Journal of Zhejiang University SCIENCE
ISSN 1009-3095
http://www.zju.edu.cn/jzus
E-mail: jzus@zju.edu.cn

Bipolaron in different configuration of quantum confinement*

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Received Feb. 18, 2004; revision accepted Apr. 15, 2004

Abstract: The authors used Landau-Pekar variational method to investigate a strong-coupling singlet optical bipolaron in different configuration of quantum confinement. Numerical and analytical results showed that when configuration changes from quantum dot and wire to well, confinement shows different effect on the formation of a bipolaron. In contrast to a bipolaron in a quantum dot or wire, the binding energy of a bipolaron in a quantum well increases with increasing confinement, indicating that confinement favors bipolaron formation in a quantum well.

Key words: Bipolaron, Quantum confinement, Quantum wire, Quantum well, Quantum dot, Binding energy
Document code: A **CLC number:** O469; O488

INTRODUCTION

The bipolaron in bulk materials has been extensively discussed in the past decade. With more recent advances in the creation of nanocrystal with strong ionic coupling (Hudgins *et al*., 1997), one natural problem is that the effect of bipolarons in some quantum low dimensional systems with effective electron-phonon coupling is very strong owing to the confinement. This problem is also relevant to the proposal of the bipolaronic mechanism for pairing in CuO_2 plane (quasi-two-dimensionality) in high T_c cuprates (Emin, 1989; Alexandrov and Kornilovitch, 2002; Lee *et al*., 2003). Such investigations are helpful for better understanding of the role of the electron-LO phonon interactions in low-dimensional structures.

More recently, with the development in microfabrication technology, such as molecular-beam epitaxy and nanolithography, it became possible to fabricate synthetic polar semiconductor or ionic structures with low dimensionality. Such structures as quantum wire, quantum dot and quantum well (Kuntscher *et al*., 2003; Lorke *et al*., 1990; Tanaka *et al*., 2003), where the electrons are confined by different configuration of quantum confinement. These systems have attracted substantial attention due to the novel physical effects from their quasi-0D (quantum dot), quasi-1D (quantum wire) or quasi-2D (quantum well) dimensionality.

Some researches (Mukhopadhyay and Chatterjee, 1996; Chen *et al*., 1998; 2001; Pokatilov *et al*., 2000; 2003; Ruan *et al*., 2002; 2003) had been devoted to the study of a polaron or bipolaron in quantum potential by taking account of the electron-longitudinal-optical (LO) phonon interactions. The general consensus is that the polaronic correction is very important and cannot be neglected. A systematic study of bipolaron in different configuration of quantum confinement is very important because it may stimulate more experimental work on some materials with low dimensionality. Within

* Project (No. 19804009) supported by the National Natural Science Foundation of China

the framework of the Laudau-Pekar variational method, we obtained the ground-state (GS) energy of a bipolaron in different configuration of quantum confinement.

ESTIMATES FOR THE BIPOLARON ENERGY IN A QUANTUM WIRE

A system of two electrons interacting with a phonon field and confined in a quantum wire is described by Frohlich Hamiltonian (in units of $m=\hbar=\omega_{LO}=1$)

$$H=\sum_{i=1,2}[\frac{\boldsymbol{p}_i^2}{2}+\sum_{\boldsymbol{k}}(v_{\boldsymbol{k}}a_{\boldsymbol{k}}\mathrm{e}^{i\boldsymbol{k}\boldsymbol{r}_i}+v_{\boldsymbol{k}}^*a_{\boldsymbol{k}}^+\mathrm{e}^{-i\boldsymbol{k}\boldsymbol{r}_i})] +U(|\boldsymbol{r}_1-\boldsymbol{r}_2|)+\sum_{\boldsymbol{k}}a_{\boldsymbol{k}}^+a_{\boldsymbol{k}}+V_{\mathrm{conf}}(\rho) \quad (1)$$

where $\boldsymbol{r}_i=[\rho_i(\boldsymbol{x}_i,\boldsymbol{y}_i),\boldsymbol{z}_i]$ and $\boldsymbol{p}$ are the position and momentum operators of the jth electron (j=1, 2), $U(\boldsymbol{r})=U/\boldsymbol{r}$ is the Coulomb interaction potential with strength U.

$$V_{\mathrm{conf}}(\rho)=\frac{1}{2}\Omega_\rho^2\rho_1^2+\frac{1}{2}\Omega_\rho^2\rho_2^2 \quad (2)$$

is the confining potential for two electrons in a quantum wire, with Ω_ρ being in units of ω_{LO}, measuring the confining strength of the parabolic potential. $a_{\boldsymbol{k}}^+$ and $a_{\boldsymbol{k}}$ are, respectively, the creation and annihilation operators of the LO phonons with the wave vector $\boldsymbol{k}=(\boldsymbol{k}_\rho,\boldsymbol{k}_z)$,

$$|v_{\boldsymbol{k}}|^2=\frac{2\sqrt{2}\pi\alpha}{v_{\boldsymbol{k}}^2},\ \alpha=\frac{\mathrm{e}^2}{\sqrt{2}}(\frac{1}{\varepsilon_\infty}-\frac{1}{\varepsilon_0}), \quad (3)$$

with v being the crystal volume and α being the electron-phonon coupling constant. Here we should mention that the impurity-phonon interactions had already been eliminated. $\varepsilon_\infty(\varepsilon_0)$ is the high-frequency (static) dielectric constant and the non-screened electron Coulomb repulsion strength is given by $U=\mathrm{e}^2/\varepsilon_\infty$, so that we can easily deduce that

$$U=\frac{\sqrt{2}\alpha}{1-\eta},\ \eta=\frac{\varepsilon_\infty}{\varepsilon_0}. \quad (4)$$

Since the bipolaron is a composite particle, we introduce a center-of-mass coordinate $\boldsymbol{R}=(\boldsymbol{r}_1+\boldsymbol{r}_2)/2$ $[\boldsymbol{P}=\boldsymbol{p}_1+\boldsymbol{p}_2]$ and a relative coordinate $\boldsymbol{r}=\boldsymbol{r}_1-\boldsymbol{r}_2$ $[\boldsymbol{p}=(\boldsymbol{p}_1-\boldsymbol{p}_2)/2]$ for the electron pair, so that the Hamiltonian of a bipolaron in a quantum wire can be rewritten as

$$H=\frac{\boldsymbol{P}^2}{4}+\boldsymbol{p}^2+\sum_{\boldsymbol{k}}a_{\boldsymbol{k}}^+a_{\boldsymbol{k}}+\frac{U}{\boldsymbol{r}}+2\sum_{\boldsymbol{k}}\cos(\frac{\boldsymbol{k}\cdot\boldsymbol{r}}{2})(v_{\boldsymbol{k}}a_{\boldsymbol{k}}\mathrm{e}^{i\boldsymbol{k}\cdot\boldsymbol{R}} +h.c.)+\frac{\Omega_\rho^2}{4}(4R_\rho^2+r_\rho^2), \quad (5)$$

$$R_\rho=(\rho_1+\rho_2)/2, R_z=(z_1+z_2)/2,$$
$$r_\rho=\rho_1-\rho_2, r_z=z_1-z_2.$$

To obtain a variational estimate of the bipolaron energy, we seek a variational solution of Eq.(5) for a singlet bipolaron in the strong-coupling limit and therefore chose the trial wavefunction in Mukhopadhyay and Chatterjee (1996):

$$|\Psi_{BP}\rangle=|\Phi(\boldsymbol{r}_1,\boldsymbol{r}_2)\rangle\exp\{\sum_{\boldsymbol{k}}(f_{\boldsymbol{k}}a_{\boldsymbol{k}}^+-f_{\boldsymbol{k}}^*a_{\boldsymbol{k}})\}|0\rangle|\xi\rangle, \quad (6)$$

where $f_{\boldsymbol{k}}$ are to be obtained variationally, $|0\rangle$ is the unperturbed zero-phonon state satisfying $a_{\boldsymbol{k}}|0\rangle=0$ for all $\boldsymbol{k}$, $|\xi\rangle$ is the antisymmetric spin function for two electrons corresponding to the singlet pairing, and $|\Phi(\boldsymbol{r}_1,\boldsymbol{r}_2)\rangle=\Phi(\boldsymbol{R},\boldsymbol{r})$ is a symmetric two-electron wave function that can be expressed as:

$$\Phi(\boldsymbol{R},\boldsymbol{r})\sim\exp(-(w_\rho R_\rho^2+w_z R_z^2))\cdot \sqrt{r_\rho^2+r_z^2}\exp\{-(u_\rho r_\rho^2+u_z r_z^2)\}, \quad (7)$$

with w_ρ, w_z, u_ρ and u_z being variational parameters to be determined.

This trial wave function had been shown to be the best one in the estimate of GS in free bipolaron systems (Verbist *et al.*, 1992). It should be more suitable in the presence of the confinement, because the confinement also contributes to the binding of two electrons.

To obtain variational upper estimate of the bipolaron self-energy, we averaged the Hamiltonian Eq.(5) over the trial wave-function $|\Psi_{BP}\rangle$ and obtained the GS energy of a bipolaron in a quantum wire as $E_{BP}=\langle\Psi_{BP}|H|\Psi_{BP}\rangle/\langle\Psi_{BP}|\Psi_{BP}\rangle$. Then minimization of E_{BP} with respect to $f_{\boldsymbol{k}}$ can be done analytically to obtain:

$$f(\boldsymbol{k})=-2v_{\boldsymbol{k}}^{*}[1-(a\boldsymbol{k}_{\rho}^{2}+b\boldsymbol{k}_{z}^{2})]\exp(-\frac{g\boldsymbol{k}_{\rho}^{2}}{2}-\frac{e\boldsymbol{k}_{z}^{2}}{2}),\quad(8)$$

Here

$$a=\frac{u_z}{32u_\rho u_z+16u_\rho^2},b=\frac{u_\rho}{32u_z^2+16u_\rho u_z},$$
$$g=\frac{1}{16u_\rho}+\frac{1}{4w_\rho},e=\frac{1}{16u_z}+\frac{1}{4w_z},\quad(9)$$

$$\sum_{\boldsymbol{k}}f^{*}(\boldsymbol{k})f(\boldsymbol{k})=\frac{2\sqrt{2}\alpha}{\sqrt{\pi}}\int_0^\infty \mathrm{e}^{-\tau}\mathrm{d}\tau\{\sqrt{\frac{1}{e-g}}\ln(\sqrt{\frac{e}{g}-1}+\sqrt{\frac{e}{g}})+\frac{3a^2}{4}[\frac{2e^{\frac{3}{2}}}{3(e-g)^2g^2}-\frac{5\sqrt{e}}{3g(e-g)^2}+\frac{1}{(e-g)^2\sqrt{e-g}}\ln(\sqrt{\frac{e}{g}-1}+\sqrt{\frac{e}{g}})]+\frac{3b^2}{4}[-\frac{4}{3e^{1/2}(e-g)^2}+\frac{g}{3e^{3/2}(e-g)^2}+\frac{1}{(e-g)^2\sqrt{e-g}}\ln(\sqrt{\frac{e}{g}-1}+\sqrt{\frac{e}{g}})]-a[\frac{\sqrt{e}}{g(e-g)}-\frac{1}{(e-g)\sqrt{e-g}}\ln(\sqrt{\frac{e}{g}-1}+\sqrt{\frac{e}{g}})]-b[\frac{1}{\sqrt{e}(g-e)}+\frac{1}{(e-g)\sqrt{e-g}}\cdot\ln(\sqrt{\frac{e}{g}-1}+\sqrt{\frac{e}{g}})]+\frac{3}{2}ab[\frac{\sqrt{e}}{3(e-g)^2g}+\frac{2}{3\sqrt{e}(e-g)^2}-\frac{1}{(e-g)^2\sqrt{e-g}}\cdot\ln(\sqrt{\frac{e}{g}-1}+\sqrt{\frac{e}{g}})]\},\quad(10)$$

$e>g$.

Finally inserting $f(\boldsymbol{k})$ and $f^{*}(\boldsymbol{k})$ into E_{BP}, we obtained the GS energy of the bipolaron in a parabolic quantum wire as:

$$E_{BP}=\frac{w_\rho}{2}+\frac{w_z}{4}+\frac{2u_z^2+2u_\rho^2+3u_\rho u_z}{2u_z+u_\rho}+\sqrt{\frac{2u_\rho}{\pi}}(\frac{2Uu_z}{2u_z+u_\rho})\cdot\{\sqrt{\frac{u_\rho}{u_z}}+\ln(\sqrt{\frac{u_\rho}{u_z}}+\sqrt{\frac{u_\rho}{u_z}-1})/\sqrt{\frac{u_\rho}{u_z}-1}\}+\frac{\Omega_\rho^2}{2w_\rho}+\frac{\Omega_\rho^2}{8u_\rho}\frac{4u_z+u_\rho}{2u_z+u_\rho}-\sum_{\boldsymbol{k}}f^{*}(\boldsymbol{k})f(\boldsymbol{k}),\quad(11)$$

with w_ρ, w_z, u_ρ and u_z being variational parameters to be determined.

ESTIMATES FOR THE BIPOLARON ENERGY IN A QUANTUM WELL OR DOT

Within the same framework of the Landau-Pekar method, we obtained the GS energy of a bipolaron in a quantum well confinement:

$$E_{BP}=\frac{w_\rho}{2}+\frac{w_z}{4}+\frac{2u_z^2+2u_\rho^2+3u_\rho u_z}{2u_z+u_\rho}+\sqrt{\frac{2u_z}{\pi}}(\frac{2Uu_z}{2u_z+u_\rho})\cdot\{\frac{u_\rho}{u_z}+\arctan(\sqrt{\frac{u_z}{u_\rho}-1})/\sqrt{\frac{u_z}{u_\rho}-1}\}+\frac{\Omega_z^2}{4w_z}+\frac{\Omega_z^2}{16u_z}\cdot\frac{2u_z+3u_\rho}{2u_z+u_\rho}-\sum_{\boldsymbol{k}}f^{*}(\boldsymbol{k})f(\boldsymbol{k}),\quad(12)$$

$$\sum_{\boldsymbol{k}}f^{*}(\boldsymbol{k})f(\boldsymbol{k})=\frac{2\sqrt{2}\alpha}{\sqrt{\pi}}\int_0^\infty \mathrm{e}^{-\tau}\mathrm{d}\tau\{\sqrt{\frac{1}{g-e}}\arctan(\sqrt{\frac{g}{e}-1})+\frac{3a^2}{4}[\frac{2e^{\frac{3}{2}}}{3(e-g)^2g^2}-\frac{5\sqrt{e}}{3g(e-g)^2}+\frac{1}{(e-g)^2\sqrt{g-e}}\arctan(\sqrt{\frac{g}{e}-1})]+\frac{3b^2}{4}[-\frac{4}{3e^{1/2}(e-g)^2}+\frac{g}{3e^{3/2}(e-g)^2}+\frac{1}{(e-g)^2\sqrt{g-e}}\arctan(\sqrt{\frac{g}{e}-1})]-a[\frac{\sqrt{e}}{g(e-g)}-\frac{1}{(e-g)\sqrt{g-e}}\arctan\cdot$$

$$(\sqrt{\frac{g}{e}-1})]-b[\frac{1}{\sqrt{e}(g-e)}+\frac{1}{(e-g)\sqrt{g-e}}\cdot$$
$$\arctan(\sqrt{\frac{g}{e}-1})]+\frac{3}{2}ab[\frac{\sqrt{e}}{3(e-g)^2 g}+\frac{2}{3\sqrt{e}(e-g)^2}$$
$$-\frac{1}{(e-g)^2\sqrt{g-e}}\arctan(\sqrt{\frac{g}{e}-1})]\}, \quad (13)$$
$$e<g,$$

with w_ρ, w_z, u_ρ and u_z being variational parameters to be determined. g and e are defined as in Eq.(9).

An expression for GS energy of a bipolaron in a quantum dot can be further obtained as that obtained by Mukhopadhyay and Chatterjee (1996).

DISCUSSIONS AND RESULTS

To obtain the stability criterion for bipolaron, we should calculate the binding energy (E_B) of the system

$$E_B = 2E_P - E_{BP}, \quad (14)$$

where E_P is the single polaron GS energy in the same approximation. For the sake of completeness we now briefly present our result for the quantum wire polaron problem as following expression:

$$E_P = \frac{\lambda_\rho}{2}+\frac{\lambda_z}{4}+\frac{\Omega_\rho^2}{2\lambda_\rho}-\alpha\sqrt{\frac{\lambda_\rho\lambda_z}{\pi(\lambda_\rho-\lambda_z)}}\ln(\sqrt{\frac{\lambda_\rho}{\lambda_z}-1}+\sqrt{\frac{\lambda_\rho}{\lambda_z}}),\ \lambda_\rho>\lambda_z. \quad (15)$$

Within the framework of prefer Landau-Pekar method, we briefly present the GS energy of a polaron in a quantum well or dot as

$$E_P = \frac{\lambda_\rho}{2}+\frac{\lambda_z}{4}+\frac{\Omega_z^2}{4\lambda_z}-\alpha\sqrt{\frac{\lambda_\rho\lambda_z}{\pi(\lambda_z-\lambda_\rho)}}\arcsin(\sqrt{1-\frac{\lambda_\rho}{\lambda_z}}),$$
$$\lambda_\rho<\lambda_z. \quad (16)$$

or

$$E_P = \frac{3}{4}\lambda+\frac{3\Omega^2}{4\lambda}-\alpha\sqrt{\frac{\lambda}{\pi}}. \quad (17)$$

The condition under which a bipolaron can exist as a stable state is that E_B should be positive. Note that the binding energy is related to three parameters: the ratio of dielectric constants η($\eta=\varepsilon_\infty/\varepsilon_0$), the effective confinement radius of the quantum dot ($R=1/\sqrt{\Omega}$) or quantum wire ($R_\rho=1/\sqrt{\Omega_\rho}$), the length of the quantum well l_z($l_z=1/\sqrt{\Omega_z}$), and the electron-phonon coupling constant α.

Fig.1 is the plot of the variation of E_B as a function of confinement such as quantum dot (R), quantum wire (R_ρ) and quantum well (l_z) at η=0.01 and α=8.0. It is interesting to note that E_B increases as l_z decreases in the quantum well. With the decrease of l_z($l_z=1/\sqrt{\Omega_z}$), with the electronic confinement becoming stronger, the strengthened effective electron-phonon coupling results in the increasing degree of E_B. In contrast to that of quantum well, E_B of quantum wire or quantum dot decreases with the increase of confinement.

With increasing degree of confinement (i.e., with increasing Ω_ρ and/or Ω_z), the Coulomb repulsion is steadily strengthened due to the fact that particles are squeezed to get closer; and the rate at

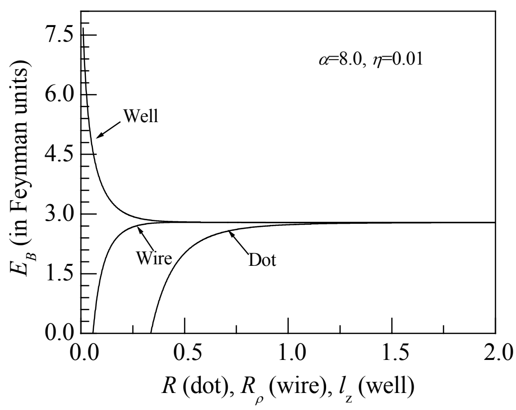

Fig.1 E_B as a function of confinement such as quantum dot (R), quantum wire (R_ρ) and quantum well (l_z) at η=0.01 and α=8.0

which this happens should be most prominent in the Q0D-configuration where the electrons are pushed towards one another from all radially inward directions. The feature was depicted explicitly by Mukhopadhyay and Chatterjee (1996) for bipolarons in quantum dots. However in the Q1D- and Q2D-configurations, the electrons are free to expand and relax themselves respectively in either one or two directions, thus resulting in comparatively weaker Coulomb repulsion.

On the other hand, in confined system, the Coulomb repulsion strength is pronounced, and the phonon-coupling is pseudo-enhanced, leading to a more effective and deeper polaronic binding to oppose and counterbalance the repulsive forces. The competitive interrelation between the two aspects of the problem determines the figures. As in quantum well (Q2D), the very pronounced attractive electron-electron interaction mediated by phonons is favorable for bipolaron formation.

The dimension of confinement of quantum wire (Q1D) is in-between that of quantum well (Q2D) and quantum dot (Q0D), which result in the phase of quantum wire being in-between that of quantum well (Q2D) and quantum dot (Q0D). On the other hand, when confinement changes to zero ($R\rightarrow\infty$), the binding energy of different confinement configuration tends to the same value. These findings showed that our results are reasonably convincing.

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