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Second-order perturbative treatment for confined polarons in low-dimensional polar semiconductors

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Abstract. Within the framework of second-order Rayleigh–Schrödinger perturbation theory, the effects of the interaction of the electrons and longitudinal optical phonons in low-dimensional semiconducting heterostructures can be investigated in a unified way. As a result, the ground-state energy for polarons confined in a general potential can be explicitly expressed as a one-dimensional integral. Moreover, some interesting problems, such as those of polarons in quantum wells, quantum wires, and quantum dots, can be readily addressed just by taking different limits. Finally, in a general sense, it is shown on the basis of numerical calculations that the polaronic effect is enhanced with lowering dimensionality and increasing asymmetry.

1. Introduction

Recent developments in microfabrication technology, such as molecular-beam and lithographic deposition, have created a variety of opportunities for the fabrication of synthetic semiconductor structures with reduced dimensionality [1–9], such as quasi-zero-dimensional quantum dots, quasi-one-dimensional quantum wires, and quasi-two-dimensional quantum wells. These heterostructures have attracted substantial attention due to the novel physical effects arising from their low dimensionality, which are very useful in device applications.

A large amount of the literature [10–32] available on such topics has been devoted to the effects of electron–phonon interaction on the energy levels and effective masses of electrons confined in these low-dimensional systems. Polarons in low-dimensional quantum structures are remarkably different from those in bulk material due to the presence of confining potentials, which may also give rise to a rich variety of phonon modes [15–21], such as confined bulk phonon and interface phonon modes. The self-energy expressions obtained taking the various phonon modes into account always look rather complicated. On the other hand, there are also a number of authors [12, 22–23] who avoided including the coupling of the electrons to the confined phonon modes as well as interface phonon modes by adopting bulk phonon approximations, and consequently presented a relatively clear picture of the sole effect arising from the interaction of the electrons and longitudinal optical (LO) phonons.

More recently, Mukhopadhyay and Chatterjee [12] derived a very simple closed-form analytical expression for the ground-state energy of polarons in parabolic quantum dots in two and three dimensions by applying second-order Rayleigh–Schrödinger perturbation theory (RSPT). Since the value of the electron–phonon coupling constant is very small for some important heterostructure materials (e.g. CdS: $\alpha = 0.527$; CdSe: $\alpha = 0.460$; CdTe: $\alpha = 0.315$; GaAs: $\alpha = 0.068$), the RSPT is helpful for dealing with such cases. Motivated by this beautiful work, we extend their method to study the Hamiltonian that describes the interaction of electrons and LO phonons in quantum dots with general parabolic potentials in three dimensions. The confining potential that we choose here is only axially symmetric (i.e. symmetrical in the xy-plane). Consequently, we can obtain a concise expression for the ground-state energy of the polarons in the general system. More interestingly, it is only a one-dimensional integral. If we stopped at this point, this might appear to be a trivial result. Fortunately, it is more interesting than that. From this general expression, by taking different special limits, we can obtain results for almost all interesting cases, such as those of polarons in quantum wells, quantum wires, quantum dots in three dimensions, and quantum dots in two dimensions. We are aware that the interaction matrix for purely one-dimensional polarons diverges due to the Coulombic nature. If this were not the case, we could even extend our discussion to quantum dots in one dimension. As 'by-products', we can obtain the free-polaron ground-state energies in both two and three dimensions for weak coupling. Thus, starting from just one general Hamiltonian, by a standard method, we can clearly provide unifying insight into the electron–LO phonon interactions in many important systems. This is precisely the purpose of the present paper.

This paper is organized as follows. In section 2, we derive the energy expressions for polarons in a general potential within the framework of the second-order RSPT. In section 3, we present our numerical results for reasonably wide ranges of the confinement potential. Finally, we give a summary of the paper.

2. Formulation

We start with Fröhlich's Hamiltonian for an axially symmetric low-dimensional semiconductive quantum structure, generalized for the interaction of an electron with LO phonons:

$$H = -\frac{1}{2}\nabla_r^2 + \frac{1}{2}[\omega^2(x^2+y^2) + \omega_z^2 z^2] + \sum_q a_q^+ a_q + \sum_q [\xi_q \exp(-\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}) a_q^+ + \mathrm{h.c.}] \tag{1}$$

where: all vectors are in three dimensions and the units have been chosen as $\hbar = m = \omega_0 = 1$ (Feynman units); ω_0, the optical phonon frequency, is assumed to be dimensionless; $\boldsymbol{r}$ is the position vector of the electron; $\omega = \omega_h/\omega_0$ and $\omega_z = \omega_{zh}/\omega_0$, where ω_h and ω_{zg} measure the confining strength of the parabolic potential in the xy-plane and the direction $\boldsymbol{z}$, respectively; and a_q^+ (a_q) is the creation (annihilation) operator for a LO phonon of wave vector $\boldsymbol{q}$. Also, ξ_q for the three-dimensional systems is given by [29]

$$|\xi_q|^2 = \frac{2^{3/2}\pi}{vq^2}\alpha \tag{2}$$

where v is the volume of the three-dimensional crystal and α is the electron–phonon coupling constant. The value of α is so small for most semiconductive materials (generally $\alpha < 1$) that the weak-coupling approximation is certain to be appropriate. In addition, we must assume, even in the weak-coupling limit, that the confining potential is stronger than the electron–phonon interaction, in order to ensure that perturbation theory is valid.

On making the second-order RSPT correction to the GS electron self-energy for the polaronic interaction, we obtain

$$\Delta E = -\sum_j \sum_q \frac{|\langle\phi_j(r)|\xi_q \exp(-\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r})|\phi_0(r)\rangle|^2}{E_j - E_0 + 1} \tag{3}$$

where

$$\left\{-\frac{1}{2}\nabla_r^2 + \frac{1}{2}[\omega^2(x^2+y^2) + \omega_z^2 z^2]\right\}\phi_j(\boldsymbol{r}) = E_j\phi_j(\boldsymbol{r}) \tag{4}$$

$$\phi_j(\boldsymbol{r}) = \left(\frac{\omega\omega_z^{1/2}}{\pi^{3/2}2^{j_1+j_2+j_3}j_1!j_2!j_3!}\right)^{1/2} H_{j_1}(\sqrt{\omega x})H_{j_2}(\sqrt{\omega y})H_{j_3}(\sqrt{\omega_z z}) \times \exp\left[-\frac{1}{2}\omega(x^2+y^2) - \frac{1}{2}\omega_z z^2\right] \tag{5}$$

$$E_j = (j_1 + j_2 + 1)\omega + \left(j_3 + \frac{1}{2}\right)\omega_z \tag{6}$$

where $H_n(\omega x)$ is the Hermite polynomial of order n. This expression (equation (3)) is similar to what was obtained as the variational ground-state energy of a Coulomb impurity-bound polaron in a Feynman–Haken path integral calculation with the effective trial action of a harmonic oscillator [25–28].

Using the Slater sum rule for the Hermite polynomials, and the transformation

$$\frac{1}{E_j - E_0 + 1} = \int_0^\infty \exp[-(E_j - E_0 + 1)t]\,\mathrm{d}t \tag{7}$$

one can easily perform the summations over j_x, j_y, and j_z in (3).

Now we introduce two new variables, $\boldsymbol{u}$ and $\boldsymbol{v}$ in place of $\boldsymbol{r}$ and $\boldsymbol{r}'$, in a familiar way:

$$\boldsymbol{u} = \frac{1}{2}(\boldsymbol{r} + \boldsymbol{r}') \qquad \boldsymbol{v} = \frac{1}{2}(\boldsymbol{r} - \boldsymbol{r}'). \tag{8}$$

Integrating over the new variable $\boldsymbol{u}$, we obtain a simple integral for the second-order RSPT correction to the polaron self-energy, which only contains the relative coordinates $\boldsymbol{v}$ and t:

$$\Delta E = -\frac{\alpha\omega\sqrt{\omega_z}}{\pi^{3/2}}\int_0^\infty \mathrm{d}t\,\frac{\mathrm{e}^{-t}}{(1-\mathrm{e}^{-\omega t})\sqrt{(1-\mathrm{e}^{-\omega_z t})}} \times \int \frac{1}{|\boldsymbol{v}|}\exp\left\{-(v_x^2+v_y^2)\left[\omega + \omega\coth\left(\frac{1}{2}\omega t\right)\right] - v_z^2\left[\omega_z + \omega_z\coth\left(\frac{1}{2}\omega_z t\right)\right]\right\}\mathrm{d}\boldsymbol{v}. \tag{9}$$

The solutions of this integral depend on the difference between $\omega[1+\coth(\frac{1}{2}\omega t)]$ and $\omega_z[1+\coth(\frac{1}{2}\omega_z t)]$. Since the function $f(x) = x[1+\coth(\frac{1}{2}xt)]$ increases with increasing x, the value of $\omega[1+\coth(\frac{1}{2}\omega t)]$ is larger than that of $\omega_z[1+\coth(\frac{1}{2}\omega_z t)]$ when ω is larger than ω_z, and vice versa. So, if we changed the relative values of ω and ω_z, different results would be obtained. We will proceed to discuss three different cases.

2.1. $\omega_z = \omega$

With $\omega_z = \omega$, we could give the second-order RSPT correction to the polaron self-energy as follows:

$$\Delta E = -\frac{\Gamma(1/\omega)}{\Gamma(1/\omega + 1/2)}\frac{\alpha}{\sqrt{\omega}}. \tag{10}$$

Here, Γ represents the normal Gamma function.

Defining $l = 1/\sqrt{\omega}$ and rewriting the above equation, we have

$$\Delta E = -\frac{\Gamma(l^2)}{\Gamma(l^2 + 1/2)}\alpha l. \tag{11}$$

Here, l is the dimensionless confinement length and is given by $l = l_0/r_0$, where l_0 and r_0 are defined as

$$l_0 = \left(\frac{\hbar}{m\omega_h}\right)^{1/2} \qquad \text{and} \qquad r_0 = \left(\frac{\hbar}{m\omega_0}\right)^{1/2}.$$

In fact, this just coincides with the case of 3D symmetrical quantum dots with parabolic potentials, discussed by Mukhopadhyay, Chatterjee and others [12, 29–30]. Thus, the results are naturally the same as theirs.

2.2. $\omega_z > \omega$

When the confinement in the direction z is stronger than that in the other two directions ($\omega_z > \omega$), we can easily simplify the form of the polaron GS energy correction to read as follows:

$$\Delta E = -\alpha\sqrt{\frac{\omega_z}{\pi}}\int_0^\infty \mathrm{d}t\, \frac{\mathrm{e}^{-t}}{\sqrt{1-\mathrm{e}^{-\omega_z t}}}\frac{\arctan\sqrt{x-1}}{\sqrt{x-1}} \tag{12}$$

with

$$x = \frac{\omega_z[1+\coth(\frac{1}{2}\omega_z t)]}{\omega[1+\coth(\frac{1}{2}\omega t)]}.$$

We note here that if the confinement in the xy-plane is at the weak-confinement limit (i.e. $\omega \to 0$), then the above equation will obviously yield the results for quantum wells with parabolic potentials. One can easily show that

$$\lim_{\omega\to 0}\omega\left(1+\coth\frac{1}{2}\omega t\right) = \frac{2}{t}.$$

Hence the form of the polaron GS energy can be reduced to

$$\Delta E = -\alpha\sqrt{\frac{\omega_z}{\pi}}\int_0^\infty \mathrm{d}t\, \frac{\mathrm{e}^{-t}}{\sqrt{1-\mathrm{e}^{-\omega_z t}}}\frac{\arctan\sqrt{x't-1}}{\sqrt{x't-1}} \tag{13}$$

where $x' = \frac{1}{2}\omega_z t[1+\coth(\frac{1}{2}\omega_z t)]$. The properties of quantum wells are reflected perfectly by the above formula. Below, we will discuss the other cases derived from the above equation by taking different limits.

From equation (13), we can obtain $\Delta E = -\alpha$, if we also make the direction z a weak-confinement one, like the others. This form corresponds to the limit of a 3D free polaron. The result is also the same as those produced by using second-order perturbative theory [22], the Feynman path integral method [27–28], Lee–Low–Pines variational calculations [31, 32], etc.

Moreover, since it also includes the other two limits:

$$\lim_{\omega_z\to\infty}\left(1+\coth\frac{1}{2}\omega_z t\right) = 2$$

$$\lim_{\omega_z\to\infty}\arctan\sqrt{x-1} = \frac{\pi}{2}$$

then if the direction z is a strong-confinement-limit one ($\omega_z \to \infty$) too, we have

$$\Delta E = -\frac{\Gamma(1/\omega)}{\Gamma(1/\omega+1/2)}\frac{\alpha}{\sqrt{\omega}}\frac{\pi}{2}. \tag{14}$$

This is just another important case of low-dimensional semiconductive quantum structure: 2D symmetrical quantum dots. Similarly, it should again be emphasized here that all of these expressions, and the following ones, are derived just from the general 3D Hamiltonian, and that the results are the same as those derived by others, who dealt with these problems using ND (N-dimensional) or 2D Hamiltonians [10–13].

Furthermore, if we take another limit, $\omega \to 0$, in equation (14), we obtain the ground-state energy of pure 2D polarons: $\Delta E = -(\pi/2)\alpha$, which is the same as the result obtained by many different methods in the literature.

2.3. $\omega_z < \omega$

As the confinement in the direction z is weaker than those in the other two directions ($\omega_z < \omega$), the form of the polaron GS energy correction (9) becomes

$$\Delta E = -\frac{\alpha}{2}\sqrt{\frac{\omega_z}{\pi}}\int_0^\infty \mathrm{d}t\ \frac{\mathrm{e}^{-t}}{\sqrt{1-\mathrm{e}^{-\omega_z t}}}\frac{1}{\sqrt{1-x}}\ln\left(\frac{1+\sqrt{1-x}}{1-\sqrt{1-x}}\right). \tag{15}$$

In the weak-confinement limit ($\omega_z \to 0$) for the direction z, we can easily deduce a new expression for the GS energy for quantum wires from equation (15), which can be written as

$$\Delta E = -\frac{\alpha}{2\sqrt{\pi}}\int_0^\infty \mathrm{d}t\ \frac{\mathrm{e}^{-t}}{\sqrt{t(1-x)}}\ln\left(\frac{1+\sqrt{1-x}}{1-\sqrt{1-x}}\right) \tag{16}$$

where

$$x = \frac{2}{\omega[1+\coth(\frac{1}{2}\omega t)]t}.$$

Equation (16) is related to the case of quantum wires, one of the most important cases of low-dimensional semiconductive quantum structures. Moreover, it certainly exhibits the nature of quantum wires.

Next, we will continue by discussing another case corresponding to a different limit of equation (15). After carefully investigating all of the cases that may arise, we find that if we take the limits $\omega \to 0$ and $x \to 1$, we obtain

$$\lim_{x\to 1}\frac{1}{\sqrt{1-x}}\ln\left(\frac{1+\sqrt{1-x}}{1-\sqrt{1-x}}\right) = 2.$$

Finally, we can obtain $\Delta E = -\alpha$. This just corresponds to the GS energy of the free 3D polarons in the weak-coupling limit. These results are also consistent with what can be inferred from other theory [22, 26–28, 31, 32].

From the above discussion, we notice something particularly interesting: if the effective confinement potential satisfies $\omega \to \infty$ and $\omega_z \to \infty$, the corresponding model is that of a 0D polaron. However, from the limit

$$\lim_{\omega\to\infty}\omega\left(1+\coth\frac{1}{2}\omega t\right) \to \infty$$

we can see that all of the results will diverge. This corresponds to the divergence of the interaction coefficient [12]

$$\lim_{N\to 1}|\xi_q|^2 = \lim_{N\to 1}\frac{\Gamma\left(\frac{1}{2}(N-1)\right)2^{N-3/2}\pi^{(N-1)/2}}{v_N q^{N-1}}\alpha \to \infty. \tag{17}$$

So, renormalization of the coupling constant α is usually called for. More interestingly, we can also easily derive the case of 3D symmetric quantum dots from the case of quantum wells, equation (13), and the case of quantum wires, equation (15), when the confinement potential satisfies $\omega \to \omega_z$ or $\omega_z \to \omega$.

We stress that, starting just from the general 3D Hamiltonian (1), by standard methods, we have obtained an overall picture of low-dimensional semiconductive systems. It not only includes the three most important cases, namely quantum wells, quantum dots, and quantum wires, but also contains a great deal of information on these systems, which can be accessed merely by taking certain limits of the above equations.

3. Numerical results and discussion

In order to calculate the polaronic energy correction, we need to know the ground-state energy in the absence of electron–phonon interaction. It is known that the ground-state energy corresponding to the harmonic Hamiltonian

$$H = P^2 + \frac{1}{2}\omega^2 r^2$$

in N dimensions is given exactly by $E_{hm} = (N/2)\omega$. Hence, the polaronic corrections to the ground-state energy for quantum wells, quantum wires, and 3D asymmetric quantum dots read $\Delta E = E_{hm} - \omega$, $\Delta E = E_{hm} - \frac{1}{2}\omega_x$, and $\Delta E = E_{hm} - \omega - \frac{1}{2}\omega_z$, respectively. As usual, the dimensionless confinement length of the semiconductor quantum structures is defined by $l_x = 1/\sqrt{\omega_x}$, $l_y = 1/\sqrt{\omega_y}$.

Now, we will present some numerical results for the polarons in semiconductor quantum structures for arbitrary coupling constants and broad ranges of the confinement lengths of these structures, obtained by means of equations (12)–(17).

Firstly, as stated before, from equation (12), we are certain to obtain the expression for the case of quantum wells (equation (13)) and the case of quantum wires (equation (16)), when the confinement length l or l_z is increased ($l \to \infty, l_z \to \infty$). After numerically solving equations (13) and (16), we plotted the polaronic correction $(-\Delta E)$ to the ground-state energy as a function of the confinement length l (or l_z); this is shown in figure 1. From this figure, we can obtain a great deal of information about the quantum well and quantum wire systems. It is very obvious that the electron–optical phonon interaction has a pronounced effect on the electronic energy when the confinement length l (or l_z) is sufficiently small. As the sizes of these two quantum systems increase, the polaronic correction increases and asymptotically

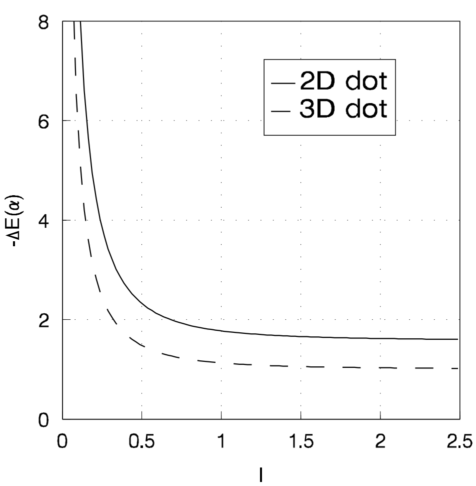

Figure 1. The polaronic corrections $-\Delta E$ (Feynman units) to the GS energy of an electron in a parabolic quantum dot as a function of the confinement length l (Feynman units), for both two and three dimensions.

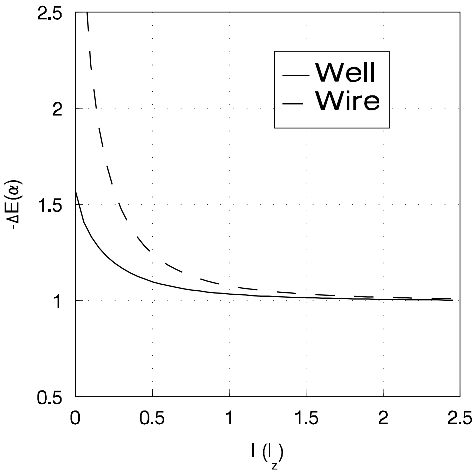

Figure 2. The polaronic corrections $-\Delta E$ (Feynman units) to the GS energy of an electron in a parabolic quantum well and that of an electron in a quantum wire as functions of the confinement lengths l and l_z (Feynman units).

assumes a constant value, exactly as stated by Hai, Peeters, and Devreese [24], and many other authors [12, 22, 31, 32]. Moreover, we also find that, for given values of the electron–phonon coupling constant α and the confinement length l, it is evident from the figure that the polaronic effect for quantum wires is stronger than that for quantum wells. In other words, the polaronic effect is more pronounced in smaller systems.

When the confinement length for the directions x and y, namely l, is equal to l_z, the confinement length for the direction z, we can derive the cases for 3D symmetric quantum dots. In exactly the same way, when the confinement length for the direction z (l_z) satisfies $l_z \to 0$, we can also obtain the case for 2D quantum dots. The information for the 2D quantum dot and 3D quantum dot is more clearly shown in figure 2. From the data shown in the figure, we conclude that the electron–optical phonon interaction also has a pronounced effect on the electronic energy when the dot size is sufficiently small. Like for the other two quantum cases, the polaronic correction increases and asymptotically assumes a constant value as the size of the dot increases for both 2D and 3D systems. Also indicated are features of low-dimensional quantum systems. In addition, it may be noticed from the figure that, for given values of the electron–phonon coupling constant α and the confinement length l, the polaronic effect is stronger for a 2D dot than for a 3D dot.

Furthermore, we can display all of the above cases in a three-dimensional plot. In figure 3, we have plotted the polaronic corrections ($-\Delta E$) to the ground-state energy in three dimensions as a function of the confinement lengths l and l_z. It is quite clear from the figure that we can observe all of the cases listed above in this configuration. We can also deduce the different confinements for the quantum wells, quantum wires, and quantum dots, and the general trends exhibited by these low-dimensional quantum systems corresponding to the different relative sizes of such structures, just as we discussed above. It is emphatically also the case that we can derive further important results from this figure. We can see by inspection the general behaviour of the low-dimensional quantum systems. When the confinement of the quantum wires is increased, the scenario tends to the case of the 1D polaron; when the confinement of the quantum well is increased, it tends to the case of the 2D polaron. Furthermore, if we compare all of these cases of low-dimensional quantum systems, we can observe the relationship between

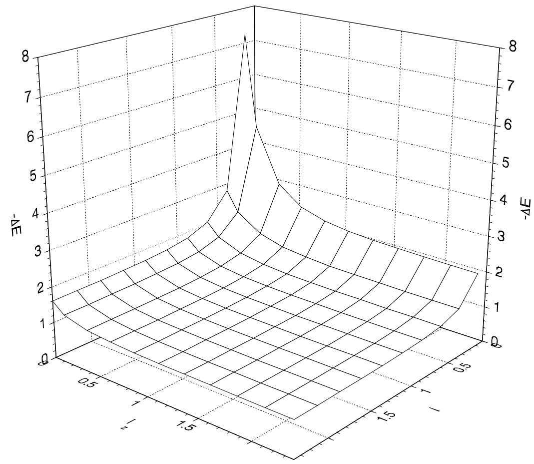

Figure 3. The polaronic corrections $-\Delta E$ (Feynman units) to the GS energy of an electron in a parabolic low-dimensional semiconductive quantum system as a function of the confinement lengths l and l_z (Feynman units).

the degree of confinement and the polaronic correction. It may be noted from the figure that, for given values of the electron–phonon coupling constant α and the confinement length l, the polaronic effect is stronger for the smaller and less symmetric quantum systems. For example, the polaronic effect for a 2D quantum dot is stronger than that for a 3D quantum dot; the polaronic effect for a quantum wire is weaker than that for a 2D quantum dot; the polaronic effect for a quantum wire is weaker than that for a quantum dot but stronger than that for a quantum well. Moreover, when the extent of the confinement increases, the polaronic corrections for the quantum well and the quantum wire will asymptotically assume the same constant value as that for the 3D quantum dot.

In addition, the results show that there is a ‘bulk’ limit. These conclusions are certainly consistent with what has been deduced elsewhere in the literature.

4. Conclusions

Some simple expressions for the ground-state energies of low-dimensional semiconductive quantum systems can be derived from just one general Hamiltonian within the framework of the RSPT. After considering all possible cases for low-dimensional quantum structures, we were able to establish general and individual characteristics of these low-dimensional semiconductive quantum systems. Then we unified them in a three-dimensional plot. As a result of numerical calculations, we were able to state that for given values of the electron–phonon coupling constant α and the confinement length l, the polaronic effect is enhanced with lowering dimension and increasing asymmetry.

It is also shown that there is an important relationship between the polaronic effect and the extent of the confinement. We can derive a relationship among the systems, from figures 1–3, as follows. The polaronic effect for a 2D quantum dot is stronger than that for a 3D quantum dot. The polaronic effect for a quantum wire is weaker than that for a quantum dot, but stronger than that for a quantum well. When the confinement length increases, the polaronic corrections for the quantum well and the quantum wire asymptotically assume the same constant value as that for the 3D quantum dot.

Finally, it should be pointed out that the present theory is also suitable for addressing other more complicated problems. Such extensions are in progress.

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