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Hartree-Fock approximation of bipolaron state in quantum dots and wires

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Abstract. The bipolaronic ground state of two electrons in a spherical quantum dot or a quantum wire with parabolic boundaries is studied in the strong electron-phonon coupling regime. We introduce a variational wave function that can conveniently conform to represent alternative ground state configurations of the two electrons, namely, the bipolaronic bound state, the state of two individual polarons, and two nearby interacting polarons confined by the external potential. In the bipolaron state the electrons are found to be separated by a finite distance about a polaron size. We present the formation and stability criteria of bipolaronic phase in confined media. It is shown that the quantum dot confinement extends the domain of stability of the bipolaronic bound state of two electrons as compared to the bulk geometry, whereas the quantum wire geometry aggravates the formation of stable bipolarons.

1 Introduction

Last three decades or so have witnessed a hectic pace of activity in the area of ultra-low-dimensional semiconductors the realization of which has now become possible with the advent of sophisticated micro-fabrication techniques. These structures provide a tiny laboratory to test the predictions of quantum mechanics and also have tremendous potentiality for applications in micro-electronic devices such as single-electron transistors, quantum-dot lasers, ultra-fast quantum computers and so on. As a natural consequence, extensive investigations have been carried out in this area in recent years, both theoretical and experimental, and a great deal of literature with extremely rich data for electronic, optical and magnetic properties has already piled up [1–6]. Recently, the role of electron-longitudinal- optical (LO) phonon interaction on various electronic properties of polar semiconductor quantum dots has been studied by a number of authors (see [7] and references therein) and one of the most important observations that has been made in this context is that the polaronic effects are extremely important in small dots and should therefore be taken into account when making devices with them. Several investigations have also been made to study the formation and stability of bipolarons in a quantum dot. A bipolaron is a bound pair of two polarons with a common cloud of virtual phonons and in the context of a quantum dot the bipolaron problem was first investigated by Mukhopadhyay and Chatterjee [8]. It was shown that in the strong coupling limit the confining potential of the quantum dot reduces the stability of the bipolaron. Essentially similar results were also observed by Senger and Erçelebi [9]. Pokatilov et al. [10] have investigated the stability of bipolarons in a spherical quantum dot with parabolic confinement by applying Feynman's variational principle and calculated the bipolaron binding energy, number of phonons in a bipolaron cloud and the bipolaron radius. They have shown that in a quantum dot bipolaron states are possible even for intermediate values of the electron-phonon coupling constant, α ($\alpha \sim 2$). They have also shown that the binding energy passes through a maximum for a certain value of the confinement length. Because of the conflicting conclusions obtained by different groups, the bipolaron problem in quantum dots has thrown up a new challenge to the theorists. Motivated by this controversy, Mukhopadhyay and Chatterjee (MC) [11] took up this problem and investigated the bipolaronic stability in realistic quantum dots using a variational method based on the Lee-Low-Pines-Huybrechts (LLPH) technique [12] and showed for the first time that stable bipolarons can indeed form in GaAs, CdS, CdSe and CdTe dots. One of the most important aspects of the investigation of MC is the incorporation of the Coulomb interaction between the two individual polarons in the unbound phase which was hitherto neglected. They have shown that inclusion of Coulomb correlation energy in the unbound phase brings in a dramatic and qualitative change in the results. Of course the

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exact form of the interaction potential for the two individual polarons in the unbound phase in a quantum dot is not known and MC had to resort to some approximation. Senger and Ercelebi [13] have made an investigation on the stability of a bipolaron in spherical quantum dots using a single hamiltonian which seems to be the correct way of dealing with the bipolaron problem in a confined system. They used a variational method and have obtained a broader range of stability than shown in [8]. The analysis of Senger and Ercelebi [13] however involves Hartree-like wave function for the two-electron system which includes the Coulomb correlation but does not incorporate the Pauli correlation. In the present paper we purport to make an improved calculation based on the Hartree-Fock approximation which takes into account the Coulomb correlation and also the proper antisymmetric behaviour of the two-electron wave function.

2 Theory

Using convenient polaron units $(m^\star = \hbar = \omega_{\rm LO} = 1)$ the Hamiltonian describing a confined electron pair coupled to LO-phonons of the medium is given by

$$H = H_e + \sum_Q a_Q^\dagger a_Q + \sum_{j=1,2}\sum_Q V_Q \left(a_Q e^{i\boldsymbol{Q}\cdot\boldsymbol{r}_j} + a_Q^\dagger e^{-i\boldsymbol{Q}\cdot\boldsymbol{r}_j}\right) \tag{1}$$

where

$$H_e = \sum_{j=1,2}\left[\frac{p_j^2}{2} + V_{\rm conf}(\boldsymbol{r}_j)\right] + \frac{U}{|\boldsymbol{r}_1 - \boldsymbol{r}_2|}. \tag{2}$$

In the above, a_Q ($a_Q^\dagger$) is the phonon annihilation (creation) operator, $\boldsymbol{r}_j$ and $\boldsymbol{p}_j$ $(j = 1, 2)$ are the positions and the momenta of the electrons. The interaction amplitude is related to the phonon wavevector $\boldsymbol{Q}$ through $V_Q = (2\sqrt{2}\pi\alpha)^{1/2}/Q$. It should be noted that using the polaron units corresponds to scaling all energy and length quantities respectively by $\hbar\omega_{\rm LO}$ and $(\hbar/m^*\omega_{\rm LO})^{1/2}$, where $\omega_{\rm LO}$ is the dispersionless frequency of LO phonons, and m^* is the effective mass of the electron. The dimensionless constants of the Coulomb interaction U and of the electron-phonon coupling α are related by the equation

$$U = \frac{e^2}{\epsilon_\infty} = \frac{\alpha\sqrt{2}}{1-\eta} \tag{3}$$

in which parameter $\eta = \epsilon_\infty/\epsilon_0$ is the ratio of the high frequency and static dielectric constants of the dot material.

We model the quantum dot with an anisotropic harmonic oscillator having cylindrical symmetry

$$V_{\rm conf}(\boldsymbol{r}) = \frac{1}{2}\left(\Omega_\rho^2\rho^2 + \Omega_z^2 z^2\right), \tag{4}$$

which describes a spherical dot for $\Omega_\rho = \Omega_z$ and conforms to a quantum wire when Ω_z is set to zero.

In the adiabatic approximation of the polaron theory the total wave function of the two polarons is written separable in the phononic and electronic parts,

$$\Psi_{\rm bipol} = \Phi(\boldsymbol{r}_1, \boldsymbol{r}_2)\, e^{\mathsf{S}}\,|0\rangle, \quad \text{with } \mathsf{S} = \sum_Q f_Q(a_Q - a_Q^\dagger) \tag{5}$$

where $|0\rangle$ denotes the phonon vacuum state, and the displaced-oscillator transformation e^{S} yields the most efficient lattice distortion around the electrons through the variational terms f_Q. Minimization of $\langle\Psi_{\rm bipol}|H|\Psi_{\rm bipol}\rangle$ with respect to f_Q yields $f_Q = V_Q s_Q$, where

$$s_Q = \frac{1}{2}\left\langle \Phi \left| \sum_{i=1,2}\left(e^{i\boldsymbol{Q}\cdot\boldsymbol{r}_i} + e^{-i\boldsymbol{Q}\cdot\boldsymbol{r}_i}\right)\right| \Phi \right\rangle, \tag{6}$$

such that the ground state energy of the two polaron complex is calculated by optimizing the expression

$$E_{\rm g} = \langle\Phi|H_e|\Phi\rangle - \sum_Q V_Q^2 s_Q^2 \tag{7}$$

in terms of the variational parameters contained in Φ.

The form of the electronic part of the trial wave function is chosen flexible enough to describe alternative ground state configurations of the two polaron system at once. These are the bipolaronic bound state, the state of two spatially separated individual polarons, and in particular for the case of small quantum dots, the state of two nearby polarons that are not in the bipolaronic bound state but yet confined within the same dot. Compatible with the adiabatic approximation and the form of the confinement potentials we write the two-electron wave function in terms of Gaussian functions. For the ground state since the electrons are assumed to be in spin singlet state the spatial part of the two-body wave function with the correct exchange symmetry has the form:

$$\begin{aligned}\Phi(\boldsymbol{r}_1, \boldsymbol{r}_2) \sim |\boldsymbol{r}_1 - \boldsymbol{r}_2| \Big[& G_{a,b}\left(\boldsymbol{r}_1 - \frac{1}{2}\boldsymbol{r}_0\right) G_{a,b}\left(\boldsymbol{r}_2 + \frac{1}{2}\boldsymbol{r}_0\right) \\ & + G_{a,b}\left(\boldsymbol{r}_1 + \frac{1}{2}\boldsymbol{r}_0\right) G_{a,b}\left(\boldsymbol{r}_2 - \frac{1}{2}\boldsymbol{r}_0\right)\Big]\end{aligned} \tag{8}$$

where we combine two anisotropic Gaussian functions $G_{a,b}(\boldsymbol{r}) = e^{-a^2\rho^2 - b^2 z^2}$ centered symmetrically around the origin at $+\frac{1}{2}\boldsymbol{r}_0$ and $-\frac{1}{2}\boldsymbol{r}_0$, where a and b are variational parameters. The factor $|\boldsymbol{r}_1 - \boldsymbol{r}_2|$ sets up a Coulomb correlation between the electrons which gets more important for nearby polarons. The separation $|\boldsymbol{r}_0|$ between the centers of the two Gaussians serves as a measure of the distance between the two electrons. Without loss of generality the electrons can be assumed to be located along the z-axis, i.e. $\boldsymbol{r}_0 = r_0\hat{z}$, when the confinement potential is weaker along z $(\Omega_\rho \geq \Omega_z)$. We expect to retrieve an upper bound to the ground state of the two polarons by tracing the value of r_0 from zero to infinity. For instance in bulk, the state of two independent polarons is achieved by $r_0 \to \infty$ and $a = b$. As we start decreasing the separation between the polarons the total energy is expected to increase due

to the Coulomb repulsion between the electrons until the polarization fields of the polarons start to overlap. Depending on the material parameters if a bound state of the electrons is favorable, the energy of the system will display a local minimum at a finite (or zero) value of r_0, where the energy of the bipolaronic state will be lower than twice the energy of a single polaron. The current formulation of the problem will also provide an answer regarding the structural form of the bipolaronic ground state; whether it leads to a one-center (like He atom) or a two-center (like H_2 molecule) charge distribution of the electrons in the bipolaronic state.

Transforming to center-of-mass, $\boldsymbol{R} = (\boldsymbol{r}_1 + \boldsymbol{r}_2)/2$, and relative, $\boldsymbol{r} = \boldsymbol{r}_1 - \boldsymbol{r}_2$, coordinates the wave function can be rewritten in the form

$$\begin{aligned} \Phi(\boldsymbol{R}, \boldsymbol{r}) &= \psi(\boldsymbol{R})\phi(\boldsymbol{r}) \qquad (9)\\ \psi(\boldsymbol{R}) &= N_R\, e^{-2a^2 R_\rho^2}\, e^{-2b^2 R_z^2} \\ \phi(\boldsymbol{r}) &= N_r\, r\, e^{-\frac{1}{2}a^2\rho^2} \left(e^{-\frac{1}{2}b^2(z+r_0)^2} + e^{-\frac{1}{2}b^2(z-r_0)^2} \right) \end{aligned}$$

where N_R, N_r are normalization constants. The transformed forms of electronic part of the Hamiltonian and the s_Q terms are written as

$$H_e = -\frac{1}{4}\nabla_R^2 - \nabla_r^2 + \frac{U}{r} + V_{\mathrm{conf}}(R, r) \qquad (10)$$

$$s_Q = \langle\psi|e^{\pm i\boldsymbol{Q}\cdot\boldsymbol{R}}|\psi\rangle\, \langle\phi|(e^{i\boldsymbol{Q}\cdot\boldsymbol{r}/2} + e^{-i\boldsymbol{Q}\cdot\boldsymbol{r}/2})|\phi\rangle \qquad (11)$$

The minimization of the variational energy E_{g} (7) with respect to the parameters a and b provides us with an upper bound of the ground state energy as a function of the separation of polarons, r_0.

3 Results and discussion

In the adiabatic approximation of the polaron problem all energy values can be scaled by α^2, which corresponds to scaling the lengths by $1/\alpha$ (a measure of polaron size). In the following we find that scaling convenient to display our numerical results [13].

We start with the bulk description of the problem. In Figure 1 we plot the energy profiles of the two-polaron complex in the bulk medium for three selected values of the material parameter η. Each curve presents two local minima as functions of the interpolaron distance r_0. The one at smaller values of r_0 is identified as the bipolaron state. The state of two independent polarons shows up as the asymptotic minimum occurring for diverging r_0, with the well-known energy, $E_{\mathrm{g}}/\alpha^2 = -2/3\pi = -0.2122$. The two minima are separated by an energy barrier, which shows that formation of bipolaron in a metastable state is possible even if its energy is higher than the energy of two polarons.

The present Landau-Pekar strong coupling theory is known to yield ground state energies proportional to α^2 for both polaron and bipolaron states therefore it does not provide a critical α value for the bipolaron stability. Effective strength of the Coulomb interaction (η) has a

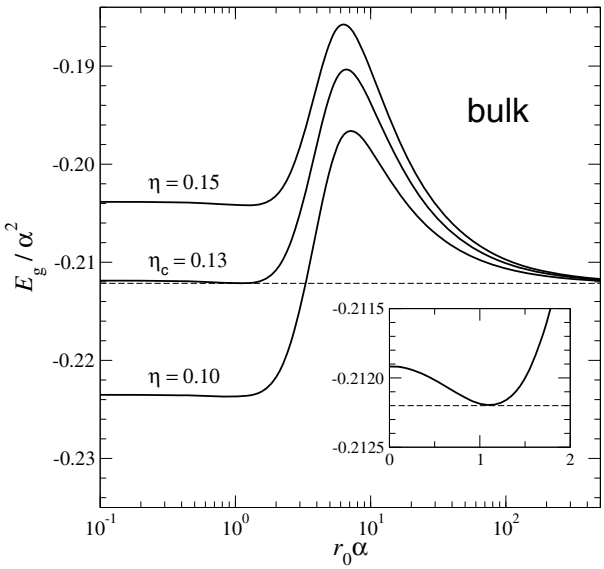

Fig. 1. Variation of the ground state energy of the two-polaron system as a function of the interpolaron distance, in bulk. The inset magnifies the bipolaronic minimum region in linear scale.

critical value determined by the comparison of the energies of bipolaron bound state and state of two polarons, where the condition for a stable bipolaron state is $\eta < \eta_c$. In Figure 1 we determine that critical value as $\eta_c = 0.130$, which is slightly less than the value we calculated previously (0.131) using a simpler wavefunction [9,13]. It is also close to value of 0.115 that was obtained in a variational analysis of an intermediate-coupling bipolaron [14].

The inset in Figure 1 portrays the variation of ground state energy around the bipolaronic minimum. The minimum occurs around a point where the centers of Gaussian charge distributions of the polarons are separated by about a polaron size, $r_0 \sim 1/\alpha$. Therefore we have a two-center configuration of bipolaron state. One should keep in mind that even though the variational principle is expected to provide reasonably accurate energy upper bounds, validity of predictions regarding the form of the wavefunction and hence the charge density is limited by the approximations made to the exact wavefunction. In our previous treatments of bulk and two-dimensional bipolaron ground states, the variational energy upper bound to the ground state energy was obtained for $r_0 = 0$, which corresponds to a one-center configuration [13,15]. The origin of the apparent discrepancy between the conclusions of our previous and present calculations on the symmetry of the ground state lies in the degree of flexibility introduced to the variational wavefunctions. In the former cases, after transforming the bipolaron wavefunction constructed from one-electron Gaussian functions to the center-of-mass and relative coordinates (as in obtaining Eq. (9) starting from Eq. (8)), the number of variational parameters commonly appearing in both the center-of-mass and relative coordinate components of the wavefunction were doubled by assigning independent parameters in each component. This allowed a further flexibility to the variational wavefunction which lowers the energy values in the small r_0 domain,

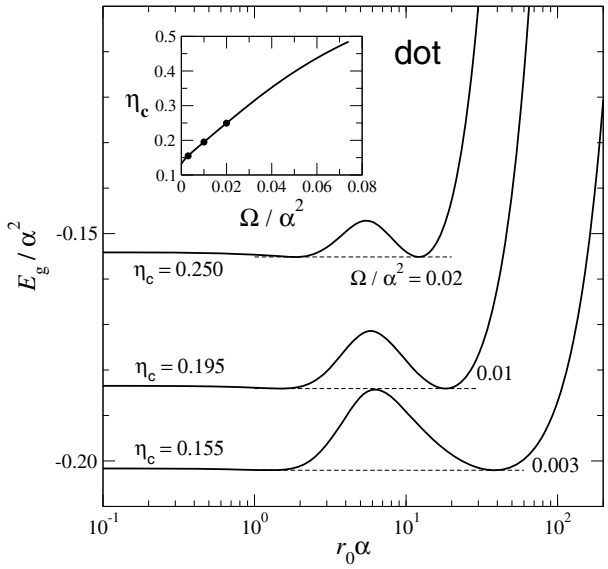

Fig. 2. Variation of the ground state energy of the two-polaron system as a function of the interpolaron distance, in a spherical dot. The scaled strength of the confining potential and the corresponding critical η values are explicitly written. The inset shows the variation of η_c with confinement strength, where the dots on the curve corresponds to three cases displayed in the figure.

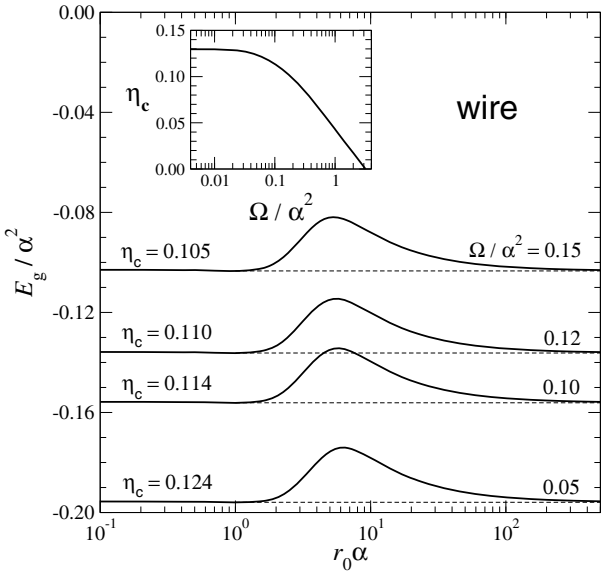

Fig. 3. Variation of the ground state energy of the two-polaron system as a function of the interpolaron distance, in wire. The scaled strength of the confining potential and the corresponding critical η values are explicitly shown. The inset shows the variation of η_c with wire size.

smearing out the local energy minimum at finite separations, and eventually leading to a one-center bipolaron state. A plausible argument in this context has been provided in reports by Mukhomorov [16,17], where the bipolaron ground state has been found in a two-center form as in the present work. The argument is based on the necessity of imposing additional constraints (such as virial theorem) in the variational optimization process [17], and leads to a conclusion that a one-center bipolaron ground state is unstable. In view of this argument, the form of the variational wavefunctions introduced in [13] and [15] might not be compatible with the virial theorem. The two-center structure of the bipolaron ground state is also supported by an analysis of experimental findings [16].

Next we consider the effect of a spherical parabolic confinement potential ($\Omega_\rho = \Omega_z = \Omega$) on the stability of bipolaron state. A parabolic potential of the form $\frac{1}{2}\Omega^2 r^2$ imposes a perfect confinement for the two polarons within the quantum dot for all nonzero values of Ω. In Figure 2, the energy profiles as functions of r_0 for various Ω values lack the asymptotic minimum corresponding to two unbound polarons. Instead of that now we have an energy minimum belonging to a state of two polarons repelled by their Coulomb interaction but kept at a finite separation by the barriers of the dot potential. Technically that bound state can be viewed as a bipolaron with a larger radius, however, it may be more appropriate to describe it as "the state of two interacting polarons confined within the same dot", which has the limit of "state of two noninteracting polarons separated to infinity" as Ω tends to zero. The attractive interaction between the polarons due to the overlap of their phonon fields rapidly decreases as the interpolaron distance gets larger than the polaron size. The critical value of Coulomb repulsion strength η for a given Ω is calculated by using the condition that the energies of the two local minima at different r_0 values are equal. Energy of that state with a large but still finite r_0 is certainly higher than that of two unbound polarons in bulk. Therefore, compared to bulk the confinement potential provides a larger η_c value for stable bipolarons through elevating the energies of the both alternative states of the two polarons in a spherical dot. For instance, the η_c value of 0.250 for $\Omega/\alpha^2 = 0.02$ almost doubles over its bulk value of 0.130. The inset of Figure 2 where we plot the calculated variation of η_c with the scaled potential strength shows the broadened domain of stable bipolarons in a quantum dot confinement.

Unlike quantum dot confinement quantum wire geometry ($\Omega_z = 0$, $\Omega_\rho = \Omega$) reduces the range of η values that admits a stable bipolaronic state. As shown in Figure 3 the energies of the bipolaron state and the state of the unbound polarons in a quantum wire levels at smaller and smaller values of η as the effective confinement strength is increased. η_c monotonically decreases with Ω and eventually vanishes for $\Omega/\alpha^2 = 3.199$ (see the inset of Fig. 3).

4 Conclusions

We have considered the problem of bipolaron formation and stability in parabolic quantum dots and wires by using a variational method in the limit of strong electron-phonon coupling, and a wave function that can conform to both the bipolaronic bound state and the state of two separated polarons. The variational wavefunction we propose admits a bipolaronic bound state at a finite separation

(about a polaron size) of two polarons. The quantum dot confinement extends the domain of stability of the bipolaronic bound state of two electrons as compared to the bulk geometry, whereas the quantum wire geometry aggravates the formation of stable bipolarons.

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