Novel Isotope Effects on the Pairing Pseudogap in High-$T_c$ Cuprates: Evidences for Polaronic Metal and Precursor BCS-Like Pairing of Large Polarons

S. Dzhumanov, O.K. Ganiev, and Sh.S. Dzumanov
Institute of Nuclear Physics, 100214, Ulughbek, Tashkent, Uzbekistan

We have studied the novel isotope effects on the pairing pseudogap in underdoped and optimally doped cuprates within the large-polaron model and two non-standard BCS-like approaches. We have shown that in the intermediate-coupling regime the precursor pairing of large polarons occurs at a mean-field temperature $T^* > T_c$ and the near-absent, sizable and very large oxygen and copper isotope effects on $T^*$ exist in cuprates with small and large Fermi surfaces. Our results for $T^*$, isotope shifts and exponents in slightly underdoped and optimally doped cuprates are in quantitative agreement with existing experiments and explain the discrepancy between various experiments.

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Superconductivity in ordinary and polaronic metals is the result of two distinct quantum phenomena (as argued in Refs. [5–8]): pairing of charge carriers at a characteristic temperature $T^*$ and condensation of bound pairs into a superfluid state at the superconducting (SC) transition temperature $T_c$. In conventional superconductors, pairing of electrons and superfluidity of Cooper pairs occur simultaneously at $T^* = T_c$. In high-$T_c$ cuprates, however, the pairing of charge carriers may occur at a higher temperature $T^*$ than the $T_c$ at which the pre-formed Cooper pairs condense into a superfluid (or SC) state [9–13]. As a consequence, below $T^*$ a pseudogap (PG) appears in cuprates, as seen in many experiments (NMR, ARPES, optical conductivity, specific heat, etc.) (see Refs. [6–8] for a review). Most of the theories proposed for the high-$T_c$ cuprates are based on different electronic mechanisms of pairing and ignore effects arising from the electron-phonon interactions. However, a number of experiments revealing the peculiar isotope effects on $T_c$, $T^*$ and other physical quantities in cuprates [9–13] have shown that the electron-phonon interactions play a major role in these materials. In particular, some experiments showed that the oxygen and copper isotope effects on $T^*$ in Y- and La-based cuprates are absent or very small [9–13] and sizable [9–13, 14]. While other experiments revealed the presence of large negative oxygen and copper isotope effects on $T^*$ in Ho-based cuprates [11, 13]. The origin of the novel isotope effects on $T^*$ in cuprates has especially been the subject of controversy [9, 11]. Although some theories [10, 15] were used to describe the oxygen isotope effect on $T^*$ in high-$T_c$ cuprates, they cannot give an answer to the puzzling question why such an isotope effect is sizable or very large in some cuprates and absent in others. Also, there is no explanation of the observed copper isotope effect on $T^*$ in cuprates.

In this Letter, we address the above issues of the isotope effects on the pairing PG in the cuprates by proposing two BCS-based approaches extended to the intermediate-coupling regime, which allow to describe the precursor pairing of large polarons above $T_c$ in underdoped and optimally doped cuprates with small and large Fermi surfaces. We derive new expressions for the mean-field pairing temperature $T^*$ and the isotope exponents $\alpha_T$. Remarkably, our results for $T^*$, isotope shifts and exponents in slightly underdoped and optimally doped cuprates are in quantitative agreement with existing experiments and resolve the controversy between various experiments. We also make quantitative predictions for the isotope effects on $T^*$ in the deeply underdoped cuprates.

In conventional superconductors the mass of free electrons is independent of the ionic mass $M$. In contrast, the charge carriers in polar materials are self-trapped and form large polarons (quasiparticles dressed by lattice distortions) [19]. In the polaronic model, the mass of polarons $m_p$ depends on the longitudinal-optical (LO) phonon-frequency $\omega_{LO}$ which in turn depends on $M$. The observed effective mass of charge carriers in cuprates is about $2m_e$ (where $m_e$ is the free electron mass) [20] and the relatively small binding energies $E_p = 0.06 eV$ and $E_p = 0.12 eV$ of the polaron are observed in optimally doped and underdoped cuprates, respectively [21]. These and other experiments [20, 22] prove that the charge carriers in cuprates are large polarons.

Here we consider the real physical situations in cuprates, namely, the cases of small and large Fermi surfaces. In slightly underdoped and optimally doped cuprates with large Fermi surfaces, the new situation arises when the polaronic effect exists and the attractive interaction mechanism (e.g., due to exchange of static and dynamic phonons) between the carriers operating in the energy range $\{- E_p - \hbar \omega_{LO}, (E_p + \hbar \omega_{LO})\}$ is more effective than in the simple BCS picture. The energies $E$ of polarons are measured from the polaronic Fermi energy $E_F$. It was argued [10, 19] (see also Ref. [23]) that the extension of the BCS theory to the intermediate coupling regime describes the precursor pairing of carriers above $T_c$. At $\varepsilon_F > E_p + \hbar \omega_{LO} >> k_B T^*$, using a similar BCS-based approach, we obtain a new and more general...
expression for the mean-field pairing temperature $T^*$:

$$k_B T^* = 1.14 (E_p + \hbar \omega_{LO}) \exp[-1/\lambda^*],$$  \hspace{1cm} (1)

where $\lambda^* = \lambda_{p}(1 + \mu^{-1/4}) - \lambda_c (1 + \mu^{-1/4})/U_c(\mu), \quad U_c(\mu) = 1 + \lambda_c (1 + \mu^{-1/4})\ln B_c(\mu), \quad \lambda_{ph} = [2m^* \hbar^2 (3\pi^2 n)^{2/3}] V_{ph}, \quad \lambda_c = [2m^* \hbar^2 (3\pi^2 n)^{2/3}] V_c, \quad B_c(\mu) = \zeta_c/A\mu^{-1/4} (1 + a\mu^{-1/4}), \quad A = \frac{c^2}{\tau} \sqrt{\frac{\mu}{2\hbar}} (2\beta)^{1/4}, \quad a = \hbar \xi \frac{2n}{\mu} (2\beta)^{1/4}/c^2, \quad b = 1/6a, \quad \xi = \varepsilon/\varepsilon_0, \quad \mu = MM'/M+M'),$ is the reduced mass of ions.

Equations (4) and (6) allow us to calculate the PG temperature $T^*$ and the exponents $\alpha_T^*$ and $\alpha_F^*$ of the oxygen and copper isotope effects on $T^*$. In our numerical calculations, we take $m^* \simeq m_e$ \textsuperscript{20}, $\varepsilon_\infty = 3 - 5 \textsuperscript{20}, \varepsilon_0 = 22 - 30 \textsuperscript{20, 24}$ and $\hbar \omega_{LO} = 0.04 - 0.07 eV \textsuperscript{17, 20}$, typical values for the cuprates. Then the values of $\xi$ and $\alpha_F$ are $\xi \simeq 3.33 - 6.47$ and $\alpha_F = 2.15 - 5.54$ (which correspond to the intermediate electron-phonon coupling regime). Notice that in discussing the experimental data for isotope effects on $T^*$ in cuprates we have taken the fact that the physical situations (doping levels, dielectric constants, $T^*$) in various experiments are rather different. The magnitude of $\beta$ is kept at the value estimated for the oxygen and copper unsubstituted compound using a given value of $\hbar \omega_{LO}$. Since the quantity $\zeta_c$ is of the order of $\varepsilon_F$, the logarithm in $B_c(\mu)$ will be small, so that the Coulomb pseudopotential $V_c$ is of the order of bare Coulomb potential $V_c$. The results of numerical calculations of $T^*$ and $\alpha_T^*$ according to Eqs. (5) and (6) at different values of $\hbar \omega_{LO}$, $\varepsilon$, $n$, $\lambda_{ph}$ and $\lambda_c$ are shown in Figs.1 - 3.

Our results provide a consistent picture of the existence of crossover temperature $T^*$ above $T_c$ and peculiar isotope effects on $T^*$ in cuprates. They explain why the small positive (see Fig.1) and very large negative (see Figs.2 and 3) oxygen isotope effects and the large negative and near-absent copper isotope effects on $T^*$ are observed in various experiments. The obtained $T^*$ is plotted in the insets of Figs. 1 - 3 as a function of $\varepsilon$ and $n$ for different values of $\lambda_{ph}$ and $\lambda_c$. The values of $\lambda^*$ varies from 0.3 to 0.5 and $T^*$ increases with decreasing $n$. We have verified that $T^*$ decreases with in-
increasing $\tilde{\varepsilon}$ for $\lambda_{ph} < 0.35$ and $\lambda_c < 0.1$ (Fig.1). In contrast, $T^*$ increase with increasing $\tilde{\varepsilon}$ for $\lambda_{ph} > 0.4$ and $\lambda_c > 0.25$ (Fig.2). The existing experimental data on $T^*$ and $\alpha_{T^*}$ could be fitted with an excellent agreement using Eqs. (1) and (3), and adjusting the parameters $\hbar \omega_{LO}$, $\tilde{\varepsilon}$, $n$, $\lambda_{ph}$ and $\lambda_c$ for each cuprate superconductor. If we choose $\hbar \omega_{LO} = 0.05eV$, $n = 0.92 \cdot 10^{21} cm^{-3}$, $\tilde{\varepsilon} = 4.841 - 5.045$, $\lambda_{ph} \simeq 0.297$ and $\lambda_c \simeq 0.077$, we see that $T^* = 150 - 161 K$ and $\alpha_{T^*}$ is very small (i.e., $\alpha_{T^*} = (0.0058 - 0.0098) < 0.01$), which are consistent with the experimental data of Ref. [9]. Further, using other sets of parameters $n = 0.94 \cdot 10^{21} cm^{-3}$, $\tilde{\varepsilon} = 5.904 - 6.119$, $\lambda_{ph} = 0.311 - 0.313$ and $\lambda_c = 0.067 - 0.070$, we obtain $T^* \approx 150 K$ and $\alpha_{T^*} \approx 0.0525 - 0.0623$, which are in good agreement with the measured values: $T^* = 150 K$ and $\alpha_{T^*} = 0.052 - 0.061$ for YBa$_2$Cu$_4$O$_8$ (with $T_c = 81 K$) [10]. Figure 1 illustrates the predicted behaviors of $\alpha_{T^*}$ and $T^*$ as a function of $\tilde{\varepsilon}$ for $\lambda_{ph} < 0.4$ and $\lambda_c < 0.1$. We see that $\alpha_{T^*}$ decreases slowly with decreasing $\tilde{\varepsilon}$. Relatively strong electron-phonon and Coulomb interactions (i.e., $\lambda_{ph} > 0.5$ and $\lambda_c > 0.5$) change the picture significantly and cause $\alpha_{T^*}$ to decrease rapidly with decreasing $\tilde{\varepsilon}$. In this case the value of $\alpha_{T^*}$ is negative and becomes very large negative with decreasing $\tilde{\varepsilon}$ or $T^*$. The pictures shown in Figs. 2 and 3 are likely realized in some cuprates (which exhibit a large negative isotope exponent $\alpha_{T^*}$) and explain another important puzzle of the cuprates [11]: the huge oxygen-isotope effect on $T^*$ observed in HoBa$_2$Cu$_4$O$_8$, whose characteristic PG temperature $T^*$ increases significantly upon replacing $^{16}$O by $^{18}$O. Indeed, with fitting parameters, $n = 0.88 \cdot 10^{21} cm^{-3}$, $\tilde{\varepsilon} = 5.088$, $\lambda_{ph} = 0.975$ and $\lambda_c = 0.82$, one can ex-
plain the observed experimental data of Ref. [11]. In this case, we obtain \( T^\ast \{O^{18}\} \simeq 170.2K, T^\ast \{O^{16}\} \simeq 220.2K, \Delta T^\ast \simeq T^\ast \{O^{18}\} - T^\ast \{O^{16}\} \simeq 50K \) and \( \alpha_T^\ast \simeq -2.54 \), which are in remarkably good agreement with the experimental data \( T^\ast \{O^{18}\} \simeq 170K, T^\ast \{O^{18}\} \simeq 220K, \Delta T^\ast \simeq 50K \) and \( \alpha_T^\ast \simeq -2.2 \pm 0.6 \) [12]. The above predicted behaviors of \( T^\ast \) and \( \alpha_T^\ast \) could be checked experimentally in other slightly underdoped and optimally doped cuprates. We have also performed similar calculations for the copper isotope effect on \( T^\ast \) in slightly underdoped HoBa\(_2\)Cu\(_2\)O\(_4\) (where the electron-phonon and Coulomb interactions seem to be much stronger than in YBa\(_2\)Cu\(_3\)O\(_7\)).

For the orthorhombic La\(_2\)Sr\(_2\)Cu\(_3\)O\(_7\) (the precursor pairing of large polarons above \( T_c \)) and the peculiar isotope effects on the PG temperature \( T^\ast \) in high-\( T_c \) cuprates with small and large Fermi surfaces. Our results show that the oxygen isotope effect on \( T^\ast \) in optimally doped cuprates with moderate electron-phonon coupling strength \( \lambda_{ph} < 0.4 \) and weak Coulomb repulsion \( \lambda_c < 0.1 \) is small positive. At \( \lambda_{ph} > 0.5 \) and \( \lambda_c < 0.5 \) the negative oxygen and copper isotope effects on \( T^\ast \) in the same compound \( \lambda_{ph} \approx 0.7, \lambda_c \approx 0.8 \) and \( \lambda_{ph} > 0.5 \), respectively. The existing experimental data and discrepancies between experiments measuring the isotope effects on \( T^\ast \) in various cuprates are consistently explained by the proposed general BCS-like pairing model. We predict that the isotope effects on \( T^\ast \) in the deeply underdoped cuprates are sizable and negative.

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\[ \frac{1}{4}(1 + \frac{\lambda}{2\pi})(1 + b_{ph}^{1/4}) \left( 1 - \frac{1}{\lambda^s(\mu)} \right). \]

where \( \lambda^s(\mu) = (\lambda_{ph} - \lambda_c)(1 + b_{ph}^{1/4}) \).

The dependences \( \alpha_T^{\ast}(\varepsilon) \) and \( T^\ast(\varepsilon) \) for \( \hbar \omega_{LO} = 0.05eV, \lambda_{ph} - \lambda_c = 0.3 \) and \( n = 0.6 \cdot 10^{21}cm^{-3} \) are presented in Fig.4. As seen from Fig.4, the \( \alpha_T^{\ast} \) is rather small and negative. The \( \alpha_T^{\ast} \) is also negative and nearly four times smaller than \( \alpha_T^{\ast} \).

In conclusion, we have developed two new BCS-based approaches extended to the intermediate coupling regime to describe the precursor pairing of large polarons above \( T_c \) and the peculiar isotope effects on the PG temperature \( T^\ast \) in high-\( T_c \) cuprates with small and large Fermi surfaces. Our results show that the oxygen isotope effect on \( T^\ast \) in optimally doped cuprates with moderate electron-phonon coupling strength \( \lambda_{ph} < 0.4 \) and weak Coulomb repulsion \( \lambda_c < 0.1 \) is small positive. At \( \lambda_{ph} > 0.5 \) and \( \lambda_c < 0.5 \) the negative oxygen and copper isotope effects on \( T^\ast \) in the same compound \( \lambda_{ph} \approx 0.7, \lambda_c \approx 0.8 \) and \( \lambda_{ph} > 0.5 \), respectively. The existing experimental data and discrepancies between experiments measuring the isotope effects on \( T^\ast \) in various cuprates are consistently explained by the proposed general BCS-like pairing model. We predict that the isotope effects on \( T^\ast \) in the deeply underdoped cuprates are sizable and negative.

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