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1989 J. Phys.: Condens. Matter 1 1999

(http://iopscience.iop.org/0953-8984/1/11/007)

J. Phys.: Condens. Matter 1 (1989) 1999–2007. Printed in the UK

Polaron properties of the Wannier exciton in a quantum-well confinement

A Erçelebi and U Özdinçer
Department of Physics, Middle East Technical University, 06531 Ankara, Turkey

Received 28 March 1988, in final form 25 July 1988

Abstract. On the basis of a perturbative–variational approach, the polaron effect on the ground-state energy of the two-dimensionally confined exciton is analysed in the overall range of the parameters characterising the problem. With particular attention devoted to the GaAs-based quantum structure, it is seen that the phonon-induced effects on the binding are rather noticeable and should not be ignored.

1. Introduction

Recently, the study of quasi-two-dimensional Wannier excitons has become a subject of interest in the context of quantum-well confinement of bound states in heterostructure-type semiconductor complexes. With particular emphasis given to the GaAs–GaAlAs system, the common conclusion reached in the relevant works is that the excitonic energy levels become deepened by appreciably large factors over the corresponding bulk values (Bastard *et al* 1982, Greene and Bajaj 1983, Greene *et al* 1984, Jiang 1984). For the case of infinite confining barriers for the electron and the hole, it has been well established that the binding energy approaches four times the bulk effective rydberg when the exciton motion is strictly two-dimensional.

A further, yet interesting, aspect that deepens the binding is the contribution coming from exciton–phonon coupling. Recent calculations performed along this line (Erçelebi and Özdinçer 1986, Degani and Hipólito 1987a, b) have revealed that the combined effect of phonon coupling together with the confinement achieved by the reduction in the effective dimensionality of the system leads to rather noticeable polaronic enhancements in the binding. In all the aforementioned work the calculations have been restricted solely to parameter values pertinent to GaAs, disregarding a more complete view of the polaron effect beyond that given for the GaAs-based quantum structure. We therefore would like to look at the problem somewhat differently using a perturbation–variation approach previously used by Devreese *et al* (1982) in their study of a bound polaron. This method has the advantage of being applicable in the overall range of Coulomb binding and phonon coupling parameters. An adiabatic (strongly coupled) excitonic state combined with a first-order perturbative correction is used as a variational wavefunction by which it is possible to achieve a smooth extrapolation towards the weak-coupling regime.

Obviously, with the inclusion of electron(hole)–phonon interactions, the problem becomes much more complicated compared to the bare exciton case. The effect of the

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lattice polarisation field brings about interesting and distinctive features in the various regimes of the problem characterised by the strength of phonon coupling and the Coulomb potential, and further by the well width, all of which enter the theory in rather involved and inter-related manners. The quantitative analysis of the problem thus becomes somewhat challenging, and therefore we are tempted to utilise an artificially simple model where the exciton wavefunction is taken to be purely two-dimensional. As such, the primary and essential approximation adopted in this work is to take into account only the two-dimensional nature of the dynamical behaviour of the electron–hole complex in a thin quantum well, and thus eliminate the extra difficulty in the mathematical structure of the problem comprised by the third dimension. This, besides facilitating our calculations, provides a means by which one can study the significance of, or at least the order of magnitude of, the polaron effect when the exciton gets squeezed between the confining barriers. In fact, the strict two-dimensional approximation for the spatial extent of the electron (coupled to bulk phonons) has already been used for the free- and bound-polaron problems in order to give some insight into the qualitative aspects of the polaron effect in confined structures (Das Sarma and Mason 1985, Mason and Das Sarma 1986). Following the same line, we thus take the particle part of the trial state to depend on the x and y coordinates only, and leave out the third spatial dimension. At this point we feel that regarding the z coordinate as being completely separable (as confirmed by Bastard *et al* (1982) for bare excitons in narrow quantum wells), or totally ignoring it, is not expected to result in drastic alterations in the qualitative fundamentals of the problem. Meanwhile, we cannot still deny the possibility that, as the phonon coupling is turned on, the transverse coordinates may become incorporated with the z direction. This, in fact, may lead to further interesting features, which we do not want to discuss at this point. For the present, we shall be content with the two-dimensional formulation of the problem, and retain the discussions pertaining to finite well widths until the last section of this paper.

In summary, the model we use consists of a purely two-dimensional characterisation of the electron–hole pair immersed in the field of bulk LO phonons of the relevant well material. It should be emphasised that for the present we have refrained from including the coupling of the exciton to the interface phonon modes because only then can a direct correspondence of our two-dimensional predictions be made with the results derived previously for the bulk case (Pollmann and Büttner 1977) where the interface phonons are totally absent. More explicitly, our main concern is primarily to give a clear view of the bulk-phonon effects stripped from all other perturbing quantities. Apart from omitting the contribution that may come from the interface optical phonons, we have also ignored any screening effects and further complications such as those due to the band non-parabolicity or the loss of validity of both the effective-mass approximation and the Fröhlich continuum Hamiltonian in ultra-thin microstructures. Nevertheless, in view of all these simplifying assumptions we still bear the hope that our results should shed some light on the polaron effect on the exciton–phonon system in a quantum-well confinement. We furthermore believe that the formalism followed in this work provides an appropriate way to account for the interesting mutual influence of the polarisation of the electron and the hole whose crucial significance has already been raised by Pollmann and Büttner (1977) in three dimensions.

2. Theory

The Hamiltonian describing a Wannier exciton confined in two dimensions and interacting via the Fröhlich Hamiltonian with the bulk LO phonons can be expressed, using

the centre of mass $\boldsymbol{R}$ and relative coordinates $\boldsymbol{r} = \boldsymbol{r}_e - \boldsymbol{r}_h$, as

$$H = -[\sigma/(\sigma+1)]\nabla_R^2 - (\sigma+1)\nabla_r^2 - (\beta/r) + \sum_Q a_Q^\dagger a_Q + \sum_Q V_Q(a_Q\, e^{i\boldsymbol{q}\cdot\boldsymbol{R}}\, e^{is_1\boldsymbol{q}\cdot\boldsymbol{r}} - cc) - \sum_Q V_Q(a_Q\, e^{i\boldsymbol{q}\cdot\boldsymbol{R}}\, e^{-is_2\boldsymbol{q}\cdot\boldsymbol{r}} - cc) \tag{1}$$

where $s_2 = m_h/(m_e + m_h)$, $s_2 = m_e/(m_e + m_h)$ and $\sigma = m_e/m_h$ is the electron–hole effective-mass ratio. The interaction amplitude for the exciton and the phonon field is related to the phonon wavevector $\boldsymbol{Q}\,(\boldsymbol{q}, q_z)$ and the coupling constant α through $V_Q = (4\pi\alpha)^{1/2}/Q$. The strength of the Coulomb potential between the electron and the hole is given by

$$\beta = \frac{e^2}{\varepsilon_0}\left(\frac{2m_e}{\hbar^3\omega_0}\right)^{1/2} \tag{2}$$

in terms of which the two-dimensional effective rydberg and the corresponding Bohr radius are

$$R_{2D} = (1+\sigma)^{-1}\beta^2 \qquad a_{2D} = (1+\sigma)\beta^{-1}. \tag{3}$$

In the above, energies have been scaled by $\hbar\omega_0$ and lengths by $(\hbar/2m_e\omega_0)^{1/2}$ with ω_0 being the LO phonon frequency.

Since the Hamiltonian is invariant under translations of the centre of mass and the lattice distortion together, it is possible to transform to a representation in which the centre-of-mass coordinates do not appear. Under the unitary transformation

$$U = \exp\left(-i\sum_Q a_Q^\dagger a_Q \boldsymbol{q}\cdot\boldsymbol{R}\right) \tag{4}$$

the Hamiltonian takes the form

$$H' = H_0 + \sum_Q \gamma_Q a_Q^\dagger a_Q + \sum_Q V_Q(a_Q\, e^{is_1\boldsymbol{q}\cdot\boldsymbol{r}} - cc) - \sum_Q V_Q(a_Q\, e^{-is_2\boldsymbol{q}\cdot\boldsymbol{r}} - cc) + [\sigma/(\sigma+1)]\sum_Q\sum_{Q'} \boldsymbol{Q}\cdot\boldsymbol{Q}' a_Q^\dagger a_{Q'}^\dagger a_Q a_{Q'} \tag{5}$$

$$H_0 = -(\sigma+1)\nabla_r^2 - \beta/r$$

with $\gamma_Q = 1 + [\sigma/(\sigma+1)]Q^2$. The phonon correlation term will be omitted in the following formulation of the theory (Wang and Matsuura 1974).

Regardless of the parameter values characterising the exciton–phonon system, the starting step is the usual canonical transformation of the strong-coupling formalism. The theory is based on utilising a product wavefunction of the form (Devreese *et al* 1982)

$$\psi = \left\{c|0\rangle + \sum_Q V_Q g_Q \eta_Q^* a_Q^\dagger|0\rangle\right\}\varphi(\boldsymbol{r}) \tag{6}$$

together with the displaced phonon state operator

$$D = e^S \qquad S = \sum_Q (V_Q\sigma_Q/\gamma_Q)(a_Q - a_Q^\dagger) \tag{7}$$

in which

$$\sigma_Q = \langle\varphi(\boldsymbol{r})|(e^{is_1\boldsymbol{q}\cdot\boldsymbol{r}} - e^{-is_2\boldsymbol{q}\cdot\boldsymbol{r}})|\varphi(\boldsymbol{r})\rangle. \tag{8}$$

In the above, $|0\rangle$ is the phonon vacuum, $\varphi(\boldsymbol{r})$ is the exciton internal wavefunction, c is a constant which serves for normalisation, and

$$\eta_Q = e^{is_1\boldsymbol{q}\cdot\boldsymbol{r}} - e^{-is_2\boldsymbol{q}\cdot\boldsymbol{r}} - \sigma_Q. \tag{9}$$

The remaining symbol g_Q in the trial state is to be determined variationally and gives the fractional admixture of the strong- and weak-coupling counterparts of the theory.

At this point it should be mentioned that g_Q bears a crucial importance for the present problem since the weak-coupling regime is readily attained even for strong electron(hole)–phonon interactions. In tightly bound electron–hole pairs (i.e. for not too weak β), the mutual interaction between the oppositely charged particles is expected to result in partial annihilation of the lattice polarisations created by either particle. Thus, even for a large α, the influence of the lattice polarisation on the interacting electron–hole pair may become considerably reduced and therefore, without the correction term in the trial state (6), one cannot achieve a satisfactory description of the polaronic aspects of the coupled exciton–lattice system. The same is true also for the case of equal-mass particles ($s_1 = s_2$), which, in the strict adiabatic treatment, exhibit the properties of a bare exciton.

An optimal fit to g_Q is obtained by minimising the expected value of $D^{-1}H'D$ in the state (6) subject to the constraint

$$\langle\psi|\psi\rangle = c^2 + \sum_Q V_Q^2 g_Q^2 \delta_Q = 1 \tag{10}$$

in which

$$\delta_Q = 2[1 - \langle\varphi(\boldsymbol{r})|\cos(\boldsymbol{q}\cdot\boldsymbol{r})|\varphi(\boldsymbol{r})\rangle] - \sigma_Q^2. \tag{11}$$

For the energy we then have

$$E = e_0 - \chi_0 + \chi. \tag{12}$$

Here, χ is a Lagrange multiplier which implicitly depends on α, β, σ and $\varphi(\boldsymbol{r})$ through the transcendental equation

$$\chi = \sum_Q V_Q^2 (g_Q/c)\delta_Q \tag{13}$$

where

$$g_Q/c = -\delta_Q/[\delta_Q(\gamma_Q - e_0 + 2\chi_0 - \chi) + e_Q - h_Q] \tag{14}$$

with

$$e_0 = \langle\varphi(\boldsymbol{r})|H_0|\varphi(\boldsymbol{r})\rangle \tag{15}$$

$$\chi_0 = \sum_Q V_Q^2 \sigma_Q^2/\gamma_Q \tag{16}$$

$$e_Q = \langle\varphi(\boldsymbol{r})|\eta_Q H_0 \eta_Q^*|\varphi(\boldsymbol{r})\rangle \tag{17}$$

$$h_Q = \sum_{Q'} 2V_{Q'}^2 \sigma_{Q'}/\gamma_{Q'}\langle\varphi(\boldsymbol{r})|\eta_Q[\cos(s_1\boldsymbol{q}'\cdot\boldsymbol{r}) - \cos(s_2\boldsymbol{q}'\cdot\boldsymbol{r})]\eta_Q^*|\varphi(\boldsymbol{r})\rangle. \tag{18}$$

At this stage we adopt a variational form for the exciton internal wavefunction $\varphi(\boldsymbol{r})$. As the simplest reasonable approximation, we shall use the two-dimensional 1s state, which we believe is well suited for the calculation of good upper bounds to the ground-state energy of the exciton–phonon system:

$$\varphi(\boldsymbol{r}) = (2/\pi)^{1/2}\lambda^{-1}\exp(-r/\lambda) \tag{19}$$

wherein λ is a further adjustable parameter governing the spatial extent of the exciton wavefunction. With this choice we obtain

$$\sigma_Q = H^3(s_1 q) - H^3(s_2 q) \tag{20}$$

where we have defined

$$H(x) \equiv [1 + (\lambda/2)^2 x^2]^{-1/2}. \tag{21}$$

Furthermore, for the set of equations (15)–(18) we have the following functional forms:

$$e_0 = (\sigma + 1)/\lambda^2 - 2\beta/\lambda \tag{22}$$

$$\chi_0 = \alpha \int_0^\infty \mathrm{d}q \sigma_Q^2 \{1 - q/[q^2 + (\sigma + 1)/\sigma]^{1/2}\} \tag{23}$$

$$e_Q = 2e_0[1 - H(q)] + (3e_0 + 4\beta/\lambda)\sigma_Q^2 - 4(e_0 + \beta/\lambda)\sigma_Q[H(s_1 q) - H(s_2 q)] + q^2\{(\sigma^2 + 1)/(\sigma + 1) - \tfrac{1}{2}[(\sigma - 1)^2/(\sigma + 1)]H^3(q)\} \tag{24}$$

and

$$h_Q = 2\chi_0(2 + \sigma_Q^2) - (8/\pi)\alpha \int_0^\infty \mathrm{d}q' \sigma_{Q'}\{1 - q'/[q'^2 + (\sigma + 1)/\sigma]^{1/2}\} F_{Q,Q'}(s_1, s_2) \tag{25}$$

in which

$$F_{Q,Q'}(s_1, s_2) = f(1, s_1) - f(1, s_2) + \sigma_Q[f(s_1, s_1) + f(s_2, s_2) - f(s_1, s_2) - f(s_2, s_1)] \tag{26}$$

with

$$f(x, x') \equiv E(m)/\mu_+ \mu_-^2 \tag{27}$$

where

$$\mu_\pm = [1 + (\lambda^2/4)(qx \pm q'x')^2]^{1/2} \tag{28}$$

and $E(m)$ is the complete elliptic integral of the second kind with parameter

$$m = \sin^2[\lambda(qq'xx')^{1/2}/\mu_+]. \tag{29}$$

Finally, for the transcendental equation (13) we have

$$\chi = -\alpha \int_0^\infty \mathrm{d}q(\delta_Q/\Delta_Q)\{1 - q/[q^2 + ((\sigma + 1)/\sigma)\Delta_Q]^{1/2}\} \tag{30}$$

where

$$\Delta_Q = 1 + (e_Q - h_Q)/\delta_Q - (e_0 - 2\chi_0 + \chi). \tag{31}$$

Because of the analytic complexity, the optimal fits to χ and λ can only be achieved by numerical techniques.

3. Results and discussion

With the inclusion of phonon coupling, the exciton problem becomes extended to the case of two interacting polarons where the properties of the electron–hole pair become strongly modified by the polarisation field. In this part of the work we analyse the phonon effects on the ground-state property as a function of the parameters characterising the exciton–phonon system.

Before doing so, however, we would like to make a small digression and give a

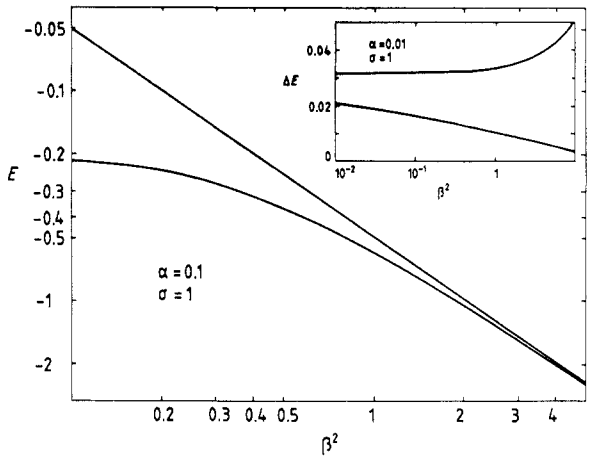

Figure 1. The ground-state energy versus the Coulomb strength. The lower (upper) curve is for the case with (without) phonon coupling. The inset (ΔE versus β^2) provides a comparison of the present theory (lower curve) with the approximate formula (32) (upper curve).

comment on the basic distinction that sets the present approach apart from the strong-coupling theory. It should be noted that, omitting the perturbative correction χ in equation (12), we arrive at the strong-coupling results for the ground-state energy with $\Delta E = \chi_0$. Regardless of the values of α and β, we trivially obtain $\sigma_Q = 0$ and hence $\Delta E = 0$ for equal-mass particles since, when $\sigma = 1$, the net charge density of the oppositely charged particles is exactly zero. This, however, is an artifact of the adiabatic approximation where the lattice is taken to be responding to the average charge-density fluctuations of the pair rather than the individual kinetics of the particles. We thus readily note that the essential role which parameter χ plays in the theory becomes very prominent in all cases where the adiabatic theory becomes inadequate. It is in fact through this parameter that the theory sets up a weighted average incorporating the weak- and strong-coupling aspects of the problem. Particularly for the exciton problem where mostly the weak-coupling aspect is dominant, the present formulation proves to be superior in all respects to the strong-coupling approximation and yields comparatively deeper binding.

Studying first the α-dependence in equation (12), we find that the polaronic shift in the binding energy, $\Delta E = |E - E(\alpha = 0)|$, increases almost linearly with the coupling constant. Taking $\beta = 1$ and $\sigma = 1$, for instance, we obtain $\Delta E \simeq 0.0197$, 0.0495 and 0.0986, respectively, for $\alpha = 0.02$, 0.05 and 0.10, amounting to a percentage deepening in the binding by 3.9%, 9.9% and 19.7%. We also note that the linear character in the α-dependence of the polaron effect is not altered by changing the mass ratio or the strength of the Coulomb potential.

Confining our discussions to the β-dependence, we observe that ΔE decreases steadily with increasing β, since for a small-size complex the lattice polarisations interfere destructively and eventually become totally annihilated. This feature is displayed explicitly in figure 1 where we plot the ground-state energy profile for both the bare ($\alpha = 0$) and polaronic ($\alpha = 0.1$) excitons. We find that the phonon coupling contributes to the ground-state energy by considerably large factors for weak Coulomb potentials. With $\sigma = 1$, the energy becomes lowered by factors of about 2.4 and 1.5 for $\beta^2 = 0.2$ and 0.5, respectively. For stronger binding the phonon contribution rapidly loses its prominence and becomes as small as 2% for $\beta^2 = 5$.

At this point it should be noted, however, that the aforementioned large factors by

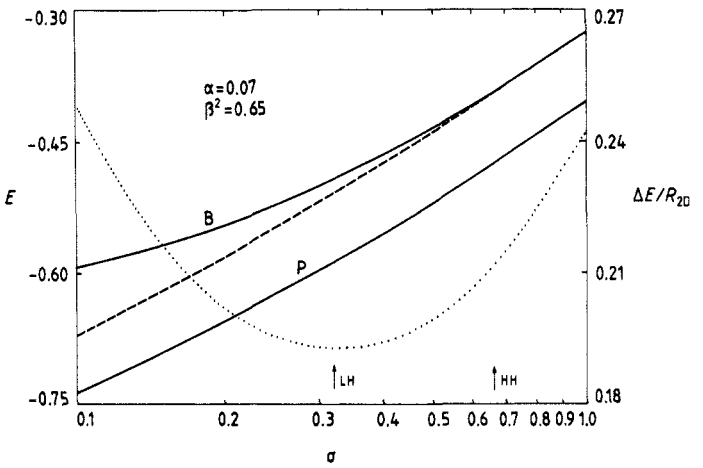

Figure 2. The ground-state energy versus the mass ratio. The solid curves B and P are for the bare and polaronic excitons, respectively. The broken curve displays the results of the strong-coupling polaron theory. The dotted curve is for the relative phonon shift. The arrows are aimed at the values for the light ($\sigma = 0.32$) and heavy ($\sigma = 0.66$) hole excitons in GaAs. The scale on the left (right) is for the ground-state energy (the relative polaron shift).

which the energy deepens may be misleading since ΔE includes implicitly some fraction of the self-energies of the electron and hole polarons. In fact the percentage of the self-energy $(-\pi\alpha/2)$ of either polaron that plays a significant role in the binding and the percentage that can be regarded as separable are governed mostly by β, and further by σ.

For the case when the inter-particle separation is taken to be artificially large, the qualitative features of the complex become considerably simplified. In this case the electron and hole polarons can be visualised as orbiting about one another with almost no overlapping parts and, therefore, for the polaron shift we write

$$\Delta E \simeq \pi\alpha[1 + \tfrac{1}{8}(1+\sigma)^{-1}\beta^2] \tag{32}$$

wherein we have scaled the bare rydberg (3) by replacing the electron mass by the polaron mass $m_p = (1 + \pi\alpha/8)m_e$. Clearly, the approximate formula given above is intended to work fairly well only when the polarons are thought to be completely separated. With growing β, it becomes no longer valid and exhibits a drastically large deviation from the actual behaviour as exemplified by the present theory (cf the inset of figure 1). This in fact indicates that the overlapping polaron effects are extremely important and alter the qualitative aspects of the phonon contributions to the problem.

A further point of view giving more impact into the partial overlap between the polaron clouds is achieved by examining the ground-state energy as a function of the mass ratio. In figure 2 we make plots for the cases with and without phonon coupling, including as well the results of the adiabatic theory for completeness. In our numerical computations we have selected $\alpha = 0.07$ and $\beta^2 = 0.65$, appropriate for GaAs where $\hbar\omega_0 \simeq 35$ meV is the unit of energy. A careful examination of the curves B and P, corresponding to the bare and polaronic excitons, reveals the following salient features. We at once observe that, as the mass ratio is varied from a small value to unity, the phonon-induced shift in the ground-state energy becomes reduced monotonically. This follows essentially from the fact that, with increasing σ, the size of the complex grows larger, resulting in weaker binding and hence in a weaker polaron effect. The consequences brought about by the decrease in the degree of localisation of the system are

however twofold. A contrasting aspect is that, as the exciton size is made larger, the destructive interference effects become less significant and, as a result, the self-energies of the polarons take part in the binding in a more efficient way. Thus, the two competitive aspects bring out a further interesting feature in the profile of the polaron effect as a function of the mass ratio. Considering the relative shift $\Delta E/R_{2D}$ rather than ΔE itself (cf the dotted curve in figure 2), we see that the phonon-induced contribution goes through a minimum at about $\sigma = 0.3$. Starting from $\sigma = 1$, the effect of the overlap between the polaron clouds dominates first leading to a reduction in $\Delta E/R_{2D}$. Meanwhile, as the mass ratio is shifted down to small values the effect of localisation starts to show up and for $\sigma < 0.3$ the exciton gets sufficiently localised so as to result in an overall enhancement in the relative phonon shift in the binding.

Having retrieved the basic qualitative aspects of the problem in two dimensions, we now make some correspondence with the GaAs-based quantum structure. It should be evidently clear that a totally satisfying comparison of our results with the experimental observations is rather impossible at this stage. The main reason for this is that the strictly two-dimensional approximation we have adopted is only interesting from a formal point of view and cannot produce an accurate physical picture for finite-width quantum wells. However, some insight can still be achieved from a qualitative viewpoint providing a conformation of the two-dimensional results to finite well widths. Starting from the two-dimensionally localised case and going to the bulk limit, the effective phonon coupling is expected to decrease significantly, since relaxing the system in the third spatial dimension leads to comparatively weaker binding. The important question at this point is whether or not the polaron effect falls off rapidly as the well width is introduced into the problem as a further parameter. Such a possibility has in fact been realised previously for the donor complex (Erçelebi and Süalp 1987), where it has been observed that the polaron shift in the ground level lies far below its two-dimensional value and is even smaller than in the bulk except for too narrow wells. We feel that this feature should not totally reflect the present case since the exciton problem has a distinguishing counterpart which has already been mentioned in the discussion for the σ-dependence. In the meantime ΔE tends to become smaller owing to the reduction in the degree of confinement, and the amount by which the polaron clouds overlap diminishes, resulting in a greater fraction of the polaron self-energies taking part in the binding. We thus expect that the rate at which the polaron effect loses its significance should not be as rapid as in the donor case.

With parameter values appropriate for GaAs we find that the polaronic deepening in the two-dimensional ground-state energy is as large as 20%, giving an indication in favour of a non-negligible polaron effect in thin quantum wells. For the heavy(light)-hole exciton we take the reduced mass in the xy plane to be $\mu = 0.04$ (0.05). It then follows that $\sigma = (m_e/\mu) - 1 = 0.66\,(0.32)$, yielding $E \simeq -13.8\,(-17.3)$ meV for the bare exciton and $E \simeq -16.7$ (-20.7) meV when phonon coupling is included. It should be emphasised that the effective transverse-mass values employed in the calculation are those which are commonly used in the literature for bulk GaAs. There is in fact some degree of uncertainty in our values for the binding energy owing to the ambiguities in the relevant parameter values brought about by quantum confinement (see, for instance, Maan *et al* 1984, Rogers *et al* 1986). Yet another aspect that casts further doubt on correlating our results with experiment is the uncertainty in the percentage enrolment of the polaron self-energies which actually contributes to the observable binding energy. Nevertheless, in spite of these drawbacks we still feel that the results of the present work give enough evidence to account for some part of the large discrepancy between the experimental binding energy and the theoretical predictions made for the bare exciton case.

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