Comments on “Translation-invariant bipolarons and the problem of high-temperature superconductivity”

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Abstract
We comment on the recent results of Refs. [1, 2] on the bipolaron problem derived using an approximation of Gross – Tulub. It is proved that, contrary to the claim made in Refs. [1, 2], the bipolaron ground state energy calculated there in the strong-coupling approximation has not been shown to constitute a variational upper bound.

Keywords:
Bipolarons, Polaron, Fröhlich Hamiltonian

1. Introduction

Bipolarons play a role in the study, e.g., of electronic and magnetic properties of polar solids [3, 4, 5, 6, 7, 8, 10, 11, 12, 13]. Bipolarons have, e.g., been invoked in studies of high-$T_c$ superconductivity [9, 10]. It was shown in [6, 7] (see also Ref. [3]) that bipolarons can exist only in a highly restricted stability-region of the $(U, \alpha)$ plane (where $\alpha$ is the electron-phonon coupling constant and $U$ is the strength of the Coulomb repulsion) and that some of the high-$T_c$ oxides belong to this stability region, whereas conventional polaron-materials (alkali-halides, Silver-halides etc...) have $(U, \alpha)$ parameters far away from the bipolaron stability region. Bipolarons are only stable at sufficiently large $\alpha$, so that intermediate and strong coupling are relevant for the present discussion. Importantly for the study of high-$T_c$ superconductivity, it was found [6, 7] that a bipolaron binds more easily in 2D than in 3D and that the average pair-radius is a few Angstroms. The groundstate properties of bipolarons were studied further, e.g., in Refs. [14, 15].

Recently, the large-bipolaron problem was approached in Refs. [1, 2] using the Gross – Tulub (GT) approximation [14, 15] in the strong-coupling limit. Surprisingly low upper bounds to the bipolaron groundstate energy were arrived at in [2]. In the limiting case for the ratio of the dielectric constants $\eta \equiv \varepsilon_\infty/\varepsilon_0 = 0$ and the Fröhlich coupling constant $\alpha \gg 1$, the bipolaron groundstate energy proposed as an upper bound in [2] is $\varepsilon_{bip}(\alpha \gg 1, \eta = 0) \approx -0.414125\alpha^2$, significantly lower than any other result in the literature.

In the present communication we analyse the method and the results of Refs. [1, 2] and we demonstrate that the approximation for the bipolaron strong coupling groundstate energy arrived at in [2] has not been shown – contrary to the claim in [2] – to constitute an upper bound.

2. General treatment in Refs. [1, 2]

Consider the two-polaron (bipolaron) system with the Hamiltonian
\begin{align}
\hat{H} &= \frac{\hbar^2}{2m}\Delta_1 - \frac{\hbar^2}{2m}\Delta_2 + \frac{e^2}{\varepsilon_\infty |\mathbf{r}_1 - \mathbf{r}_2|} \\
&+ \sum_{\mathbf{k}} \hbar \omega_0 a_\mathbf{k}^\dagger a_\mathbf{k} + \sum_{\mathbf{k}} V_{\mathbf{k}} \left( a_\mathbf{k} + a_\mathbf{k}^\dagger \right) \left( e^{i \mathbf{k} \cdot \mathbf{r}_1} + e^{i \mathbf{k} \cdot \mathbf{r}_2} \right).
\end{align}

with the coupling parameters
\begin{align}
V_{\mathbf{k}} &= \frac{\hbar \omega_0}{k} \left( 2 \sqrt{2\pi m} \right)^{1/2} \left( \frac{\hbar}{m \omega_0} \right)^{1/4},
\end{align}

Here, $\alpha$ is the Fröhlich polaron coupling constant, $\varepsilon_\infty$ is the high-frequency dielectric constant. The system of units is chosen with $2m = 1$, $\hbar = 1$, and the LO-phonon frequency $\omega_0 = 1$. After the transformation to the center-of-mass and relative coordinates
\begin{align}
\mathbf{R} &= \frac{\mathbf{r}_1 + \mathbf{r}_2}{2}, \quad \mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2,
\end{align}
a first Lee-Low-Pines (LLP)-type canonical transformation
\begin{align}
\hat{S}_1 &= \exp \left( -i \mathbf{R} \cdot \sum_{\mathbf{k}} \mathbf{k} a_{\mathbf{k}}^\dagger a_{\mathbf{k}} \right),
\end{align}

and the averaging of the transformed Hamiltonian with a trial wave function $\varphi(r)$ for the relative motion (in the oscillatory form as in Refs. [1, 2]),
\begin{align}
\varphi(r) &= \frac{1}{(\pi \rho^2)^{3/4}} \exp \left( -\frac{r^2}{2\rho^2} \right)
\end{align}
we arrive at the reduced Hamiltonian

\[
H = \frac{1}{2} \sum_k k a_k^+ a_k + \sum_k a_k^+ a_k + \frac{4}{\sqrt{\pi}} \frac{\alpha}{\sqrt{1 - \eta}} \rho^2 (a_k + a_k^+) + \frac{4\alpha}{\sqrt{\pi} \sqrt{1 - \eta}} \rho^2.
\]  

(6)

The second canonical transformation

\[
S_2 = e^{-\frac{\sqrt{\rho}}{2} a_k (a_k^+)},
\]

with the trial phonon shifts \(f_k\) chosen as in Ref. [2] with the variational parameter \(\mu\),

\[
f_k = -4 \left( \frac{\pi \alpha}{k^2 V} \right)^{1/2} \exp \left( -\frac{1}{2 \mu} k^2 \right),
\]

and the generalized Bogoliubov transformation used in Ref. [15] result in the following variational functional for the bipolaron groundstate energy

\[
E_{bip} = E_R + \frac{3}{16} \sum_k k a_k^+ a_k + \frac{4}{\sqrt{\pi}} \frac{\alpha}{\sqrt{1 - \eta}} \rho^2
\]

\[
+ \frac{8 \sqrt{2\pi}}{\sqrt{\rho^2 + \frac{2}{\mu}}} \frac{1}{\sqrt{\rho^2 + \frac{2}{\mu}}} \Omega(\alpha,\mu)
\]

\[= \frac{3}{16} \sum_k k a_k^+ a_k + \frac{4}{\sqrt{\pi}} \frac{\alpha}{\sqrt{1 - \eta}} \rho^2,
\]

(9)

with the parameters

\[
\tilde{\alpha} = 4 \sqrt{2\alpha}, \quad a = \frac{2 \sqrt{2}}{\sqrt{\rho^2 + \frac{2}{\mu}}},
\]

and the recoil energy

\[
E_R = \frac{3}{16} \sum_k k a_k^+ a_k + [1 + \Omega(\tilde{\alpha}, \alpha)],
\]

(11)

which results from the recoil term \(\frac{3}{16} \sum_k k a_k^+ a_k\) in the Hamiltonian (6). The function \(Q(\alpha, a)\) is given by the integral expression [15]

\[
Q(\alpha, a) = \frac{2}{\sqrt{\pi}} \int_0^{\infty} \frac{e^{-y^2} \left[ 1 - \Omega(\gamma) \right] dy}{\left[ \frac{1}{y^2} + v(y) \right]^2 + \frac{2}{\pi} e^{-2y^2}},
\]

(12)

with the functions

\[
v(y) = 1 - ye^{-y^2} \int_0^y e^{-z^2} dz - \xi \int_y^{\infty} e^{-z^2} dz, \quad \Omega(\gamma) = 2\gamma^3 \left[ 1 + 2\gamma^2 \right] \int_y^{\infty} e^{-z^2} dz - y,
\]

(13)

(14)

and the parameters

\[
\lambda = \frac{4\alpha}{3 \sqrt{2\pi} a}, \quad \xi = \sqrt{y^2 + \frac{4}{a^2}},
\]

(15)

3. Using the approximation of Ref. [15] in Ref. [2]

In Ref. [15], the function \(Q(\alpha, a)\) [denoted in [15] as \(q(1/\lambda)\)] has been replaced by the value \(Q_{\infty} \approx 5.75\). If the recoil energy (11) is calculated using \(Q(\alpha, a) = Q_{\infty}\), one arrives at the result of Ref. [2] for the bipolaron energy (Eq. (15) of Ref. [2]). However, this approximation does not guarantee a variational upper bound for the polaron and bipolaron groundstate energies for the following reason.

For finite parameters \(\alpha\) and \(a\), \(Q(\alpha, a)\) is an increasing function of both \(\alpha\) and \(a\), as shown in Fig. 1. Also in the strong-coupling limit, when both \(\alpha\) and \(a\) tend to infinity, the asymptotic expression for this function,

\[
Q(\alpha, a)_{\infty} \approx 2 \sqrt{\frac{2 \sqrt{2}}{\pi} \alpha a},
\]

(16)

increases monotonically. The value \(Q_{\infty} \approx 5.75\) has been obtained in Ref. [15] assuming a finite cutoff for the phonon wave vectors. Indeed, the phonon wave vectors are restricted by a short-wavelength cutoff at the boundary of the Brillouin zone. Consequently, in the strong-coupling limit \(\alpha \to \infty\), the position of the steep maximum mentioned in Ref. [15] can lie beyond the integration range. However, as far as the calculation of the (bi)polaron energy is performed within the continuum approach, it is only consistent either to avoid a cutoff all together (in the continuum formalism), or to introduce a cutoff from the very beginning of the calculation. A consistent treatment of the polaron problem in the Gross – Tulub approximation using a cutoff was performed in Ref. [16]. It was found that a cutoff leads to the appearance of additional positive terms in the polaron recoil energy. These terms were not found in Ref. [15]. Being positive, they definitely lead to an increase of the groundstate energy. Therefore missing these terms can lead

\[\text{As written in Ref. [15] on p. 4, } “\text{It is of interest to note that as } \lambda \to \infty \text{ the integrand in (2.12) has a steep maximum at } y^2 = 3/4; \text{ however, if we take into account that the domain of integration over } y \text{ is in fact limited and if we use the values } g^2 = 10 \text{ considered in the following, this singularity does not arise.”}\]
to the violation of the variational principle. Hence the bipolaron groundstate energy arrived at in Refs. [1, 2] is incorrectly claimed to constitute an upper bound for the groundstate energy of the bipolaron.

4. Bipolaron variational energy in the continuum approach

Let us consistently consider the polaron and bipolaron groundstate energies within the continuum approximation using the complete recoil energy (i.e., without a cutoff). In the strong-coupling limit, using (9) with the asymptotic expression (16), the polaron and bipolaron groundstate energies are then found to be

\[ E_{\text{pol}}(\alpha) \approx -0.31683\alpha^{4/3}, \] (17)

\[ E_{\text{bip}}(\alpha \gg 1, \eta = 0) \approx -0.868509\alpha^{4/3}. \] (18)

Therefore, when the (bipolaron) groundstate energy is consistently calculated within the continuum GT approach, an incorrect dependence \( E \propto \alpha^{4/3} \) results in the strong-coupling limit. This problem, however, was not realised by the author of Refs. [1, 2].

5. Conclusion

In the present communication we have proved that the strong-coupling expression for the bipolaron groundstate energy calculated in Refs. [1, 2] contrary to what is claimed in those works is not justified as upper bound for the bipolaron groundstate energy. A contribution to the recoil energy due to a momentum cutoff (16) within the GT scheme is missed in [1, 2]. Thus the results of Refs. [1, 2] have been obtained using the incomplete recoil energy.

We have also worked out the GT scheme using the complete recoil energy within the continuum approach as is necessary for a consistent theory. This leads to an incorrect dependence \( E_{\text{bip}} \propto \alpha^{4/3} \) (instead of \( E_{\text{bip}} \propto \alpha^2 \)) for the polaron groundstate energy in the strong-coupling limit. A consistent calculation within the continuum approach, however, was not performed in Refs. [1, 2].

Note also that for intermediate \( \alpha \), the continuum GT method leads to a bipolaron groundstate energy higher than twice the groundstate energy for a single polaron. Therefore this method fails to describe a bound bipolaron state for any \( \alpha \).

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Figure 2: Bipolaron variational groundstate energy calculated using the complete recoil energy within the Gross–Tulub scheme in the continuum approach. For comparison, the bipolaron groundstate energy calculated in Ref. [6] and twice the groundstate energy of a single polaron from Ref. [13] are plotted in the same graph.

In order to check whether the continuum variational GT approach is adequate for intermediate \( \alpha \), we have calculated the bipolaron groundstate energy using the expression (9) with \( Q(\alpha, \alpha) \) given by (12) instead of the value \( Q_{\infty} \approx 5.75 \) used in Refs. [1, 2]. In Fig. 2, this bipolaron groundstate energy is compared with the variational result of Ref. [6] and with twice the energy of a single polaron calculated using the path-integral variational method of Feynman [17]. It is seen that for intermediate \( \alpha \), the bipolaron groundstate energy calculated using the variational GT scheme in the continuum approach lies above twice the energy of a single polaron.