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1996 J. Phys.: Condens. Matter 8 4017

(http://iopscience.iop.org/0953-8984/8/22/006)

J. Phys.: Condens. Matter **8** (1996) 4017–4029. Printed in the UK

Formation and stability of a singlet optical bipolaron in a parabolic quantum dot

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Received 22 November 1995, in final form 4 April 1996

Abstract. The stability of a strong-coupling singlet optical bipolaron is studied for the first time in two- and three-dimensional parabolic quantum dots using the Landau–Pekar variational method. It is shown that the confining potential of the quantum dot reduces the stability of the bipolaron.

Much effort has lately gone into exploring and understanding the various physical properties of ultra-low dimensional semiconductors [1] in which the motion of the charge carriers is confined in all spatial directions. The realization of such structures is now possible with the development of many sophisticated micro-fabrication techniques such as molecular beam epitaxy and nanolithography. These structures are typically of the order of a few nanometres in size and are usually referred to as zero-dimensional objects or, more technically, as quantum dots. Because of their fully quantized energy spectra, the quantum dots of polar semiconductors exhibit many new physical effects [2] which are quite different from their bulk counterparts and are extremely interesting from the point of view of both fundamental physics and microelectronic device applications. Consequently, extensive investigations [3] have been carried out in this area in recent years, both theoretical and experimental, and a great deal of literature with extremely rich data involving, in particular, electronic properties has already piled up.

More 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 [4] 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. To our knowledge, however, nobody has explored the possibility of bipolaron formation in quantum dots. The purpose of the present paper is to make an attempt in this direction.

A bipolaron is a bound pair of two electrons dressed with a cloud of virtual phonons. Normally two conduction band electrons would repel each other because of their repulsive Coulomb interaction, but in polar materials there is an additional interaction between electrons mediated by virtual phonons which is attractive. If this phonon mediated attractive interaction can overcome the mutual Coulomb repulsion then the electrons can form a bipolaronic bound state. This idea was first introduced in the polaron literature by Pekar [5] in the early fifties and various aspects of the bipolaron have subsequently been investigated by several authors [6]. The bipolaron problem is interesting for both academic reasons and for its practical importance in polar semiconductors and semiconducting glasses [7].

0953-8984/96/224017+13$19.50

However, above all, the discovery of high temperature superconductivity [8] in CuO_2 based layered ceramic materials and the subsequent proposal of the bipolaronic mechanism [9] for pairing has made the bipolaron problem most fascinating and has brought it to the forefront of current research.

For the sake of simplicity in calculation we shall neglect here the size quantization of phonons and model the relevant phonon modes by the corresponding bulk modes. The same approximation has also been used by several other authors. For instance, Zhu and Gu (1992) [4] employed this approximation in their second-order RSPT calculation of the polaron self-energy and Schmitt-Rink *et al* [4] and Bockelmann and Bastard [4] have studied the optical phonon broadening treating the phonons as bulk phonons. The bulk phonon approximation was also used earlier, quite successfully, in the case of quantum wells [10]. Recently Fomin and Smondyrev [6] and Klimin *et al* [11] have shown that interface phonons do have significant influence on the polaronic phenomena in nanostructures. We therefore believe that the incorporation of size quantization of phonons and the electron–interface phonon interaction into our analysis will certainly improve our results, but we expect that the essential qualitative features will remain more or less the same.

Quantum dots can be fabricated in both two and three dimensions. A quantum dot embedded in a three-dimensional (3D) material, with the motion of the dot electron confined in all the three spatial directions, will be called a 3D quantum dot, while that embedded in a purely two-dimensional (zero thickness) system with the electron's motion confined in two spatial dimensions will be referred to as a two-dimensional (2D) quantum dot. Thus, in an ideal 2D quantum dot the confinement lengths are small but finite in two spatial directions, while that in the third direction is zero. In reality, however, no systems can be purely 2D, but if the thickness of the system is much smaller than the confinement lengths in the two spatial directions, the corresponding dot may be referred to as a quasi-2D quantum dot. In the present work, our analysis will be restricted to 3D and purely 2D quantum dots. We shall make an N-dimensional formulation for the sake of generality and obtain results for both two- and three-dimensional dots as special cases.

The Hamiltonian for a system of two electrons moving in an N-dimensional parabolic quantum dot and interacting with LO phonons of the system can be written as

$$H' = -\frac{\overline{h}}{2m}\nabla^2_{r'_1} - \frac{\overline{h}}{2m}\nabla^2_{r'_2} + \frac{e^2}{\epsilon_\infty|\boldsymbol{r}'_1 - \boldsymbol{r}'_2|} + \frac{m}{2}\sum_{j=1}^{2}\sum_{i=1}^{N}\omega^2_{pji}{x'_{ji}}^2$$
$$+\overline{h}\omega_0\sum_{\boldsymbol{q}'} b^\dagger_{\boldsymbol{q}'}b_{\boldsymbol{q}'} + \sum_{i=1}^{2}\sum_{\boldsymbol{q}'}\left[{\xi'}_{q'}\mathrm{e}^{-\mathrm{i}\boldsymbol{q}'\boldsymbol{r}'_i}b^\dagger_{\boldsymbol{q}'} + h.c.\right] \tag{1}$$

where all vectors are N dimensional. The first two terms refer to the kinetic energy of the two electrons, the third term to their mutual Coulomb repulsion and the fourth term to the parabolic confining potential, $\boldsymbol{r}'_1$ and $\boldsymbol{r}'_2$ denote the position vectors of the electrons, m their Bloch effective mass, β the strength of the Coulomb interaction and ω_{pi} is the frequency of the confining potential for the motion in the ith direction. The fifth term describes the unperturbed phonon Hamiltonian with ω_0 denoting the dispersionless LO phonon frequency and $b^\dagger_{\boldsymbol{q}'}$ ($b_{\boldsymbol{q}'}$) the creation (annihilation) operator for a phonon of wavevector $\boldsymbol{q}'$. Finally the sixth term gives the interaction of the two electrons with LO phonon field, $\xi_{q'}$ being the electron–phonon interaction coefficient for which we shall follow the prescription of Peeters *et al* [12]. We shall use the Feynman units in which the energy is scaled by $\overline{h}\omega_0$, length by r_0 where r_0=q_0^{-1}, q_0 being an inverse length defined by $(\overline{h}^2q_0^2/m) = \overline{h}\omega_0$, volume by r_0^N and wavevector by q_0. Such scalings are equivalent to putting $\overline{h} = \omega_0 = m = 1$. In these

units, the Hamiltonian (1) reads

$$H = -\frac{1}{2}\nabla_{r_1}^2 - \frac{1}{2}\nabla_{r_2}^2 + \frac{\beta}{r_{12}} + \frac{1}{2}\sum_{j=1}^{2}\sum_{i=1}^{N}\omega_{ji}^2 x_{ji}^2 + \sum_{\boldsymbol{q}} b_{\boldsymbol{q}}^\dagger b_{\boldsymbol{q}} + \sum_i \sum_{\boldsymbol{q}} \left[\xi_q \mathrm{e}^{-\mathrm{i}\boldsymbol{q}\boldsymbol{r}_i} b_{\boldsymbol{q}}^\dagger + h.c.\right] \quad (2)$$

where

$$\boldsymbol{r}_1 = \boldsymbol{r}_1'/r_0 \qquad \boldsymbol{r}_2 = \boldsymbol{r}_2'/r_0 \qquad \boldsymbol{q} = \boldsymbol{q}'/\boldsymbol{q}_0$$
$$\beta = \frac{\left(\frac{e^2}{\hbar\omega_0\epsilon_\infty}\right)}{\left(\frac{\hbar}{m\omega_0}\right)^{1/2}}\omega_i = \frac{\omega_{p,i}}{\omega_0} \quad (3)$$

with the following prescription [12]

$$|\xi_q|^2 = \frac{\Gamma\left(\frac{N-1}{2}\right)2^{N-\frac{3}{2}}\pi^{\frac{N-1}{2}}}{V_N q^{N-1}}\ \alpha \quad (4)$$

where V_N is the volume of the N-dimensional crystal and α the dimensionless electron–phonon coupling constant. In what follows we shall consider a symmetric dot so that $\omega_1 = \omega_2 = \ldots = \omega_N = \omega$.

We seek a variational solution of (2) for a singlet bipolaron in the strong-coupling limit and therefore choose a trial wavefunction of the form

$$|\Psi_{BP}\rangle = |\Phi(\boldsymbol{r}_1, \boldsymbol{r}_2)\rangle \exp\left[\sum_{\boldsymbol{q}}\left(f_{\boldsymbol{q}} b_{\boldsymbol{q}}^\dagger - f_{\boldsymbol{q}}^* b_q\right)\right]|0\rangle|\zeta\rangle \quad (5)$$

where f_q are to be obtained variationally, $|0\rangle$ is the unperturbed zero-phonon state satisfying $b_{\boldsymbol{q}}|0\rangle = 0$ for all $\boldsymbol{q}$, $|\zeta\rangle$ is the antisymmetric spin function for the two electrons corresponding to the singlet pairing and $|\Phi(\boldsymbol{r}_1, \boldsymbol{r}_2)\rangle$ is a symmetric two-electron wave function which we write as

$$|\Phi(\boldsymbol{r}_1, \boldsymbol{r}_2)\rangle = \phi(\boldsymbol{r}_1)\phi(\boldsymbol{r}_2)g(r_1, r_2, |\boldsymbol{r}_1 - \boldsymbol{r}_2|) \quad (6)$$

with ϕ as one-electron functions and g the Coulomb correlation factor. For $\phi(\boldsymbol{r})$ we try a Gaussian function and for the correlation factor g we choose a Jastraw type function so that (6) can be written as

$$\Phi(\boldsymbol{r}_1, \boldsymbol{r}_2) = \left[\frac{\lambda^N(\lambda^2 - b)^{\frac{N}{2}+1}}{N\pi^N}\right]^{1/2} r_{12}\exp\left[-\frac{\lambda^2}{2}(r_1^2 + r_2^2)\right]\exp\left[\frac{b}{4}r_{12}^2\right] \quad (7)$$

where λ and b are variational parameters. Variation of the energy $E_{BP} = \langle\Psi_{BP}|H|\Psi_{BP}\rangle$ with respect to f_q yields

$$f_q = -2\xi_q\rho_q \quad (8)$$

where

$$\rho_q = \left[1 - \frac{q^2}{4N(\lambda^2 - b)}\right]\exp\left[-\frac{q^2}{8}\left\{\frac{(1-a)}{\lambda^2} + \frac{1}{\lambda^2 - b}\right\}\right]. \quad (9)$$

The variational energy (E_{BP}), the size of the bipolaron (R_{BP}) and the number of virtual phonons in the bipolaron cloud (N_{BP}) then take on the following expressions

$$E_{BP} = \frac{N}{4}\lambda^2 + \frac{N}{4}t^2\lambda^2 - \frac{1}{2}\left(1 - \frac{2}{N}\right)t^2\lambda^2 + \frac{\sqrt{2}\beta\Gamma\frac{(N+1)}{2}t\lambda}{N\Gamma(N/2)}$$
$$+\frac{1}{4l^4}\left[\frac{N}{\lambda^2} + \frac{(N+2)}{\lambda^2 t^2}\right] - 2\sqrt{2}\frac{\Gamma(\frac{N-1}{2})}{\Gamma(\frac{N}{2})}$$
$$\times(t^2+1)^{1/2}t\lambda\alpha\left[1 - \frac{1}{N(t^2+1)} + \frac{3}{4N^2(t^2+1)^2}\right] \quad (10)$$

$$R_{BP} = \langle \Psi_{BP} | r_{12} \Psi_{BP} \rangle = 2\sqrt{2} \frac{\Gamma\left(\frac{N+3}{2}\right)}{N\Gamma\left(\frac{N}{2}\right)} \left(\frac{1}{t\lambda}\right) \tag{11}$$

$$N_{BP} = -2\sqrt{2} \frac{\Gamma\left(\frac{N-1}{2}\right)}{\Gamma\left(\frac{N}{2}\right)(t^2+1)^{1/2}} t\lambda\alpha \left[1 - \frac{1}{N(t^2+1)} + \frac{3}{4N^2(t^2+1)^2}\right] \tag{12}$$

where $l = 1/\sqrt{\omega}$ and $t^2 = (\lambda^2 - b)/\lambda^2$ is the new variational parameter in place of b. The variational parameters λ and t have to be obtained numerically by solving the following equations

$$\frac{\partial E_{BP}}{\partial \lambda} = 0 \qquad \frac{\partial E_{BP}}{\partial t} = 0. \tag{13}$$

It may be noted that equation (10) reduces to the GS energy of a corresponding strong-coupling bulk bipolaron (Chatterjee and Sil (1993) [6]) in the limit $l \to 0$ which is expected in view of the correspondence principle. To obtain the stability criteria we have to find out the binding energy (BE) of the bipolaron which is given by

$$BE = 2E_P - E_{BP} \tag{14}$$

where E_P is the single polaron GS energy in the same approximation. For the sake of completeness we now briefly present our calculation for the quantum dot polaron problem which can be described in Feynman units by the Hamiltonian

$$H = -\frac{1}{2}\nabla_{\vec{r}}^2 + \frac{1}{2}\omega^2 r^2 + \sum_{\vec{q}} b_{\vec{q}}^{\dagger} b_{\vec{q}} + \sum_{\vec{q}} \left[\xi_q \mathrm{e}^{-\mathrm{i}\vec{q}\vec{r}} b_{\vec{q}}^{\dagger} + h.c.\right] \tag{15}$$

where the notations have exactly the same meaning as in (2) for a symmetric dot. In the strong-coupling approximation the polaron wave function for the GS may be chosen as

$$|\Psi_P\rangle = \left(\frac{\mu}{\pi^{1/4}}\right)^N \exp\left(-\frac{\mu^2 r^2}{2}\right) \exp\left[\sum_{\vec{q}} \left(g_{\vec{q}} b_{\vec{q}}^{\dagger} - g_{\vec{q}}^{*} b_{\vec{q}}\right)\right] |0\rangle \tag{16}$$

where μ, and g_q are variational parameters. Minimization of $E_P = \langle \Psi_P | H | \Psi_P \rangle$ with respect to g_q can be done analytically to obtain

$$g_q = -\xi_q \exp\left[-\left(\frac{q^2}{4\mu^2}\right)\right] \tag{17}$$

and thus the variational energy E_P assumes the following expression

$$E_P = \frac{N}{4}\mu^2 + \frac{N}{4}\left(\frac{1}{l^4\mu^2}\right) - \frac{\alpha}{2}\frac{\Gamma\left(\frac{N-1}{2}\right)}{\Gamma\left(\frac{N}{2}\right)}\mu \tag{18}$$

where μ has to be obtained numerically from

$$\frac{\partial E_P}{\partial \mu} = 0. \tag{19}$$

It may again be noted that equation (18) assumes the Landau–Pekar expression for the bulk polaron in the limit $l \to \infty$. We have performed numerical calculations for the binding energy (BE) and the size (R_{BP}) of the bipolaron and for the number of virtual phonons (N_{BP}) in the bipolaron cloud for both 2D and 3D dots. We have also determined the bipolaron stability criteria which are obtained by demanding that the binding energy of the bipolaron be positive. We find that binding energy of a strong-coupling bipolaron in a quantum dot depends on two parameters. These parameters are $\eta = \epsilon_\infty/\epsilon_0$ and the

confinement length l. In figure 1 we show the variation of the bipolaron binding energy (BE) as a function of l for a few values of η ($\eta = 0.01, 0.05, 0.1$) for a 3D quantum dot. It is clear that the binding energy decreases with the decrease in the confinement length l. The variation is extremely rapid below a certain value of the confinement length and indeed the bipolaron becomes unstable if l is made smaller than a critical value l_c. At such confinement lengths a bipolaron breaks up into two individual polarons. The reason for this instability of the bipolaron is not difficult to understand. The decrease in the confinement length of the dot potential will lead to an enhancement in the average Coulomb repulsion between the electrons. When this Coulomb repulsion increases considerably with a corresponding significant decrease in dot size, it may then dominate the phonon mediated attractive electron–electron interaction, thereby inhibiting the formation of stable bipolarons. However, when l is large, the bipolaron binding energy does not change much with l and hence the bipolaron stability becomes more or less independent of l. This is essentially the bulk limit. One may furthermore observe from figure 1 that the bipolaron binding energy increases with a decrease in η, which is again not quite unexpected. Since $\eta = (1 - (\sqrt{2}\alpha/\beta))$ for a given value of α, a decrease in η means a decrease in β, and hence a decrease in the strength of the electron-electron Coulomb repulsion, which in turn implies an increase in the effective electron–electron attractive interaction. Thus a decrease in η would lead to an enhancement in the bipolaron binding energy favouring the formation of stable bipolarons. Figure 2 shows the variation of the binding energy of the bipolaron with l for $\eta = 0.01$, 0.05 and 0.1 for a 2D quantum dot. The variations are qualitatively similar to those observed in 3D dots. Quantitatively however, the polaronic interactions are stronger in 2D than in 3D and consequently, for the same value of η, the bipolaron binding energy is larger in a 2D dot than in its 3D counterpart. Naturally, for a given value of η the critical confinement length l_c is smaller for a 2D quantum dot than that for the corresponding 3D dot. In figures 3 and 4 we show explicitly the behaviour of the binding energy of a bipolaron as a function of η for a few values of l. As expected, BE decreases as η increases and thus for a given value of l, η has to be smaller than some critical value η_c for the bipolaron to be stable. It is clear that the value of η_c increases with increasing l. Comparison between figures 3 and 4 shows that for a given confinement length η_c is larger for a 2D quantum dot than that for the corresponding 3D dot. One interesting observation that one can make from figure 3 is the crossing of binding energy curves corresponding to $l = \infty$ and $l = 0.447$ at a small value of η. The physical explanation of the crossing is not completely clear. Interestingly however, this crossing is not present in the case of 2D quantum dots.

In figure 5 we plot the bipolaron radius (R_{BP}) as a function of l for $\eta = 0.01, 0.05$ and 0.1 for a 3D quantum dot. The corresponding results for a 2D dot are shown in figure 6. Again the results for the 2D and 3D dots are qualitatively similar. The bipolaron size increases quite rapidly with l at small values of l. The variation is however very slow when l is large. This is quite interesting behaviour, and is opposite to what is observed in the bulk bipolaron problem where a large binding energy is associated with a smaller radius of the bipolaron. Furthermore we find that, for a given value of l, the bipolaron size increases with η, which is expected. However, if the confinement length is sufficiently small, the η dependence of the bipolaron radius becomes essentially insignificant. Comparison between figures 5 and 6 shows that the radius of a strong-coupling bipolaron is smaller in a 2D quantum dot than that in the corresponding 3D dot.

Figures 7 and 8 give the plots of the number of virtual phonons (N_{BP}) in the bipolaron cloud in 2D and 3D quantum dots respectively. Here we also find that, for the same values of η and l, N_{BP} is much larger in a 2D dot than in a 3D dot. In both cases we find that

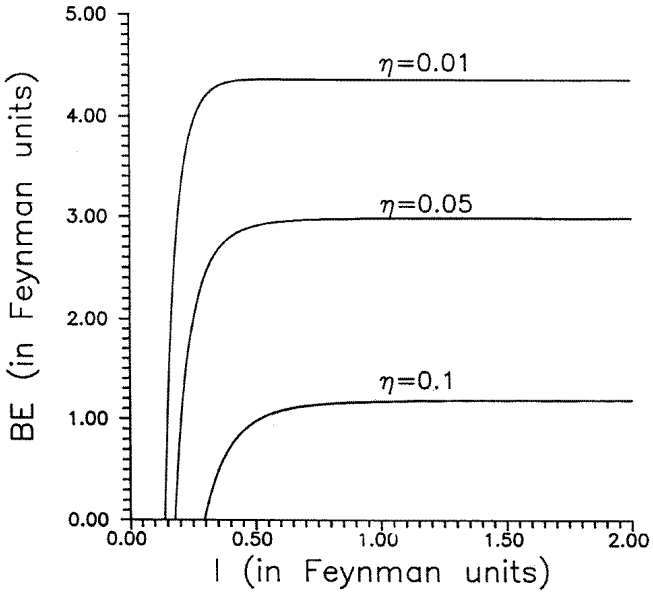

Figure 1. Binding energy of the bipolaron (BE) (in Feynman units) as a function of the confinement length l (in Feynman units) for different values of η ($\eta = 0.01,\ 0.05,\ 0.1$) in a 3D quantum dot.

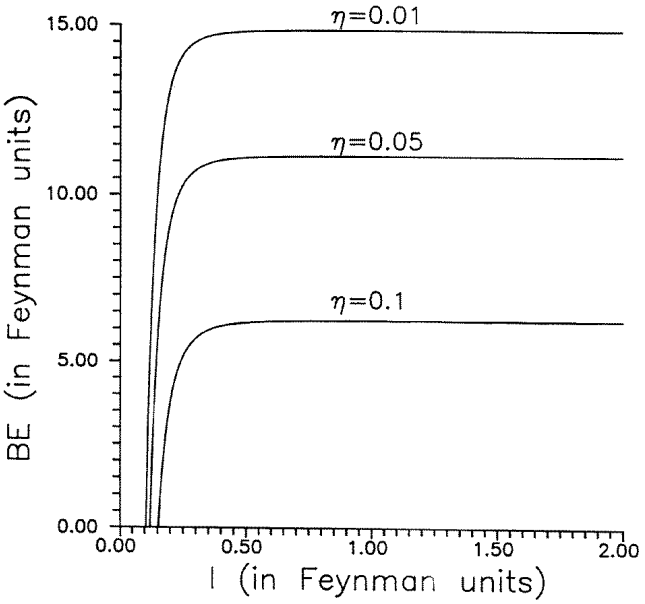

Figure 2. Binding energy of the bipolaron (BE) (in Feynman units) as a function of the confinement length l (in Feynman units) for different values of η ($\eta = 0.01,\ 0.05,\ 0.1$) in a 2D quantum dot.

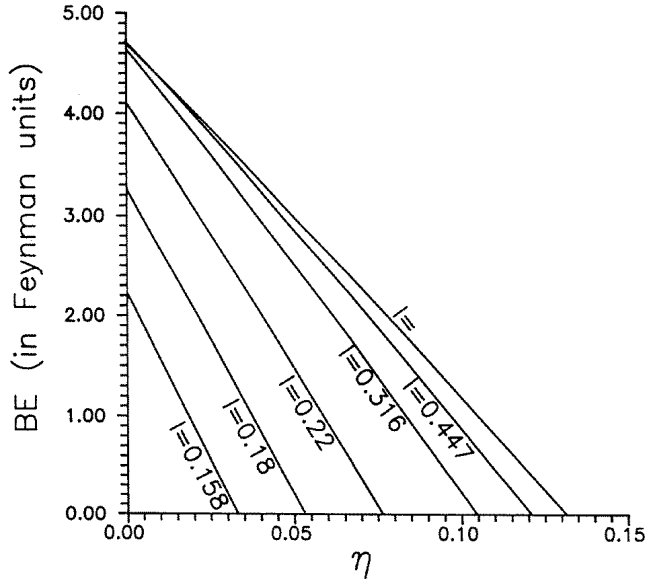

Figure 3. Binding energy of the bipolaron (BE) (in Feynman units) as a function of η for different values of the confinement length l (in Feynman units) in a 3D quantum dot.

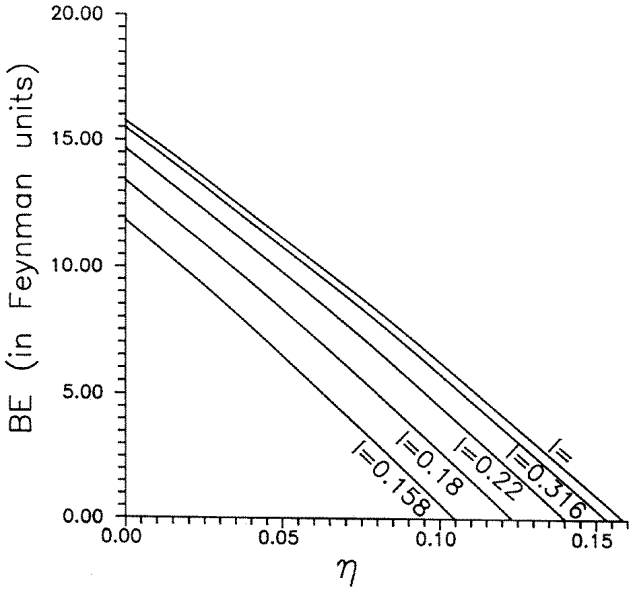

Figure 4. Binding energy of the bipolaron (BE) (in Feynman units) as a function of η for different values of the confinement length l (in Feynman units) in a 2D quantum dot.

N_{BP} decreases quite rapidly with increasing l when l is small. When l exceeds a certain value the decrease of N_{BP} with l becomes extremely slow, eventually saturating to the bulk value. It may again be noted here that in the conventional bipolaron problem (in a bulk crystal), a reduction in N_{BP} is associated with a decrease in the binding energy, which is

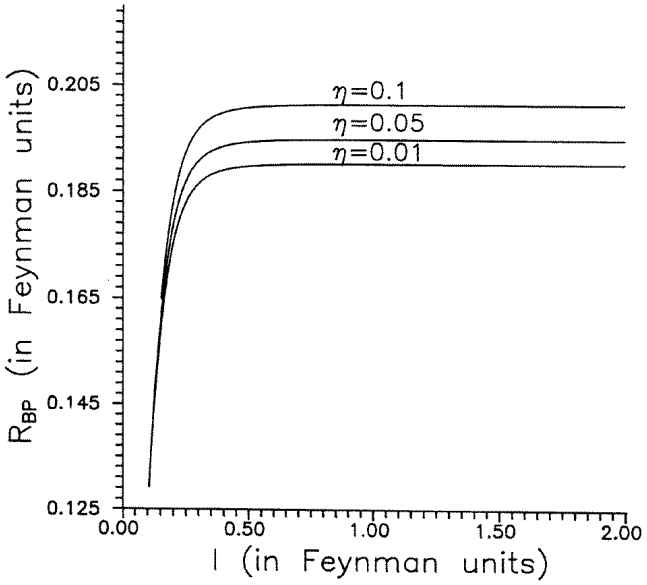

Figure 5. Size of the bipolaron R_{BP} (in Feynman units) as a function of the confinement length l (in Feynman units) for different values of η ($\eta = 0.01,\ 0.05,\ 0.1$) in a 3D quantum dot.

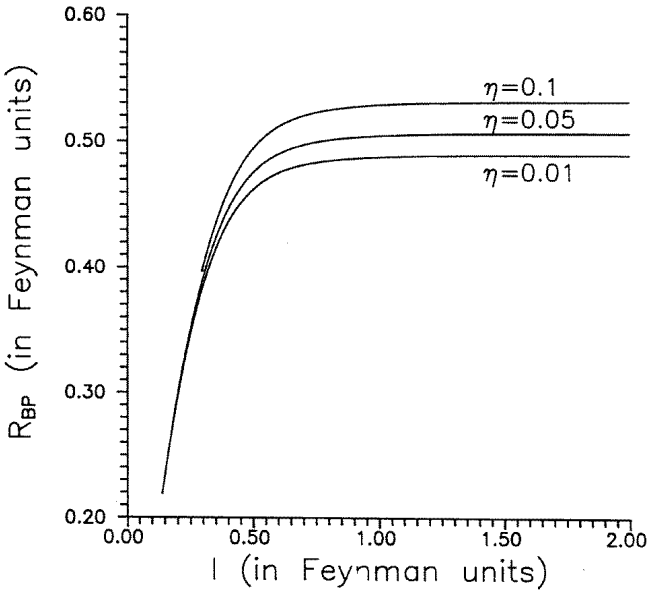

Figure 6. Size of the bipolaron R_{BP} (in Feynman units) as a function of the confinement length l (in Feynman units) for different values of η ($\eta = 0.01,\ 0.05,\ 0.1$) in a 2D quantum dot.

quite opposite to what we observe here. For a given l, however, the number of phonons in the bipolaron cloud are found to increase with η which is, of course, expected behaviour.

In figures 9 and 10 we show the variation of η_c explicitly as a function of the confinement length for 3D and 2D dots respectively. The figures show that in both cases η_c rises very

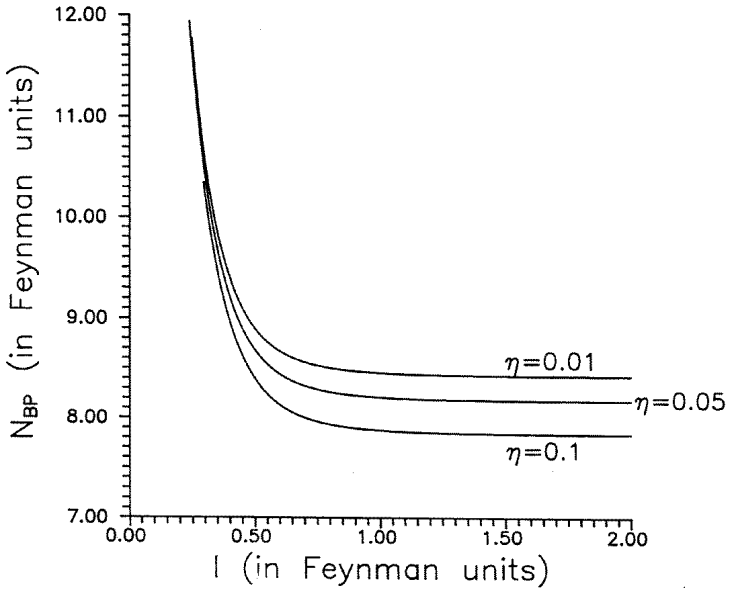

Figure 7. Number of phonons in the bipolaron cloud N_{BP} (in Feynman units) as a function of the confinement length l (in Feynman units) for different values of η ($\eta = 0.01,\ 0.05,\ 0.1$) in a 3D quantum dot.

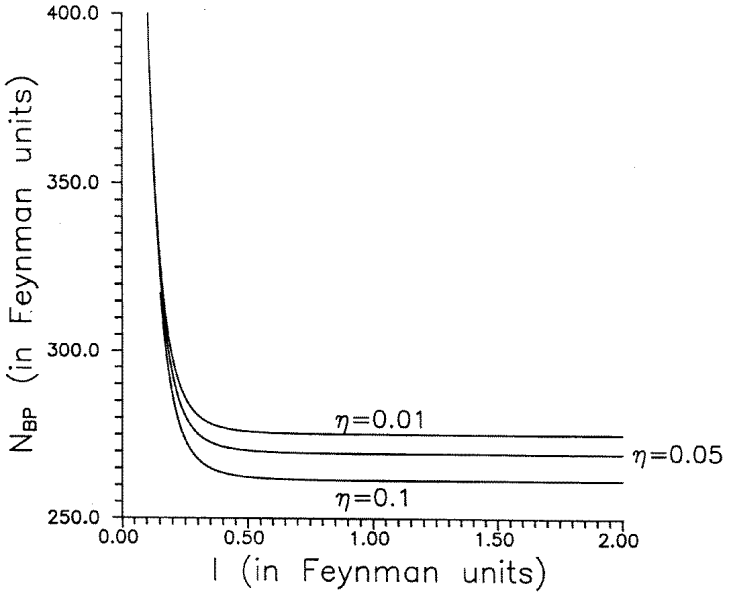

Figure 8. Number of phonons in the bipolaron cloud N_{BP} (in Feynman units) as a function of the confinement length l (in Feynman units) for different values of η ($\eta = 0.01,\ 0.05,\ 0.1$) in a 2D quantum dot.

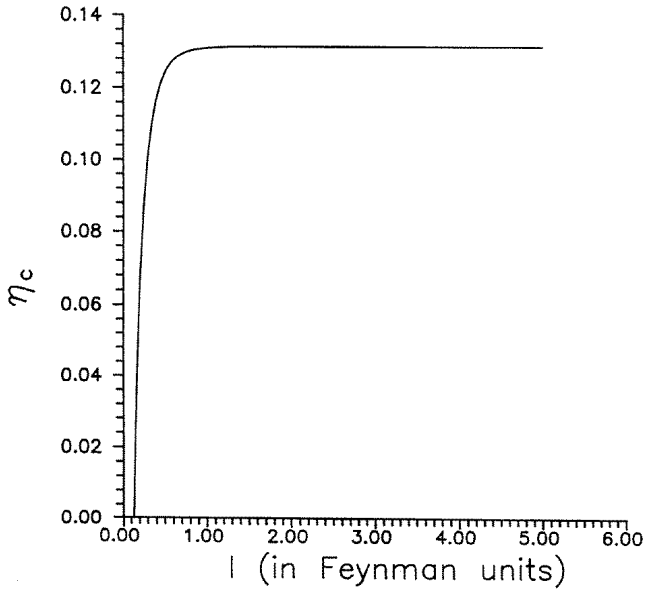

Figure 9. η_c as a function of the confinement length l (in Feynman units) in a 3D quantum dot.

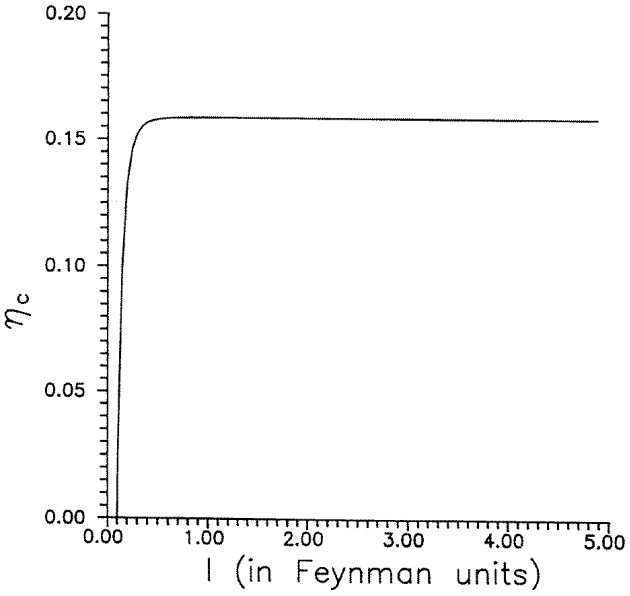

Figure 10. η_c as a function of the confinement length l (in Feynman units) in a 2D quantum dot.

rapidly with l at small l and then saturates to the corresponding bulk value. One may furthermore observe that stable bipolarons can indeed exist, even in a sufficiently small dot, if the value of η for such a dot is smaller than a certain critical value $\eta_c(l)$. However, since $\eta_c(l)$ is always less than η_c $(l \to \infty)$, the bipolaronic stability criteria are, in general, less favourable in quantum dots than in bulk crystals.

In conclusion, we have investigated the possibility of formation and stability of a bipolaron in two- and three-dimensional symmetric quantum dots with parabolic confinement. We have shown that the bipolaron binding energy decreases with the decrease in the confinement length in both two- and three-dimensional dots and below a certain critical value of the confinement length (l_c) which depends on η and the dimensionality of the dots, a bipolaron becomes unstable and breaks up into two individual polarons. We have furthermore shown that there also exists a critical value of $\eta(\eta_c)$ (depending on the confinement length and the dimensionality of the dot) below which the GS of a strong electron–phonon system is a bipolaronic state. However, $\eta_c(l)$ is less than $\eta_c(\infty)$ for any finite value of l and therefore the bipolaronic stability criteria are apparently less favourable in quantum dots than in bulk crystals. The present analysis however neglects the size quantization of phonons and the effect of interface phonons. The electron–interface phonon coupling will give rise to another phonon mediated attractive interaction which will also favour the bipolaron formation and may enhance the stability region of the bipolaron. Thus we expect that the incorporation of interface phonons will make the bipolaron stability criteria a little more favourable than that obtained from our present calculation. The role of size quantization and the electron–interface phonon interaction on the bipolaron stability will be investigated quantitatively in a forthcoming paper.

Acknowledgment

One of the authors (SM) wishes to thank the University Grants Commission, India for financial support.

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