Comment on "Experimental and Theoretical Constraints of Bipolaronic Superconductivity in High T_c Materials: An Impossibility"

Recently Chakraverty *et al.* [1] raised objections to the bipolaron model of superconducting cuprates [2]. They claimed that (a) Bose-Einstein condensation (BEC) is not realized for any preformed pairs because this scenario cannot account for the observed value of T_c, (b) bipolarons are ruled out as superconducting pairs because they are too heavy, (c) the coherence length in cuprates is such that pairs are strongly overlapped, and (d) the bipolaron theory is incompatible with photoemission experiments. Here I show that their objections are the result of an incorrect approximation for the bipolaron energy spectrum and misuse of our theory.

It is commonly accepted that new superconductors lie close to BEC. We can now assess how close. The bipolaron energy spectrum has been derived for perovskites [3]. It consists of two energy bands $E_{\mathbf{k}}^{x,y} = 2[t\cos(k_{x,y}a) + t'\cos(k_{y,x}a) + t_\perp \cos(k_z d)]$, where a and d are the lattice constants, and t, t' ($\simeq -t/4$) and $t_\perp$ ($\ll t$) the bipolaron hopping integrals. By expressing the band-structure parameters through the in-plane and out-of-plane penetration depths $\lambda_{ab} = [m_x m_y c^2/8\pi n_B e^2(m_x + m_y)]^{1/2}$, $\lambda_c = [m_c c^2/16\pi n_B e^2]^{1/2}$, one readily obtains T_c from the density sum rule [2],

$$k_B T_c = \frac{dc^2\hbar^2}{20\lambda_{ab}^2 e^2[1 + \ln(16\pi n_B k_B T_c e^2 \lambda_c^2 d^2/\hbar^2 c^2)]}. \tag{1}$$

Here $m_x = \hbar^2/2ta^2$, $m_y = 4m_x$, and $m_c = \hbar^2/2|t_\perp|d^2$. This expression provides an unambiguous parameter-free estimate of T_c. In particular, we obtain $T_c \simeq 100$ K for $YBa_2Cu_3O_7$ ($\lambda_{ab} = 1600$ Å [1], $\lambda_c = 12\,600$ Å [4], and the bipolaron density $n_B = 6 \times 10^{21}$ cm^{-3}) assuming that all $2x$ holes are bound into bipolarons in the doped Mott insulator $YBa_2Cu_3O_{6+x}$. The measured value is $T_c = 92$ K [4]. Hence, the consideration, which takes into account the multiband energy structure clearly indicates that cuprates are in the BEC regime, while the erroneous approximation [1] yields T_c about 3 times higher.

We now check to what extent our expression for the bipolaron hopping integral, $t = 0.25 t_{\mathrm{band}} e^{-g^2}$ [3] corresponds to the value of the penetration depth. Here $g^2 \equiv \gamma\alpha^2$ with $\gamma = \sum_{\mathbf{q}} \gamma^2(\mathbf{q})[1 - \cos(q_x a)]/\sum_{\mathbf{q}} \gamma^2(\mathbf{q})$ and $\alpha^2 = \epsilon_p/\hbar\omega$. A strong interaction of carriers with c-axis longitudinal phonons (the frequency $\hbar\omega \simeq 74$ meV) has been established experimentally in $YBa_2Cu_3O_{6+x}$ while the coupling with other phonons is relatively weak [5]. Because of a low c-axis conductivity this interaction is the Fröhlich unscreened interaction with a $\mathbf{q}$-dependent matrix element $\gamma(\mathbf{q}) \sim q_z/q^2$ [3], which yields $\gamma = 0.162$. As a result, one obtains $m_x = 12m_e$ with the same polaron level shift $\epsilon_p = 250$ meV and bare hopping $t_{\mathrm{band}} = 150$ meV as in Ref. [1]. This bipolaron mass is close to $m_x \simeq 14m_e$ in $YBa_2Cu_3O_7$ obtained from the experimental λ_{ab}. It is also close to $m^* \simeq 13m_e$ found in those optical experiments [6], which distinguish between incoherent and Drude contributions. Mott and I always emphasized that small bipolarons in cuprates are *intersite* pairs [2] as a result of *unscreened* interaction [3]. By considering *on-site* bipolarons or by leaving out the coefficient γ in our expression for intersite bipolaron hopping Chakraverty *et al.* misuse the theory and overestimate the small bipolaron mass by *2 orders* of magnitude.

The coherence length of the charged Bose gas has nothing to do with the size of a boson. It can be as large as in the BCS superconductor [2]. Hence, one cannot distinguish the BCS and BEC by arguing (c) that the coherence area is large enough to accommodate many holes. What is really conclusive is the critical behavior. All experiments so far unambiguously show that the upper critical field and the specific heat near the transition are those of charged bosons [2]. The claim (d) that with bipolarons "one should expect essentially $\mathbf{k}$-independent spectral functions showing a $\mathbf{k}$-independent gap" [1] is not correct either. When an electron is emitted from the sample, the resulting hole propagates in the polaron band. It is the dispersion of this band which is measured by angle-resolved photoemission spectroscopy. The normal state gap is also $\mathbf{k}$ dependent owing to the polaron and bipolaron band dispersion. In fact, the bipolaron theory describes all major features of the tunneling and photoemission spectra [7].

To the best of our knowledge there are no unambiguous experimental facts so far which are qualitatively in disagreement with the bipolaron theory. Of course, new facts may disagree. What is clear, however, is that any theory, beautiful or not, cannot be destroyed by "ugly" artifacts as those in Ref. [1].

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