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Theory of D^- centres in GaAs/GaAlAs multi-quantum well systems

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Abstract. Details of a theoretical model to describe the properties of the D^- centre in GaAs/GaAlAs multi-quantum well (MQW) systems in the presence of a magnetic field are presented. It is based on a matrix diagonalization procedure that was developed by the present authors earlier for the D^0 centre in the same MQW systems. The results obtained are compared with published experimental data obtained from far-infrared magnetotransmission and photoconductivity measurements. Good agreement is obtained with these data for samples with relatively wide well widths. The calculations show that the dominant transition is between the $|1s^2)$ ground state and the $|1s2p_+)$ excited state, and indicate that the original suggestion that the main transition is of an ionizing type involving a Landau level is probably invalid. Qualitative agreement is also obtained between the calculations and the results of experiments performed on samples with narrower well widths.

1. Introduction

The D^- donor centre is formed when a second electron attaches itself to the neutral D^0 donor. There is currently much experimental and theoretical interest in D^- centres in quasi-two-dimensional systems, as shown by the many publications that have appeared within the last year. There are several reasons for this interest. Probably the most important is that the D^- donor in a quasi-two-dimensional environment is the simplest system in which many electron problems may be studied (see e.g., Cheng *et al* 1993a, b). Also it closely resembles the problem of an H^- atom in an intense magnetic field such as that existing in star clusters but in fields that are accessible in a laboratory. Besides these more fundamental reasons, there is the need to correctly identify donors in device material such as GaAs/GaAlAs multi-quantum wells (MQWs). This is important as, for example, the current through a semiconductor heterojunction device is ultimately determined at the level of individual impurity atoms (Dellow *et al* 1992).

The first observations of a D^- ion in a semiconductor occurred in the late 1960s; for example, Dean *et al* (1967) identified the ion in Si and Larsen (1979) in both Si and Ge. It was later found in some specially prepared GaAs samples by Armistead *et al* (1985) while Najda *et al* (1989) describe comprehensive infrared magneto-optical experiments relating to the centre in GaAs, InP and InSb. A list of other experiments is also given in the latter publication. The existence of D^- centres in GaAs/GaAlAs MQW systems was first reported by Huant *et al* (1990) but identification could not be regarded as definitive at that time.

Very recently, a number of other experiments has been undertaken from which a positive identification of the D^- donor at the centre of a GaAs QW in a GaAs/GaAlAs MQW system could be made. In these experiments, both the barrier and well are δ doped with Si (called double-planar doping, Huant *et al* 1990, 1992) so that, initially, neutral D^0 donors are present

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in both the well and barrier. Subsequently, electrons move from donors in the barrier to donors in the well and so produce D^- ions in the well. Also it is possible to alter the D^- populations by optical pumping (Mandray *et al* 1992, Cheng *et al* 1993a) provided that the relative initial populations are correct. These authors were able to identify transitions related exclusively to the D^- centre in the well using far-infrared (FIR) magneto-transmission experiments with applied magnetic fields of up to 13 T. The effects of optical pumping were also investigated in these experiments, and the impurity centres were confirmed to be confinement related by tilting the magnetic field. Previously, Mueller *et al* (1991, 1992) identified the D^- centre in wide GaAs QWs by an analysis of the dependence of the observed photoconductivity spectrum on the applied magnetic field and sample orientation. At the same time, theoretical investigations of the main transition within the spin singlet ground state of the D^- ion were undertaken. It was concluded that no behaviour in the resulting spectra could be obtained from a D^0 atom in either the well or the barrier. In a further development, Dzyubenko (1993) has extended the earlier work of Bychkov *et al* (1981) to a systematic study of D^{2-} ions in this system. However, it is deduced that such ions are thermodynamically unstable against the separation of the extra electron as all associated states have an energy higher than that of the D^- ground state.

Another set of FIR magnetotransmission and photoconductivity experiments has been undertaken independently by Holmes *et al* (1992a, b) and Cheng *et al* (1993a, b). These used specially designed samples with very wide barriers (of 600 Å) δ doped with Si donors at the centre of both the wells and the barriers. The number density of electrons in the wells was controlled by the accumulated illumination from an *in situ* red light emitting diode. Such photon dose experiments in a single sample controlled a progressive electron occupation of the various states of donors in the well, namely D^+ ions, neutral D^0 donors and D^- ions and also of the free electron Landau levels. They also ruled out any possibility that the peaks in the observed spectra are due to a preferential positioning of the D^0 impurity in the well. Another independent study of the same system but with different geometries using magnetophotoconductivity experiments has been undertaken by Chang *et al* (1992). Transitions related to D^- are again identified and clearly correlate with those observed by other authors. These latter authors also attribute the dramatic changes observed in the spectrum between low and high fields to a magnetic field induced metal insulator transition.

Despite the increasing amount of experimental data available on D^- centres, there has been little theoretical work reported which can be used to describe D^- centres in wells of more than 100 Å and over a range of magnetic fields. Some modelling was undertaken by Mueller *et al* (1992) to support their data. The only other relevant theoretical studies known to us are those by Pang and Louie (1990) and Xia and Quinn (1992). Pang and Louie used a diffusion Monte Carlo method but direct comparison with the field dependent data is very limited due to the lack of comparable data (see Mueller *et al* 1992). Xia and Quinn solved the Schrödinger equation numerically including the mixing of the subband wave functions. The D^- ions were investigated using the local density functional method and the dominant transition energy was calculated using the transition state technique. Good agreement with the data of Huant *et al* (1990) was demonstrated.

Another approach to the D^- problem in a MQW has been to consider the states of these ions to be entirely two dimensional. With this approximation, Larsen and McCann (1992a) have calculated excited states analytically in the limits of high and low fields while in a subsequent paper (Larsen and McCann 1992b) used variational techniques applicable to a range of field strengths. McDonald (1992) has also calculated analytically the energies of a two-dimensional D^- ion in the strong field limit. However, these methods are all only

suitable for narrow wells, and so cannot be used to describe the data of Mueller *et al* (1992) and Holmes *et al* (1992a, b), for example.

We note that other recent theoretical work on the D^- donor has been described in papers by Adamowski and Bednarek (1988), who calculate the influence of the electron–phonon coupling, Zhu (1992), who calculates the field dependence of the binding energies and the electron correlation effects for a two-dimensional D^- ion, Sandler and Proetto (1992), performing a variational calculation of the bound states in two dimensions and Zhu *et al* (1992), who refer specifically to a D^- centre in a spherical quantum dot. However, this last group of papers cannot be applied readily to the experimental work described above.

The object of this paper is to describe our own theoretical calculations for transitions within a D^- donor in a MQW system as a function of magnetic field. The method is different from all other theoretical calculations described above, and is *not* a variational one. It is based on an extension of our earlier work (Dunn and Pearl 1991), adding in the Coulomb interaction between electrons using the D^0 donor states obtained previously for the same system. The transition energies are obtained from a numerical diagonalization of the *total* Hamiltonian of the system using as basis states a product of the square well function and Gaussian adapted, hydrogen like orbital states as in our earlier works. The results obtained will be compared to the experimental data of Mueller *et al* (1992) and Holmes *et al* (1992a, b) where the two dimensional models are not appropriate.

2. The model for a D^0 impurity

Among the most extensively studied and reported MQW structures are those consisting of alternate GaAs and $Ga_{1-x}Al_xAs$ layers. As there are discontinuities in the conduction band at the interfaces between the layers, the GaAs and GaAlAs layers act as QWs and barriers respectively. Si impurity atoms are introduced into the GaAs layers and behave as hydrogenic D^0 donors in their unionized state. It is usual to use a dimensionless Hamiltonian to describe the system. This is the same as that used for D^0 centres in bulk GaAs but modified to include a potential energy term $V_B(z)$, defined to have the value zero in the wells and the value V_0 in the barriers. The Hamiltonian is thus

$$\mathcal{H} = -\nabla^2 + \gamma^2\rho^2/4 + \gamma l_z - 2/r + V_B(z). \tag{2.1}$$

The effective Rydberg R_y (= 5.83 meV) for bulk GaAs is used as the unit of energy, the effective Bohr radius a_0 (= 98.7 Å) for GaAs as the unit of length, and γ is a dimensionless unit of magnetic field B (which is related to B by $\gamma = 0.148B$ where B is in tesla). The effective mass m^* (= $0.067m_e$) for bulk GaAs is used for the electron in the well but in the barriers the value is taken to be $(0.067 + 0.083x)m_e$ (Casey and Pannish 1978), for an Al concentration x. The z direction is chosen to lie along B, which coincides with the direction of growth of the MQW (i.e. perpendicular to the layers). Also r is the distance of the electron from the impurity such that

$$r = \sqrt{\rho^2 + (z - z_I)^2} \qquad \rho^2 = x^2 + y^2 \tag{2.2}$$

where z_I is the distance of the impurity ion from the origin at the centre of the well containing the donor. Based on experimental evidence, V_0 is taken to be 60% of the total band gap ΔE_g estimated from the empirical expression (Lee *et al* 1980)

$$\Delta E_g = \left(1.155x + 0.37x^2\right) \text{eV}. \tag{2.3}$$

Many attempts at solving the Hamiltonian (2.1) have been made for D^0 donor impurities both in the bulk and in MQWs (see the review by Zawadzki 1991). All of the methods are

approximate, and many of them invoke variational techniques. Different forms of solution have been obtained dependent on whether a low-field (hydrogen like) or high-field (Landau like) approximation to the solutions is required. Most work has focused on the hydrogen like solutions because they give good descriptions of the lowest-energy states of the system in the magnetic fields applied experimentally. These will be the solutions investigated here.

Of the procedures used for D^0 impurities, the variational method of Greene and Bajaj (1985) and Greene and Lane (1986) gives good agreement with experiment for the 1s to $2p_{\pm}$ transitions in the MQWs of interest here. Their method uses hydrogenic basis states in which the Slater type exponentials are expanded into a set of Gaussians. These are better behaved numerically, and also reproduce some of the Landau type behaviour required in strong magnetic fields. Two of us (Dunn and Pearl 1991) later used a matrix diagonalization procedure (MDP) with basis states of the same form as those used in the above method. This gives similar results for the 1s to $2p_{\pm}$ transitions, but can also be used to calculate transitions to further excited states. This method (which is not variational) is outlined below.

The eigenstates Ψ for the donor in an MQW were written as a product of bulk hydrogenic states ψ_{nlm} and states $f(z)$ that are solutions of the standard square problem. That is

$$\Psi = f(z)\psi_{nlm} \tag{2.4}$$

where $f(z)$ is given by

$$fz = \begin{cases} \cos kz & \text{for } -L/2 + n(L+b) < z < L/2 + n(L+b) \\ A\exp(\kappa z) + B\exp(-\kappa z) & \text{for } L/2 + n(L+b) < z < -L/2 + (n+1)(L+b) \end{cases} \tag{2.5}$$

and where L and b are the well and barrier widths respectively and n is an integer labelling the wells. The parameters k, κ, A and B were determined using periodic boundary conditions and the requirements that $f(z)$ and $(1/m^*)\partial f/\partial z$ be continuous across the MQW boundaries.

The Slater type exponential terms in the hydrogenic states were replaced by a sum of Gaussian functions, both for numerical convenience and to help reproduce the Landau type behaviour required for strong magnetic fields. The total wavefunction was thus expressed in cylindrical coordinates as a linear combination of basis functions ψ_i, where

$$\psi_i = f(z)(z - z_1)^{q_i}\rho^{|m_i|}\exp(im_i\phi)\exp\left[-\alpha_i(z - z_1)^2\right]\exp\left[-(\beta_i + \delta)\rho^2\right]. \tag{2.6}$$

The coefficients α_i and β_i were taken from the A_t sets of Huzinaga (1965) with the restriction

$$\alpha_i = A_t \Rightarrow \beta_i = A_t \text{ or } A_{t\pm 1}. \tag{2.7}$$

Thus with an A_t set of say five values, 13 basis states were obtained. δ is introduced as an additional field dependent parameter to allow for the constriction of the wavefunctions in a magnetic field. Following Greene and Lane (1986), we choose $\delta = 0.1\gamma$, although the results are found to be somewhat insensitive to this choice. q_i is a parity-dependent factor for the state i that has a value of zero for even z parity and unity for odd z parity, and $m_i = m$ for state i.

In the MDP, approximate eigenstates and eigenvalues of $\mathcal{H}$ were found by solving numerically the generalized eigenvalue equation

$$H\Psi = EU\Psi \tag{2.8}$$

where $\mathcal{H}$ and U are the Hamiltonian and overlap matrices respectively between the basis states given above. As m is a good quantum number, states with different m_i values are not coupled together. The z type parity π_z $(= (-1)^{l+m})$ is also a good quantum number for impurities located at the centre of the well. Hence states with differing m and q values can be treated as separate eigenvalue problems.

The values of the various matrix elements and overlaps were found by evaluating the relevant integrals. The main advantage of the method over others is that the diagonalization procedure automatically gives many valid excited states which are orthogonal to states within the same matrix equation. The results obtained by this method† were found to be in very good agreement with the experimental data for the D^0 donor and transitions to many of the excited states were identified. A subsequent letter (Pearl *et al* 1992) showed that all the remaining transitions that could not be explained by hydrogenic type states could be explained as transitions to so called metastable states. It is not necessary to consider these further in this paper.

3. The model for a D^- ion

3.1. General considerations

If the two electrons in the D^- ion are at the points r_1, r_2, the dimensionless Hamiltonian $\mathcal{H}(r_1, r_2)$ describing the system can be written in the form

$$\mathcal{H}(r_1, r_2) = \mathcal{H}(r_1) + \mathcal{H}(r_2) + \mathcal{H}_{12} \quad . \tag{3.1}$$

where $\mathcal{H}(r_k)$ is the Hamiltonian describing the kth electron as in (2.1) for a D^0 ion, and $\mathcal{H}_{12}$ is the term describing the repulsive Coulomb interaction between the two electrons, of the form

$$\mathcal{H}_{12} = 2/|r_1 - r_2| = 2/r_{12}. \tag{3.2}$$

There are two possible situations. The first is the case where the electrons have parallel spins, to form a spin triplet state, and the second is the case where the electrons have anti-parallel spins producing a spin singlet state. As it is very difficult, experimentally, to obtain transitions between singlet and triplet states, we have two virtually independent sets of states.

The state with the lowest energy is that in which the electrons have opposite spins, and is thus a spin singlet whose orbital wavefunction has the form

$$|1s^2\rangle = |\psi_{1s}(r_1)\psi_{1s}(r_2)\rangle. \tag{3.3}$$

Transitions to excited spin singlet states only will thus dominate the transitions observed in experiments. We also suppose that the dominant transition will be to an excited state in which one of the electrons is in the $2p_\pm$ state as the $1s$–$2p_+$ transition dominates all others in the case of D^0. Thus this excited state is written as

$$|1s2p_\pm\rangle = (1/\sqrt{2})|\psi_{1s}(r_1)\psi_{2p_\pm}(r_2) + \psi_{2p_\pm}(r_1)\psi_{1s}(r_2)\rangle. \tag{3.4}$$

† We note that further detailed investigations of many of the data points considered originally by Dunn and Pearl (1991) have revealed that the values quoted for the well and barrier thicknesses were inaccurate. New calculations (Barmby *et al* 1993) have shown that the revised calculations using the correct parameters and an isotopic mass are in even better agreement with the experimental points.

3.2. Application of the MDP to the D^- ion

We choose basis states for the $|1s^2\rangle$ state from products of the 1s basis states used for the D^0 ion. Similarly, basis states for $|1s2p_\pm\rangle$ are chosen from combinations of the 1s and $2p_\pm$ basis states of the D^0 ion. These basis states do not need to be normalized as the overlap matrix is introduced in subsequent calculations.

We introduce a contracted notation

$$|j,l\rangle = \sqrt{1/2}|\psi_j(r_1)\psi_l(r_2) + \psi_j(r_2)\psi_l(r_1)\rangle \tag{3.5}$$

for any of the spin singlet basis states, where j and l are components of the D^0 basis states given in (2.6). (As the states need not be normalized, this form can also be used for the cases $j = l$). As $|j,l\rangle = |l,j\rangle$ it is only necessary to take the states with $l \geqslant j$ into account. Thus with N D^0 basis states we have N^2 possible D^- basis states, but only $\frac{1}{2}(N^2+N)$ of these states need be taken into account. In the calculations described here, 20 D^0 basis states are taken from the relevant matrices in the article by Dunn and Pearl (1991) giving a possible 210 D^- basis states. In practice, as the amount of computing time needed is large, this number is reduced considerably using further considerations, as described in section 4.

The principles of the method to be used here are the same as those used earlier for D^0. Thus we need to evaluate the relevant overlap and matrix elements of the total Hamiltonian (3.1) between all the components of the two-electron states appearing in the matrix. The overlap integrals are given by

$$\langle j,l|k,n\rangle = U_{jk}U_{ln} + U_{jn}U_{lk} \tag{3.6}$$

where U_{jk}, U_{ln}, U_{jn} and U_{lk} are the same as the D^0 overlap integrals. (The labels r_1, r_2 drop out of the calculation.) In a similar way, the matrix elements of the Hamiltonian $\mathcal{H}(r_1, r_2)$ are given by

$$\langle j,l|\mathcal{H}(r_1) + \mathcal{H}(r_2) + \mathcal{H}_{12}|k,n\rangle = H_1 + H_2 + H_{12} \tag{3.7}$$

where

$$H_1 = H_2 = \tfrac{1}{2}\big(H_{jk}U_{ln} + H_{ln}U_{jk} + H_{lk}U_{jn} + H_{jn}U_{lk}\big) \tag{3.8}$$

with H_{jk} being the same matrix elements as those occuring in the D^0 problem. The Coulomb term H_{12} is given by

$$H_{12} = 2\Big(H_{j,l}^{k,n} + H_{j,l}^{n,k}\Big) \tag{3.9}$$

(as the labels 1 and 2 can be interchanged without altering the results) where

$$H_{j,l}^{k,n} = \int\int \psi_j^*(r_1)\psi_l^*(r_2)\frac{1}{r_{12}}\psi_k(r_1)\psi_n(r_2)\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2 \tag{3.10}$$

and similarly for $H_{j,l}^{n,k}$. To evaluate these sixfold integrals, we use cylindrical coordinates and employ the expansion of $1/r_{12}$ given by (see e.g., Slater 1960)

$$\frac{1}{r_{12}} = \sum_{k'=0}^{\infty}\sum_{s=-k'}^{k'}\frac{(k'-|s|)!}{(k'+|s|)!}\frac{r_a^{k'}}{r_b^{k'+1}}P_{k'}^{|s|}(\cos\theta_1)P_{k'}^{|s|}(\cos\theta_2)\exp\big[\mathrm{i}s(\phi_1-\phi_2)\big] \tag{3.11}$$

where r_a is the smaller of r_1 and r_2, and r_b is the larger. The $P_{k'}^{|s|}(\cos\theta)$ are Legendre polynomials. Taking the two cases $r_2 < r_1$ and $r_1 < r_2$ separately, we define

$$I_1 = H_{j,l}^{k,n}(r_2 < r_1) \qquad \text{and} \qquad I_2 = H_{j,l}^{k,n}(r_1 < r_2) \tag{3.12}$$

so that

$$H_{j,l}^{k,n} = I_1 + I_2. \tag{3.13}$$

For the $|1s^2\rangle$ ground state, the only terms in the variables ϕ_1 and ϕ_2 come from the expansion (3.11), which integrates to give $4\pi^2$ if $s = 0$ and zero otherwise. This thus reduces the integral to four dimensions and the sum in (3.11) to one over k' only. Although extra angular factors appear when considering the $|1s2p_\pm\rangle$ excited state, the integrals can be reduced in a similar manner. The most complicated part of the problem is to evaluate the remaining integrals for the relevant states of the system. Although the details depend upon the states in question, there are a few general points applicable to all states. All of the integrals can be reduced to three dimensions by analytically integrating over one of the ρ variables. Each integral can also be expanded into a sum of four integrals taking different ranges for z_1 and z_2 related to the square well functions $f(z)$ given in (2.5). Thus, for example, I_1 is expanded to give

$$I_1 = I_{WW1} + I_{WB1} + I_{BW1} + I_{BB1} \tag{3.14}$$

where 'WW' indicates that both z_1 and z_2 range over the wells only, and 'WB' indicates that z_1 ranges over the wells while z_2 ranges over the barriers.

As a first approximation we then take

$$I_1 \simeq I_{WW1} \text{ etc} \tag{3.15}$$

as the wavefunctions are generally of larger magnitudes in the wells and therefore the integrals are larger. (We have confirmed numerically that the other integrals are indeed much smaller.) Also, I_{WW1} is approximated further by limiting the sum to three wells only in order to save computing time. This is a reasonable approximation for relatively wide wells since the electrons in the D^- centre will have the highest probability of being found in the central well. The details of the calculation of the integrals I_1 are given in appendix C of the thesis by Pearl (1993); the types of integral that appear and their evaluation follow closely those outlined in the article by Dunn and Pearl (1991).

3.3. The diagonalization

The energies of the various states are obtained by solving the generalized eigenvalue equation (2.8) for the states of interest. The calculation of the triple integrals such as I_1 involves a significant amount of computer time. In order to reduce the number of integrals required, only some of the 210 basis states are used. The selection is made as follows: initially, solutions to the generalized eigenvalue problem are obtained using all 210 basis states but neglecting the Coulomb part H_{12} in (3.7). Then, with the eigenvector associated with the lowest energy eigenvalue written in the form

$$(c_1, c_2, c_3, \ldots, c_{210})^T \tag{3.16}$$

basis states associated with the highest absolute values of the coefficients c_i only are chosen. The eigenvalue problem is then solved using this smaller number of basis states but using the complete Hamiltonian, to give a first estimate of the ground state energy. More states are then added into the problem (by selecting states with the next-largest values of $|c_i|$) until the energy obtained converges to a limiting value. For example, figure 1 shows the energy obtained for the $|1s^2\rangle$ ground state using different numbers of basis states. The figure clearly shows that the energy converges to a value very close to -5.02 cm^{-1}. We find that about 35 states are needed in the calculation of the energy of this state for the range of magnetic field used experimentally. Similar calculations are undertaken for the $|1s2p_\pm\rangle$ and $|1sN\rangle$ states, for each set of values for L and b.

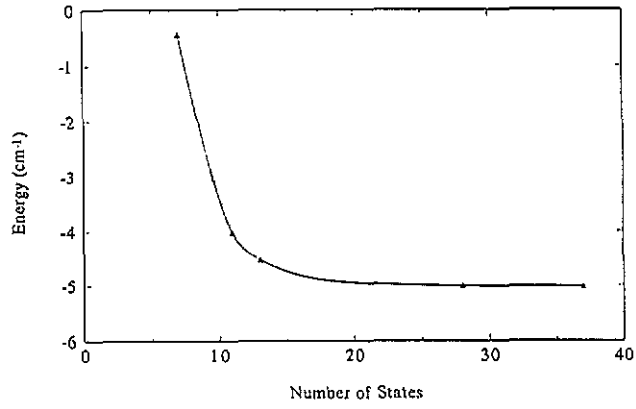

Figure 1. Ground state energy in 510 Å/350 Å MQWs for $B = 5$ T, plotted against the number of basis states used.

3.4. Transitions to an ionizing state

In the original study by Huant *et al* (1990) and also in the theoretical work of Pang and Louie (1990), it was supposed that the D^- transition observed was such that one of the 1s electrons undergoes an ionizing transition to a Landau level ψ_N. Although we do not believe that this is the main transition observed, it is straightforward to undertake such a calculation for comparison purposes using the method described here. The excited state wavefunction is given by

$$|1sN\rangle = (1/\sqrt{2})|\psi_{1s}(r_1)\psi_N(r_2) + \psi_N(r_1)\psi_{1s}(r_2)\rangle \tag{3.17}$$

where

$$\psi_N(r) = N_L f(z)\sqrt{\gamma/2\pi}\exp(im\phi)\exp(-\gamma\rho^2/4)(\gamma\rho^2/2)^{|m|/2}P_{Nm}(\gamma\rho^2/2). \tag{3.18}$$

P_{Nm} is the associated Laguerre polynomial and N_L is a normalization constant. The selection rules require that any transitions from 1s must be to levels with $m = \pm 1$.

The generalized eigenvalue problem can be solved after calculation of the relevant overlaps and matrix elements as before. The matrix elements of $\mathcal{H}(r_1, r_2)$ to a Landau level with $m = 1$ are given by

$$E_{1s} + E_N - 2\langle\psi_N|(1/r)|\psi_N\rangle + 2\langle 1sN|(1/r_{12})|1sN\rangle \tag{3.19}$$

(if the states ψ_{1s} and ψ_N are normalized) where E_{1s} and E_N are the energies of the 1s state and Landau level N of D^0 respectively. (The 1s state and a Landau level with $m = \pm 1$ are orthogonal.) The calculation of the two matrix elements in this equation follows the same formalism as above.

4. Application to real systems

We look first at the data of Mueller *et al* (1991) who performed magnetooptical experiments on 510 Å/350 Å MQWs with $x = 0.2$. (The notation used for the MQW gives the values of the well and barrier widths respectively in the form L/b.) Figure 2 shows our calculated energies corresponding to transitions from the $|1s^2\rangle$ ground state to the $|1sN(=1)\rangle$ and $|1s2p_+\rangle$ excited states at magnetic fields of 3 T and 5 T, together with the experimental data. It is clear that the predicted energies of transitions to the $|1s2p_+\rangle$ state agree with the experimental data extremely well. However, the predicted energy corresponding to transitions to the $|1sN(=1)\rangle$

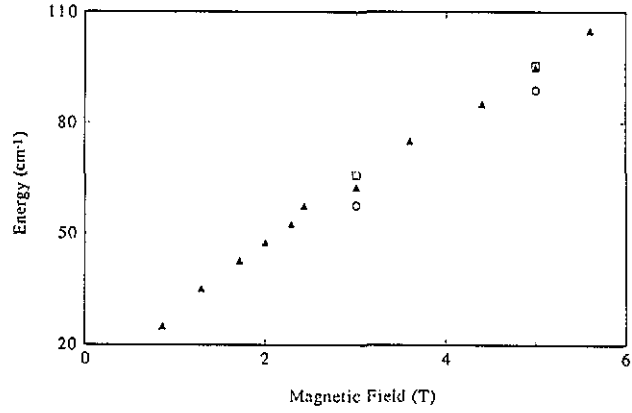

Figure 2. Transition energies from the $|1s^2\rangle$ ground state to the $|1s2p_+\rangle$ (□) and $|1sN(=1)\rangle$ (○) excited states in 510 Å/350 Å MQWs together with the experimental data of Mueller *et al* (1991) (▲).

state is slightly lower than that indicated experimentally. Therefore, it is reasonable to conclude that the experimental points correspond to transitions to the $|1s2p_+\rangle$ state, as would seem to be more acceptable from a physical point of view.

We next examine the magneto-optical data of Holmes *et al* (1992a) and Cheng *et al* (1993a) on a 200 Å/600 Å GaAs/GaAlAs MQW for which $x = 0.3$. Our calculated energies for transitions to the $|1s2p_+\rangle$ (□), $|1s2p_-\rangle$ (◇), $|1sN(=1)\rangle$ (○) and $|1sN(=0)\rangle$ (▽) excited states are shown in figure 3, together with the experimental data (▲). It can be seen that the predicted transition energies both to the $|1s2p_\pm\rangle$ levels and to the Landau levels are all fairly close to those observed in the experiments. However, although the energy of the $|1s2p_-\rangle$ state agrees with the data slightly better than that of the $|1sN(=0)\rangle$ state, the agreement is not close enough to definitively distinguish between the different excited states.

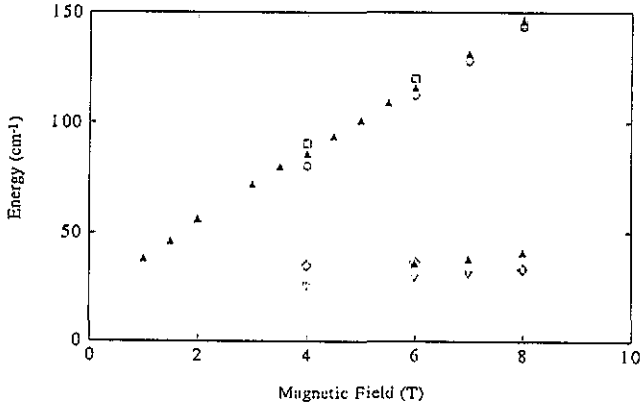

Figure 3. Transition energies from the $|1s^2\rangle$ ground state to the $|1s2p_+\rangle$ (□), $|1s2p_-\rangle$ (◇), $|1sN(=1)\rangle$ (○) and $|1sN(=0)\rangle$ (▽) excited states in 200 Å/600 Å MQWs. Also shown are the experimental data of Holmes *et al* (1992a) and Cheng *et al* (1993a) (▲).

Holmes *et al* (1992a) and Cheng *et al* (1993a) also performed magneto-optical experiments on samples similar to those described above but with a well width of only

100 Å. The results of our calculations for these samples, plus the experimental results, are shown in figure 4 (using the same symbols as in figure 3). Unfortunately, it can be seen that none of the predicted transition energies agree with the experimental data. This suggests that the approximations used for the calculation of the energies of the states are not sufficiently accurate. In both of the MQW systems discussed above, the wells, in addition to the barriers, were relatively wide. Here, the wells are much narrower so that the wavefunctions will spread much more into the adjoining well and barrier material. This means that it is reasonable to assume that extra terms are needed in the integral calculations, which were not needed in the previous case. Thus, despite the large increase in computing time required, the calculations have been repeated with the more accurate approximations

$$I_1 \simeq I_{WW1} + I_{WB1} + I_{BW1} \tag{4.1}$$

at fields of 2 T and 6 T. These results are also displayed in figure 4 (using filled symbols equivalent to the open symbols used for the simpler approximation). It can be seen that the correction has reduced the discrepancy between experiment and theory. However, the agreement is still only qualitative, and no comment can be made on the nature of the excited states involved in the transitions. This suggests that maybe we have not reached a true minimum in the calculation of the energy of the $|1s2p_{\pm}\rangle$ states and more basis states are needed in the calculation. It may also be necessary to include the terms I_{BB1} etc (from equation (3.14)) in the calculation of the integrals although it is unlikely that the energy would increase significantly since these extra terms have much smaller magnitudes than those already included. However, neither of these improvements have been possible with our current computational resources.

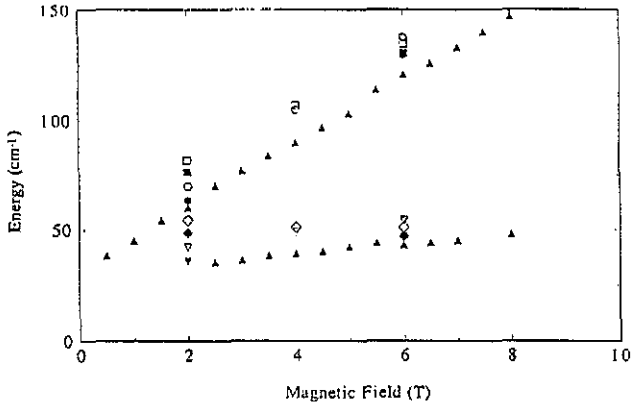

Figure 4. Transition energies from the $|1s^2\rangle$ ground state to the following excited states, using approximations (3.15) and (4.1) respectively: $|1s2p_+\rangle$ (□ and ■), $|1s2p_-\rangle$ (◇ and ◆), $|1sN(=1)\rangle$ (○ and ●), $|1sN(=0)\rangle$ (▽ and ▼). Also shown are the experimental data of Holmes *et al* (1992a) and Cheng *et al* (1993a) (▲).

We mention here that Huant *et al* (1990) performed far-infrared magnetotransmission and magnetophotoconductivity experiments on 100 Å/100 Å MQWs with an Al concentration $x = 0.25$. We have calculated the transition energies predicted for MQWs of this size using the simple approximation (3.15), but no agreement with the experimental results is obtained. This is to be expected, as both the wells and barriers are relatively narrow. We would expect better agreement to be obtained using the more accurate approximation (4.1), although not complete agreement for the same reasons mentioned above for the data of Cheng *et al*

(1993a). Also, the validity of these data has itself been doubted (Mueller *et al* 1992). Therefore we have not performed the improved calculations for this sample size.

5. Discussion and conclusions

Our calculated energy for transitions to the $|1s2p_+\rangle$ excited state for the wide-well, wide-barrier system investigated by Mueller *et al* (1991) is in good agreement with the experimental data, and indicates that the dominant transition for the D^- impurity is not to the $|1sN(=1)\rangle$ state as originally proposed by Huant *et al* (1990). Qualitative agreement between the calculations and experiment is also obtained for the 200 Å /600 Å system studied by Holmes *et al* (1992a) and Cheng *et al* (1993a), and also with the 100 Å well system studied by the same authors when additional terms are included in our model. However, exact agreement is not obtained due to the approximations that it is necessary to introduce into our model due to computational limitations. The model is more accurate for wide wells, where the wavefunction is predominantly localized in the central well, than for narrow wells, where the spreading into adjacent wells is much more important.

For every MQW system considered here, a common trend is found in the results as the magnetic field increases. The relative slope with field of the transition energy to the $|1s2p_+\rangle$ state is smaller than that of the $|1sN(=1)\rangle$ state (and similarly for the $|1s2p_-\rangle$ state and $|1sN(=0)\rangle$ states). This results in a crossing of the transition energies at a particular value of magnetic field. This can be understood by considering what happens to the two outer electrons of the D^- centre as the field increases, and hence the orbit size decreases. For the situation where the outer electrons are in the 1s and $2p_+$ states, the electrons become much closer together. This increases the Coulomb repulsion between them, and thus reduces the effects of the magnetic field. For the case where the two outer electrons occupy the 1s and the $N = 1$ Landau level, the Coulomb repulsion between the two electrons does not have such a great effect since the electrons are further apart, and hence the effect of the magnetic field dominates. This result for D^- is in contrast to the case of the D^0 centre where transitions to the $|2p_+\rangle$ state always lie below transitions to the $N = 1$ Landau level at all fields (Pearl *et al* 1992). Furthermore, we have found that calculations using different barrier widths but the same well widths give different values for the magnetic field at which the $|1s2p_+\rangle$ and $|1sN(=1)\rangle$ lines cross, indicating that the width of the barrier is also an important quantity.

An additional way to decide on the nature of the excited state involved in the observed transitions is to look at transition probabilities. Using the usual perturbation mechanism for an electric dipole transition (see, for example, Barmby *et al* 1993), calculations have been performed for the D^0 centre in a 150 Å/150 Å MQW system with $x = 0.33$. Comparing transitions at a field $B = 5$ T, for example, it is found that the probability of an electron making a transition to a $2p_+$ state is 18 times greater than that of an electron making a transition to the $N = 1$ Landau level. By analogy with the D^0 centre, we assume that the outer electron of the D^- centre has a higher probability of making a transition to the $2p_\pm$ states than to the $N = 0$, 1 Landau levels. In making this analogy we have neglected the repulsive Coulomb interaction between the two electrons, which should strictly be included for D^- centres.

It is important to point out here that the observed splitting between the two components in the experimental data of Cheng *et al* (1993a) for the 200 Å/600 Å MQWs (and those of Huant *et al* (1990)) is of the order of 1.5% less than 2γ, due to the Coulomb interaction between the two electrons. Thus in the calculations, it is necessary to explicitly calculate the transition energies for the $|1s2p_-\rangle$ states rather than simply subtracting 2γ from the $|1s2p_+\rangle$ transition energies.

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