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On the stability of Fröhlich bipolarons in spherical quantum dots

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Abstract

In the strong-electron–phonon-coupling regime, we retrieve the stability criterion for bipolaron formation in a spherical quantum dot. The model that we use consists of a pair of electrons immersed in a reservoir of bulk LO phonons and confined within an isotropic parabolic potential box. In this particular quasi-zero-dimensional geometry, where the electrons do not have any free spatial direction to expand indefinitely, a plausible approach would be to treat the electrons either to form a bipolaronic bound state or enter a state of two close, but individual polarons inside the same dot. The confined two-polaron model adopted here involves the polaron–polaron separation introduced as an adjustable parameter to be determined variationally. It is found that the fundamental effect of imposing such a variational flexibility is to modify the phase diagram to a considerable extent and to sustain the bipolaron phase in a broader domain of stability.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Over the last few decades, proceeding the progress in the growth technology of mesoscopic nanostructures and the discovery of high-temperature superconductors, there has appeared a revived and extensive interest in the Fröhlich bipolaron concept of Pekar [1] and Vinetskii and Gitterman [2]. As one of the pioneering works published within the context of two-polaron systems, we should also mention the paper by Bishop and Overhauser [3] where they study the phonon-mediated interaction between a pair of electrons and show that, for ionic materials, the effective electron–electron potential may lead to an attractive deep potential well with a minimum occurring for particle separations as close as a few tens of Ångstrom units. In selective materials, provided the effective Coulomb repulsion is not larger than a critical strength and the electron–phonon coupling is not smaller than a critical value, the lattice effects may account for a considerable part of the energy of the electron–electron pair which consists of attracting the particles against their Coulomb repulsion. Thus, the fundamental condition under which a bipolaronic bound state can form is that the repulsive Coulomb interaction should

not be so strong as to dominate over and hence break up the phonon-mediated binding which holds the particles together. Among the numerous papers devoted to the study of the formation and stability of bipolarons, we cite some examples [4–31] which are relevant to the long-range Fröhlich interaction with the optical branch of the phonon spectrum. Adopting different models and approximating theories, several aspects of the polaron–bipolaron transition have been studied rather extensively as functions of the Coulomb and phonon coupling strengths in three, two [20–24, 30], and even lower dimensionalities pertaining to quantum wires and dots [25–31]. For an overview of the polaron concept and a brief discourse on bipolaron formation the reader is referred to the review article by Devreese [19].

Of particular relevance to the content of the present article are the recent solutions for the bipolaron state in quantum dots [27–31]. In most of the preceding works, including the study of bipolarons in quantum dots, the phase boundary of the bipolaron stability region is determined by the inequality

$$E_{\text{bipol}} - 2E_{\text{p}} = \varepsilon \leqslant 0, \tag{1}$$

where E_{bipol} and E_{p} denote, respectively, the bipolaron and one-polaron ground-state energies calculated in identical quantum dots. In statistical arguments [29], if one were to imagine an ensemble of n_0 quantum dots and n electrons ($n_0 > n$) in thermodynamical equilibrium, the inequality $\varepsilon < 0$ would conceptually mean a higher population of quantum dots with two electrons (bipolarons) than those containing only one electron (polarons). Relying on the stability criterion based essentially on this inequality, Mukhopadhyay and Chatterjee [27] and the present authors [30] have reached the conclusion that for strong-coupling polarons the role of the geometric confinement is to disfavour the bipolaron stability in small quantum dots. An outcome along the same lines has been reported by Pokatilov *et al* [29] under the Feynman variational principle. In their plots of the electron–phonon coupling constant versus the dot size we note that, even for fairly weak Coulomb coefficients, the value of the coupling constant below which the bipolaron phase is unfavourable increases inevitably when the dot radius is made smaller than the size of a polaron. This qualitative trend has been re-emphasized by Chatterjee and Mukhopadhyay [31], where they explicitly note that at strong coupling, below a critical value of the confinement length, the bipolaron becomes unstable and breaks up into two individual polarons. Finally, as a counter-view to the outcomes of these papers, we should necessarily include in our reference list the contravening paper by Wan *et al* [28] treating the zero-dimensional bipolaron problem within the framework of the path-integral formulation and leading to the finding that the confinement effects act in favour of bipolaronic stability in the zero-dimensional configuration. Moreover, it has been argued that, even at weak phonon coupling, bipolarons can form in quantum dots under strong confinement.

In view of the diverging results outlined above we are tempted to review the effect of the geometric confinement on the two-polaron problem where we make a small but qualitatively distinguishing digression in the approach to the determination of the domain of bipolaron formation. Instead of demanding that one must have a deeper energy for the bipolaron state than for two noninteracting polarons in similar individual quantum dots, we base our comparison of the bipolaron energy with reference to the energy of two off-centre polarons contained essentially in the same quantum dot by the ever-growing parabolic barriers of the confining potential. Stating this alternatively, we demand that one must distinguish between whether the electron pair is localized within the dot dominantly by the phonon-induced attraction or else dominantly by the geometric confinement. We therefore think that a sensible approach will be to find out whether the polarons confined in the quantum dot are ‘on-centre’ or ‘off-centre’ polarons. The underlying reason which makes us tempted to modify the stability criterion in such a sense is that in the dot-type confinement the electrons do not have any free

spatial direction to expand and this intrinsic feature sets the zero-dimensional case apart from all the higher-dimensional geometries where the electrons are free along at least one spatial direction. Thus, in the peculiar case of an electron pair in a quantum dot the alternative of the bipolaronic bound state is the case in which the phonon-mediated binding breaks up and the electrons are repelled away in opposite directions from their common centre of mass, yet still held in the close proximity of one another by the geometric confinement. Consequently, one has a description consisting of two individual but nearby polarons constrained to orbit within the dimensions allowed by the barriers of the dot potential. Therefore, unlike the higher-dimensional configurations, the polarons can by no means be projected apart indefinitely, but remain in finite separation with optimal relative position coordinates which we intend to determine 'variationally' in the following calculations. In what follows we review the problem of the two-polaron complex in a quantum dot through a distinctive discourse diverting from the commonly used methodology founded on contrasting the bipolaron state with the state of one single polaron in a similar dot of the same size and of identical material parameters. Clearly, for shallow confinement potentials ($\Omega \approx 0$) the present treatment is expected to duplicate the same physical issue as tackled formerly [30] by the approach which uses equation (1) as the stability criterion.

We adopt an oversimplified model of a pair of electrons immersed in a reservoir of bulk LO phonons and confined within a three-dimensional isotropic box of parabolic boundary strengths, given by $V_{\rm conf}(r) = \frac{1}{2}\Omega^2 r^2$, in which the dimensionless frequency Ω serves for the measure of the degree of confinement of the electrons. The particular parabolic form of the potential used here is appealing in the sense that the static depletion fields achieved in microstructures such as quantum wires and dots which are laterally confined by Schottky gates exhibit nearly parabolic potentials. Besides this, the usage of the quadratic confinement potential greatly facilitates the calculations and leads to tractable analytic expressions due to its compatibility with the harmonic charge density fluctuations of the electrons in the field of phonons. The peculiar trait posed by the confinement potential model used here is that the unlimited increasing barriers of the parabolic potential restrict the pair of polarons to remain persistently inside the same dot rather than allowing the electrons to make transitions to individual one-polaron states in bulk medium or in separate dots juxtaposed in the material. The present model could therefore be considered as aiming to provide presumably an appropriate insight into the problem of two deeply bound electrons localized in a dot intended to form a bipolaron state.

We should emphasize that the fundamental approach followed in this work is to take into account solely the generic low-dimensional aspect of the dynamical behaviour of the confined electrons and visualize them as interacting with the medium and with one another through exchange of virtual phonons. Thus, adopting the bulk-phonon approximation and leaving out all the other complicating detailed features, we exemplify an alternative viewpoint to the criterion for bipolaron stability where we choose to search for the variational energy minima pertaining to the distinguishing states of the so-named on- or off-centre polarons and via this distinctive approach we obtain a tracking of the polaron–bipolaron phase boundary as a function of the degree of confinement.

In our treatment of the Fröhlich interaction we choose to use the strong-coupling-polaron approximation, since realization of bipolarons requires large values of the electron–phonon coupling constant so as to sustain the phonon-mediated binding and withstand against the strong repulsive Coulomb interaction. Fortunately, the adiabatic theory we follow is motivated by the recent advances in producing nanocrystals with strong ionic coupling [32].

2. Theory

The Hamiltonian describing the confined electron pair system coupled to LO phonons is given in the usual polaron units ($m^\star = \hbar = \omega_{\rm LO} = 1$) by

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

where

$$H_e = \frac{1}{2}\sum_{j=1,2}(p_j^2 + \Omega^2 r_j^2) + \frac{U}{|\vec{r}_1 - \vec{r}_2|}. \tag{3}$$

In the above, a_Q ($a_Q^\dagger$) is the phonon annihilation (creation) operator and $\vec{r}_j$ ($j = 1, 2$) are the positions of the electrons. Similarly, p_j ($j = 1, 2$) denote the respective momenta. The interaction amplitude is related to the phonon wavevector $\vec{Q}$ through $V_Q = (2\sqrt{2}\pi\alpha)^{1/2}/Q$. The dimensionless constants of the Coulomb interaction U and of the electron–phonon coupling α are related by the equation [19]

$$U = \frac{e^2}{\epsilon_\infty} = \frac{\alpha\sqrt{2}}{1-\eta} \tag{4}$$

in which parameter η is the ratio of the high-frequency and static dielectric constants of the material, given by

$$\eta = \epsilon_\infty/\epsilon_0 < 1. \tag{5}$$

In the present variational approximation we adopt the conventional trial ansatz of the adiabatic polaron theory, where we write the total wavefunction to consist of a part relevant to the phonon variables and a part which contains the particle coordinates only, i.e.

$$\Psi_{\rm bipol} = \Phi(\vec{r}_1, \vec{r}_2)|0\rangle, \tag{6}$$

where $|0\rangle$ denotes the phonon vacuum state.

Here, we need to use an appropriate form for the electronic part of the trial wavefunction (6) that is suitable to reflect a unifying description of the two-polaron complex over a range uncovering both the bipolaronic bound state and the state of two unbound but nearby polarons. As a sensible approximation, we construct $\Phi(\vec{r}_1, \vec{r}_2)$ in a product form consisting of two similar 'shifted' one-electron Gaussian wavefunctions, i.e.

$$\Phi(\vec{r}_1, \vec{r}_2) = g(|\vec{r}_1 - \vec{r}_2|)G_\lambda(\vec{r}_1 - \tfrac{1}{2}\vec{r}_0)G_\lambda(\vec{r}_2 + \tfrac{1}{2}\vec{r}_0), \tag{7}$$

where

$$G_\lambda(\vec{x}) = N_\lambda \exp(-\tfrac{1}{2}\lambda^2 x^2), \tag{8}$$

and $g(|\vec{r}_1 - \vec{r}_2|)$ is the Coulomb correlation function of Jastrow type. Parameters λ and $\vec{r}_0$ ($\sim$the distance between the centres of the Gaussians) will be treated as adjustable parameters.

The bipolaron state, $\Psi_{\rm bipol}$, thus constructed, is to be used in concert with two successive displaced-oscillator transformations intended to yield the most efficient lattice distortions centred at $\pm\frac{1}{2}\vec{r}_0$, about which the electrons are assumed to exhibit rapid harmonic charge density fluctuations. We thus set

$$\begin{aligned} &\mathsf{H} \rightarrow S_2^{-1}S_1^{-1}\mathsf{H}S_1S_2 \\ &S_n = \exp\sum_Q V_Q s_Q\{a_Q \mathrm{e}^{(-1)^n \mathrm{i}\frac{1}{2}\vec{Q}\cdot\vec{r}_0} - a_Q^\dagger \mathrm{e}^{-(-1)^n \mathrm{i}\frac{1}{2}\vec{Q}\cdot\vec{r}_0}\}, \end{aligned} \tag{9}$$

in which s_Q is calculated variationally to yield the optimal value

$$s_Q = \left\langle \Phi(\vec{r}_1, \vec{r}_2) \left| \tfrac{1}{2} \sum_{j=1,2} (\mathrm{e}^{\mathrm{i}\vec{Q}\cdot\vec{r}_j} + \mathrm{e}^{-\mathrm{i}\vec{Q}\cdot\vec{r}_j}) \right| \Phi(\vec{r}_1, \vec{r}_2) \right\rangle. \tag{10}$$

A further optimization of

$$E_g \equiv \langle \Phi(\vec{r}_1, \vec{r}_2) | H_e | \Phi(\vec{r}_1, \vec{r}_2) \rangle - \sum_Q V_Q^2 s_Q^2 \tag{11}$$

with respect to the variational parameters contained in $\Phi(\vec{r}_1, \vec{r}_2)$ corresponds to the self-trapping description of the polarons where the charge density of the electrons and the lattice polarization influence each other in such a way that a stable relaxed state of the 'two-electron + phonon' complex is eventually attained. In the following we shall identify the case for which $\vec{r}_0$ turns out to be zero as referring to the bipolaron phase where the two Gaussians map identically on one another, positioned centrally at the origin and clothed entirely in common by the lattice deformation.

Transforming to a representation in the centre of mass and relative coordinates,

$$\vec{R} = \tfrac{1}{2}(\vec{r}_1 + \vec{r}_2), \qquad \vec{r} = \vec{r}_1 - \vec{r}_2, \tag{12}$$

with corresponding momenta $\vec{P} = \vec{p}_1 + \vec{p}_2$ and $\vec{p} = \frac{1}{2}(\vec{p}_1 - \vec{p}_2)$, the product wavefunction (7) conforms to a form separable in the centre of mass and relative coordinates, each part having an oscillator-type waveform with distinctive spatial extents, given by

$$\Phi(\vec{R}, \vec{r}) = g(r) G_a(\vec{R}) G_b(\vec{r} - \vec{r}_0). \tag{13}$$

We should remark that the oscillator–oscillator wavefunction that we have adopted proves to be a feasible approximation [22] which, on the other hand, lends a great amount of computational simplification due to its compatibility with the quadratic barriers of the confining potential.

In this representation the essential terms H_e and s_Q involved in equation (11) are rewritten as

$$H_e = \frac{1}{4} P^2 + p^2 + \Omega^2 R^2 + \frac{1}{4} \Omega^2 r^2 + \frac{U}{r} \tag{14}$$

and

$$s_Q = \langle \Phi(\vec{R}, \vec{r}) | 2 \cos(\tfrac{1}{2} \vec{Q} \cdot \vec{r}) \exp(\pm \mathrm{i} \vec{Q} \cdot \vec{R}) | \Phi(\vec{R}, \vec{r}) \rangle. \tag{15}$$

In our calculations we set the correlation function simply as $g(r) = r$. Such a choice for the Jastrow factor has proved to yield a comparatively lower variational upper bound to the ground-state energy for the bipolaron [22]. The explicit analytic form of the variational energy E_g (11) is given through a series of closed but somewhat lengthy and tedious expressions which we refrain from listing here. It should be noted that in the process of obtaining the optimal fits to a, b and $\vec{r}_0$ the variational theory is expected to set up a detailed fractional admixture of all the contributions from each single parameter (U, α and Ω) characterizing the system.

3. Results and conclusions

Since analytical minimization of E_g (11) is not possible, the determination of the optimal fits to the variational parameters a, b and $\vec{r}_0$ requires numerical treatment. In the following discussions we shall devote special emphasis to the role which the particular parameter $\vec{r}_0$ plays in the theory.

With regard to the strong-coupling expansion to leading order in α, i.e. $\mathcal{O}(\alpha^2)$, we would like to draw attention to the fact that, if in equations (2) and (3) the energies are scaled by α^2

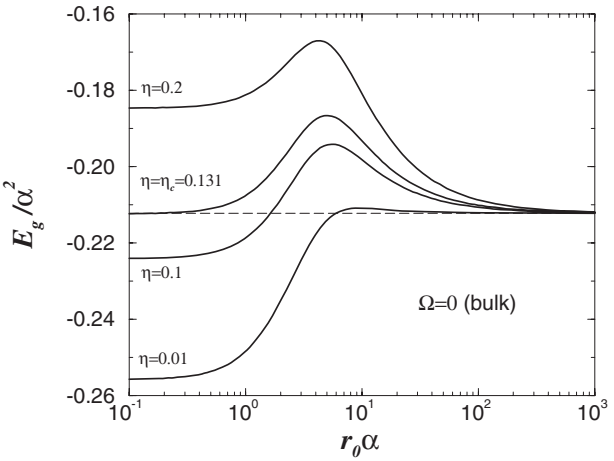

Figure 1. The ground-state energy of the two-polaron complex as a function of the variational parameter $\vec{r}_0$ for a succession of different η values in the bulk ($\Omega = 0$) limit. The dashed line represents the corresponding energy of two strongly coupled free polarons: $E_g/\alpha^2 = -2/(3\pi)$.

and lengths by α the only modification in the Hamiltonian would be to replace the confining parameter Ω by Ω/α^2 and the Coulomb coefficient U by U/α. Thus, in a representation where the critical value of parameter η is plotted as a function of Ω/α^2, rather than bare Ω, we find that, irrespective of the value of α, one can display the phase boundary on a single universal curve for any degree of confinement. In every case, whatever the value of Ω is, the ground-state energy is seen to be proportional to the square of the coupling constant, i.e. $E_g = -\mathcal{C}\alpha^2$, where the corresponding coefficient of proportionality bears a functional relation solely to Ω/α^2. Therefore, in the foregoing particular plots for which the confinement parameter Ω is expressed in the ratio Ω/α^2, one can conveniently assign α any arbitrary value with no loss in generality.

To visualize the essential role which the variational parameter $\vec{r}_0$ in the shifted oscillator waveform (13) plays in the theory, we find it instructive first to provide a comprehensive display of the variation of E_g over a broad range of this parameter in the bulk limit, which is easier to understand. Setting $\Omega = 0$ and holding η (and hence U) fixed at four distinctive values, we plot the ground-state energy of the two-polaron complex as a function of r_0 (figure 1). Following the series of plots drawn for each fixed η, we note that there show up two asymptotic minima in the energy profiles located at $r_0 = 0$ and $r_0 \gg 1$, the former yielding an indication of a bipolaron state, the latter representing the state of two infinitely separated noninteracting free polarons each with energy

$$\frac{E_g}{\alpha^2} = -\mathcal{C}\left(\frac{\Omega}{\alpha^2}\right) = -\frac{1}{3\pi} \approx -0.1061.$$

Very recently, energy profiles of similar qualitative nature, separating one state from the other by a barrier, have been reported by Mukhomorov [18] in a study of the bulk-bipolaron in the intermediate-coupling regime, where it has been concluded that the spatial structure of the bipolaron is that of an axially symmetric quasi-molecular dimer. However, within the framework of the adiabatic calculation adapted in the present work, the bipolaron ground state is found to be spherically symmetric, characterized by the vanishing value of the parameter r_0. We should also note that the current upper bounds for the bipolaron ground-state energy are observed to lie lower than those given in [18].

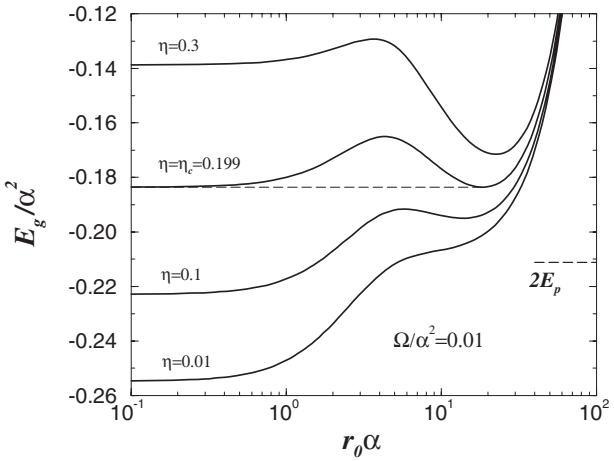

Figure 2. The ground-state energy of the two-polaron complex as a function of the variational parameter $\vec{r}_0$ for a succession of different η values in the parabolic quantum dot where $\Omega/\alpha^2 = 0.01$. The short dashed line by the right margin refers to twice the total energy of one polaron in a similar dot derived from the paper by Yıldırım and Erçelebi [33].

Within the content of figure 1 we observe further that the minima on the left margin which lie below the dashed horizontal line ($E_g/\alpha^2 = -0.2122$) provide evidence in favour of energetically stable bipolaron states, and those which lie above are to be recognized as representing metastable states of the bipolaron. Tuning η to even larger values, this corresponding minimum starts to lose its prominence and eventually diminishes, signifying that beyond a certain strength of the Coulomb coefficient the only configuration the electrons can form is the distantly spaced state of free polarons. We find that the particular value of η for which the two minima have the same level along the r_0 axis is $\eta_c^{(3D)} = 0.131$, as calculated previously in [22] and [30] in three dimensions using the same oscillator–oscillator-type waveform for the electrons.

Imposing the geometric confinement, one again encounters in general two minima in the ground-state energy profiles, one of which is attained asymptotically at $r_0 = 0$, reflecting the bipolaron phase, i.e. the on-centre configuration of the two polarons. Referring to the curves in figure 2 plotted for $\Omega/\alpha^2 = 0.01$ and for not too weak η, we observe the second minimum to occur not at infinity as in three dimensions but, instead, at a finite value of r_0, somewhere near $r_0\alpha \simeq 20$. For this ratio of Ω to α^2, the critical value of η at which the ground-state energies of the on-centre and off-centre configurations cross over is found to be $\eta_c = 0.199$, a value somewhat above the three-dimensional value due to the pseudo-enhancement of the effective phonon coupling under confinement. At this critical η the distance between the mean coordinates of the electrons in the off-centre configuration is calculated to be $r_0 \simeq 20/\alpha$, amounting to a value of two when $\alpha = 10$. The value of r_0 is seen to increase slightly as η is adjusted to larger values, since the strengthened Coulomb repulsion separates the particles with greater repulsion against the confining potential barriers. A careful glance at the series of plots in the figure reveals that there are three major domains of interest. In the opposite regimes of large and small η, one encounters only one of the two minima pertaining to solely either the off-centre (two polarons: PP) or the on-centre (bipolaron: BP) phases. For intermediate values of η the two phases are seen to coexist where either the bipolaron state is energetically more stable and the off-centre two-polaron configuration is metastable, or vice versa, depending on whether $\eta < \eta_c$ or $\eta > \eta_c$.

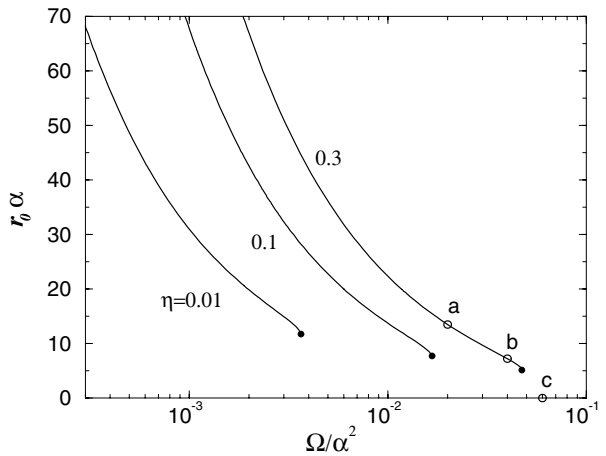

Figure 3. The optimal value of $\vec{r}_0$ as a function of the confining parameter Ω for $\eta = 0.01, 0.1, 0.3$. The bold dots at the tip of each curve give the respective points at which the minima relevant to the off-centre configuration of the polarons diminish. The open circles a, b and c represent the respective points for which $\Omega = 0.5$, 1.0 and 1.5 when $\alpha = 5$ and $\eta = 0.3$.

As an additional note in regard to the content of figure 2, we would like to include the remark that if the barriers of the confining potential, instead of growing indefinitely in the radial directions, were of limited height, then a further configuration in which the electrons could possibly relax themselves would be in the form of separate polarons in the bulk medium outside the dot. Moreover, in an alternative structure of an array of dots separated by high potential barriers, it might also turn out to be likely to have the electrons make transitions into the neighbouring dots and form one-polaron bound states with one electron in one dot. Such a possibility is depicted by the fact that the corresponding ground-state energy of a pair of polarons in similar separate dots (shown by the dashed line on the right margin) may turn out to be the deepest of all the configurations (see, for instance, the curve for $\eta = \eta_c$). This particular situation has already been mentioned by Pokatilov *et al* [29]. Such cases, however, lie beyond the content of the present study, where we take the electron pair as confined strictly within the increasingly steeper barriers of the parabolic potential box.

In figure 3 we plot three simultaneous curves describing the effect of confinement on the optimal value of $\vec{r}_0$ (other than $\vec{r}_0 = 0$) for $\eta = 0.01, 0.1$ and 0.3. Comparing the three curves in a sequence from small to larger values of η we infer that, for a given Ω, the stronger the Coulomb coefficient is, the larger is the equilibrium distance between the polarons. Holding α fixed at any desired value and increasing the degree of confinement, we note that parameter r_0 displays in common a decreasing trend due to the simultaneous effects of the geometric confinement and the pseudo-enhanced phonon-mediated attraction between the electrons, forcing them toward one another against their Coulomb repulsion. A salient feature met here is that as Ω grows to a critical value (shown by a bold dot at the tip of each curve) the local minimum relevant to the off-centre configuration of the PP state diminishes and conforms to an inflection point. Beyond this critical degree of confinement, the only minimum which E_g can attain is relevant solely to the bipolaron state characterized by the on-centre ($\vec{r}_0 = 0$) configuration of the polarons. Thus, within the framework of the present model of a harmonic confining potential, the effect of confinement is seen to favour the formation of bipolarons and, for quantum dots of small size, the bipolaron state may even turn out to be the only and unique ground state of the two-polaron assembly. A more apparent description of this

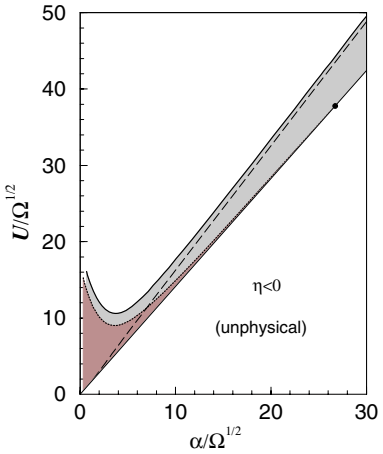

Figure 4. The phase diagram for bipolaron formation in the space of the Coulomb coefficient and the electron–phonon coupling constant. The portion shaded in light grey is the region where the bipolaron phase can be realized with dominance over the off-centre configuration. In the darker region solely the on-centre configuration will form. The bottom solid line ($\eta = 0$) is the boundary of the unphysical region. The triangular area lying above this boundary and below the dashed line gives the bipolaronic phase as calculated in [30].

feature can be achieved in the inset of figure 5, where E_g is plotted as a function of r_0 as an example for $\alpha = 5$ and $\eta = 0.3$ in the three different degrees of confinement: (a) $\Omega = 0.5$, (b) $\Omega = 1.0$ and (c) $\Omega = 1.5$. For correspondence, we designate the appropriate locations of these particular Ω-values by open circles in figure 3.

In figure 4 we plot a phase diagram in the 'U–α' plane, picturing the domain of stability of the bipolaron phase (the complete region shaded in light and dark grey). Scaling both the abscissa and the ordinate in units of $\Omega^{1/2}$ allows us to display the stability region for any desired value of Ω. For the purpose of comparison, the figure also includes the phase boundaries (the dashed and the bottom solid lines) obtained in a previous paper [30] of the present authors pertaining to exactly the same model, where the only major distinction is that the bipolaron formation was tested on the basis of the inequality (1) ($\varepsilon < 0$), which contrasts the bipolaron energy against the sum of the energies of two free polarons in similar separate dots. In order to lay out the basic qualitative trait which sets the present work apart from the former we need to summarize some of our previous results [30], which we duplicate in figure 4.

In the phase picture the space lying below the shaded region corresponds to $\eta_c < 0$, and is therefore unphysical. The upper space bounded from below by the dashed line gives the unstable region where the polarons choose to remain separated. Thus, it is only the narrow triangular area bounded by the dashed and the bottom solid lines in which the polarons can be found in a bound state forming a stable bipolaron and the vertex at which these lines join defines a critical minimum: $\alpha_c = 1.31\Omega^{1/2}$ for the coupling constant.

Comparing these outcomes with those of the present study we observe significant alterations in the phase diagram of both quantitative and qualitative nature: in particular, in the strong-confinement limit. We first note that the region in which bipolarons can remain stable broadens and this feature is seen to be most prominent under strong confinement. In this extreme ($\alpha/\Omega^{1/2} < 10$) one readily notes that the phonon-mediated binding can withstand stronger Coulomb repulsion. In the portion shaded light grey the bipolaron (on-centre) configuration is observed to have a deeper binding than the off-centred form of two

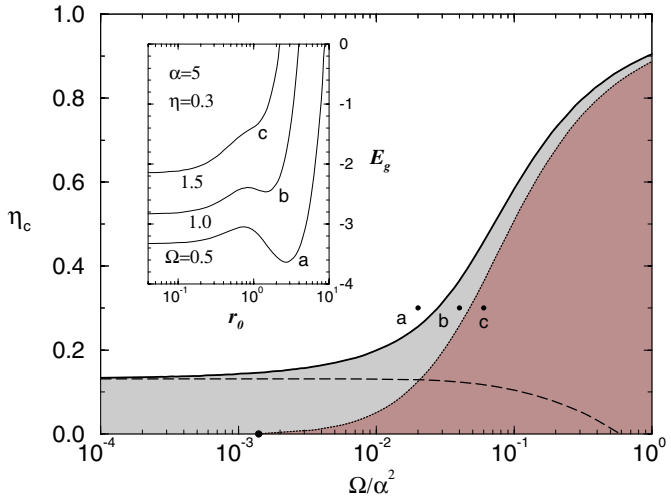

Figure 5. The critical ratio η_c as a function of the confining parameter Ω. The solid curve is the phase boundary above (below) which the off- (on-) centre configuration occurs more favourably. In the dark-grey region bounded from above by the dotted line the two-polaron complex is found to take solely the bipolaron configuration. The dots labelled by a, b and c locate the respective points for which the relevant plots of E_g versus r_0 are given in the inset for $\Omega = 0.5$, 1.0 and 1.5 when $\alpha = 5$ and $\eta = 0.3$. The dashed curve is the phase boundary separating the bipolaron state from the configuration consisting of two polarons in individual dots (duplicated from [30]).

polarons. In the portion shaded dark grey the bipolaron state shows up uniquely. The bold dot indicates the point at which the dark-shaded region terminates.

What is also peculiar in regard to the present approach is that we do not meet here a critical lower bound for the coupling constant. Instead, no matter how small the ratio $\alpha/\Omega^{1/2}$ might be set, there is always a considerably wide range of stability even for not too strong α, provided one has a sufficiently high degree of confinement. Thus, contrary to the implications of our former paper, we find that the confinement effects act in favour of bipolaronic stability in the zero-dimensional configuration and, for not too strong electron–phonon coupling, bipolarons can even form in quantum dots under strong confinement.

A more complete trace of the critical condition on the Coulomb strength as a function of Ω is given in figure 5, where we provide a layout of the alternative related parameter η against Ω/α^2 plotted universally for any given value of α. The solid curve which starts at value $\eta_c^{(3D)} = 0.131$ on the left margin is the main boundary ($\eta = \eta_c$) along which the two minima corresponding to the off-centre (PP) and the on-centre (BP) configurations have the same level of energy. Above and below this boundary one respectively has either the PP or the BP state occuring more favourably, dominating over the other. In following the horizontal direction to the right along the sequence of the three dots representing the particular cases ((a)–(c)) exemplified in the inset for $\alpha = 5$ and $\eta = 0.3$, one readily notes that entering the intermediate region shaded in light grey we encounter the configuration where the BP state dominates over the PP state (see, for instance, the relevant energy profile in plot (b) in the inset). Tuning Ω to even larger values, one crosses a second boundary (the dotted curve), beyond which the variational calculation yields $r_0 = 0$ as the unique optimal fit to this parameter. In this region, shaded in dark grey, the polarons are thus realized to conform totally to the on-centre configuration yielding a stable bipolaronic bound state.

The phase boundary given by the dashed curve is a duplicate of figure 3 in our previous article [30], which, as an alternative to the bipolaron state, assumes two individual polarons in separate quantum dots rather than two off-centre polarons in the same dot. Once again, we observe that the critical η values calculated in the former work show a strong digression and lie considerably below those of the present approach. Contrary to the predictions of the present model, the dashed curve is seen to decrease monotonically and eventually intersect the horizontal axis ($\eta = 0$) at $\Omega/\alpha^2 = 1/(1.31)^2 = 0.583$. The qualitative meaning attributed to this 'contrasting' aspect is that, in the dot-type confinement, no matter how weak the Coulomb strength might be set, there is always an upper bound for Ω beyond which the bipolaron breaks up into two individual polarons and increased values of α can only support the bipolaron to conserve its stability at correspondingly higher degrees of confinement. Let us stress once again that the results of the present work do not support any of the above aspects of our previous article and the qualitative discrepancy between the outcomes of the two approaches is very striking in the small-size limit of the quantum dot.

In this article we have reconsidered the problem of formation of bipolarons in quantum dots within the framework of the strong-coupling adiabatic approximation where we have treated the electrons to be strictly confined within the dot either to form a bipolaron state or enter a state of two close, but individual polarons. The confined bipolaron model adopted here involves the polaron–polaron separation introduced as a free parameter which one determines variationally under the competitive and interrelated roles of the parameters (U, α and Ω) which characterize the system. It is observed that the essential consequence of imposing the interpolaron separation as a variational parameter is to enlarge the region of bipolaron formation. In view of the results thus attained, we feel that the present variational approach might lend some evidence in favour of expecting somewhat stronger values of the Coulomb repulsion the bipolaron state can withstand without breaking up into two separate polarons.

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