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1995 J. Phys.: Condens. Matter 7 4489

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Variational studies of the quasi-two-dimensional D^- centre in magnetic fields

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Received 14 July 1994, in final form 7 November 1994

Abstract. We present a simple variational scheme for calculating the binding energies and the lowest dipole optical transition energies of D^- impurities in the middle of quantum wells at moderate and high magnetic fields. The objective was to make the model and the method as simple as possible but to be able to explain many of the experimental features observed. The simple form of the electronic wavefunctions used enables physical insight into the properties of the D^- system in quantum wells. The scheme, based on the parabolic approximation for the conduction band, gives results compatible with those of more sophisticated approaches. A simple renormalization procedure makes the results compatible also with the experimental results for GaAs quantum wells for which band non-parabolicity proved to be important. The energies for the lowest dipole optical transitions were calculated for a broad range of magnetic fields and well widths.

1. Introduction

Negatively charged shallow donors (D^- centres) were identified in several semiconductors many years ago [1–6]. Nevertheless, the observation of the signature of these centres in the optical spectra of bulk, homogeneous semiconductors continues to be an experimental challenge. Stringent requirements concerning both sample preparation and experimental conditions have to be satisfied in order to obtain a significant population of negatively charged donors in bulk semiconductors.

Quasi-two-dimensional D^- centres have a much shorter history, since the first evidence for their identification was reported only a few years ago [7]. It has already become clear, however, that, in contrast to the bulk case, the quasi-2D D^- centres in multi-quantum wells (MQW) can show up in relatively standard optical and magneto-optical experiments, even those aimed at different targets. There are several factors favouring the observation of D^- centres in MQW as compared to the case of bulk semiconductors [8]. First, the binding energies of electrons to centre-of-well donors are much higher than to donors situated in the barrier. Secondly, whatever the position of their parent donor ions, the electrons are practically confined to the well regions. This confinement, together with the lateral confinement resulting from the magnetic field perpendicular to the wells, increases the binding energies of all impurity states. The effect is more pronounced for the second electron bound to the centre-of-well donor D^- than for the single electron of the neutral centre-of-barrier donor D^0 . For high enough magnetic fields one can reach in MQW significant

equilibrium populations of centre-of-well donors D^- at the expense of nearby barrier donors, which lose their electrons [9].

In MQW the optical excitations of the D^- centres can fall in the same spectral region as those of neutral impurities. Unfortunately, in contrast to the bulk case, where the impurity excitation energies depend on the magnetic field only, here these energies depend also on the geometry of the structure and to some extent on the height of the barriers. The convincing identification of D^- features in a given experimental spectrum is therefore not straightforward and has to rely on comparisons with quantitative theoretical predictions for the particular MQW. These theoretical results are up to now rather scarce.

Pang and Louie [10] using the diffusion quantum Monte Carlo method calculated the ground-state binding energy of D^- at the centre of a single 100 Å wide quantum well formed by GaAs/Ga_{0.75}Al_{0.25}As heterostructures. They performed the calculations only for three different values of the parameter $\gamma = \hbar\omega_c/2Ry^*$ defining the strength of the external magnetic field perpendicular to the well ($\hbar\omega_c$ being the cyclotron energy and Ry^* the effective Rydberg), namely for $\gamma = 0, 1$ and 3 . A few extra points calculated by Pang and Louie are reported in [8].

More complete, variational calculations of a few low-energy states of D^- were reported by Mueller *et al* [11] for a 510 Å centre-doped GaAs/(Ga,Al)As quantum well. These calculations performed for three different directions and several values of the magnetic field allowed definite identification of the optical signature of the D^- centres in GaAs quantum wells. In the variational procedure trial wavefunctions of Chandrasekhar [12] type with seven free parameters were used for each D^- state, with numerical calculations of the multidimensional integrals.

Xia and Quinn [13] used the local-spin-density formalism, proved to be effective in atomic spectra calculations, to determine the binding and the optical transition energies for D^- centres in two different quantum wells at three magnetic field strengths. Their results for binding energies seem to converge to those of Pang and Louie [10] for high magnetic fields, being considerably lower for low and intermediate magnetic fields.

The fourth type of calculations for D^- centres in quantum wells was due to Dzyubenko and Sivachenko [14]. These authors constructed the wavefunctions of the D^- centre from properly chosen and symmetrized products of the free-electron solutions for the few lowest Landau levels from the lowest electrical sub-band. Sometimes as many as 100 basis states were used. In spite of the strict rigidity of thus constructed wavefunctions along the direction of the field, the results of this method, according to the authors, coincide within a few per cent (up to 8%) accuracy with the results of Pang and Louie [10].

Yet another variational method with linear parameters only was proposed by Dunn *et al* [15]†. To construct the D^- wavefunctions these authors used the one-electron wavefunctions containing the sums of several Gaussian-like eigenfunctions of the angular momentum component L_z , with several fixed decay constants. No correlation factors were used. This approach gave rather good agreement with the experimentally observed optical transition energies in wide quantum wells, but failed to reproduce the data for wells of widths smaller than about two effective Bohr radii.

All the types of calculations of the D^- spectrum mentioned above seem to require quite a lot of computational effort, this is why no detailed studies of the evolution of the spectrum as functions of the well width, barrier heights, donor position, etc were performed. Our experience with the variational calculations of the D^- spectra in bulk semiconductors

† The authors of this paper seem to distinguish between their $1s2p$ and $1s N = 0$ ($M = -1$) states of the D^- system, whereas in reality both represent the same state but in low- and high-field notation, respectively. The two different energies obtained in their paper result simply from the use of two different trial functions.

and parabolic quantum wells [16] suggests, however, that at moderate and strong magnetic fields quite accurate results can be obtained with little computational effort. When one is interested in low-energy excitations involving only the outer electron of the D^- system, few per cent accuracy can be obtained with the use of very simple Gaussian trial wavefunctions. This is because the outer, weakly bound electron, whose wavefunction is determined mostly by the magnetic field, does not feel the fine details of the charge distribution of the inner electron. The same should be true in the case of quasi-2D D^- centres, in spite of the fact that in this case the Coulomb interactions are enhanced. The enhancement is, however, a secondary effect resulting from the quasi-2D confinement. It is the well potential and the magnetic field, and not the Coulomb interaction, that determine in the first instance the shape of the wavefunction of the outer electron. We expect that it is only for the ground singlet state of the D^- ion, where the wavefunctions of both electrons strongly overlap, that electron-electron correlations play a significant role.

The purpose of the present paper is to work out a simple computational scheme based on the above intuitions and to test it profiting from the existence of results of previous, more elaborate calculations. The scheme will then be applied for the analysis of the evolution of the lowest excitation energy of D^- as a function of the magnetic field and the width of the well.

In section 2, we discuss the singlet ground-state energies and the extension of the envelope wavefunctions of the inner and outer orbitals for different quantum well widths and as a function of magnetic field. In section 3 the analysis of the singlet p^- -like excited state is done and the transition energies from the singlet ground state to this state are reported.

2. Singlet ground state of the D^- ion

We shall consider the MQW structure as a set of independent quantum wells and, as a consequence, the case of a single well sandwiched between barriers of finite height will be treated. If the interfaces are perpendicular to the z axis, then a square-well potential can be written in the form

$$V(z) = \begin{cases} 0 & \text{for } |z| < l/2 \\ V_0 & \text{for } |z| > l/2 \end{cases}$$

where l denotes the width of the well and V_0 the barrier height (for a GaAs well sandwiched by $\text{Ga}_{1-x}\text{Al}_x\text{As}$ barriers, V_0 is given by $V_0 = 0.65 \times 1.247x$ eV [17]). We shall consider the case when the ionized donor is placed in the centre of the well. For the D^- ion in a magnetic field parallel to the z axis, our dimensionless orbital Hamiltonian in the effective-mass approximation has the form

$$\mathcal{H} = H(1) + H(2) + \frac{2}{|\mathbf{r}_1 - \mathbf{r}_2|} \tag{1}$$

where

$$H(i) = -\nabla_i^2 - 2/r_i + \frac{1}{4}\gamma^2\rho_i^2 + V(z_i) + \gamma L_{zi} \quad i = 1, 2 \tag{2}$$

\mathbf{r}_1 and \mathbf{r}_2 give the positions of both electrons relative to the nucleus; $\rho_i^2 = x_i^2 + y_i^2$; and L_{z1} and L_{z2} are the z components of the electronic orbital angular momentum operators. The

energy is measured in effective Rydbergs $Ry^* = m^*e^4/2\hbar^2\varepsilon^2$, and the effective Bohr radius $a_B^* = \hbar^2\varepsilon/m^*e^2$ is the unit of distance ($-e$ and m^* are the charge and effective mass of an electron, respectively, and ε the static dielectric constant). Being interested in the electronic states bound to donors at the centre of the well (i.e. states only weakly penetrating in the barriers), we use the same values of m^* and ε for the well and the barrier regions.

The binding energy of the D^- -ion singlet ground state is defined as

$$E_B(\gamma) = E_{D^0}(\gamma) + E_e + \gamma - E_{D^-}(\gamma) \quad (3)$$

where $E_{D^0}(\gamma)$ denotes the ground-state energy of a neutral donor; $E_{D^-}(\gamma)$ is the ground-state energy of the D^- ion; E_e is the energy of the bottom of the lowest sub-band for an electron in the quantum well in the absence of the magnetic field; and γ is a measure of the magnetic field, which at the same time is the magnetic part of the ground-state energy of a free electron in a quantum well, far from the donor ion. In this definition of the binding energy, we assumed the same spin state for the initial and final states.

In the following we limit our interest to the region of intermediate and high magnetic fields and we choose the following Chandrasekhar-type [12] variational wavefunction for the singlet ground state of the D^- ion:

$$\Psi(\mathbf{r}_1, \mathbf{r}_2) = (1/\sqrt{2})[\Psi(\mathbf{r}_1)\Psi'(\mathbf{r}_2) + \Psi'(\mathbf{r}_1)\Psi(\mathbf{r}_2)](1 + A|\rho_1 - \rho_2|^2 + C|z_1 - z_2|^2) \quad (4)$$

where the one-electron wavefunction Ψ is defined by

$$\Psi = N \exp \left[- \left(\frac{\rho^2}{4a_{\perp}^2} + \frac{z^2}{4a_{\parallel}^2} \right) \right] f(z). \quad (5)$$

Here a_{\perp} and a_{\parallel} are variational parameters of the Gaussian-like envelope wavefunction, $f(z)$ is the ground-state eigenfunction of the square-well potential problem, and N is the normalization constant. The function Ψ' is defined by the same equation (5) but with different set of parameters (a'_{\perp} and a'_{\parallel}). This means that both electrons are put in orbitals of form similar to that of the ground-state wavefunction of an electron at D^0 centre in a quantum well and magnetic field. We have chosen a very simple form of the one-electron trial functions with only two variational parameters. The form (5) seems to be well suited for the outer, loosely bound electron, whose wavefunction should be formed to a large extent by the external magnetic field. In contrast, the function (5) is not flexible enough to reproduce correctly the fine details of the charge distributions of the inner electron of D^- as well as of the single electron of D^0 centre. In our study of D^- in bulk semiconductors in a magnetic field [16] we have proved, however, that the systematic errors resulting from the finite flexibility of the trial functions cancel to a large extent when one calculates the binding energy of the outer electron, provided the same approximations for both D^- and D^0 systems are used. Similar cancellation should be expected when one is interested in the energies of optical transitions involving the outer electron.

The repulsive interaction between the two electrons is accounted for in equation (4) by the correlation factor

$$(1 + A|\rho_1 - \rho_2|^2 + C|z_1 - z_2|^2). \quad (6)$$

The simple forms of both the envelope functions and the correlation factor considerably simplify the calculations of the expectation value of the Hamiltonian (1), reducing the six-dimensional integrals to double integrals over z_1 and z_2 variables.

All numerical calculations reported below were performed for a given depth $V_0 = 34.9Ry^*$ of the well (which corresponds to the composition $x = 0.25$ in GaAs/Ga $_{1-x}$ Al $_x$ As structures). The binding (and the excitation) energy proved to be, however, a very slowly varying function of the well depth and our results apply, within 2% accuracy, for all well depths in the range $20Ry^* < V_0 < 50Ry^*$. It is only for narrow wells, low barriers and high magnetic fields at the same time that the accuracy is somewhat lower (about 4% for $l = 0.5a_B^*$, $\gamma = 5$, $V_0 = 20Ry^*$).

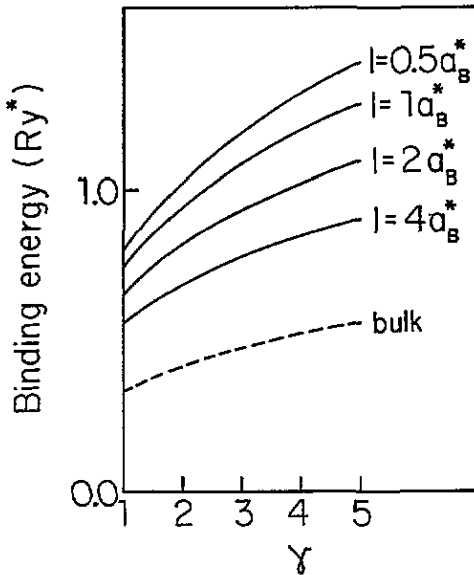


Figure 1. Binding energy of D^- ion in the ground state for various quantum wells as a function of magnetic field.

In figure 1 we present the calculated binding energy of the D^- centre in the middle of the quantum well for intermediate and strong magnetic fields. Four different well widths were studied: $l = 0.5, 1, 2$ and 4 effective Bohr radii a_B^* . The bulk binding energy of the D^- obtained with similar Gaussian trial functions are also plotted for comparison. Despite the simplicity of the trial functions (4), our results compare favourably with the available results of more elaborate calculations. For $\gamma = 1$, $l = 1a_B^*$, our result ($0.734Ry^*$) is only slightly lower than the energy $0.747Ry^*$ obtained variationally by Mueller *et al* [11], and the energy $0.77 \pm 0.02Ry^*$ obtained by Monte Carlo (MC) method by Pang and Louie [10] (Xia and Quinn [13] got $0.71Ry^*$). At $\gamma = 3$, our result ($1.08Ry^*$) is lower than the MC result [10] ($1.13 \pm 0.02Ry^*$) by about 5% but is still better than that of Dzyubenko and Sivachenko [14] (8% lower than MC result) or that of Xia and Quinn [13] ($1.07Ry^*$).

One can see from figure 1 that even for relatively wide wells the effect of the confinement is quite strong. The binding energy for the well width $l = 4a_B^*$ is still much larger than for D^- centres in bulk materials. The binding energy increases rapidly with decreasing width of the well, the effect being more pronounced at high magnetic fields. The binding energy in the most narrow well ($l = 0.5a_B^*$) at the highest field $\gamma = 5$ is almost three times larger than in bulk semiconductors.

To get some insight into the physical origin of the increased binding for D^- impurities in wells, we calculated the expectation value of the Hamiltonian (1) using the wavefunction (4) with fixed parameters: for the inner electron we used the parameters obtained from

the minimization of the energy of the neutral impurity D^0 , and for the outer electron the parameters of the free electron localized by the magnetic field at the position of the impurity ion (the correlation factor was neglected). Such a procedure may be considered as a first-order perturbation calculation, with the only difference that the variational and not the exact wavefunction of D^0 is used. The results show that in our range of magnetic fields and well widths the unperturbed wavefunctions account for the dominant part of the binding energy of the outer electron. The binding energies thus calculated amount for $\gamma = 1$ and $l = 4a_B^*$ to about 85% of the variational results and even more for larger fields and narrower wells. Such a perturbation procedure evidently fails to reproduce the binding energy in the case of the D^- system in the bulk and in very wide wells since the free-electron wavefunction does not describe the Coulomb-induced localization of the outer electron in the z direction. Two physically different contributions to the perturbationally calculated binding energies can be distinguished. The first contribution with classical origin comes from the balance between the Coulomb attraction to the positive centre and the Coulomb repulsion from the inner electron. The attraction dominates since the $1s$ type of electronic states allows for the close approach of the outer electron to the positive centre. The origin of the other contribution is purely quantum, related to the indistinguishability between the two electrons occupying similar overlapping states in the vicinity of the ion. This quantum contribution would vanish in two limiting cases, when the overlap was zero or when it was unity. The relative importance of the classical contribution increases with increasing magnetic field, from about 40–50% of the calculated binding energy at $\gamma = 1$ to about 90% for $\gamma > 5$ and $l = a_B^*$ and about 80% for $\gamma > 5$ and $l = 4a_B^*$.

In figures 2 and 3 we plot the variational parameters a_{\perp} and a_{\parallel} defining the localization of the inner electron, and the transverse parameter a'_{\perp} of the outer electron of the D^- system, respectively. The former are compared with the corresponding parameters of the inner electron in the D^- system in bulk materials, the latter with the free-electron parameter $\gamma^{-1/2}$ (the magnetic length) and with the outer-electron parameters from the D^- in the bulk case.

Concerning the inner orbital we note that the stronger confinement of the electron in narrower wells decreases the role of the longitudinal parameter a_{\parallel} of the Gaussian for the localization in the z direction, while enhancing the localization in the plane perpendicular to the field. In our widest well the parameters tend to the corresponding values for the bulk case, while the binding energy is still quite different. We remark here that the variational parameters of the inner orbitals of D^- are considerably smaller than those of the D^0 system, despite the fact that the perturbational binding energies are close to the variational ones. This confirms that the binding of the outer electron is only weakly affected by the details of the charge distribution of the inner electron, and it is not worth putting much effort into the improvement of the trial wavefunction of the inner electron.

For the outer orbital, the transverse parameter a'_{\perp} differs significantly from the free-electron value (and the bulk-case value) only in our low-field region and for narrow wells. The longitudinal parameter a'_{\parallel} is not plotted, since the binding energy is very weakly dependent on its value and almost the same binding energy can be obtained when putting $a'_{\parallel} = \infty$. It was only for the widest well and for high fields that we found minor differences. When comparing the variational parameters with those used in the perturbational calculations mentioned above, we state again that the wavefunction of the outer electron is essentially formed by the well and the magnetic field. It is the inner electron that adjusts its wavefunction to the presence of the second electron in the D^- system, but this adjustment has little effect on the binding of the outer electron.

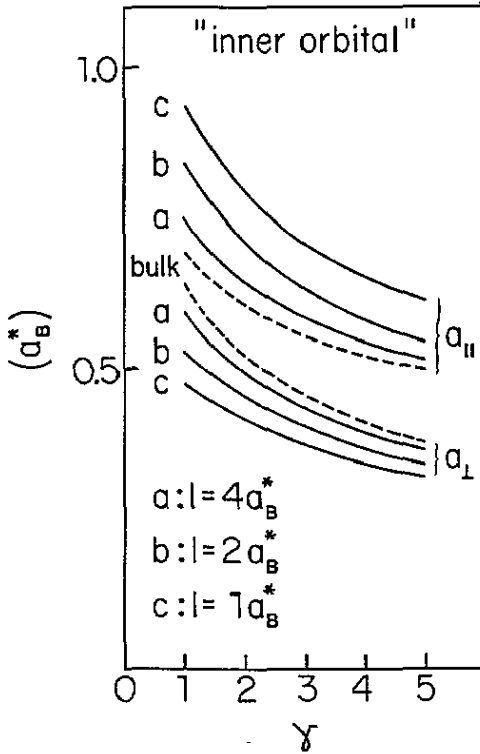


Figure 2. Optimum variational parameters a_{\perp} and a_{\parallel} for the Gaussian function used for the inner orbital as a function of the magnetic field γ , for various well widths. The broken curves correspond to the bulk case.

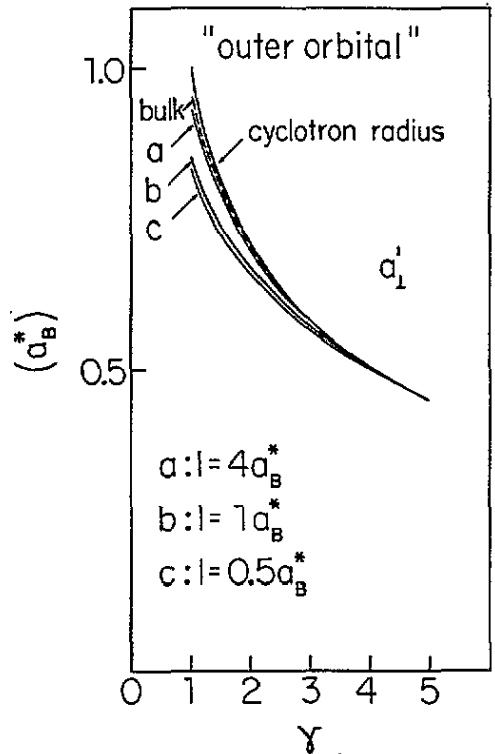


Figure 3. Optimum variational parameters a'_{\perp} for the Gaussian function used for the outer orbital as a function of the magnetic field γ , for various well widths. The broken curve represents the a'_{\perp} for the bulk case.

3. Singlet p^- -like excited state and optical transition energies

It was shown by Larsen and McCann [18] and Dzyubenko [19] that in strong magnetic fields the final states of dipole magneto-optical transitions from the ground state of quasi-2D D^- impurities are excited singlet p -like levels with energies above the successive Landau levels. We are interested in this section in the lowest-energy dipole transition to the p^- -like excited state. In the case of GaAs wells this transition was experimentally observed by Huan *et al* [7] and more recently by Holmes *et al* [20]. Its energy was found to differ by approximately $\hbar\omega_c$ from the energy of the dominant transition to the excited p^+ -like level (provided the region of resonance polar interaction is not reached). For the lowest singlet p^- -like excited state of the D^- ion we use the trial wavefunction of the form (4) with the p -like outer-electron wavefunction $\Psi'(r)$ defined by:

$$\Psi'(r) = N'(x - iy) \exp \left[- \left(\frac{\rho^2}{4a_{\perp}^{\prime 2}} + \frac{z^2}{4a_{\parallel}^{\prime 2}} \right) \right] f(z). \tag{7}$$

The orthogonality of this function to the $1s$ -like function (5) guarantees the orthogonality of the total two-electron wavefunctions of the ground and excited states and the vanishing of the overlap of the outer and inner orbitals for the excited state.

Table 1. The dipole optical transition energies $\Delta E(R\gamma^*)$ from the ground to the p^- -like excited state for various values of magnetic field γ and well widths l .

γ	$l(a_B^*)$			
	0.5	1	2	4
1	0.95	0.89	0.78	0.63
2	1.32	1.20	0.99	0.77
3	1.55	1.38	1.10	0.83
4	1.73	1.51	1.18	0.89
5	1.86	1.61	1.24	0.94

The calculated energies of the dipole optical transitions from the ground to the lowest p^- -like excited state are reported in table 1. These energies for all wells and all magnetic fields are larger than the corresponding ground-state binding energies for the outer electron—the energies of the p^- -like excited states being higher than the Landau level energies in the absence of the impurity ion. The Coulomb repulsion between the inner and outer electrons is in this case stronger than the Coulomb attraction of the outer electron to the positive centre. In contrast to the ground-state case the p -like character of the outer orbitals prevents the close approach of the outer electron to the positive centre. The repulsion dominates over the attraction whatever, prolate or oblate, is the shape of the charge distribution of the inner electron. The electron is still very close to the ion and the arguments based on the multipole expansion [18] of the charge distribution are not yet applicable.

One can see from table 1 that the transition energy ΔE is a very smooth function of the two arguments, γ and $\ln l$, so that its values at intermediate γ and/or $\ln l$ can be determined, with reasonable accuracy, by linear interpolations between the points given in table 1. Bearing in mind that for parabolic bands the energies of the two lowest excited singlet states of the D^- system differ by the cyclotron energy 2γ (outside the region of the resonant polaron coupling), we hope that table 1 will prove useful for identifying the optical spectra of the D^- system in quantum wells.

Surprisingly enough the simple renormalization procedure proposed by Huant *et al* [8] makes the results from table 1 (obtained for parabolic bands) agree well even with the experimental results obtained for narrow GaAs wells, for which band non-parabolicity proved to be important [21]. The renormalization procedure consists of attributing to the electrons bound in a D^- system the Ekenberg [21] mass m_{\parallel}^* , proper for the electron from the first electric sub-band of a given GaAs quantum well in its motion parallel to the interfaces. It is the mass m_{\parallel}^* that was used to define the effective Rydberg, the effective magnetic field γ and the effective Bohr radius when comparing the results of table 1 with experiment.

In table 2 we compare the experimental results of Huant *et al* [8] (as read from figure 3 in [8]) and of Holmes *et al* [20] (as read from figure 1 in [20]) with the renormalized results of our calculations. ΔE was obtained variationally whereas ΔE^i by interpolating the data from table 1, for the appropriate γ and l/a_B^* . One can see that for all wells and for both values of the magnetic field our results, both calculated and interpolated, differ from the experimental values of Huant *et al* [8] by at most 5% and usually less. It is only for the lowest magnetic field and the most narrow well that the difference approaches 15%; the origin of this discrepancy might be both experimental (as the errors given by Huant *et al* [8] for this particular well are much higher than for others) and theoretical (as the value of γ is already as low as 0.71). The comparison with the data of Holmes *et al* [20], quite favourable at higher magnetic fields, is less satisfactory at lower fields, where the discrepancy approaches 10%. For low fields our trial wavefunctions are evidently less

Table 2. Comparison of the experimental results of Huan *et al* [8] and Holmes *et al* [20] (ΔE^{exp}) with the calculated dipole transition energies (ΔE) and with interpolated data from table 1 (ΔE^{i}) (both types of theoretical energies corrected to account for non-parabolicity).

l (\AA)	B (T)	ΔE^{exp} (meV)	ΔE (MeV)	ΔE^{i} (meV)
58	6	6.0 [8]	5.10	
	9	6.5 [8]	6.25	6.2
95	6	5.1 [8]	4.88	
	9	5.75 [8]	5.91	5.8
144	6	4.6 [8]	4.57	
	9	5.25 [8]	5.44	5.35
194	6	4.35 [8]	4.30	
	9	4.8 [8]	5.04	4.95
373	6	3.6 [8]	3.56	
	9	4.0 [8]	4.06	4.0
100	6	5.3 [20]	4.83	
	9	6.2 [20]	5.89	5.75
200	6	4.5 [20]	4.29	
	9	5.15 [20]	5.02	4.9

applicable; one can see that at 6 T our results are systematically lower than the experimental data, whereas at 9 T our calculated transition energies are in all cases but one slightly higher than those reported in [8], although still lower than the energies found by Holmes *et al* [20]. In general the agreement is surprisingly good in view of the simplicity of our calculation scheme and seem to prove the usefulness of our results for predicting and interpreting the transition energies of D^- in quantum wells. note that the experimental results for nominally very similar quantum wells may well differ by several per cent, as seen from table 2. This seems to impose limits on the desired accuracy of the theoretical results and to justify the use of simplified approaches.

4. Summary

We demonstrated that a very simple variational procedure with Gaussian-like trial wavefunctions, avoiding the numerical calculation of multidimensional integrals, gives surprisingly accurate results for the binding and optical transition energies of the outer electron of the negatively charged donor D^- in quantum wells at moderate and high magnetic fields. In these conditions the shape of the wavefunction of the outer electron is determined primarily by the well confinement potential and by the magnetic field and not by the Coulomb interactions in the system. Our results agree within a few per cent accuracy with the results of more sophisticated and elaborate calculations, as well as with the experimental data. For narrow GaAs/GaAlAs quantum wells, simple scaling of the results was employed to account for non-parabolicity. Our results, tabulated for a broad range of well widths and magnetic fields, may thus be used as a guide in magneto-optical experiments with almost arbitrary quantum wells.

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