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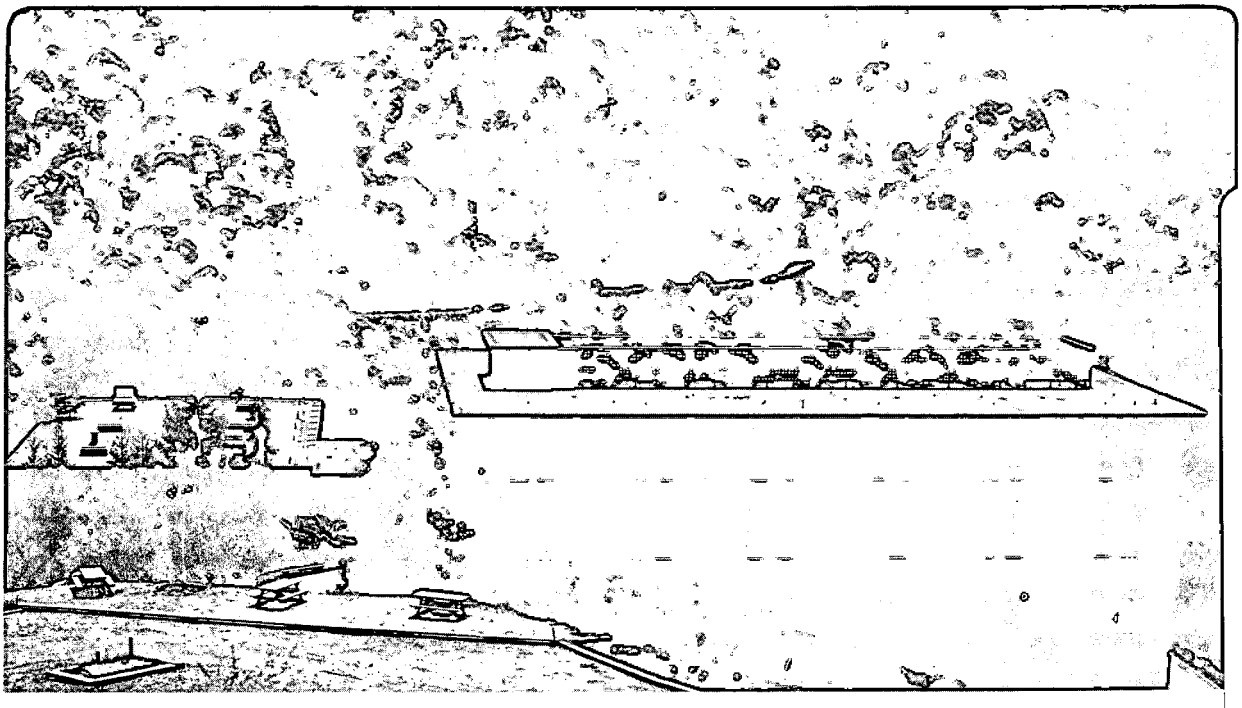

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D^- CENTERS IN HIGH MAGNETIC FIELDS AND QUANTUM WELLS

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ABSTRACT

The ground-state and optical properties of negative donor (D^-) centers in strong magnetic fields and quantum wells are studied using the effective mass theory. The exact ground-state binding energy is calculated to within statistical noise by a diffusion quantum Monte Carlo method. Excited state and optical transition energies are computed by carrying out variational Monte Carlo calculations. Comparison with available magneto-optical data of D^- centers in bulk GaAs and in GaAs/$Al_xGa_{1-x}As$ quantum wells shows good agreement between theory and experiment over a range of magnetic field strengths.

1. Introduction

A D^- center in semiconductors is formed by a neutral donor center (D^0), such as a Si atom in GaAs, trapping an extra electron [1]. For bulk semiconductors, such as GaAs, InP, or InSb, states associated with the D^- centers can be thought as states of two electrons of effective mass m^* under a central Coulomb potential screened by the host dielectric constant [1]. This effective mass model has been successful in understanding neutral shallow donors. For example, results from variational calculations (such as the peak position of the calculated far-infrared absorption lines for D^0 centers in GaAs/$Al_xGa_{1-x}As$ quantum wells) are in good agreement with experimental observations [2]. However, owing to the electron-electron correlation effects in D^- centers, earlier variational schemes did not give very accurate results for the energetics of D^- centers in magnetic fields [1]. Most recently, D^- states have been identified experimentally in

$GaAs/Al_xGa_{1-x}As$ quantum wells with various widths [3,4]. Magneto-optical measurements were carried out with an applied field perpendicular to the well structures. The results show that D^- centers are observable at fields larger than 4 T.

We present in this paper results on calculations of the ground-state properties using a diffusion quantum Monte Carlo (DQMC) method and of both the ground-state and excited-state properties using a variational quantum Monte Carlo (VQMC) method. The system is treated within the effective mass theory. Our results show a strong enhancement in the binding energy of D^- centers in quantum wells compared to that in bulk. In addition, they support the interpretation that the experimentally observed optical transition [3] is from a D^- ground state to a localized but unbound D^- excited state [4]. The DQMC approach in principle provides the exact ground-state properties of D^- centers, whereas the VQMC approach gives a good estimate on the excited state properties.

The remainder of this paper is arranged as follows. In Section 2, we present our effective mass model for D^- centers in quantum wells. In Section 3, the basic aspects of the DQMC and VQMC methods are discussed. In Section 4, our theoretical results for D^- in quantum wells of various widths are presented together with comparison with the available experiment results. In Section 5, some implications of the numerical results are discussed.

2. Effective Mass Model for D^- Centers

For a D^- center located at the center of a single $GaAs/Al_xGa_{1-x}As$ quantum well and with an applied uniform magnetic field along the z-direction perpendicular to the well, the Hamiltonian in the effective mass model is given by:

$$H = H(1) + H(2) + V(r_{12}) + \frac{\gamma}{2}L_z \tag{1}$$

with

$$H(i) = -\frac{1}{2}\nabla_i^2 + \frac{\gamma^2}{8}(x_i^2 + y_i^2) + U(r_i) + V_Q(z_i), \tag{2}$$

where $\gamma = \hbar\omega_c/2Ry^*$ is the magnetic field expressed in the effective atomic units with ω_c the cyclotron frequency and Ry^* the effective Rydberg; $V(r)$ and $U(r)$ are the screened interaction between two electrons and the interaction of an electron with the central impurity charge, respectively; L_z is the angular momentum component along the field direction; and $V_Q(z)$ is the quantum well potential:

$$V_Q(z) = \begin{cases} V_0, & |z| > d/2 \\ 0, & |z| \le d/2 \end{cases} \tag{3}$$

with V_0 the potential barrier height and d the well width.

Since the magnetic field is along the z direction, L_z is conserved with quantum number M. The ground state has $M = 0$. Excited states may have $M = 0, \pm 1, \ldots$, etc. As for the optical transition observed in the magneto-optic experiments [3], selection rule dictates that the transition is from the ground state to the $M = +1$ state in the $N = 1$ Landau level which is unbound but localized. The $M = +1$ final state has a $M = -1$ replica in the $N = 0$ Landau level which is lower in energy by exactly γ in atomic unit. Therefore, for the excited states, we will only present results for this $M = -1$ state unless otherwise specified.

3. Diffusion and Variational Quantum Monte Carlo Methods

For our variational studies, we adopt a wavefunction of the following form for the D^- center:

$$\Phi(\mathbf{r}_1, \mathbf{r}_2) = f(r_{12})[g_1(r_1)g_2(r_2) + g_1(r_2)g_2(r_1)] \prod_{i=1}^{2} \phi(z_i), \tag{4}$$

where $\phi(z) = cos(kz)$ in the well and $cos(kd/2)exp[\lambda(d/2 - |z|)]$ outside the well (k and λ are determined by solving for the solution of a single electron in such a quantum well), $f(r) = exp[ar/(1 + (C_\rho^2\rho^2 + C_z^2 z^2)^{1/2})]$, and the single particle orbital g_i is the same form as that used by Larsen [4], which is

$$g_i(r) = \rho^{|M|}\, exp(-\eta_i\rho^2 - \kappa_i\sqrt{\rho^2 + \alpha_i z^2}) \tag{5}$$

with M the z-component angular momentum of the orbital.

There are total of eight variational parameters $(C_\rho, C_z, \eta_1, \eta_2, \kappa_1, \kappa_2, \alpha_1, \alpha_2)$. Total energies are minimized with respect to these parameters to get the variational eigenvalues of the Hamiltionian in equation 1. A correlated-walk scheme is used to search for the energy minimum with the guidance of total energy derivatives with respect to each parameter. The search stops when these derivatives are less than a certain value (usually 10^{-3} a.u.). The set of configurations is re-generated using the new set of parameters to avoid bias in the initial set of configurations. This process is repeated two or three times until all biases are eliminated. To obtain the binding energy, the D^0 ground state energies are also calculated using the form of $g(r)\phi(z)$ in equation 4 and 5. For the transition energy, both the excited-state and ground-state energies are calculated using the VQMC method.

A more sophisticated and numerically involved DQMC method [5] has also been used to obtain the exact ground-state energy of equation 1 and 2. In the DQMC approach, for a two-electron system, one takes a variational wavefunction $\Phi(\mathbf{R})$ with $\mathbf{R} = (\mathbf{r}_1, \mathbf{r}_2)$ as a

trial wavefunction for the ground state and constructs a time dependent probability density, $F(\mathbf{R},t)$:

$$F(\mathbf{R},t) = \Psi(\mathbf{R},t)\Phi(\mathbf{R}), \tag{6}$$

with

$$\Psi(\mathbf{R},t) = exp[-Ht]\ \Phi(\mathbf{R}). \tag{7}$$

As long as $\Phi(\mathbf{R})$ is not orthogonal to the ground state of H, $\Psi(\mathbf{R},t)$ will approach the ground state as t goes to infinity. Defining a time dependent expectation value $E(t)$:

$$E(t) = \frac{<\Phi(\mathbf{R})|H|\Psi(\mathbf{R},t)>}{<\Phi(\mathbf{R})|\Psi(\mathbf{R},t)>} = \frac{\int d\mathbf{R}F(\mathbf{R},t)\epsilon(\mathbf{R})}{\int d\mathbf{R}F(\mathbf{R},t)}, \tag{8}$$

where $\varepsilon(\mathbf{R}) = \Phi^{-1}(\mathbf{R})H\Phi(\mathbf{R})$ is the local energy in configuration space, then $E(t)$ becomes the exact ground-state energy of H as t goes to infinity [6]. The DQMC approach provides an exact result within statistical noise of the calculation and an excellent test of our ground-state variational wavefunction. However, it may not be used straightforwardly for excited-state calculations.

4. Simulation results

In figure 1, the DQMC binding energies [5] together with results from two earlier calculations [7,8] using variational methods are compared to the experimental results for the case of Si doped GaAs [1]. Due to the improved form of our variational wavefunctions, our new VQMC results are in fact virtually indistinguishable with our DQMC results if plotted in the same figure. The excellent agreement of the DQMC results with the experimental data indicates that the effective mass model describes the D^- centers in GaAs quite well. In addition, this figure shows that the discrepancies of earlier variational calculations [7,8] with experiment [1] are purely due to the inadequacy of the trial wavefunctions previously used.

We have also calculated the excited-state energies using VQMC method for the bulk case. We found that the $M = -1$ state described in Section 2 is only slightly unbound. Therefore, whether the final state of the transition observed [1] is the vacuum level or the $M = -1$ state makes no difference in the comparison of theory and experiment.

We also examined the limiting case of a D^- center in 2D. Due to a complete confinement on the $z = 0$ plane, the attractive Si impurity centers becomes much more significant in 2D than in 3D. Binding energies of the ground state are enhanced for both D^0 and D^- centers. The D^- state binding energy in 2D (0.511 Ry^*) is an order of magnitude larger than that in 3D (0.056 Ry^*) according to our DQMC calculations [9]. The qualitative behavior of a two-orbital structure in the electron charge density distribution of 2D D^- center in low field

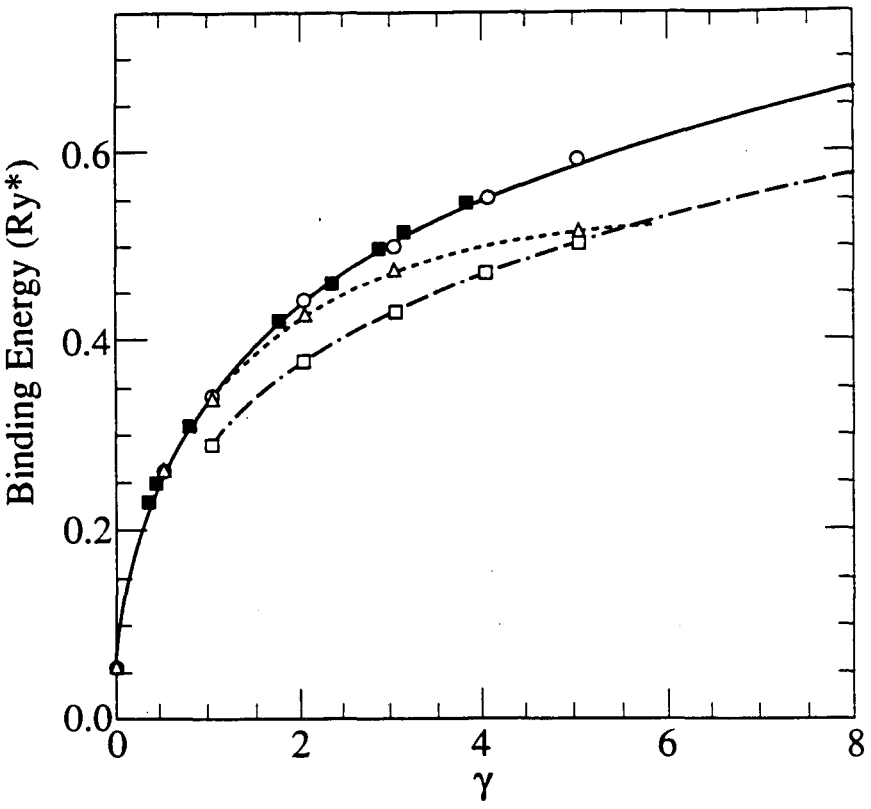

Figure 1. Binding energies of D^- centers in GaAs calculated by DQMC [5] (○) compared with the experimental data of Ref. [1] (●) and earlier calculations of Ref. [7] (□) and Ref. [8] (△). The error bars of DQMC results are within the circles.

is found to be similar to the case in 3D. However, the excited states of the D^- center shows a much different behavior in 2D. At some distance away from the origin, the defect center may be over-screened. Therefore, the electron in the outer orbital sees an average repulsive potential if it is far away from the center (which is the case when $M \neq 0$). In the strong field limit, Larsen *et al* [10] was able to show that the binding energy of 2D D^- center in the $M = -1$ state is negative and its absolute value is $\sim 71\%$ of the ground-state binding energies. However, this percentage decreases gradually with decreasing magnetic field strength and becomes $\sim 12\%$ at $\gamma = 1$ based on our VQMC calculation.

Quantum wells of experimental interest are, of course, a case in between 2D and 3D with regard to the confinement along the z-direction. We expect that the binding energies of the ground state and the $M = -1$ excited state will also be in between that of 2D and 3D. Our studies were carried out for quantum wells with Al concentration $x = 0.25$ and various well widths. The barrier height is chosen as $0.65\Delta E_g$ with $\Delta E_g = 1.247x$ eV as the band gap mismatch between GaAs and $Al_xGa_{1-x}As$. The effective masses are taken from experimental

Table 1. Comparison of DQMC and VQMC results for D^- binding energies for the $d = 100\mathring{A}$ quantum wells. Error bars are 0.02 Ry^*.

	DQMC	VQMC
$\gamma = 0$	0.29	0.26
$\gamma = 1$	0.77	0.74
$\gamma = 3$	1.13	1.11

Table 2. VQMC and experimental results for D^- center in $GaAs/Al_xGa_{1-x}As$ quantum well of various widths at $\gamma = 1$. Listed are the binding energies, transition energies from ground state to the $M = -1$ excited state, and the experimental measurements [3].

	Binding Energy	Transition Energy	Experiment
2D	1.12	1.25	...
$d = 58\mathring{A}$	0.81	0.95	0.94
$d = 95\mathring{A}$	0.76	0.88	0.87
$d = 194\mathring{A}$	0.67	0.77	0.75
$d = 373\mathring{A}$	0.59	0.63	0.63
3D	0.34	0.34	0.34

measurements [3]. The dielectric constant in the well as well as in the barrier are chosen as those of bulk GaAs ($\epsilon = 12.53$).

At zero magnetic field, the theory shows a dramatic increase, by a factor of five, for the binding energy of D^- in the 100 Å quantum well as compared to that of 3D. In Table 1, the ground state binding energies for the $d = 100\mathring{A}$ quantum well are compared between the DQMC and VQMC methods for $\gamma = 0, 1$ and 3. We find excellent agreement between DQMC and VQMC results illustrating the effectiveness of our variational wavefunctions.

In Table 2, we list results at $\gamma = 1$ for quantum wells of various widths ($d = 95\mathring{A}, 194\mathring{A}$ and $373\mathring{A}$). VQMC results for the D^- ground state binding energies and transition energies (from ground state to $M = -1$ excited state) are presented together with experimental results [3]. The ground state to $M = -1$ excited state transition energies agree well with the experiment while the ground-state binding energies are consistently lower by 5-15%. The discrepancy however diminishes as the well width increases. This agrees well with that the final state of the transition is the $M = -1$ state instead of the vacuum level. The over-screening effect in

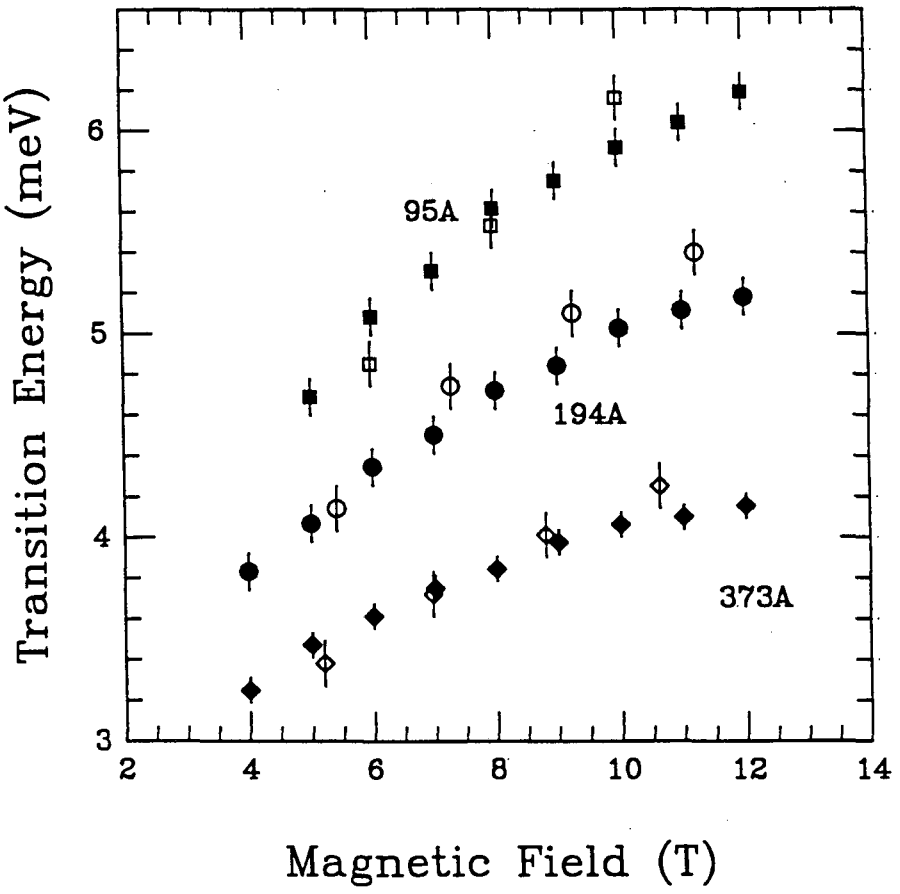

Figure 2. VQMC transition energies are presented together with corresponding experimental results [3] for three quantum well sizes ($d = 95\mathring{A}$, $194\mathring{A}$, and $373\mathring{A}$). Solid and open circles are experimental (•) and theoretical (○) results for $d = 95\mathring{A}$ quantum wells. Squares (■ and □) and diamonds(◆ and ◇) are for $d = 194\mathring{A}$ and $373\mathring{A}$ wells, respectively.

In figure 2, the VQMC transition energies are presented together with the corresponding experimental results [3] for three quantum well sizes ($d = 95\mathring{A}$, $194\mathring{A}$, and $373\mathring{A}$). Again, we see a good agreement, although there is some noticeable deviation of theory from experiment at very high fields. We attribute the deviations partially to the VQMC approach used. At strong magnetic field, the absolute value of the total energy increases sharply with magnetic fields. It is more difficult to obtain transition energy accurate to the order of 0.02 Ry^* at high fields than at low fields. Further, our $M = -1$ excited state and the ground state variational wavefunctions may fare differently with increasing fields, resulting in a different level of error cancellations. It is likely that with improved wavefunctions, the agreement of theory and experiment at the high end of the magnetic field would improve.

5. Summary

We have discussed some of the properties of D^- centers in quantum wells and magnetic fields obtained using the DQMC and VQMC approaches. It is found that D^- states in bulk semiconductors such as Si doped GaAs are well described by the effective mass model. Electron-electron correlation effects are important in these systems. We find that although simple trial wavefunctions used previously would not give a complete treatment of these correlation effects, improved wavefunctions are capable of yielding very accurate results in VQMC calculations. On the other hand, the DQMC method applied here in principle has treated electron correlations exactly in the ground state. For D^- centers in quantum wells, our theoretical results re-enforce the identification made in Ref. 3 that the observed special feature in the magneto-optical spectra indeed arises from D^- centers in the quantum wells. Our results further quantitatively showed that the observed optical transition is from the D^- ground state to an unbound but localized D^- excited state with M = 1 in the $N = 1$ Landau level.

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