π ($h = 0.5$), 0) at 10 K, and (b) the spectral weight shift from the upper to lower branches as a function of doping, x [72].

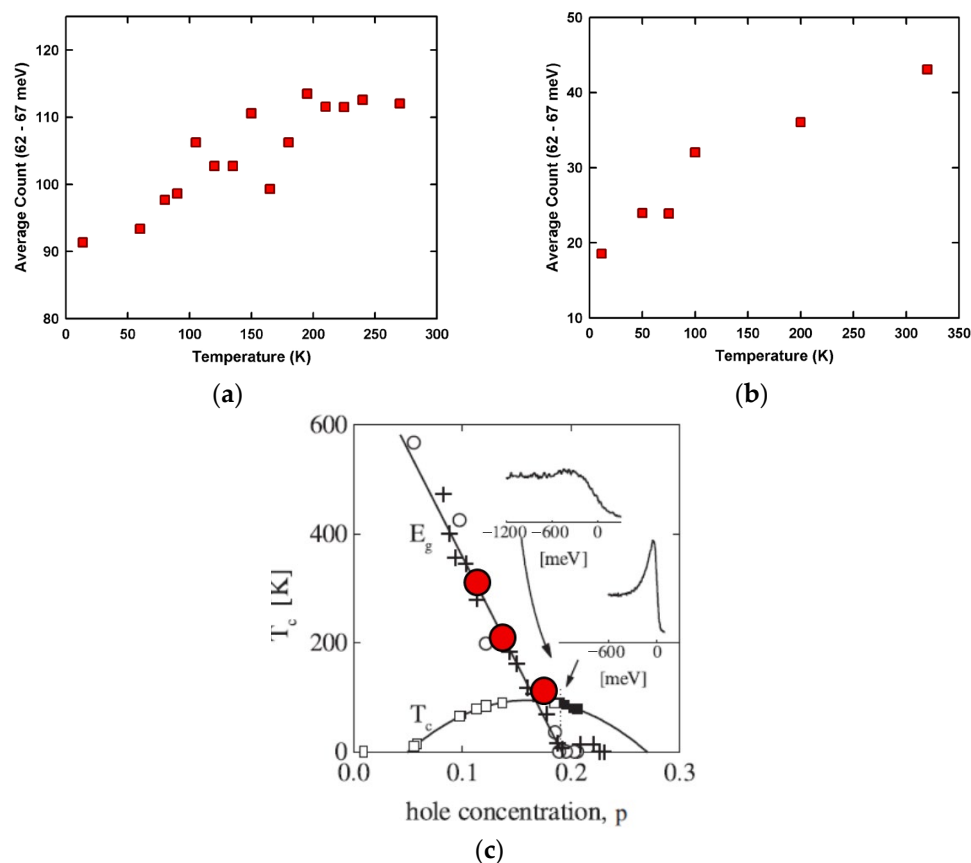


Figure 6. The average phonon intensity (62–67 meV) of the half-breathing phonon mode of $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ at $h = 0.5$, (a) for $x = 0.8$ ($T_c = 80$ K and $T^* = 200$ K), and (b) for $x = 0.6$ ($T_c = 60$ K and $T^* = 300$ K). (c) Hole concentration dependence of the pseudogap energy E_g (open circle by ARPES, cross by specific heat) expressed as T^* [76] and the starting temperature of the phonon spectral weight shift (red circle, added by the author).

Our direct diagonalization calculation on the two-band 1D Hubbard model with frozen phonon shows strong effects of the half-breathing phonon modes on doped holes [36]. Electron-phonon coupling was introduced through the Su-Schrieffer-Heeger model, in which phonons modify the electron transfer integral t_{ij} , and periodic boundary conditions were used. Displacements of oxygen toward or away from Cu induce strong charge transfer between the O and Cu. The strength of charge transfer is conveniently expressed in terms of the Born effective charge, $Z^* = Z + Z(a/u)$, of oxygen, where Z is the ionic charge, ΔZ is the amount of charge transfer induced by phonon, u is the phonon amplitude, and a is the interatomic distance. As shown in Figure 7, the change in Z^* due to doping reaches about 2 at $h = 0.25$ for $x = 0.25$, almost canceling the intrinsic charge of oxygen. The accumulated charge transfer within the phonon wave packet creates a local hole wave packet. The value of Q at the maximum in Z^* ($h = 0.25$ for $x = 0.25$) corresponds to a state with about one hole per wavelength. An $h = 0.25$ wave packet covers two oxygen sites along one direction, or more likely, two Zhang-Rice sites [77]. When a local pair is formed, it involves four contiguous Zhang-Rice sites on a Cu square with 12 oxygen ions and 4 Cu ions.

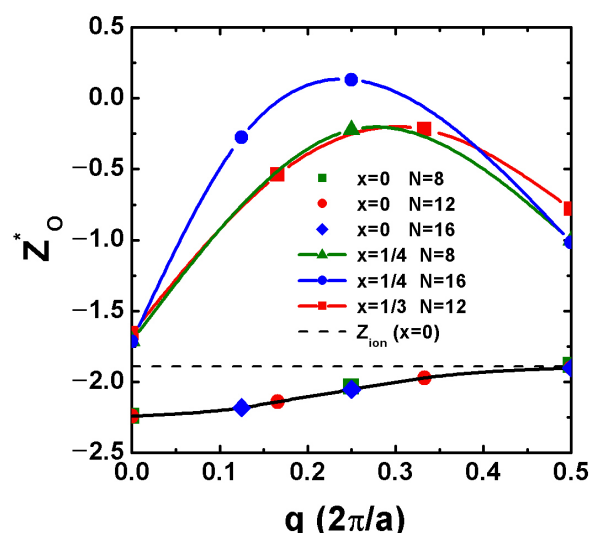


Figure 7. Born effective charge of oxygen, Z^* , of the two-band 1D Hubbard model [36]. x is the doping level, N is the number of atoms in the chain.

5. Electron Correlation and High-Temperature Superconductivity

Interestingly, even though the cuprates are strongly correlated electron systems, there have been very few discussions regarding the direct effects of electron correlation on the HTSC phenomenon. The effects of strong correlation have been discussed mostly in terms of the Hubbard U , and they enter the argument only indirectly. However, as the correlation energy is so large, even subtle effects can have very significant consequences. As the case for beryllium illustrates, electron dynamics significantly affect electron correlation; as such, massive changes in electron dynamics associated with BEC may have important effects.

To the best of our knowledge, only J. B. Goodenough has extensively discussed the possible direct effects of electron correlation on the HTSC [78]. He argued that changes in the Cu–O distance due to phonons modulate the polaronic band width, W , affecting the balance between W and the correlation energy U . As a result, the phonon wave-packet produces a correlation bag, and binds a pair of electrons. This argument suggests that strong electron correlation can enhance the polaronic pairing force.

As discussed earlier, in regular BCS superconductors, Cooper pairs strongly overlap with each other, and the real-space correlations among Cooper pairs are irrelevant to superconductivity. In the HTSC cuprates, however, Cooper pairs are most likely localized in space, forming vibrons. In such a case, our observation on superfluid He becomes relevant. It is possible that in the cuprates Cooper pairs become further apart upon BE condensation, thus reducing Coulomb repulsion energy. This energy reduction adds to the driving force for BEC [65,79]. As the repulsion energy, such as the Hubbard U , is so large, on the order of 10 eV, even a small change in the repulsion energy would amount to a significant driving force. Although it is not likely that the value of U actually changes upon BEC, a slight change in the p - d hybridization would result in the same effect.

It is difficult to estimate the magnitude of the reduction in the repulsion energy. In a very crude model, we may assume Cooper pairs form a 2D hexagonal lattice to estimate the average distance between them. For a hole density of $x = 0.2$, the average distance between the Cooper pairs is 13.25 Å. In a simple point-charge model, for six neighbors, this translates to a Coulomb repulsion energy of 0.13 eV even when assuming a fairly strong dielectric constant of $\epsilon = 50\epsilon_0$ [80]. If the distance changes by 10% upon BEC as in ^4He , the change in the repulsion energy is $-13 \text{ meV} = -151 \text{ K}$. This crude estimate demonstrates that the order of the magnitude of the effect may be comparable to T_c . Thus, the reduction in repulsion energy upon BEC can have a significant impact on T_c .

This scenario is also consistent with the T_c diagram shown in Figure 6c. As x increases, the average distance between the Cooper pairs in 2D decreases as $\sim 1/(x - x_c)^{\frac{1}{2}}$, where x_c is the phase boundary between the AFM phase and the SC phase. Thus, the gain in the repulsion energy will be $\sim (x - x_c)^{\frac{1}{2}}$, which is consistent with the increase in T_c with x . Above the optimum concentration, T^* decreases, and so does T_c . Furthermore, if the pairing temperature determines T_c , it should decrease with T^* beyond the optimum concentration. The fact that it does not suggests the existence of an additional driving force for T_c other than pairing, such as the reduction in Coulomb energy due to BEC.

6. Conclusions

Right after the discovery of the HTSC, many theories were proposed to explain this remarkable phenomenon. Most of these are “spin-only” theories, as it did not appear that the BCS or the phonon-based strong coupling theory could explain the phenomenon. It was recognized right away that the strong electron correlation in Cu is key to this mechanism, and researchers focused on the Hubbard model or the t - J Hamiltonian. However, due to the intrinsic difficulties with the many-body nature of the Hubbard model, progress has been slow. In the meantime, evidence of strong lattice effects and phonon involvement has accumulated. To save space, I have summarized only those observations made by my group. It appears that many phonons form a wave-packet to trap a hole in the form of a vibron, assisted by spins to localize holes, thus creating real-space hole pairs.

If pairing occurs in real space, our earlier observation on superfluid He becomes relevant. Using inelastic neutron scattering and energy-resolved dynamic PDF analysis, we found that the distance between BE-condensed He atoms is 10% longer than that between non-condensed atoms. In BECs, atoms are in the ground state with no momentum and thus have a lower probability of colliding with each other, increasing the interatomic distance. If the same situation applies to the Cooper pairs in the cuprates, the increase in the pair-pair distance upon BEC reduces the Coulomb repulsion between the pairs and adds to the driving force for the HTSC. In this case, HTSC is achieved, not due to a strong pairing force but rather by the increased BE condensation temperature.

At the moment, this scenario remains pure speculation without direct experimental support. However, for a long time, researchers have focused on pairing, whereas BEC for an ideal gas has been assumed without scrutiny. I suggest that it is time to reconsider the direct effect of strong electron repulsion on BE condensation of real-space Cooper pairs.

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Conflicts of Interest: The author declares no conflicts of interest.

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