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Electronic structure of donor–impurity complexes in GaAs/Ga_{1-x}Al_xAs quantum wells

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Abstract. The ground state of the two-electron donor–impurity complexes D^- and D_2^0 confined in a quantum well is analysed by using a variational procedure. A model approximation that can be used in the two-electron problem in order to separate the variables is proposed, and it is shown that, for the D^- negative ion and the D_2^0 complex, the electron–electron interaction may be eliminated, in this approximation, by introducing an additional effective charge at a centre of symmetry. The D^- binding energy is calculated as a function of the GaAs/Ga_{1-x}Al_xAs quantum-well width for different magnetic field strength values, whereas the D_2^+ and D_2^0 dissociation energies are calculated as functions of the spacing between the impurities in the complexes, and for different well widths. The results for the first ($D_2^0 \rightarrow D_2^+ + e$) and the second ($D_2^+ \rightarrow D^+D^+ + e$) ionization binding energies as functions of the well width are presented for different separations between impurities. Finally, the scheme that we propose is extremely simple and provides a realistic description of few-particle ground-state electronic structures confined in a quantum well.

1. Introduction

Low-dimensional semiconductor microstructures with neutral and charged impurities and impurity complexes exhibit new phenomena which present a lot of challenges to both theoretical and experimental physics. A considerable amount of work has been carried out on the shallow-neutral-donor electronic properties of GaAs/Ga_{1-x}Al_xAs quantum-well (QW) structures [1–3]. It has been shown [4–6] that the dimensionality decreases with the atomic confinement, and this leads to a change in the symmetry from spherical to planar and to an increase in the ground- and excited-state binding energies. Shallow negative D^- donors (neutral shallow donors that each bind an additional electron) have been experimentally observed [7, 8] in GaAs/Ga_{1-x}Al_xAs QWs. It has also been demonstrated [9, 10] by means of variational calculations that the two-dimensional D^- binding energy is about ten times greater than the three-dimensional one, and that the impurity mean-value radius is reduced by half when its form is changed from spherical to planar. For this reason, optical excitations of the D^- complex may fall in the same spectral [11] region as the D^0 state in a QW. The identification of the D^- features in a given experimental spectrum is therefore not straightforward, and this suggests the need for a careful theoretical analysis. Any calculation of the D^- spectrum has to take into account the electronic correlation, which is responsible for the system's stability. Variational [10–12] and Monte Carlo [13] methods have been

used to find the D^- binding energy in a QW. Each of these two types of calculation requires a set of adjustable parameters, and quite long computational times (for example, the Chandresekhar-type [14] trial function with seven variational parameters [12] has been used). Due to the computational cost of these types of calculation, no detailed studies of the evolution of the spectrum as functions of the well width, barrier height, donor position, etc, have been performed. Similar difficulties should occur in D_2^0 -complex (two-electron problem) analysis.

The linear and non-linear optical properties of GaAs/Ga_{1-x}Al_xAs QWs depend on shallow levels dominated by excitons, which represent another few-particle-complex example. Excitons and biexcitons (analogous to hydrogen atoms and molecules, respectively) are the best-known examples of such complexes. The trions predicted by Lampert in 1958 [15], the negatively charged exciton (X^- : two electrons bound to a hole), and the positively charged exciton (X^+ : two holes bound to an electron) are much less familiar [16–18]. The trion binding energy is substantially increased in a GaAs/Ga_{1-x}Al_xAs QW [16, 19]. It has also been demonstrated that as the electron–hole effective-mass ratio m_e^*/m_h^* is reduced, excitonic complexes become more stable [20]. As $m_e^*/m_h^* \rightarrow 0$, the X^- , X^+ , and biexciton complexes are reduced to the D^- negative ion, D_2^+ complex, and D_2^0 molecules, respectively.

Each of the above-mentioned problems refers to one kind of few-particle-complex problem in a non-integer space dimension changing between two and three. In dimensional-scaling theory, it was shown [21] that the two-dimensional Schrödinger equation for few-particle systems presents one special case in which the correlation effect becomes very pronounced. For this condition, it is reasonable to use a symmetrical-structure [22] model with fixed-electron relative positions, which was successfully used to calculate the binding energy of a two-dimensional biexciton.

In this work we propose a similar model scheme for the ground-state electronic structure of the D^- centre and D_2^0 covalently bound molecule. We will then show that the D^- and D_2^0 (two-electron problem) Hamiltonians become separable and are reduced to the D^0 and D_2^+ (one-electron problem) Hamiltonians with an additional negative effective charge at a symmetry centre.

This work is organized as follows. In section 2 we introduce the theoretical scheme used throughout the study of the D_2^+ , D^- , and D_2^0 ground-state properties. Section 3 presents our calculated results and discussion, and section 4 gives the conclusions.

2. The theoretical framework

As commented in the introduction, the symmetrical-structure model [22] provides a simple framework for the treatment of few-particle systems. We choose to study the ground state of the two-electron complexes D^- and D_2^0 , which are conveniently reduced, via the symmetrical-structure model, to the problem of D^0 and D_2^+ centres (a one-electron problem). In the case of the D^- centre, we include the effects of applied magnetic fields, in order to compare our results with previous theoretical calculations and experimental data.

2.1. The ground state of the bound D_2^+ complex

First we study the bound D_2^+ complex as a final state of the D_2^0 molecular complex in the ionization process for which the binding energy depends on the difference between the D_2^0 and D_2^+ ground-state energies. Additionally, the D_2^+ complex is one of the simplest few-particle models, with two positive ions interchanging their only electron. We assume,

for the sake of simplicity, that the two positive ions with separation R are located in the centre region of a GaAs/Ga_{1-x}Al_xAs QW at the $(0, 0, 0)$ and $(R, 0, 0)$ positions. Within the framework of the effective-mass approximation, the dimensionless Hamiltonian (without nuclear repulsion) for the ground state of the D_2^+ complex may be written as

$$H = -\frac{1}{\rho_1} \frac{\partial}{\partial \rho_1} \rho_1 \frac{\partial}{\partial \rho_1} - \frac{\partial^2}{\partial z^2} - \frac{1}{\rho_1^2} \frac{\partial^2}{\partial \theta^2} + V(z) - \frac{2}{r_1} - \frac{2}{r_2} \quad (2.1)$$

where ρ_1 , z , and θ are the electron cylindrical coordinates, and r_1 and r_2 are the distances from the electron to ions 1 and 2, respectively, with

$$r_1^2 = \rho_1^2 + z^2$$

and

$$r_2^2 = \rho_2^2 + z^2 = \rho_1^2 + R^2 - 2R\rho_1 \cos \theta + z^2.$$

The barrier potential $V(z)$ is taken to be a square well of height V_0 and width L .

As the distance between the ions becomes very large ($R \rightarrow \infty$), the Hamiltonian (2.1) reduces to the well-known case of the isolated hydrogenic impurity [5], and as $R \rightarrow 0$, it reduces to the case of the He⁺-atom-like impurity. To obtain the ground-state energy corresponding to the above Hamiltonian, we choose a linear combination of Bastard-type [5] trial envelope wave functions, with an explicit dependence on the lowest-energy solution $f_k(z)$ for the quantum square well, i.e.,

$$\begin{aligned} \Psi(\rho_j, z) &= f_k(z)[G(\rho_1, z) + G_2(\rho_2, z)] \\ G_j(\rho_j, z) &= \exp[-\alpha(\rho_j^2 + z^2)^{1/2}] \quad j = 1, 2 \end{aligned} \quad (2.2a)$$

with

$$f_k(z) = \begin{cases} \cos kz & |z| \leq L/2 \\ Ae^{-\kappa|z|} & |z| \geq L/2 \end{cases} \quad (2.2b)$$

where the quantities A and κ are determined by the matching conditions at the interface, and α is a variational parameter. As in our model the two ions are localized in the same plane, parallel to the QW interfaces, the same variational parameter must be used for both ion centres in the trial envelope function (2.2a). The ground-state energy is determined by numerically minimizing with respect to α the expression

$$E(\alpha, R, L) = \langle \Psi | H | \Psi \rangle / \langle \Psi | \Psi \rangle$$

and the D_2^+ binding energy E_b for the ionization process $D_2^+ \rightarrow D^+D^+ + e^-$ is therefore given by

$$E_b(R, L) = k^2 - \min E(\alpha, R, L) \quad (2.3)$$

where k^2 is the lowest subband energy of the square well.

2.2. The model for the negative D^- ion

In the case of the negative D^- centre in a GaAs/Ga_{1-x}Al_xAs QW in the presence of a magnetic field applied perpendicular to the interfaces, the Hamiltonian may be represented as [11]

$$H = \sum_{i=1}^2 H_0(i) + U(1, 2) \quad (2.4a)$$

with

$$H_0(i) = -\nabla_i^2 + \gamma L_{z_i} + \frac{1}{4} \gamma^2 \rho_i^2 + V(z_i) - \frac{2Q_e}{r_i} \quad (2.4b)$$

and

$$U(1, 2) = \frac{2}{|\mathbf{r}_1 - \mathbf{r}_2|} - \frac{1}{2r_1} - \frac{1}{2r_2} \quad (2.4c)$$

where $r_i = (\rho_i^2 + z_i^2)^{1/2}$ is the position of the i th electron with respect to the fixed charge Q_e , ρ_i and z_i are the corresponding cylindrical coordinates, $Q_e = 3/4$ is the effective charge (in units of the proton charge e), and $\gamma = \hbar\omega_c/2R_y^*$ ($\hbar\omega_c$ is the cyclotron energy) is a dimensionless magnetic field intensity (the magnetic field is taken perpendicular to the interfaces). Notice that the above Hamiltonian is not separable and has no exact analytical solution. As the QW width is close to the effective Bohr radius $a_0^* = \hbar\varepsilon/e^2m^*$, the electron orbits become almost planar [5] with the mean value of the radii diminished to $a_0^*/2$, and therefore, for such a situation, we propose a geometrical model [22] of a quasi-two-dimensional D^- negative ion with both electrons instantaneously localized at the same distance with respect to the ion nucleus. Within this model, the D^- Hamiltonian becomes separable, and the effect of the electron–electron interaction is essentially taken into account by the interaction of each of the electrons with a renormalized-nucleus effective charge at the well centre. The solution is then a product of two one-particle wave functions of a hydrogenic-like atom, and the D^- ground-state energy is therefore equal to twice the one-particle eigenvalue (the solution of the D^- Hamiltonian is reduced to solving the D^0 Hamiltonian with a renormalized-nucleus effective charge at the well centre). In our calculations, we have used a three-parameter one-particle trial envelope variational wave function

$$\psi(\rho, z) = f_k(z) \exp(-\lambda^{-1} \sqrt{\rho^2 + \beta^2 z^2}) \exp(-\alpha \rho^2)$$

proposed by Muller *et al* [8] to describe the Landau levels in the limit of strong magnetic fields. As we shall see later, the use of this trial wave function provides results in excellent agreement with quantum Monte Carlo calculations [13] for both the D^0 and D^- states. In order to calculate the D^- ground-state energy, we have used the harmonic approximation for the zero-point energy correction in the total D^- energy, i.e., the energy corrections were evaluated as a function of the angle between the two electron vector positions. Therefore, it may be shown that the ionization process $D^- \rightarrow D^0 + e$ has the following binding energy:

$$E_b(D^-) = k^2 + \gamma + E_{D^0}(Q_e = 1) - 2 \left[E_{D^0}(Q_e = 3/4) + \frac{1}{2} \hbar\omega \right] \quad (2.5)$$

where E_{D^0} corresponds to the energy associated with the Hamiltonian (2.4b).

2.3. The symmetrical-structure model for the D_2^0 complex

In the framework of the effective-mass approximation, this problem becomes similar to that of the H_2 molecule, but the additional confinement presented by the QW potential reduces the system symmetry and makes the problem more complicated. On the other hand, when this confinement is considerable, the electronic motion becomes more correlated, and it is then possible to use a symmetrical-structure model [22] to analyse the D_2^0 ground state. The two-electron Hamiltonian (without nuclear repulsion) for the D_2^0 complex may be written as

$$H(D_2^0) = \sum_{i=1}^2 H_0(i) + U(1, 2) \quad (2.6a)$$

$$H_0(i) = -\nabla^2(i) - \frac{2}{|\mathbf{r}_i - \mathbf{R}_1|} - \frac{2}{|\mathbf{r}_i - \mathbf{R}_2|} - \frac{2Q_e}{r_i} + V(z_i) \quad (2.6b)$$

$$U(1, 2) = \frac{2}{|\mathbf{r}_1 - \mathbf{r}_2|} + \frac{2Q_e}{r_1} + \frac{2Q_e}{r_2} \quad (2.6c)$$

where $|\mathbf{r}_i - \mathbf{R}_j|$ is the relative distance between the i th electron and the j th ion. Similarly to the case for the previous D^- problem, the Hamiltonian becomes separable for correlated motion in a symmetrical-structure [22] model for a confined D_2^0 molecular complex. In this case, an appropriate symmetrical-structure approximation may be considered, in which both the two positive ions and the two electrons have opposite and symmetrical positions with respect to the origin, i.e., the two electrons and two ions of the D_2^0 complex are localized at the corners of a parallelogram. In our model, the electrons conserve the symmetrical positions at all times during their motion, and the ions, for the sake of simplicity, are fixed in a plane centred in the QW, and parallel to the interfaces. For this configuration, the Hamiltonian (2.6a) becomes separable when $\mathbf{r}_1 = -\mathbf{r}_2$ and $Q_e = -1/4$, as the term $U(1, 2)$ which contains the electron–electron interaction vanishes, and the effect of the electron–electron interaction is taken into account by the interaction of each of the electrons with a renormalized-nucleus effective $Q_e = -1/4$ charge at the molecule centre. The wave function for the Hamiltonian (2.6a) in this case has the form $\Psi(1, 2) = \phi(1)\phi(2)$, where the wave functions $\phi(i)$ satisfy the one-particle equation

$$H_0(i)\phi(i) = E(Q_e)\phi(i) \quad i = 1, 2. \quad (2.7)$$

The D_2^0 complex ground-state energy is $E(D_2^0) = 2E_{D_2^+}(Q_e = -1/4)$. The one-particle Hamiltonian $H_0(i)$ for $Q_e = 0$ coincides with that for the one-electron D_2^+ complex, so the ground-state D_2^+ energy is equal to $E(D_2^+) = E_{D_2^+}(Q_e = 0)$, and the binding energy for the ionization process $D_2^0 \rightarrow D_2^+ + e^-$ (the first ionization) may be found from

$$E_b(D_2^0) = k^2 + E_{D_2^+}(Q_e = 0) - 2E_{D_2^+}(Q_e = -1/4) \quad (2.8)$$

and we adopt in equation (2.7) the same Bastard-type trial envelope function, equation (2.2), as was previously used for the D_2^+ complex.

3. Results and discussion

In our calculations, the distance is measured in units of the effective Bohr radius a_0^* , and the energy in effective rydbergs, $R_y^* = e^2/2\epsilon a_0^*$, and we assume the GaAs values of $m^* = 0.067m_0$ (m_0 is the free-electron mass) and the static dielectric constant [23] $\epsilon = 12.35$ throughout both the well and the barrier.

In figure 1 we display the variation of the binding energy $E_b(R, L)$ of D_2^+ as a function of the GaAs well width for three different separations of the nuclei, $R = 500 \text{ \AA}$, 100 \AA and 50 \AA . Each of the $E_b(R, L)$ curves reaches a maximum at approximately the same critical value $L = L_c \approx 30 \text{ \AA}$. Notice that for narrow ($L < L_c$) QW widths, the 3D behaviour is restored due to the effect of the overflow of the wave function to the barrier region. For large ($L \gg L_c$) QW widths, one of course recovers the 3D behaviour. Also notice that one finds the D_2^+ binding energy as essentially the 3D value of $1 R_y^*$ (as for the isolated impurity D^0) in the case of large separations of the nuclei ($R \rightarrow \infty$) and for narrow and large well widths (cf. curve a in figure 1). For fixed QW widths, as the D_2^+ inter-nucleus separation diminishes and becomes less than the effective Bohr radius (curve c in figure 1), the D_2^+ three-particle system begins to behave rather like a He^+ atom, with increasing values of the binding energy. The results in figure 1 allow a study to be made, for different well widths,

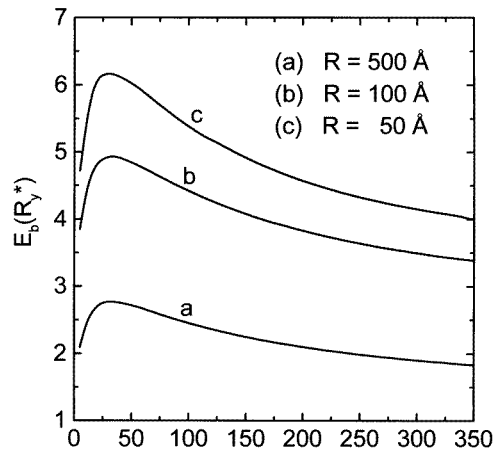


Figure 1. The ground-state variational binding energy for the D_2^+ complex centred in a GaAs/Ga_{0.7}Al_{0.3}As QW as a function of the QW width for three different values of the inter-nucleus distance R .

of the ionization mechanism $D_2^+ \rightarrow D^+D^+ + e$, as the D_2^+ complex loses the last electron (second ionization).

In order to obtain the D_2^+ -complex electronic energy or total energy (E) as a function of the inter-nucleus distance R , it is necessary to add the nuclear-repulsion energy ($2/R$), and, therefore, the total energy is

$$E = \min E(\alpha, R, L) + 2/R.$$

The results obtained for this energy E for different QW widths, $L = 1000 \text{ \AA}$, 200 \AA , and 40 \AA , are shown in figure 2. The equilibrium distance (R_e) for curve a of figure 2, for $L = 1000 \text{ \AA}$, is $R_e \simeq 2 a_0^*$ with a dissociation energy of $1.26 R_y^*$, which essentially tends

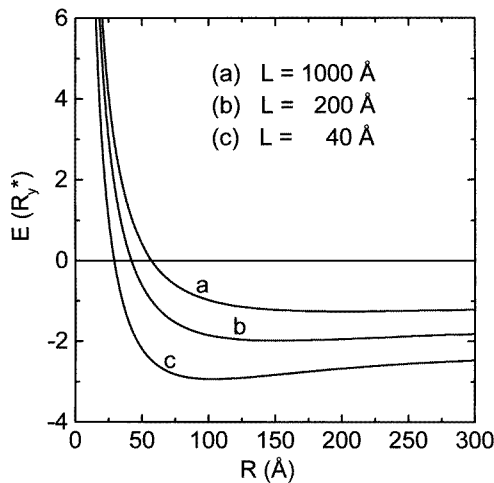


Figure 2. The total energy of the D_2^+ complex (from the variational calculation) centred in a GaAs/Ga_{0.7}Al_{0.3}As QW as a function of the inter-nucleus distance R , for different values of the well width L .

to the corresponding experimental value $1.195 R_y^*$ for the case of the H_2^+ molecule [24] (in fact our calculated value for $L = 25\,000 \text{ \AA}$ is $1.175 R_y^*$). As the QW width decreases to $L = L_c \approx 30 \text{ \AA}$, the equilibrium position changes to approximately $R_e \simeq 1 a_0^* \simeq 100 \text{ \AA}$ (curve c of figure 2), and the dissociation energy corresponds essentially to the two-dimensional D_2^+ molecular complex energy value [21].

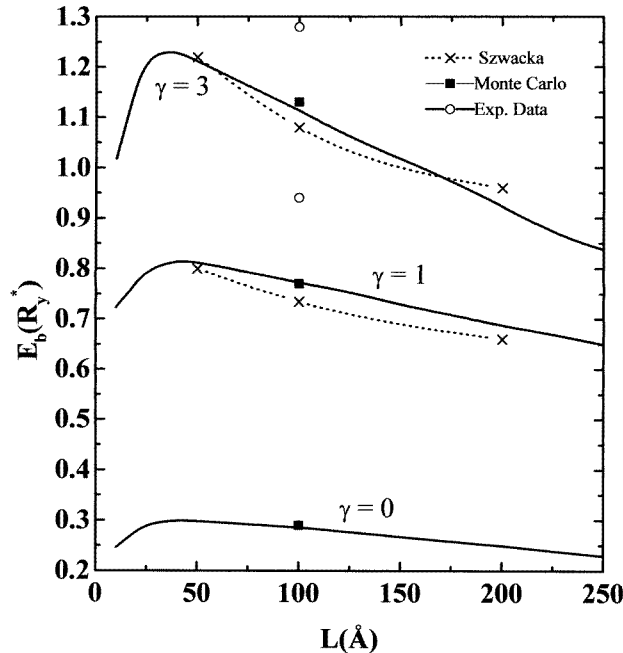


Figure 3. The ground-state binding energy for a negatively charged D^- centred in a GaAs/Ga_{0.75}Al_{0.25}As QW as a function of the well width L , and for different values of the magnetic field γ , calculated by using our model (solid curves) within a variational procedure, and including zero-point energy corrections. Also shown are the $\gamma = 1$ and $\gamma = 3$ variational results obtained by Szwacka *et al* [11] (the dotted curve is a guide to the eye), and Monte Carlo [13] values for $L = 100 \text{ \AA}$. The open dots in the figure represent experimental data from reference [7].

In figure 3, we present the results of our variational calculation (with the zero-point energy correction) for the D^- binding energy as a function of the well width, for three different magnetic field values corresponding to $\gamma = 0$, $\gamma = 1$, and $\gamma = 3$, as compared to the $\gamma = 1$ and $\gamma = 3$ results from the variational calculation by Szwacka *et al* [11]. One may compare our results for $L = 100 \text{ \AA}$ with both Monte Carlo calculations [13] and experimental data [7]. For $\gamma = 0$, our result ($0.29 R_y^*$) coincides with the one obtained by the quantum Monte Carlo method [13]. For $\gamma = 1$ and $L = 100 \text{ \AA}$, the D^- binding energies are $0.77 R_y^*$, $0.94 R_y^*$, and $0.77 R_y^*$, obtained by the Monte Carlo method [13], experiment [7], and within our model calculation, respectively. Similarly, for $L = 100 \text{ \AA}$ and $\gamma = 3$, one obtains $1.13 R_y^*$, $1.28 R_y^*$, and $1.11 R_y^*$, respectively. It is seen that our results are in good agreement with Monte Carlo calculation [13], although we underestimate the binding energy in comparison with experimental data [7]. We believe that this discrepancy may be reduced if one includes effective-mass and dielectric constant mismatches in the well and barriers.

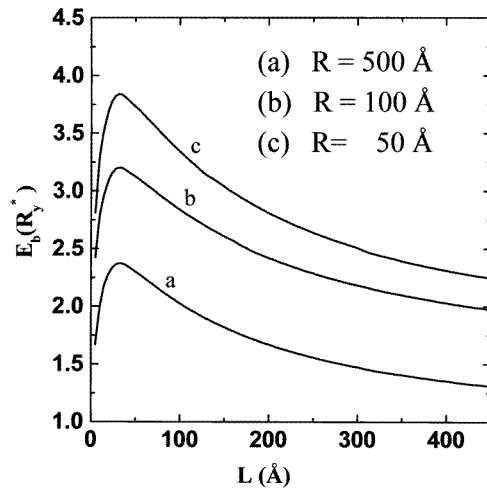


Figure 4. The ground-state binding energy for the D_2^0 complex centred in a GaAs/Ga_{0.7}Al_{0.3}As QW as a function of the well width L , for different inter-nucleus distances.

The results of our calculations for the D_2^0 binding energy as a function of the QW width for three different separations between the nuclei, $R = 50 \text{ \AA}$, 100 \AA , and 500 \AA , are presented in figure 4. It is apparent that all of the curves have similar forms with the binding energy increasing by approximately 60% as the separation between nuclei diminishes from 500 \AA to 50 \AA . Notice that, for large separation of the nuclei (curve a of figure 4), the D_2^0 complex appears as two isolated hydrogenic-like atoms, whereas for distances less than the effective Bohr radius a_0^* (curve c of figure 4), it is essentially a He-like atom, and the binding energy difference for the two separations (500 \AA and 50 \AA) tends to the ionization energy difference for hydrogenic and He-like atoms.

In figure 5 we present the total energy of the D_2^0 complex (including the nuclear-repulsion

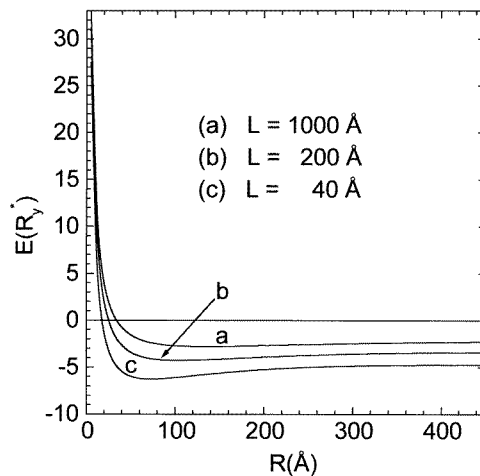


Figure 5. The total energy of the D_2^0 molecular complex centred in a GaAs/Ga_{0.7}Al_{0.3}As QW as a function of the inter-nucleus distance R , for different values of the well width L .

energy) as a function of the separation R between the nuclei, for different QW widths. In the case where $L = 1000 \text{ \AA}$ (weakly confined), our potential curve gives an equilibrium inter-nucleus distance of $R_e = 1.37 a_0^*$, in good agreement with the experimental value and the result of a simple variational [25] calculation for the H_2 molecule, which are $1.40 a_0^*$, and $1.38 a_0^*$, respectively. The results for the dissociation energy are $2.6 R_y^*$, $2.25 R_y^*$, and $2.33 R_y^*$, obtained using our model ($L = 5000 \text{ \AA}$), an improved LCAO method [25], and experimental data [25], respectively. The dissociation energy for the potential curves in figure 5 increases and the equilibrium distance diminishes as the QW width decreases to 40 \AA , and one observes the opposite trend for $L < 40 \text{ \AA}$.

4. Conclusions

In this work we have presented results for the binding and dissociation energies of D_2^+ and D_2^0 molecular complex states in GaAs/Ga_{1-x}Al_xAs QWs obtained by using a variational procedure. We have also studied the behaviour of the D^- and D_2^0 binding energies as functions of the QW width, within a model approximation, in which one is able to substitute in the electron–electron interaction by introducing an effective charge at a centre of symmetry. Finally, the scheme that we used is extremely simple and provides a realistic description of few-particle ground-state electronic structures confined in semiconducting QWs.

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