Stability of Formation of Large Bipolaron: Nonrelativistic Quantum Field Theory

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Dedicated to Herbert Spohn on the occasion of his 60th birthday

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Abstract

We are concerned with the stability of formation of large bipolaron in a 3-dimensional (3D) crystal. This problem is considered in the framework of nonrelativistic quantum field theory. Thus, the Hamiltonian formalism, as Fröhlich introduced, is employed to describe the bipolaron. We approach the problem by characterizing some sufficient or necessary conditions for the bipolaron being stable. This paper gives a full detail of the author’s talks at ESI, RIMS, and St. Petersburg State Univ. in 2005.
I. INTRODUCTION

The question whether bipolarons can be formed has attracted ever a great deal of studies since Shafroth\textsuperscript{1} in particular showed bipolaronic superconductivity takes place when the temperature is below that of Bose-Einstein condensation. Over the past few decades there have been several renewals\textsuperscript{2,3,4,5} of interest in this problem from the point of view of high-temperature superconductor. In this paper, we will focus our mind on the stability of formation of large bipolaron.

We will treat two electrons coupled with longitudinal optical (LO) phonons in a 3-dimensional crystal. Generally, the electron-phonon interaction dresses an electron in a phonon cloud. This dressed electron is the so-called polaron. We assume the electron-phonon interaction is described by the second quantization, as Fröhlich\textsuperscript{6,7} introduced. If the Coulomb repulsion between the two electrons puts them so far away from each other that each electron dresses itself in an individual phonon cloud, there is no exchange of phonons between the two. Namely, there is the only Coulomb repulsion between them and thus only two separated single polarons are formed in the crystal. On the other hand, if the distance between the two electrons is so short that the phonon-exchange occurs, there is a possibility that attraction appears\textsuperscript{8} between them and therefore that they are bound to each other. The bound two polarons is called a bipolaron.

In the light of superconductivity, many studies have been performed for the large bipolaron.\textsuperscript{5,9,10,11,12} As for small polaron,\textsuperscript{13,14,15,16,17} Alexandrov and Kornilovich\textsuperscript{18} clarified the physical properties of the small Fröhlich polaron. Alexandrov and Ranninger\textsuperscript{3} showed the possibility that small bipolarons might be superconducting. Moreover, Alexandrov and Mott,\textsuperscript{19} and Alexandrov\textsuperscript{20} pointed out the mobility of the small bipolaron as well as that of the large bipolaron. Thus, we are also interested in how the size of large bipolaron grows.

Using the classical picture described in Ref.\textsuperscript{21}, we can explain the occurrence of attraction between the two electrons in the following. Since an electron has negative charge, a first electron leaves behind a deformation trail in the crystal lattice, which affects the positions of the ion cores. This trail is associated with an increased density of positive charge owing to the ion cores. Therefore, it has an attractive effect on a second electron. Namely, the lattice deformation causes the attraction between the first and second electrons. Thus, the problem is whether the attraction is enough to bind the two electrons.
Pekar\textsuperscript{22} showed that there is no formation of the large bipolaron for any optic dielectric constant $\epsilon_\infty$ and the static dielectric constant $\epsilon_0$ in 3D systems. Also Takada\textsuperscript{23} reached the same result in 3D systems from the weak-coupling limit by studying the Bethe-Salpeter equation. Obeying the classical theory of phonon described in Ref.\textsuperscript{24}, we briefly see that two large polarons cannot be bound to each other from the point of view of binding energy

$$2E_{\text{AM}}^{\text{SP}}(r) - E_{\text{AM}}^{\text{BP}}(r),$$

where $E_{\text{AM}}^{\text{SP}}(r)$ is the total energy of a large (single) polaron in a sphere of radius $r > 0$, and $E_{\text{AM}}^{\text{BP}}(r)$ the total energy of a large bipolaron in the sphere. Because the binding energy is always negative in classical theory. More precisely, with the help of expressions of $E_{\text{AM}}^{\text{SP}}(r)$ and $E_{\text{AM}}^{\text{BP}}(r)$ given in Ref.\textsuperscript{24}, we can easily derive the equation:

$$- (2E_{\text{AM}}^{\text{SP}}(r) - E_{\text{AM}}^{\text{BP}}(r)) = \sqrt{2} \alpha \left( \frac{1}{1 - \eta} - 1 \right) \frac{1}{r}$$

for $\alpha$ (the coupling constant of electrons and the phonon field) and $\eta := \epsilon_\infty/\epsilon_0$ (the ionicity of the crystal) under the natural units. So, we can say that the binding energy is negative in the classical theory because $0 < \eta < 1$. A similar form to RHS of Eq.\textsuperscript{(1.1)} will play an important role in Secs.IV, V, and VII (see Eqs.(4.7), (5.3), (5.7), (5.13), and (5.14)). Thus, the attraction derived in classical theory is too weak to bind the two polarons.

On the other hand, Vinetskii and Giterman\textsuperscript{25} found that the bipolaron is formed only for very small ionicities $\eta$. Emin\textsuperscript{5} also pointed out a possibility of formation of large bipolaron for sufficiently large coupling constants $\alpha$ in addition to small ionicities $\eta$. Our argument using Fröhlich’s interaction in this paper is based on many studies in quantum theory by prior literature\textsuperscript{9,10,11,12} to derive the effective attraction. Bassani et al.\textsuperscript{9} showed the large bipolaron formation for large $\alpha$ and small $\eta$ by estimating the binding energy. Verbist et al.\textsuperscript{10} showed the stable region of $\eta$ for the formation of the large bipolaron in the strong-coupling limit. It is worthy of note that the Feynman path-integral approach\textsuperscript{11,26} is also useful for the bipolaron problem.

In this paper, to derive an effective attraction in the same framework as in studies,\textsuperscript{9,10,11,12} we will adopt the notion of Feynman’s classical virtual phonon\textsuperscript{27} instead of the deformation trail in the crystal lattice. In addition, we will take the image of Peeters and Devreese’s classical bipolaron\textsuperscript{28} into our argument. In their theory of the classical bipolaron, the two electrons rotate in a circle around a common fixed center. We now denote the radius of the circle by $r_{\text{PD}}$. Then, the classical bipolaron has to be localized in the closed ball $\overline{B}(r_{\text{PD}})$ of radius $r_{\text{PD}}$ centered at the fixed point. The radius $r_{\text{PD}}$ should turn out long when the Coulomb
repulsion beats the attraction caused by the phonon field. Conversely, it should turn out short when the attraction wins over the Coulomb repulsion. So, if the attraction between the two electrons is so weak that the Coulomb repulsion makes the interior of the ball $B(r_{PD})$ completely contains the whole crystal, then we cannot hope that the classical bipolaron is formed in the crystal. One of our ideas is to take this image into nonrelativistic quantum field theory by reorganizing the way we have ever done.\textsuperscript{29} Thus, we have to introduce the notion of the size of the region in which the two electrons constructing bipolaron live. For the ground state energy $E_{BP}$ of bipolaron and the ground state energy $E_{SP}$ of single polaron, we introduce an energy $E(r)$ coming from the phonon field for every $r > 0$ as:

$$2E_{SP} - E_{BP} = E(r) - \frac{U}{r}.$$  \hspace{1cm} (1.2)

That is, we express the binding energy with the energy $E(r)$ and the Coulomb repulsive potential $U/r$. Here, $r$ stands for the distance between the two electrons now. Taking account of Emin’s work\textsuperscript{30} and Salje’s\textsuperscript{31} $E(r)$ is probably non-negative. Thus, main purpose in this paper is to estimate $E(r)$ so that the stability of formation of large bipolaron can be characterized in terms of the estimated $E(r)$ and also $\eta$.

In Sec.II we will introduce the Hamiltonian of the bipolaron which has the Fröhlich interaction. In Sec.III we will show a device to introduce a parameter $\theta \geq 0$ which controls the coupling strength between the regimes of the weak- and strong-coupling theories. To find this parameter, we will develop Lieb and Thomas’ method.\textsuperscript{32} In Sec.IV we will argue spatial localization\textsuperscript{29,33} of the relative motion of bipolaron in the weak-coupling regime by estimating $E(r)$. Especially, we will estimate the distance between the two electrons in the bipolaron from below by adopting Lieb’s idea\textsuperscript{34} into nonrelativistic quantum field theory. Thus, we will understand how the ionicity raises the size of the bipolaron in the weak-coupling regime. In Sec.V combining the notion of Feynman’s virtual phonon\textsuperscript{27} and the image of Peeters and Devreese’s classical bipolaron,\textsuperscript{28} we will derive two effective Hamiltonians in quantum theory from the original one. The effective Hamiltonians describe those in the strong-coupling regime. With the help of these effective Hamiltonians we will find a sufficient condition for the bipolaron formation in terms of $E(r)$ or $\eta$ and give a lower and an upper bounds to the ground state energy $E_{BP}$. In Sec.VI we will argue the spatial localization in the strong-coupling regime. In Sec.VII we will consider a sufficient condition for the positive binding energy.
The total energy of the bipolaron consisting of two electrons coupled with the LO phonons is described by the Hamiltonian $H_{\text{BP}}$:

$$H_{\text{BP}} = H_{\text{el-el}} + H_{\text{ph}} + H_{\text{el-ph}},$$

(2.1)

where $H_{\text{el-el}}$ is the energy of two electrons with the Coulomb repulsion between them, $H_{\text{ph}}$ the free energy of the phonon field, and $H_{\text{el-ph}}$ Fröhlich’s interaction derived through the second quantization of the Coulomb long-range interaction:

$$H_{\text{el-el}} = \sum_{j=1,2} \frac{1}{2m} p_j^2 + \frac{U}{|x_1 - x_2|},$$

(2.2)

$$H_{\text{ph}} = \sum_k \hbar \omega_k a_k^\dagger a_k,$$

(2.3)

$$H_{\text{el-ph}} = \sum_{j=1,2} \sum_k \left\{ V_k e^{i k \cdot x_j} a_k + V_k^* e^{-i k \cdot x_j} a_k^\dagger \right\}.$$  

(2.4)

In Eq.(2.2), the position and momentum operators of the $j$th electron ($j = 1, 2$) of mass $m$ are denoted by $x_j$ and $p_j$, respectively, so $p_j = -i\hbar \nabla x_j$. The strength of the Coulomb repulsion is designated by the symbol $U$, so $U \equiv e^2/\epsilon_\infty$ for the electric charge $e$ and the optic dielectric constant $\epsilon_\infty$. In Eq.(2.3), $a_k$ and $a_k^\dagger$ are the annihilation and creation operators, respectively, of the LO phonon with the momentum $\hbar k$. Then $a_k$ and $a_k^\dagger$ satisfy the canonical commutation relation, $[a_k, a_{\ell}^\dagger] = \delta_{k\ell}$, because phonons are bosons. The LO phonons can be assumed to be dispersionless, $\omega_k = \omega_{\text{LO}}$. In Eq.(2.4), $V_k$ is given by $V_k := -i\hbar \omega_{\text{LO}} (4\pi \alpha r_{\text{fp}}/k^2 V)^{1/2}$ for the crystal volume $V$ and the free polaron radius $r_{\text{fp}} \equiv (\hbar/2m\omega_{\text{LO}})^{1/2}$. The dimensionless electron-phonon coupling constant is given by

$$\alpha := \frac{1}{\hbar \omega_{\text{LO}}} \frac{e^2}{2} \left( \frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_0} \right) \frac{1}{r_{\text{fp}}},$$

(2.5)

where $\epsilon_0$ is the static dielectric constant.

Concerning the Hamiltonian $H_{\text{BP}}$ of bipolaron, we make some remarks. Since the ionicity $\eta$ of the crystal is defined by $\eta := \epsilon_\infty/\epsilon_0$, it satisfies $0 < \eta < 1$. In terms of $\eta$, the strength of the Coulomb repulsion is rewritten as $U = \sqrt{2} \alpha/(1 - \eta)$. Since we consider the two-body system of large polarons, the wave vector $k$ in $\sum_k$ runs over the first Brillouin zone. This fact makes some noticeable differences between phonon and photon (cf. Table 23.4 of Ref. 35). The primitive cell is usually given by the first Brillouin zone for a crystal. However,
it is not always to chose the first Brillouin zone for another solid. When a solid has metallic properties, we have to take the Fermi surface into account. In this case, we employ a reduced zone scheme.35

We use the natural units $\hbar = m = \omega_{\text{LO}} = 1$ from now on. Using the conversion38 of sums to integrals, we estimate $\sum_k |V_k|^2$ at $\sqrt{2} \alpha K/\pi$ as:

$$\sum_k |V_k|^2 \approx \frac{N}{V_c^*} \int_{|k| \leq K} d^3 k \frac{4 \pi \alpha r_{dp}}{V k^2} = \frac{V}{(2\pi)^3} \int_{|k| \leq K} d^3 k \frac{4 \pi \alpha r_{dp}}{V k^2} = \sqrt{2} \alpha K,$$

where $V_c^*$ is the volume of the primitive cell in the reciprocal lattice and $V_c^* = (2\pi)^3 V_c$ for $V_c$, the volume of the primitive cell in the direct lattice. We denoted by $N$ the number of primitive cells which is contained in the crystal volume $V$. So, when integral $\int d^3 k$ is over the first Brillouin zone, $K$ means the radius of a sphere of the first Brillouin zone. For example, in the Debye interpolation scheme35 for a harmonic crystal, $K$ is given by $k^3_D = (3/4\pi)(2\pi)^3 N/V = 6\pi^2 N/V$. This equation says, as is well known, there is the relation between $k_D$ and $k_F$ (the radius of the Fermi surface), i.e., $k_D = (2/Z)^{1/3} k_F$, where $Z$ is the nominal valence. When the integral $\int d^3 k$ can be extended from the first Brillouin zone to the whole $k$-space, $K$ plays the role of an ultraviolet (UV) cutoff.

Using the approximation35 of the Fourier expansion, $V/(4\pi |x|) \approx \sum_k e^{ik \cdot x}/k^2$, we obtain

$$\sum_k |V_k|^2 e^{ik \cdot x} \approx \frac{\alpha}{\sqrt{2} |x|}.$$

We often use this approximation (2.7) in this paper.

We also use the coordinate of the center-of-mass, $X_1 = (x_1 + x_2)/2$, and the coordinate of the relative motion, $X_2 = x_1 - x_2$, in this paper. Each momentum is given by $P_j = -i \nabla X_j$, $j = 1, 2$. So, we have $P_1 = p_1 + p_2 = M_1 X_1$ and $P_2 = (p_1 - p_2)/2 = M_2 X_2$, respectively, where we set the masses as $M_1 = 2$ and $M_2 = 1/2$. Then, $H_{\text{BP}}$ is unitary-equivalent to

$$\tilde{H}_{\text{BP}} = \tilde{H}_{\text{el-el}} + H_{\text{ph}} + \tilde{H}_{\text{el-ph}},$$

where

$$\tilde{H}_{\text{el-el}} = \sum_{j=1,2} \frac{1}{2M_j} P_j^2 + \frac{U}{|X_2|},$$

$$\tilde{H}_{\text{el-ph}} = \sum_k c(X_2, k) \left\{ V_k e^{ik \cdot X_1} a_k + V_k^* e^{-ik \cdot X_1} a_k^\dagger \right\},$$
with
\[ c(X_2, k) = 2 \cos \frac{k \cdot X_2}{2} = e^{ik \cdot X_2} + e^{-ik \cdot X_2}. \]

As for \( H_{\text{BP}} \), we note the following. The bipolaron Hamiltonian \( H_{\text{BP}} \) and the total momentum \( \Pi_{\text{tot}} = p_1 + p_2 + \sum_k k a_k^\dagger a_k \) are commutable, i.e., \([\Pi_{\text{tot}}, H_{\text{BP}}] = 0\), because of the translation invariance of \( H_{\text{BP}} \). We can show that \( H_{\text{BP}} \) has no ground state in the standard mathematical representation because of the continuous symmetry of the translation invariance.\(^{39}\) It is known in general that if a Hamiltonian \( H \) has a continuous symmetry (that is, \([H, \Pi] = 0\) for the generator \( \Pi \) of a transformation) and moreover \( H \) has a ground state, there are two possibilities: the transformation \( e^{it\Pi} \) might leave the ground state invariant for every real number \( t \), which is called manifest symmetry, or otherwise, it might map a ground state \( \Psi_0 \) to another ground state \( e^{it\Pi} \Psi_0 \), which is called hidden symmetry. In the latter case, we can show that there are infinitely degenerate ground states.\(^{39}\) Moreover, the Nambu-Goldstone theorem suggests that the spontaneous symmetry breaking would occur and then the Nambu-Goldstone bosons would appear. We conjecture that they appear as acoustic phonons in bipolaron, taking account of the result in Ref.\(^{40}\).

To avoid such a kind of situation coming from the continuous symmetry and thus to give a possibility that \( H_{\text{BP}} \) has a ground state, we can consider, for example, the case where we restrict the electrons’ movement into the crystal and the case where we nail down the center-of-mass of the two electrons at a point \( Q \). For both cases, we only have to employ
\[ H_{\text{el-ph}}(\rho) = \rho(x_1 + x_2) \sum_{j=1,2} \sum_k \left( V_k e^{ik \cdot x_j} a_k + V_k^* e^{-ik \cdot x_j} a_k^\dagger \right) \]
instead of \( H_{\text{el-ph}} \) in \( H_{\text{BP}} \), where \( \rho(x) \) is a function satisfying \( 0 \leq \rho(x) \leq 1 \). Namely, in those cases the total Hamiltonian is \( H_{\text{BP}} = H_{\text{el-el}} + H_{\text{ph}} + H_{\text{el-ph}}(\rho) \). In the former, \( \rho(x) \) is defined so that \( \rho(x) = 0 \) outside the crystal and \( \rho(x) = 1 \) inside the crystal. In the latter, \( \rho(x) \) is defined by \( \rho(x) \equiv \rho_Q(x) \), where \( \rho_Q(x) \) is 1 if \( x = 2Q \); \( \rho_Q(x) \) is 0 if \( x \neq 2Q \). Then, we can separate the center-of-mass motion from \( H_{\text{BP}} \) and thus obtain its relative motion. Taking \( \rho(x) = 1 \) for every \( x \) restores the Hamiltonian \( H_{\text{el-el}} + H_{\text{ph}} + H_{\text{el-ph}}(\rho) \) to the original \( H_{\text{BP}} \).

When we introduce \( \rho(x) \) into \( \tilde{H}_{\text{BP}} \), we employ \( \tilde{H}_{\text{el-ph}}(\rho) \) instead of \( \tilde{H}_{\text{el-ph}} \), where \( \tilde{H}_{\text{el-ph}}(\rho) \) is given by putting \( \rho(2X_1) \) in front of \( \sum_k \) in the definition of \( \tilde{H}_{\text{el-ph}} \). When we fix the center-of-mass at \( Q \), our target Hamiltonian is \( \tilde{H}_{\text{BP}}^{\text{rel}} = \tilde{H}_{\text{el-el}} + H_{\text{ph}} + \tilde{H}_{\text{el-ph}}(\rho_Q) \). As used in Ref.\(^{9}\), there is another method, which is given by fixing the total momentum \( \Pi_{\text{tot}} \) at a real number in the spectrum of \( \Pi_{\text{tot}} \). For example, in the case where \( Q \) is the origin \( O \), we can obtain the
Hamiltonian $\tilde{H}_{BP}^{\Pi=0} := e^{iP_{ph} \cdot X_1} \tilde{H}_{BP} e^{-iP_{ph} \cdot X_1} = P_2^2 / 2M_2 + U / |X_2| + H_{ph} + P_{ph}^2 / 2M_1 + \tilde{H}_{el-ph}(\rho_0)$, where $P_{ph} := \sum_k k a_k^\dagger a_k$.

III. WEAK- AND STRONG-COUPLING REGIMES

In this section, a parameter $\theta \geq 0$ is introduced to control the strength of the coupling constant $\alpha$ in the Hamiltonian of single polaron. We will use this parameter $\theta$ for the Hamiltonian of bipolaron as well as that of single polaron throughout this paper.

Let us define $E_{BP}$, the ground state energy of bipolaron, as that of $H_{BP}$, i.e., $E_{BP} := \inf \text{Spec}(H_{BP})$. We denote the energy spectrum of a Hamiltonian $H$ by $\text{Spec}(H)$. The ground state energy of $H$ always means $\inf \text{Spec}(H)$ throughout our argument.

We denote the ground state energy of single polaron by $E_{SP}$, i.e., $E_{SP} := \inf \text{Spec}(H_{SP}^{(j)})$, for the single-polaron Hamiltonian:

$$H_{SP}^{(j)} = \frac{1}{2} p_j^2 + \sum_k a_k^\dagger a_k + \sum_k \left\{ V_k e^{ikx_j} a_k + V_k^* e^{-ikx_j} a_k^\dagger \right\}, \quad j = 1, 2. \quad (3.1)$$

Through many studies we know there are two regimes with respect to powers of $\alpha$ in $E_{SP}$: $E_{SP} \propto -\alpha$ in the weak-coupling limit and $E_{SP} \propto -\alpha^2$ in the strong-coupling limit.

When the wave vector $k$ in $\sum_k$ runs over an infinite lattice, Lieb and Thomas controlled the UV cutoff $K$ by $\alpha$ rigorously to give a lower bound to $E_{SP}$ in the strong-coupling theory. But, since our $K$ is now the radius of the first Brillouin zone, we add a device to their idea. Using a unitary operator, we introduce the preceding parameter $\theta \geq 0$ to control the coupling strength between the regimes of the weak- and the strong-coupling theories. Namely, we represent the two regimes by $\theta$ in the following. We fix $\alpha$ in the weak-coupling theory first ($\theta = 0$). As we switch on $\theta$ ($\theta > 0$) and make it large, we have the electron-phonon coupling constant $\alpha_\theta := \alpha \theta$ in the strong-coupling theory and $\alpha_\theta$ increases. The method proposed here will be basically employed to obtain effective Hamiltonians for the bipolaron in Sec. V.

We define a canonical transformation with the generator $G(\theta) := i \theta \sum_k \{ V_k a_k - V_k^* a_k^\dagger \}$. Then, our unitary-transformed Hamiltonian is $H_{SP}^{(j)}(\theta) := e^{iG(\theta)} H_{SP}^{(j)} e^{-iG(\theta)}$. We note that $E_{SP}$ is the ground state energy of $H_{SP}^{(j)}(\theta)$ since $H_{SP}^{(j)}(\theta)$ is unitary-equivalent to $H_{SP}^{(j)}$. We show that $E_{SP}$ can be estimated in the following. It is easy to show that for $\alpha$ in the weak-coupling regime

$$-\frac{\sqrt{2} \alpha}{\pi} K \leq E_{SP} \leq 0. \quad (3.2)$$
We can show that \( E_{\text{sp}} \propto -\alpha \) for small coupling constants \( \alpha \) in the weak-coupling regime by obeying a perturbative method as well known or using a nonperturbative one such as in Theorem 2 of Ref. \[43\]. Meanwhile, it is shown in this section that for so large \( \theta \) that \( \alpha_\theta := \alpha \theta \) is in the strong-coupling regime, a lower and an upper bounds to Theorem 2 of Ref.\[43\]. Meanwhile, it is shown in this section that for so large \( \theta \) that \( \alpha_\theta := \alpha \theta \) is in the strong-coupling regime, a lower and an upper bounds to \( E_{\text{sp}} \) are given as:

\[
- c_{\text{sp}} \alpha_\theta^2 - \left| 1 - \frac{1}{\theta} \right| \sqrt{2} \alpha_\theta \frac{\pi}{K} \leq E_{\text{sp}} \leq - c_{\text{sp}} \alpha_\theta^2 + \theta \sqrt{2} \alpha_\theta \frac{\pi}{K},
\]

with the approximation \((2.4)\), where \( c_{\text{sp}} = 0.108513 \cdots \). We note that the estimate \((3.3)\) in the strong-coupling regime tends to the estimate \((3.2)\) in the weak-coupling one as \( \theta \) approaches 0.

To show the inequality \((3.3)\) we need some mathematical arguments. We now introduce a mathematical parameter \( R' > 0 \) into \( H_{\text{sp}}^{(j)} \) so that \( \lim_{R' \to \infty} H_{\text{sp}}^{(j)}(R') = H_{\text{sp}}^{(j)} \) (in the norm resolvent sense\[44\]) as follows:

\[
H_{\text{sp}}^{(j)}(R') := \frac{1}{2} p_j^2 + \sum_k a_k^\dagger a_k + \gamma_1(x_j) \sum_k \left\{ V_k e^{ikx_j} a_k + V_k^* e^{-ikx_j} a_k^\dagger \right\}, \quad j = 1, 2, \quad (3.4)
\]

where \( \gamma_1(x) \) is a smooth real-valued function which satisfies \( 0 \leq \gamma_1(x) \leq 1 \) and \( \gamma_1(x) = \gamma_1(-x) \) for every \( x \). The parameter \( R' > 0 \) is introduced so that \( \gamma_1(x) = 1 \) for \( |x| \leq R'/2 \); \( \gamma_1(x) = 0 \) for \( |x| \geq R' \). We denote by \( E_{\text{sp}}(R') \) the ground state energy of \( H_{\text{sp}}^{(j)}(R') \), i.e., \( E_{\text{sp}}(R') := \inf \text{Spec}(H_{\text{sp}}^{(j)}(R')) \). Then, it automatically follows that \( \lim_{R' \to \infty} E_{\text{sp}}(R') = E_{\text{sp}} \).

In the same way as for \( H_{\text{sp}}^{(j)}(\theta) \) we define \( H_{\text{sp}}^{(j)}(\theta, R') \) by \( H_{\text{sp}}^{(j)}(\theta, R') := e^{iG(\theta)} H_{\text{sp}}^{(j)}(R') e^{-iG(\theta)} \). Then, through the approximation \((2.4)\) we approximate \( H_{\text{sp}}^{(j)}(\theta, R') \) as:

\[
H_{\text{sp}}^{(j)}(\theta, R') \approx \frac{1}{2} p_j^2 - 2\theta \gamma_1(x_j) \sum_k |V_k|^2 e^{ikx_j} + H_{\text{ph}} + \sum_k \left\{ V_k \left( \gamma_1(x_j) e^{ikx_j} - \theta \right) a_k + V_k^* \left( \gamma_1(x_j) e^{-ikx_j} - \theta \right) a_k^\dagger \right\}
\]

\[
+ \theta^2 \sum_k |V_k|^2. \]

We estimate \( E_{\text{sp}}(R') \) from below in the first step and from above in the next step. After obtaining both estimates, taking \( R' \to \infty \) yields the estimate \((3.3)\).

We arbitrarily fix a normalized phonon state \( \Psi \) and a normalized wave function \( \psi(x_j) \) of the electron satisfying \( \lim_{|x_j| \to \infty} \psi(x_j) = 0 \). The brackets \( \langle \langle \quad \rangle \rangle \) stands for an averaging over the wave function \( \psi(x_j) \). We define a unitary operator \( U_{j,\alpha\theta} \) so that \( U_{j,\alpha\theta}^* p_j U_{j,\alpha\theta} = \alpha \theta p_j \) and \( U_{j,\alpha\theta} x_j U_{j,\alpha\theta} = x_j/\alpha \theta \). Then, the term \( \langle \langle U_{j,\alpha\theta}^* H_{\text{sp}}^{(j)}(\theta, R') U_{j,\alpha\theta} \rangle \rangle \) in the equation,

\[
\left\langle \langle U_{j,\alpha\theta} \psi \right| H_{\text{sp}}^{(j)}(\theta, R') \left| U_{j,\alpha\theta} \psi \right\rangle \right. = \left. \left\langle \langle U_{j,\alpha\theta}^* H_{\text{sp}}^{(j)}(\theta, R') U_{j,\alpha\theta} \right| \psi \right\rangle,
\]
can be approximated as:

\[
\langle U_{j,\alpha \theta} H_{\text{SP}}^{(j)}(\theta, R') U_{j,\alpha \theta} \rangle \\
\approx \frac{\alpha^2 \theta^2}{2} \langle p_j^2 \rangle - 2\theta \sum_k |V_k|^2 \langle \gamma_1(x_j/\alpha \theta) e^{ik \cdot x_j/\alpha \theta} \rangle + \theta^2 \sum_k |V_k|^2 \\
+ H_{ph} + \sum_k \left\{ V_k \langle \gamma_1(x_j/\alpha \theta) e^{ik \cdot x_j/\alpha \theta} - \theta \rangle a_k + V_k^* \langle \gamma_1(x_j/\alpha \theta) e^{-ik \cdot x_j/\alpha \theta} - \theta \rangle a_k^* \right\}.
\]

Thus, it is estimated from below as:

\[
\langle U_{j,\alpha \theta} H_{\text{SP}}^{(j)}(\theta, R') U_{j,\alpha \theta} \rangle \geq \frac{\alpha^2 \theta^2}{2} \langle p_j^2 \rangle - \theta \sum_k |V_k|^2 \langle \gamma_1(x_j/\alpha \theta) e^{ik \cdot x_j/\alpha \theta} \rangle^2 \\
+ (\theta - 1) \sum_k |V_k|^2 \langle \gamma_1(x_j/\alpha \theta) e^{ik \cdot x_j/\alpha \theta} \rangle^2.
\]

Since \(|\langle \gamma_1(x_j/\alpha \theta) e^{ik \cdot x_j/\alpha \theta} \rangle| \leq 1\), we arrive at the inequality:

\[
\langle (U_{j,\alpha \theta} \psi) | H_{\text{SP}}^{(j)}(\theta, R') | (U_{j,\alpha \theta} \psi) \rangle \\
\geq \frac{\alpha^2 \theta^2}{2} \int d^3x |\nabla_x \psi(x)|^2 \\
- \theta \sum_k |V_k|^2 \int \int d^3x d^3y \gamma_1(x/\alpha \theta) \gamma_1(y/\alpha \theta) |\psi(x)|^2 |\psi(y)|^2 e^{ik \cdot (x-y)/\alpha \theta} \\
- |\theta - 1| \sum_k |V_k|^2.
\] (3.5)

Approximation (2.7) shows

\[
\sum_k |V_k|^2 \int \int d^3x d^3y \gamma_1(x/\alpha \theta) \gamma_1(y/\alpha \theta) |\psi(x)|^2 |\psi(y)|^2 e^{ik \cdot (x-y)/\alpha \theta} \\
\leq \frac{\alpha^2 \theta}{\sqrt{2}} \int \int d^3x d^3y \frac{|\psi(x)|^2 |\psi(y)|^2}{|x-y|},
\] (3.6)

where inequalities, \(0 \leq \gamma_1(x/\alpha \theta), \gamma_1(y/\alpha \theta) \leq 1\), were used in the above. Thus, the inequalities, (3.5) and (3.6), lead us to the estimate from below:

\[
\langle (U_{j,\alpha \theta} \psi) | H_{\text{SP}}^{(j)}(\theta, R') | (U_{j,\alpha \theta} \psi) \rangle \geq \alpha^2 \theta^2 \mathcal{E}_p(\psi) - |\theta - 1| \sum_k |V_k|^2,
\]

where \(\mathcal{E}_p(\psi)\) is the Pekar functional \(^{45,46,47,48}\) i.e.,

\[
\mathcal{E}_p(\psi) := \frac{1}{2} \int d^3x |\nabla_x \psi(x)|^2 - \frac{1}{\sqrt{2}} \int \int d^3x d^3y \frac{|\psi(x)|^2 |\psi(y)|^2}{|x-y|}.
\]
Lieb proved that there is a unique and smooth minimizing $\psi(x)$ in $c_{SP} := -\inf_{\psi, \langle \psi \rangle = 1} E_P(\psi)$ up to translations. Then, using the estimate (2.6) and the estimate of $c_{SP}$ shown by Miyake and by Gerlach and Löwen, we obtain

$$E_{SP}(R') \geq -c_{SP} \alpha_\theta^2 - \left| 1 - \frac{1}{\theta} \right| \frac{\sqrt{2} \alpha_\theta}{\pi} K, \quad c_{SP} = 0.108513 \cdots.$$  (3.7)

This inequality works for the upper bound in the inequality (3.3).

Let us proceed to the proof of the upper bound next. For the estimate from above, we prepare

$$H^{(j)}(\theta', R') := \frac{1}{2} p_j^2 - 2\theta \gamma_1(x_j) \sum_k |V_k|^2 e^{ik \cdot x_j} \approx \frac{1}{2} p_j^2 - \frac{\sqrt{2} \alpha_\theta}{|x_j|} + \text{error}_{r'}(x_j, \theta),$$

$$\text{error}_{r'}(x_j, \theta) := (1 - \gamma_1(x_j)) \frac{\sqrt{2} \alpha_\theta}{|x_j|},$$

by using the approximation (2.7).

For $r \geq 0$ we define a function $C_0(r)$ by

$$C_0(r) := (1 + r) e^{-r}. \quad (3.8)$$

Since $C_0(r') < C_0(r) < C_0(0) = 1$ for $0 < r < r'$ and $C_0(\infty) = \lim_{r \to \infty} C_0(r) = 0$, we take a unique point $r_*$ so that $C_0(r_*) = 1 - \sqrt{c_{SP}}$. We set $C_1(\mu, R')$ and $R_\mu$ as $C_1(\mu, R') := C_0(\mu R')$ and $R_\mu := r_* / \mu$, respectively. Then, we have a simple inequality:

$$C_1(\mu, R') \leq 1 - \sqrt{c_{SP}}, \quad (3.9)$$

provided $R_\mu \leq R'$. Using the wave function $\psi_\mu(x_j)$ defined by $\psi_\mu(x_j) := \sqrt{\mu^3/\pi} e^{-\mu |x_j|}$ for $\mu > 0$ as a trial function, we can estimate $\langle \psi_\mu | H^{(j)}_{\text{eff}}(\theta, R') | \psi_\mu \rangle$ from above:

$$\langle \psi_\mu | H^{(j)}_{\text{eff}}(\theta, R') | \psi_\mu \rangle \leq \frac{\mu^2}{2} - \sqrt{2} \alpha_\theta \mu + \sqrt{2} \alpha_\theta \mu C_1(\mu, R'), \quad (3.10)$$

where we used $0 \leq 1 - \gamma_1(x_j) \leq 1$. Evaluating RHS of the inequality $E_{SP}(R') \leq \langle \psi_\mu \Omega_{\text{ph}} | H^{(j)}_{\text{SP}}(\theta, R') | \psi_\mu \Omega_{\text{ph}} \rangle$ for the phonon vacuum $\Omega_{\text{ph}}$, it follows from the inequalities (3.9) and (3.10) that

$$E_{SP}(R') \leq \frac{1}{2} \left( \mu - \sqrt{2c_{SP}} \alpha_\theta \right)^2 - c_{SP} \alpha_\theta^2 + \theta^2 \sum_k |V_k|^2.$$
for every \( \mu > 0 \) and \( R' \geq R_\mu \). Setting the parameter \( \mu = \mu_* := \sqrt{2c_{SP} \alpha_\theta} \) and using estimate (2.6), we arrive at the following upper bound:

\[
E_{SP}(R') \leq \inf_{\mu > 0} \langle \psi_\mu \Omega_{ph} | H_{SP}^{(j)}(\theta, R') | \psi_\mu \Omega_{ph} \rangle \leq -c_{SP} \alpha_\theta^2 + \theta \frac{\sqrt{2} \alpha_\theta}{\pi} K. \tag{3.11}
\]

Both bounds (3.7) and (3.11) lead us to the conclusion that if \( R' \geq R_{\mu_*} \equiv r_*/\mu_* = r_*/\sqrt{2c_{SP} \alpha_\theta} \) and \( \theta > 0 \) then

\[
-c_{SP} \alpha_\theta^2 - \left| 1 - \frac{1}{\theta} \right| \frac{\sqrt{2} \alpha_\theta}{\pi} K \leq E_{SP}(R') \leq -c_{SP} \alpha_\theta^2 + \theta \frac{\sqrt{2} \alpha_\theta}{\pi} K. \tag{3.12}
\]

Taking the limit \( R' \to \infty \) in the above, we reach our desired estimate (3.3).

**IV. SPATIAL LOCALIZATION IN WEAK-COUPLING REGIME**

In this section, we consider the case where \( \alpha \) is in the weak-coupling regime. As noted in Sec.II, the Hamiltonian formalism in the mathematical representation does not show us the existence of any ground state in the state space because of the translation invariance of \( H_{BP} \) with \( \rho(x) \equiv 1 \). Thus, we employ \( H_{el-ph}(\rho) \) in \( H_{BP} \) so that \( \rho \) breaks the translation invariance. Then, in the weak-coupling regime, we seek a necessary condition for \( H_{BP} \) having a bound state and investigate spatial localization of bipolaron. We note that we can obtain the same results for \( \tilde{H}_{BP}^{rel} \) and \( \tilde{H}_{BP}^{\Pi=0} \) as described below. Although we do not prove this remark in this paper, it is proved in the same way as shown for \( H_{BP} \).

In Ref.29 we showed that if the quantum particle is not spatially localized, then the particle dressed in the cloud of bosons cannot exist. In that problem, divergent soft bosons for the infrared catastrophe cause the spatial nonlocalization. From this point of view, we study how the Coulomb repulsion between the two electrons arrests the spatial localization in bipolaron.

Let us proceed with the definition of spatial localization. We say that the relative motion of the bipolaron in a bound state \( \Psi_n \) is spatially localized in the closed ball \( \overline{B}(r) := \{ X_2 | |X_2| \leq r \} \) if \( \langle \Psi_n | \Psi_n \rangle^{-1} \langle \Psi_n | x_1 - x_2 | \Psi_n \rangle \leq r \). In this paper we will use the phrase that a bound state \( \Psi_n \) of the bipolaron with the size less than \( r > 0 \) does not exist when the relative motion of the bipolaron in \( \Psi_n \) is not spatially localized in \( \overline{B}(r) \). For a ground state \( \Psi_0 \) we define the distance \( d_{BP}(\Psi_0) \) between the two electrons in bipolaron by

\[
d_{BP}(\Psi_0) := \langle \Psi_0 | \Psi_0 \rangle^{-1} \langle \Psi_0 | x_1 - x_2 | \Psi_0 \rangle. \tag{4.1}
\]
To estimate $E(r)$ in Eq. (1.2) under the weak-coupling regime, we define a positive constant $E_w(\alpha)$ by

$$E_w(\alpha) := 4 \sum_k |V_k|^2 = \frac{4\sqrt{2}\alpha}{\pi} K = \frac{8\sqrt{2}\alpha}{\Lambda}$$

(4.2)

for every $\alpha > 0$, where $\Lambda$ is a wave length defined by $\Lambda := 2\pi/K$. It is easy to check $E_{BP} \leq 0$ by evaluating RHS of the inequality $E_{BP} \leq \inf_{\psi} \langle \psi_\Omega \mid \tilde{H}_{BP} \mid \psi_\Omega \rangle$ for normalized wave functions $\psi$ of the two electrons and the phonon vacuum $\Omega_{ph}$. The simple operator inequality $H_{ph} + \tilde{H}_{el-ph} \geq -E_w(\alpha)$ shows that $H_{BP} \geq -E_w(\alpha)$, making a lower bound to the ground state energy:

$$E_{BP} \geq -E_w(\alpha).$$

(4.3)

We can show $E_{BP} \propto -\alpha$ for small $\alpha$ in the weak-coupling regime by a perturbative or nonperturbative means in the same way as in the case of $E_{SP}$.

We prove in this section that if the bipolaron has a bound state with eigenenergy $E$ so that its relative motion is spatially localized in $B(r)$, then there is the energy inequality:

$$E_w(\alpha) + E \geq \frac{U}{r} = \frac{\sqrt{2}\alpha}{(1 - \eta)r}.$$  

(4.4)

Our result says that

$$d_{BP}(\Psi_0) \geq d_{BP}^{low} := \frac{U}{E_w(\alpha) + E_{BP}} \geq \frac{\Lambda}{8(1 - \eta)}$$

(4.5)

for any ground state $\Psi_0$. Thus, we realize that the distance $d_{BP}(\Psi_0)$ between the two electrons in the bipolaron grows more and more as $\eta$ turns out to be closer to 1 and also that the coupling constant $\alpha$ in the weak-coupling regime cannot stem its growth. That is not the case in the strong-coupling regime (see Sec. VII). This is a noticeable difference between the cases in the weak-coupling regime and in the strong-coupling one.

Moreover, this growth tells us that if a bipolaron has a ground state $\Psi_0$ for $\eta$ sufficiently close to 1, then $B(d_{BP}(\Psi_0))$ completely contains the whole crystal. On the other hand, since the bipolaron in the ground state $\Psi_0$ should be formed in the crystal, the two electrons in the bipolaron must exist in the crystal. Thus, we have met a contradiction. Therefore, reductio ad absurdum makes us conclude from these two facts contradicting each other that any bipolaron in a ground state cannot be formed in the crystal if $\eta$ is close to 1 in the
weak-coupling regime. We will give another lower bound to \( d_{BP}(\Psi_0) \) in the inequality (4.10) later.

Our above result also states, in particular, that the relative motion of the bipolaron in a ground state is not spatially localized in \( \overline{B}(r) \), provided that the ground state exists under the condition:

\[
E_w(\alpha) < \frac{U}{r} \quad \text{(i.e.,} \quad 0 \leq 1 - \frac{\Lambda}{8r} < \eta\text{)}. \quad (4.6)
\]

In terms of the binding energy with Eq. (1.2), the condition (4.6) means

\[
- (2E_{SP} - E_{BP}) \approx - E_w(\alpha) - E_{BP} + \frac{U}{r} \geq - E_w(\alpha) + \frac{U}{r} = \sqrt{2} \alpha \left( \frac{1}{1 - \eta} - \frac{8r}{\Lambda} \right) \frac{1}{r} > 0 \quad (4.7)
\]

by estimating \( E(r) \) at \( E_w(\alpha) + E_{BP} \). Thus, as for the relation between the spatial localization of a ground state and the binding energy, we can interpret our result as that a ground state of the bipolaron with the size of \( d_{BP}(\Psi_0) \) less than \( r \) satisfying the condition (4.6) cannot exist because of the negative binding energy (4.7).

We define the number of phonons in a ground state \( \Psi_0 \) by \( N_{ph}(\Psi_0) := \langle \Psi_0 | \sum_k a_k^\dagger a_k | \Psi_0 \rangle \). Then, it is shown in this section that \( N_{ph}(\Psi_0) \) is estimated as:

\[
\frac{U}{d_{BP}(\Psi_0)} + (2E_{SP} - E_{BP}) \leq N_{ph}(\Psi_0) \leq 2E_w(\alpha) + E_{BP} + 2\sqrt{E_w(\alpha)(E_w(\alpha) + E_{BP})}. \quad (4.8)
\]

The first inequality yields the upper bound to the binding energy:

\[
2E_{SP} - E_{BP} \leq N_{ph}(\Psi_0) - \frac{U}{d_{BP}(\Psi_0)} < N_{ph}(\Psi_0). \quad (4.9)
\]

Hence it follows that another lower bound to \( d_{BP}(\Psi_0) \) is given as:

\[
\frac{U}{N_{ph}(\Psi_0) - (2E_{SP} - E_{BP})} \leq d_{BP}(\Psi_0). \quad (4.10)
\]

Let us begin by proving the first result. We adopt Lieb’s idea into our argument to prove it. Here we assume that \( \tilde{H}_{BP} \) has a bound state \( \Psi_n \) with its eigenenergy \( E \), i.e., \( \tilde{H}_{BP}\Psi_n = E\Psi_n \), and that it is spatially localized in \( \overline{B}(r) \). Here we can suppose \( \Psi_n \) is normalized, i.e., \( \langle \Psi_n | \Psi_n \rangle = 1 \), without any loss of generality. Because we only have to take \( \langle \Psi_n | \Psi_n \rangle^{-1/2} \Psi_n \) as the normalized bound state in case that \( \Psi_n \) is not normalized. Then, since \( P_2^2/2M_1 \geq 0 \), evaluating RHS of \( 0 = \langle |X_2|\Psi_n| \tilde{H}_{BP} - E |\Psi_n\rangle \) from below yields

\[
0 \geq \frac{1}{2M_2} \langle |X_2|\Psi_n|P_2^2\Psi_n\rangle + U + \langle |X_2|^{1/2}\Psi_n|H_{ph} + \tilde{H}_{el-ph}|X_2|^{1/2}\Psi_n\rangle - E\langle \Psi_n||X_2||\Psi_n\rangle.
\]
This leads us to the inequality:

$$0 \geq \frac{1}{2M_2} \langle |X_2|\Psi_n|P_2^2\Psi_n| + U - (E + E_w(\alpha)) r, \quad (4.11)$$

where we used the spatial localization of $\Psi_0$ and the fact that $E + E_w(\alpha) \geq E_{bp} + E_w(\alpha) \geq 0$ by the inequality \textbf{(4.3)}. It follows from the same argument that

$$0 \geq \frac{1}{2M_2} \langle P_2^2\Psi_n|X_2|\Psi_n| + U - \alpha r. \quad (4.12)$$

Since $\Delta_{X_2}|X_2|^{-1} = 4\pi \delta(X_2)$, we have $(P_2^2|X_2| + |X_2|P_2^2)/2 = |X_2|P_2^2|X_2|^{-1}P_2|X_2| - 2\pi|X_2|\delta(X_2)|X_2|$. Thus, we reach the equation and inequality:

$$\frac{1}{2M_2}\langle \Psi_n|P_2^2|X_2| + |X_2|P_2^2\Psi_n\rangle = \frac{1}{2M_2}\langle P_2|X_2|\Psi_n\rangle - \alpha \geq 0. \quad (4.13)$$

Combining the inequalities (4.11)–(4.13), we arrive at the first desired inequality (4.4).

To show the inequality (4.5), we use reductio ad absurdum. As an assumption for it we suppose $d_{bp} < d_{bp}^{low}$. Then, there is a positive number $r'$ so that $d_{bp} < r' < d_{bp}^{low}$. Since $d_{bp} < r'$, the above fact already proved says $E_w(\alpha) + E_{bp} \geq U/r'$, and so $r' \geq d_{bp}^{low}$. This is not consistent with the fact that $r' < d_{bp}^{low}$. Thus, reductio ad absurdum supplies the second desired inequality.

Let us proceed with the proof of our next statement. Since $E_{bp} \leq 0$, the simple term $E_w(\alpha)$ can substitute for $E_w(\alpha) + E_{bp}$ in the inequality (4.4) for the ground state $\Psi_0$. Thus, the contraposition of the first result leads us to the third desired result on the spatial nonlocalization.

Let us estimate the number of phonons in a ground state $\Psi_0$ now. We regard $\Psi_0$ as being normalized. We define the Hamiltonian $\sum_j \tilde{H}_{sp}^{(j)}$ of the two separate single polarons for $\tilde{H}_{bp}$ by using the coordinates $X_1$ and $X_2$, i.e., $\sum_j \tilde{H}_{sp}^{(j)} := \sum_{j=1,2} P_j^2/2M_2 + 2H_{ph} + 2H_{el-ph}$. In addition, we note that the Hamiltonian of the two separate single polarons for $\tilde{H}_{bp}$ is 

$$\sum_j \tilde{H}_{sp}^{(j)} = P_j^2/2M_2 + 2H_{ph} + H_{el-ph}(\rho Q),$$

and that for $\tilde{H}_{bp}^{\Pi=0}$ is

$$\sum_j \tilde{H}_{sp}^{(j)} = P_j^2/2M_2 + 2H_{ph} + P_{ph}^2/2M_1 + H_{el-ph}(\rho Q).$$

Note that $1 = |X_2|^{-1/2}|X_2|^{1/2}$ now. Then, Schwarz’s inequality implies that $1 = \langle |X_2|^{-1/2}\Psi_0|X_2|^{1/2}\Psi_0\rangle \leq \langle \Psi_0|X_2|^{-1}\Psi_0\rangle^{1/2}\langle \Psi_0|X_2|\Psi_0\rangle^{1/2}$, making the inequality $1/d_{bp}(\Psi_0) \leq \langle \Psi_0|X_2|^{-1}\Psi_0\rangle$. Hence it follows from this inequality, together with sandwiching the equation $\sum_j \tilde{H}_{sp}^{(j)} - 2E_{sp} = \tilde{H}_{bp} - 2E_{sp} + H_{ph} - U/|X_2|$ between $\langle \Psi_0\rangle$ and $|\Psi_0\rangle$. Therefore, the lower bound in the inequality
(4.8) is obtained from this inequality. As for the upper bound, we use the simple operator inequality:

\[
C \tilde{H}_{BP} - H_{ph} \geq (C - 1) \left\{ H_{ph} + \frac{C}{(C - 1)} \tilde{H}_{el-ph} \right\} \geq - \frac{C^2}{C - 1} E_w(\alpha), \quad C > 1,
\]

to come up with \( H_{ph} \leq C \tilde{H}_{BP} + C^2 E_w(\alpha)/(C - 1) \). Putting this inequality between \( \langle \Psi_0 | \psi_0 \rangle \) and \( |\Psi_0 \rangle \) leads us to the inequality \( N_{ph}(\Psi_0) \leq \inf_{C>1} \left[ C E_{BP} + C^2 E_w(\alpha)/(C - 1) \right] \). RHS of this inequality attains the lower bound in the inequality (4.8).

V. BIPOLARON FORMATION

In this section we deal with the original \( H_{BP} \) (i.e., in the case \( \rho(x) \equiv 1 \)) in the strong-coupling regime and derive two effective Hamiltonians from \( H_{BP} \) by modifying Bogolubov's method.\(^{37}\) Our modified method is similar to Adamowski's\(^{12}\) and ours.\(^{33}\) More precisely, we seek a canonical transformation \( U_\theta \) with the parameter \( \theta \geq 0 \) so that the transformed Hamiltonian has the form of \( H_{BP}(\theta) := U_\theta^* H_{BP} U_\theta = H_{eff}(\theta) + H_{ph} + H_{el-ph}(\theta) + \Sigma_\theta \), where \( H_{eff}(\theta) \) is an effective Hamiltonian in quantum mechanics, and \( \Sigma_\theta \) a divergent energy as \( \theta \rightarrow \infty \). The canonical transformation \( U_\theta \) requires that the effective Hamiltonian \( H_{eff}(\theta) \) should gain an attractive potential \( V(\theta) \) from the phonon field as \( H_{eff}(\theta) = H_{el-el} + V(\theta) \). Thanks to this extra attractive potential \( V(\theta) \), we expect that there is a critical point \( \theta_c \) so that the Hamiltonian \( H_{eff}(\theta) \) itself or the Hamiltonian \( H_{eff}^{rel}(\theta) \) for the relative motion of \( H_{eff}(\theta) \) has a ground state if \( \theta > \theta_c \). On the other hand, it has no ground state if \( \theta < \theta_c \). Thus, we can expect that the bipolaron is stable at/near the point \( \theta_c \). Actually, according to the recent result,\(^{49}\) if we apply the above method to \( \tilde{H}_{BP}^{\Pi=0} \), then we might be able to show that \( \tilde{H}_{BP}^{\Pi=0} \) has a ground state for sufficiently large \( \theta > 0 \).

To find the canonical transformation \( U_\theta \), we adopt the notion of Feynman's virtual phonon\(^{27}\) and the image of Peeters and Devreese's classical bipolaron,\(^{28}\) being led to the classical images of the two kinds of states of bipolaron: balanced state and unbalanced state. The balanced state designates the image that the virtual phonon sits at the center of two electrons. On the other hand, the unbalanced state represents the image that the virtual phonon is not on the line segment between the two electrons. Then, two effective Hamiltonians are derived in quantum mechanics through these classical images.

It is shown in this section that the approximation \(^{27,47}\) yields an effective Hamiltonian
describing balanced state in quantum mechanics:

\[ H_{\text{eff}}(\theta) = H_{\text{el-el}} + V(\theta) = \frac{1}{2}p_1^2 + \frac{1}{2}p_2^2 + \frac{U(\theta)}{|x_1 - x_2|} \]  

(5.1)

with

\[ U(\theta) = U - \sqrt{2}\alpha\theta = \sqrt{2}\alpha \left( \frac{1}{1 - \eta} - \theta \right). \]  

(5.2)

It is clear that \( \theta_c = 1/(1 - \eta) \). Let us fix \( \alpha \) arbitrarily so that \( 0 < \alpha < 1 \). We set \( c_{BP} \) as:

\[ c_{BP} := \frac{2}{5} \left( \frac{9}{2} \frac{1}{\theta (1 - \eta)} \right)^2. \]  

(5.3)

Then, using the effective Hamiltonian \( H_{\text{eff}}(\theta) \) in Eq.(5.1), we show that an upper bound to the ground state energy \( E_{BP} \) is given as:

\[ E_{BP} \leq -c_{BP}\alpha \frac{1}{\theta} + \frac{\sqrt{2} \alpha}{\pi} \left( 4K + \frac{K^3}{3} \right), \]  

(5.4)

provided that

\[ 1 - \frac{1}{c_s}\theta > \eta. \]  

(5.5)

We note that the condition (5.5) prohibits us from taking the limit \( \theta \to 0 \) in the inequality (5.4) because

\[ \theta > \frac{1}{c_s(1 - \eta)} > \frac{1}{1 - \eta}. \]

Meanwhile, it is also shown in this section that a lower bound to \( E_{BP} \) as:

\[ E_{BP} \geq \left( \inf_{\varphi} \mathcal{E}_\theta(\varphi) \right) \alpha \frac{1}{\theta} - \left| 1 - \frac{1}{\theta} \right| \frac{4\sqrt{2} \alpha}{\pi} K, \]  

(5.6)

where

\[ \mathcal{E}_\theta(\varphi) := \frac{1}{2} \int \int d^3x_1 d^3x_2 \left[ |\nabla x_1 \varphi(x_1, x_2)|^2 + |\nabla x_2 \varphi(x_1, x_2)|^2 + \frac{2\sqrt{2}}{\theta (1 - \eta)} \frac{|\varphi(x_1, x_2)|^2}{|x_1 - x_2|} \right] \]

\[- \frac{1}{\sqrt{2}} \int \int d^3x_1 d^3x_2 \int \int d^3y_1 d^3y_2 \sum_{j, j' = 1, 2} \frac{|\varphi(y_1, y_2)|^2}{|x_j - y_{j'}|} \]  

(5.7)

is an energy functional describing unbalanced state and \( \inf_{\varphi} \mathcal{E}_\theta(\varphi) < 0 \). This functional is the almost same as Vinetskii and Giterman's, \( \mathcal{E}_\varphi(\varphi) \). Namely, for wave functions \( \varphi(x_1, x_2) \) satisfying \( \varphi(x_1, x_2) = \varphi(x_2, x_1) \), our energy functional becomes theirs. Here, we note that the
estimate (5.6) in the strong-coupling regime recovers the estimate (4.3) in the weak-coupling one as \( \theta = 0 \).

At the end of this section we show that the approximation (2.7) yields another effective Hamiltonian describing unbalanced state in quantum mechanics:

\[
H_{\text{eff}}(\theta) = \alpha^2 \theta \left[ \sum_{j=1,2} \frac{1}{2} p_j^2 - \sqrt{2} \sum_{j=1,2} \frac{1}{|x_j|} + \frac{\sqrt{2}}{\theta (1 - \eta) |x_1 - x_2|} \right].
\]  

(5.8)

Here we note that the approximation (2.7) breaks the translation invariance so that the effective Hamiltonian \( H_{\text{eff}}(\theta) \) in Eq.(5.8) describes that the virtual phonon is nailed down at the origin. Let us set \( E_s(\alpha) \) as:

\[
E_s(\alpha) := \left( \frac{2\sqrt{2}}{r} - \frac{1}{r^2} - 1 \right) \alpha \theta
\]  

(5.9)

now. Then, the effective Hamiltonian \( H_{\text{eff}}(\theta) \) in Eq.(5.8) has a ground state provided that there is an \( r > 0 \) so that

\[
\frac{U}{r} < E_s(\alpha).
\]  

(5.10)

Thus, we are led to the conclusion that \( \theta_c \leq 1/(2 - \sqrt{2})(1 - \eta) \). The condition (5.10) puts restrictions on \( \theta, \eta \) and \( r \). Namely, the sufficient condition for the inequality (5.10) is:

\[
1 - \frac{1 + \sqrt{2}}{\sqrt{2} \theta} > \eta,
\]  

(5.11)

and

\[
R_{\theta,\eta} - \sqrt{R_{\theta,\eta}^2 - 1} < r < R_{\theta,\eta} + \sqrt{R_{\theta,\eta}^2 - 1},
\]  

(5.12)

where

\[
R_{\theta,\eta} := \sqrt{2} \left( 1 - \frac{1}{2\theta (1 - \eta)} \right).
\]

Therefore, for sufficiently large \( \theta \) (i.e., \( \theta \approx \infty \)), \( 0.585 \leq r/r_p \leq 3.415 \).

Since the inequalities (3.3), (5.4) and (5.6) say that \( E(r) \geq 0 \) is dominated from below by \( \sqrt{2} \alpha \theta / r \) and/or \( E_s(\alpha) \) for sufficiently large \( \theta > 0 \), in the case of the balanced state the condition \( \theta > 1/(1 - \eta) \) makes the estimated binding energy positive:

\[
-(2E_{\text{sp}} - E_{\text{bp}}) \lesssim -\frac{\sqrt{2} \alpha \theta}{r} + \frac{U}{r} = \sqrt{2} \alpha \left( \frac{1}{1 - \eta} - \theta \right) \frac{1}{r} < 0.
\]  

(5.13)
In the case of the unbalanced state the condition (5.10) also makes the estimated binding energy positive:

\[- (2E_{el} - E_{ph}) \lesssim -E_s(\alpha) + \frac{U}{r} = \alpha_0 \left[ 1 + \frac{1}{r^2} + \sqrt{2} \left( \frac{1}{\theta(1 - \eta)} - 2 \right) \frac{1}{r} \right] < 0. \quad (5.14)\]

Thus, the appearance of \( \theta \) in Eqs. (5.13) and (5.14) is very different than it does not appear in Eqs. (1.1) and (4.7).

### A. Strategy for Effective Hamiltonians

In Ref. 33 we derived the Coulomb attractive potential from the electron-boson interaction for the model introduced by Gross.52 Similarly, we use such a canonical transformation with trial functions \( \beta_j(k, x_j), j = 1, 2 \), which have the parameter \( \theta \). By controlling the trial functions, we derive the effective Hamiltonian \( H_{\text{eff}}(\theta) \). Set \( \beta_{j,k} := \beta_j(k, x_j) \), where we assume \( \beta_j(k, x_j)^* = \beta_j(-k, x_j) \) for every \( x_j \) and \( j = 1, 2 \), i.e., \( \beta_{j,k}^* = \beta_{j,-k} \). We give the generator \( T \) by \( T := i \sum_{j=1,2} \sum_k \{ V_k \beta_{j,k} e^{ikx} a_k - V_k^* \beta_{j,k}^* e^{-ikx} a_k^\dagger \} \). Then, we obtain the canonical transformation \( U_\theta \) by \( U_\theta := e^{iT} \). Precisely writing down, we obtain each term in the following. The effective potential \( V(\theta) \) is given by

\[
V(\theta) := 2 \sum_k |V_k|^2 \left\{ \beta_{1,k}^* \beta_{2,k} - \left( \beta_{1,k}^* + \beta_{2,k} \right) \right\} e^{-ik(x_1 - x_2)}.
\]

The electron-phonon interaction \( H_{\text{el-ph}}(\theta) \) is decomposed into the two parts as \( H_{\text{el-ph}}(\theta) = H_{\text{el-ph}}^{(1)} + H_{\text{el-ph}}^{(2)} \), which are respectively defined by

\[
H_{\text{el-ph}}^{(1)} := \sum_{j=1,2} \sum_k \left\{ V_k \left( 1 - \beta_{j,k} - \frac{1}{2} D(\beta_{j,k}) \right) e^{ikx} a_k \right.
+ V_k^* \left( 1 - \beta_{j,k}^* - \frac{1}{2} D(\beta_{j,k})^* \right) e^{-ikx} a_k^\dagger \},
\]

\[
H_{\text{el-ph}}^{(2)} := \sum_{j=1,2} \left\{ p_j A_j + A_j^\dagger p_j \right\} + \frac{1}{2} \sum_{j=1,2} \left\{ A_j^2 + 2A_j^\dagger A_j + A_j^\dagger 2 \right\},
\]

where \( D(\beta_{j,k}) = -\Delta_{x_1} \beta_{j,k} - 2ik \nabla_{x_1} \beta_{j,k} + k^2 \beta_{j,k} \) and \( A_j = \sum_k V_k \left( k \beta_{j,k} - i(\nabla_{x_1} \beta_{j,k}) \right) e^{ikx} a_k \).

The divergent energy as \( \theta \to \infty \) is

\[
\sum_{\theta} := \sum_k |V_k|^2 \left\{ \left( 1 + \frac{k^2}{2} \right) (|\beta_{1,k}|^2 + |\beta_{2,k}|^2) + \frac{1}{2} \left( |\nabla_{x_1} \beta_{1,k}|^2 + |\nabla_{x_2} \beta_{2,k}|^2 \right) - \Re (\beta_{1,k} + \beta_{2,k}) \right\}.
\]
The main problem in our strategy is what is the principle to find the best effective Hamiltonian. Here we check well-known method for a while by way of trial. If we employ $\beta_j(k) = \theta(1 + k^2/2)^{-1}$ for $\theta > 0$ ($j = 1, 2$), the $e^{iT}$ as $\theta = 1$ is the very unitary operator we used for the model introduced by Gross. That is, it is the unitary operator in Tomonaga’s intermediate coupling approximation rearranged in Lee, Low, and Pines’ study of the polaron. In the intermediate coupling approximation, the variational principle works so that the self-energy turns out lowest. Thus, employing this $\beta_j, k \equiv \beta_j(k)$, with the help of the approximation of the Fourier expansion (2.7) we obtain

$$V(\theta) = -\frac{4\sqrt{2}\pi\alpha}{\sqrt{(2\pi)^3}} \left\{ \sqrt{\frac{\pi}{2}} \theta(2 - \theta) + \theta(2 - \theta) \sqrt{\frac{\pi}{2}} e^{-\sqrt{2}|x_1 - x_2|} + \theta^2 \sqrt{\frac{\pi}{2}} e^{-\sqrt{2}|x_1 - x_2|} \right\}.$$ 

Since $U = \sqrt{2}\alpha/(1 - \eta)$, the effect Hamiltonian $H_{\text{eff}}(\theta)$ can be derived as:

$$H_{\text{eff}}(\theta) = \frac{p_1^2}{2} + \frac{p_2^2}{2} + \left( \frac{1}{1 - \eta} - \theta(2 - \theta) \right) \frac{\sqrt{2}\alpha}{|x_1 - x_2|}$$

$$+ \theta(2 - \theta) \sqrt{2}\alpha e^{-\sqrt{2}|x_1 - x_2|} - \theta^2 \sqrt{2}\alpha e^{-\sqrt{2}|x_1 - x_2|}.$$ 

Unfortunately, we cannot gain enough attraction from the electron-phonon interaction because of the same reason as in Eq.(1.1). Thus, we have now lost such a principle to derive the best effective Hamiltonian as in Lee, Low, and Pine’s study. Then, we depend on the picture given by the combination of the two notions: Feynman’s of virtual phonon and Peeters and Devreese’s of the classical bipolaron. As is in Kadanoff’s explanation, Feynman considered that the electron in the phonon cloud, which forms a single polaron, is coupled to another particle with a harmonic oscillator. We call the particle a (classical) virtual phonon. We use the notion of this virtual phonon. Although Feynman assumed harmonic oscillator for the interaction between the electron and the virtual phonon, we assume the Coulomb attractive potential between each electron and the virtual phonon instead as follows: We imagine the Peeters and Devreese’s classical bipolaron first. We assume a (classical) virtual phonon sits at the center $(x_1 + x_2)/2$ of the line segment of $x_1$ and $x_2$. In this case, the two electrons can feel an attraction between themselves besides the Coulomb repulsion without noticing the existence of the virtual phonon, though the attraction is actually made by the virtual phonon. Thus, we reach the image of the balanced state. On the other hand, for the unbalanced state, the two electrons have to become aware of the existence of the virtual phonon.
B. Effective Hamiltonian for Balanced State

In this subsection, we consider how we can obtain the Coulomb attraction between the two electrons in quantum theory for the balanced state. To derive such the Coulomb attraction, we employ \( \beta_{j,k} \) simply defined by \( \beta_{1,k} = -\beta_{2,k} = \sqrt{\theta} \). Then, \( \Sigma_\theta \) is approximated to \( 2\sqrt{2}\alpha(\theta - \sqrt{\theta})K/\pi + \sqrt{2}\alpha\theta K^3/6\pi \) by the estimate (2.6), so that \( \lim_{\theta \to \infty} \Sigma_\theta/\theta^2 = 0 \). Then, the approximation of the Fourier expansion (2.7) leads us to the more effective \( V(\theta) \):

\[
V(\theta) = -\frac{\sqrt{2}\alpha\theta}{|x_1 - x_2|}.
\]

Therefore, we obtain the effective Hamiltonian \( H_{\text{eff}}(\theta) \) defined in Eq. (5.1) representing the balanced state. Using coordinates of the center-of-mass and the relative motion, we realize that \( H_{\text{eff}}(\theta) \) is unitary-equivalent to \( \tilde{H}_{\text{eff}}(\theta) \):

\[
\tilde{H}_{\text{eff}}(\theta) := \frac{1}{4}P_1^2 + \tilde{H}_{\text{eff}}^{\text{rel}}(\theta),
\]

where

\[
\tilde{H}_{\text{eff}}^{\text{rel}}(\theta) := P_2^2 + \frac{U(\theta)}{|X_2|}.
\]

At the end of this subsection, we show the upper bound (5.4) to \( E_{\text{BP}} \). We now introduce a mathematical parameter \( R > 2 \) into \( H_{\text{BP}} \) so that \( \lim_{R \to \infty} H_{\text{BP}}(R) = H_{\text{BP}} \) (in the norm resolvent sense). To introduce the parameter \( R > 2 \), we use a real-valued function \( \gamma_2(x) \) defined by \( \gamma_2(x) := \chi_R(|x|) \), where \( \chi_R(r) := 1 \) for \( r < R/2 \) and \( \chi_R(r) := 0 \) for \( r > R \) with linear interpolation. Then, it is easy to show that \( \sup_x |\nabla_x \gamma_2(x)| \leq 2/R < 1 \). We define the Hamiltonian \( H_{\text{BP}}(R) \) with the parameter \( R > 2 \) by \( H_{\text{BP}}(R) := H_{\text{el-ph}} + H_{\text{ph}}(R) \), where \( H_{\text{el-ph}}(R) \) is defined by putting \( \gamma_2(x_1 - x_2) \) in front of \( \sum_{j=1,2} \sum_k \) in the original \( H_{\text{el-ph}} \) of \( H_{\text{BP}} \).

We can define the unitary-transformed Hamiltonian \( H_{\text{BP}}(\theta, R) \) by \( H_{\text{BP}}(\theta, R) := e^{iT}H_{\text{BP}}(R)e^{-iT} \). Then, \( H_{\text{BP}}(\theta, R) \) is approximated as \( H_{\text{BP}}(\theta, R) \approx H_{\text{eff}}(\theta, R) + H_{\text{ph}} + H_{\text{el-ph}}(\theta, R) + \Sigma_\theta(R) \) through the approximation (2.7). Here the effective Hamiltonian \( H_{\text{eff}}(\theta, R) \) is given by \( H_{\text{eff}}(\theta, R) = H_{\text{eff}}(\theta) + \text{error}_R(x_1 - x_2, \theta) \) for the effective Hamiltonian \( H_{\text{eff}}(\theta) \) in Eq. (5.1) and the error term \( \text{error}_R(x_1 - x_2, \theta) \):

\[
\text{error}_R(x_1 - x_2, \theta) = \left(1 - \gamma_2(x_1 - x_2)^2\right)\frac{\sqrt{2}\alpha\theta}{|x_1 - x_2|},
\]
and the electron-phonon interaction $H_{\text{el-ph}}(\theta, R)$ is given by $H_{\text{el-ph}}(\theta, R) = H_{\text{el-ph}}^{(1)}(R) + H_{\text{el-ph}}^{(2)}$ for $H_{\text{el-ph}}^{(2)}$ given in Sec. V.A and $H_{\text{el-ph}}^{(1)}(R)$:

$$H_{\text{el-ph}}^{(1)}(R) := \sum_{j=1,2} \sum_k \left\{ V_k \left( \gamma_2 (x_1 - x_2) - \beta_{j,k} - \frac{1}{2} D (\beta_{j,k}) \right) e^{ikx_j} a_k^\dagger + V_k^\ast \left( \gamma_2 (x_1 - x_2) - \beta_{j,k}^\ast - \frac{1}{2} D (\beta_{j,k})^\ast \right) e^{-ikx_j} a_k^\dagger \right\}. $$

The self-energy $\Sigma_\theta(R)$ in $H_{\text{BP}}(\theta, R)$ is given by

$$\Sigma_\theta(R) = \sum_k |V_k|^2 \left\{ \left( 1 + \frac{k^2}{2} \right) (|\beta_{1,k}|^2 + |\beta_{2,k}|^2) + \frac{1}{2} (|\nabla_{x_1} \beta_{1,k}|^2 + |\nabla_{x_2} \beta_{2,k}|^2) - \gamma_2 (x_1 - x_2) \Re (\beta_{1,k} + \beta_{2,k}) \right\}. $$

Using coordinates of the center-of-mass and the relative motion, we realize that $H_{\text{eff}}(\theta, R)$ is unitary-equivalent to $\tilde{H}_{\text{eff}}(\theta, R) := P_1^2/4 + \tilde{H}_{\text{eff}}^{\text{rel}}(\theta, R)$, where $\tilde{H}_{\text{eff}}^{\text{rel}}(\theta, R) := \tilde{H}_{\text{eff}}^{\text{rel}}(\theta) + \text{error}_R(X_2, \theta)$.

For the real number $c_\ast$, we can take a unique point $r_{\text{BP}}$ so that $C_0(r_{\text{BP}}) = 1 - c_\ast$, where $C_0(r)$ was defined in Eq. (3.8). We set $C_2(\mu, R)$ and $R_{\text{BP}}$ as $C_2(\mu, R) := C_0(\mu R)$ and $R_{\text{BP}} := r_{\text{BP}}/\mu$, respectively. Then $C_2(\mu, R) \leq 1 - c_\ast$, provided $R_{\mu} \leq R$. Setting $r = 1/\mu$ in the definition of $\psi_\mu(x_j)$, we have the wave function $\phi_\mu(X_1)$ as $\phi_\mu(X_1) := \sqrt{\mu^3/\pi} e^{-\mu|X_1|}$. We denote that of $X_2$ by $\psi_\mu(X_2) := \sqrt{\mu^3/\pi} e^{-\mu|X_2|}$. Set $R_{\text{BP}}$ as $R_{\text{BP}} := r_{\text{BP}}/\mu$. Then $C_2(\mu, R) \leq 1 - c_\ast$, provided $R_{\text{BP}} \leq R$. It follows from simple estimates that

$$\langle \psi_\mu | \tilde{H}_{\text{eff}}^{\text{rel}}(\theta, R) | \psi_\mu \rangle = \mu^2 - \sqrt{2} \alpha \theta \left( c_\ast - \frac{1}{\theta(1 - \eta)} \right) $$

for every $R$ which satisfies $R_{\text{BP}} \leq R$. It is easily shown that $\langle \phi_\mu | P_1^2 \phi_\mu \rangle = \mu^2$. We define a wave function $\varphi(X_1, X_2)$ by $\varphi(X_1, X_2) := \phi_\mu(X_1) \psi_\mu(X_2)$. Then, for every $\mu > 0$ and $R \geq R_{\text{BP}}$, the ground state energy $E_{\text{BP}}(R) = \inf \text{Spec}(H_{\text{BP}}(\theta, R))$ of $H_{\text{BP}}(\theta, R)$ is estimated from above as:

$$E_{\text{BP}}(R) \leq \langle \varphi \Omega_{\text{ph}} | \tilde{H}_{\text{BP}}(\theta, R) | \varphi \Omega_{\text{ph}} \rangle \leq \frac{1}{4} \langle \phi_\mu | P_1^2 | \phi_\mu \rangle + \langle \psi_\mu | \tilde{H}_{\text{eff}}^{\text{rel}}(\theta, R) | \psi_\mu \rangle + \Sigma_\theta(R) \leq \frac{5}{4} \left( \mu - \frac{2\sqrt{2}}{5} \alpha \theta \left( c_\ast - \frac{1}{\theta(1 - \eta)} \right) \right)^2 - \frac{2}{5} \left( c_\ast - \frac{1}{\theta(1 - \eta)} \right)^2 \alpha^2 \theta^2 + \frac{\sqrt{2} \alpha \theta}{\pi} \left( 4K + \frac{K^3}{3} \right). $$

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Setting the parameter $\mu$ as $\mu = \mu_{BP} := (2\sqrt{2}/5)\alpha \theta (c_\ast - 1/\theta (1 - \eta)) > 0$ because of the guarantee (5.3) for it, we arrive at

$$E_{BP}(R) \leq -c_{BP} \alpha^2 + \frac{\sqrt{2} \alpha \theta}{\pi} \left( 4K + \frac{K^3}{3} \right)$$

for every $R \geq \max\{2, R_{BP}\}$. Taking $R \to \infty$ in the above, we obtain our desired upper bound.

C. Effective Hamiltonian for Unbalanced State

In this subsection, we consider how we can obtain the Coulomb attraction between each electron and virtual phonon in quantum theory for the unbalanced state.

Let us proceed with deriving the energy functional $E_\theta(\varphi)$ in Eq.(5.7) first. We arbitrarily fix a normalized phonon state $\Psi$ and a normalized wave function $\varphi(x_1, x_2)$ of the electron satisfying $\lim_{|x_j| \to \infty} \varphi(x_1, x_2) = 0$. We denote by $\langle \cdots \rangle$ an averaging over the wave function $\varphi(x_1, x_2)$. Then, in the same way as in the case of single polaron, we can estimate the term $\langle U^*_\alpha \varphi | H_{BP} | U_\alpha \varphi \rangle$ in the equation,

$$\langle (U_\alpha \varphi) | H_{BP} | (U_\alpha \varphi) \rangle = \langle \Psi | \langle U^*_\alpha H_{BP} U_\alpha \rangle | \Psi \rangle,$$

from below as:

$$\langle U^*_\alpha H_{BP} U_\alpha \rangle \geq \frac{\alpha^2 \theta^2}{2} \left( \langle p_1^2 \rangle + \langle p_2^2 \rangle \right) + \frac{\alpha \theta U}{|x_1 - x_2|} - \theta \sum_k |V_k|^2 \left| e^{ik \cdot x_1/\alpha \theta} + e^{ik \cdot x_2/\alpha \theta} \right|^2$$

$$- |\theta - 1| E_\omega(\alpha).$$

Using the approximation (2.7), in the same way as for $E_{SP}$, we reach the lower bound (5.6).

To obtain an effective Hamiltonian for the unbalanced state, we only have to employ $G(\theta)$ as $T$. Namely, set $\beta_{j,k}$ as $\beta_{j,k} \equiv \beta_j(k) = e^{-ik \cdot x_j \theta}$. Then, we have $T = G(\theta)$. Under the approximation (2.7), we approximate $H_{BP}[\theta] := U^*_\alpha e^{iG(\theta)} H_{BP} e^{-iG(\theta)} U_\alpha$ to:

$$H_{BP}[\theta] \approx H_{eff}(\theta) + H_{ph} + \sum_{j=1,2} \sum_k \left[ V_k \left( e^{ik \cdot x_j/\alpha \theta} - \theta \right) a_k + V_k^* \left( e^{-ik \cdot x_j/\alpha \theta} - \theta \right) a_k^\dagger \right] + \Sigma_\theta,$$

where $H_{eff}(\theta)$ is given by Eq.(5.8) and $\Sigma_\theta := \theta E_\omega(\alpha)/4$. We note again that the approximation (2.7) breaks the translation invariance in the original Hamiltonian $H_{BP}$.
Let us fix \( r > 0 \) arbitrarily. We denote by \( \psi_j(x_j) \) the wave functions \( \sqrt{(1/\pi r^3)} \ e^{-|x_j|/r} \) for \( j = 1, 2 \). We define the wave functions \( \psi_r(x_1, x_2) \) with the parameter \( r \) by \( \psi_r(x_1, x_2) := \psi_1(x_1) \psi_2(x_2) \). We decompose \( H_{\text{eff}}(\theta) \) into the following:

\[
H_{\text{eff}}(\theta) = \alpha^2 \left\{ \frac{1}{2} p_1^2 - \frac{\sqrt{2}}{|x_1|} + \frac{1}{2} p_1^2 - \left( 1 - \frac{1}{\theta(1-\eta)} \right) \frac{1}{|x_1|} \right. \\
+ \left. \frac{\sqrt{2}}{\theta(1-\eta)} \left( \frac{1}{|x_1 - x_2|} - \frac{1}{|x_2|} \right)^2 \right\}.
\]

It is well known (see page 89 of Ref.56) that \( \langle \psi_r | |x_1 - x_2|^{-1} - |x_2|^{-1} |\psi_r \rangle \leq 0 \) is obtained since \( \psi_1(x_1) \) is spherically symmetric. Thus, the inequality \( \langle \psi_r | \alpha^2 H_{\text{eff}}(\theta) |\psi_r \rangle \leq \sqrt{2} (1/\theta (1-\eta) - 2)/r + 1/r^2 < -1 \) is obtained from the above decomposition and the inequality. Therefore, it follows from the HVZ theorem\(^\text{56} \) that \( H_{\text{eff}}(\theta) \) has a ground state under the condition \( (5.10) \).

The restrictions on \( \theta, \eta, \) and \( r \) are obtained by solving the inequality \( (5.10) \) with respect to the variable \( r \).

VI. SPATIAL LOCALIZATION IN STRONG-COUPLING REGIME

In this section we consider the approximated \( H_{\text{bp}}[\theta] \) given in Eq.(5.16). For a ground state \( \Psi_0 \) of \( H_{\text{bp}}[\theta] \), we define the radius \( u_{\text{bp}}(\Psi_0) \) of the sphere in which the two electron lives by

\[
u_{\text{bp}}(\Psi_0) := \max_{j=1,2} \left\{ \frac{\langle \Psi_0 | |x_j|^{-1} \langle \Psi_0 | |x_j| \rangle \langle \Psi_0 | |x_j| \rangle \rangle}{\langle \Psi_0 | |x_j| \rangle \langle \Psi_0 | |x_j| \rangle \rangle} \right\}.
\]

Then, we show in this section that if the bipolaron has a ground state \( \Psi_0 \), then there is a relation:

\[
u_{\text{bp}}(\Psi_0) \geq \frac{1}{\sqrt{2}} \left\{ 1 + \left( 1 + \frac{3\theta}{4} \right) \frac{E_{\text{w}}(\alpha)}{\alpha^2 \theta} \right\}^{-1} \left( \frac{1}{\theta(1-\eta)} - 2 \right).
\]

Thus, we are led to the conclusion that, even if \( \eta \) approaches 1, \( \theta > 0 \) in the strong-coupling regime works to stem growth of the \( u_{\text{bp}}(\Psi_0) \). This is a noticeable difference from the case of the weak-coupling regime \( (1.5) \).

We can show the above result in the same way as we did in Sec.IV. Adopting Lieb’s idea\(^\text{34} \) into our argument lead us the inequality,

\[
0 \geq -\alpha^2 u_{\text{bp}}(\Psi_0) - \sqrt{2} \alpha^2 + \frac{\sqrt{2} \alpha^2}{\theta(1-\eta)} \frac{|x_j|}{|x_1 - x_2|} - (4 + 3\theta) \sum_k |V_k|^2 u_{\text{bp}}(\Psi_0).
\]
Here we used the inequalities $E_{BP} \leq 0$ and $p_{j'}^2/2 - \sqrt{2}/|x_{j'}| \geq -1$ for $j' \neq j$. Since $|x_1 - x_2| \leq |x_1| + |x_2|$, we eventually obtain

$$0 \geq -\left\{2\alpha_0^2 + \left(2 + \frac{3\theta}{2}\right)E_w(\alpha)\right\}u_{BP}(\Psi_0) + \sqrt{2}\alpha_0^2\left(\frac{1}{\theta(1 - \eta)} - 2\right),$$

which implies the inequality (6.2).

**VII. POSITIVE BINDING ENERGY**

Combining the inequalities (3.3) and (5.4), we obtain a sufficient condition for the binding energy being positive. Namely, if $c_\ast$, $\theta$, and $\eta$ satisfy $c_{BP} > 2c_{SP}$, then the binding energy is positive, i.e.,

$$E_{BP} < 2E_{SP} \quad (7.1)$$

for sufficiently large $\theta > 0$ with the condition (5.5). Here, remember $c_{SP} = 0.108513\cdots$. Then, we note that $0 < c_{BP} \leq 0.4$ and $\lim_{c_\ast \to 1, \theta \to \infty} = 0.4$ under the condition (5.5). Moreover, the condition $c_{BP} > 2c_{SP}$ is equivalent to

$$\frac{2}{5}\left(c_\ast - \frac{1}{\theta(1 - \eta)}\right)^2 > 0.217024, \quad (7.2)$$

which has the form with corrections in the inequality (12) of Ref. 51. Thus, the condition (7.2) is almost equivalent to

$$1 - \frac{1}{0.2635\theta} > c_\ast - \frac{1}{0.2635\theta} > \eta. \quad (7.3)$$

As Smodyrev and Devreese pointed out, we must keep it in mind that the inequality (7.2) is a sufficient condition for the positive binding energy (7.1), not a necessary condition. If we take the effect of not only the leading terms of $c_{SP}$ and $c_{BP}$ but also their remainders into the condition (7.3), we should control $c_\ast$ so that $\eta$ and $\theta$ meet to the results by experiments.

According to the recent result of study of the Hamiltonians $\sum_{j}^H \bar{H}_{SP}^{(j)}$ and $\bar{H}_{BP}^{P=0}$, we might be able to choose $\inf_{\varphi,\langle \psi \rangle = 1} E_\theta(\varphi)$ as $c_{BP}$ (i.e., $-c_{BP} = \inf_{\varphi} E_\theta(\varphi)$) so that $2E_{SP} - E_{BP} = -(2c_{SP} - c_{BP})\alpha_0^2 > 0$ for sufficiently large $\theta > 0$. We note that this type of equation expressing the binding energy is pointed out by Vinetskii and Giterman.24,25
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