D^- centers in quantum wells: Spin-singlet and spin-triplet magneto-optical transitions

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(Received 25 January 1993; revised manuscript received 22 July 1993)

We consider the ground and excited states of D^- centers, i.e., neutral donors D^0 trapping an extra electron, in the spin-singlet and spin-triplet states in GaAs/$Ga_{1-x}Al_xAs$ quantum wells (QW's) in quantizing magnetic fields $B > 6$ T. Taking into account the effects of band nonparabolicity and magnetopolaron corrections, we derive the energies of the dipole-allowed transitions and their line strengths. Very good agreement (e.g., within 1–3 cm^{-1} below the region of the resonant magnetopolaron splittings) with the experimental data of Huant *et al.* for the singlet transitions is found.

It is well known that both a reduction of dimensionality and strong external magnetic fields lead to the enhancement of the effects of interparticle correlations. Recently Huant *et al.* in the series of magneto-optical experiments in magnetic fields up to 21 T have shown that by selective doping of GaAs/$Ga_{1-x}Al_xAs$ quantum wells (QW's) it is possible to "engineer" a stable population of negative donor centers D^-;[1] in contrast to bulk semiconductors, it is even possible to convert all[2,3] neutral donors D^0 within a QW to D^-. A quasi-two-dimensional (quasi-2D) D^- center in a strong magnetic field is an interesting example of a few-particle correlated quantum-confined system for which total-spin-dependent *e-e* repulsion and electron-impurity attraction are strongly enhanced. Although a large amount of work, both experimental and theoretical, has been devoted to quasi-2D neutral donors D^0 in magnetic fields (e.g., Ref. 4 and references therein to earlier publications), there are only few theoretical considerations of D^- states in QW's. Pang and Louie, using a quantum diffusion Monte Carlo approach, have calculated with high accuracy[5] the binding energy of the singlet D^- *s* ground state which turns out to be systematically below the values derived from the experiments.[1] This discrepancy has been elucidated independently by Larsen and McCann[6] and Dzyubenko,[7] who considered a strictly 2D case in the strong magnetic field approximation in which D^- eigenstates can be found exactly (see also Ref. 8). They have shown that in QW's, in the presence of magnetic fields, D^- spectra are completely discrete and the magneto-optical transitions of the singlet lead to the final unbound localized D^- states (rather than to a continuum, as it was initially assumed in Refs. 1 and 5). As a result, D^- singlet transition energies turn out to exceed substantially ($\sim 20\%$) the ground state binding energy. More recently, in the work of Mueller *et al.*[9] variational calculations of the ground and several excited D^- singlet states for wide 510 Å QW's have been performed (see also Ref. 10) which are in very good agreement with their experimental results obtained in the fields $B < 6$ T. In higher fields and for narrower QW's the effects of band nonparabolicity (NP) and polaron corrections become increasingly important and a systematic treatment of D^- states is still lacking. It is the aim of the present paper to provide such a consideration of the singlet and triplet quasi-2D D^- states based on a direct diagonalization of the total Hamiltonian (with the application of the Ekenberg free electron Hamiltonian[11] to the D^- problem for a treatment of NP) and subsequent inclusion of magneto-polaron effects. Some preliminary results are contained in Ref. 12.

Throughout this work we shall consider the case of a GaAs/$Ga_{0.7}Al_{0.3}As$ QW with the width $d = 100$ Å and with the on-center impurity. The Hamiltonian of the system is of the form

$$H = H_0 - \frac{e^2}{\varepsilon r_1} - \frac{e^2}{\varepsilon r_2} + \frac{e^2}{\varepsilon|\mathbf{r}_1 - \mathbf{r}_2|}, \tag{1}$$

where $\mathbf{r} = (\boldsymbol{\rho}, z)$ and the Hamiltonian of free electrons in a perpendicular magnetic field B is given by

$$H_0 = \sum_{\sigma,n,m} \left[E_n + \frac{1}{2} g\mu_B B\sigma_z\right] a^\dagger_{nm\sigma} a_{nm\sigma}, \tag{2}$$

$$E_n = \epsilon_0 + \hbar\omega_c(n + 1/2).$$

The operator $a^\dagger_{nm\sigma}$ creates an electron in the lowest subband (with the energy ϵ_0) with the spin $\sigma = \uparrow, \downarrow$ ($\sigma_z = \pm 1$) and the wave function $\zeta_0(z)\phi_{nm}(\boldsymbol{\rho})$. Here $\phi_{nm}(\boldsymbol{\rho})$ is the in-plane factored wave function,[13] n is the Landau level number, and m is the oscillator quantum number connected with the projection of the angular momentum m_z by the relation $m = n - m_z$. $\zeta_0(z)$ and ϵ_0 are determined in a standard procedure in which we neglect the difference between electron effective masses in GaAs and in $Ga_{1-x}Al_xAs$ (thus taking $m^* = 0.067m_0$) and the barrier height is taken to be 60% of the band-gap discontinuity $\Delta E_g = 1.155x + 0.37x^2$ eV. In strong

magnetic fields $r_H \leq a_B$ [where $a_B = \varepsilon\hbar^2/m^*e^2$ is the effective Bohr radius and $r_H = (\hbar c/eB)^{1/2}$ is the magnetic length] we take account only of the lowest three to seven Landau levels (depending on B) and, since $r_H a_B \sim d^2$, we ignore the admixture of higher subbands due to the Coulomb interactions. To take into account the effect of the GaAs conduction band NP, we shall use the result of Ekenberg[11] for the Landau level energies in QW's, $E_n \to E_n + \delta E_n$, neglecting small spin-orbit splitting. Since the second order corrections are also small (as can be verified for QW's under consideration), we consider only the first order corrections [which allows us to still use the unperturbed wave functions $\phi_{nm}(\boldsymbol{\rho})$]

$$\delta E_n^{(1)} = \alpha_0\langle k_z^4\rangle + \frac{2(2\alpha_0+\beta_0)}{r_H^2}\langle k_z^2\rangle(n+1/2) + \frac{4\alpha_0}{r_H^4}(n^2+n+1/4) + \frac{\beta_0}{2r_H^4}(n^2+n+1) \qquad (3)$$

[cf. Eq. (35) of Ref. 11, which we have slightly corrected by the proper symmetrization of the kinematic momentum operators in the initial Hamiltonian]. The calculated corrections to the cyclotron energy $\hbar\omega_c$ which follow from the Ekenberg's approach are in very good agreement with the experimentally determined values.[14,12]

We construct wave functions of D^-, $\Psi_{M_z}(\mathbf{r}_1, \mathbf{r}_2)$, with the projection of the total angular momentum M_z out of the noninteracting wave functions $\zeta_0(z)\phi_{nm}(\boldsymbol{\rho})$ as a systematic expansion over symmetrized (for the singlet S) or antisymmetrized (for the triplet T) combinations involving the states with $n_1 + n_2 - m_1 - m_2 = M_z$. For, e.g., the singlet with $M_z = -2$ in $B = 6.7$ T we take account of 100 initial basis states — all in the first seven Landau levels.

In high fields it is appropriate[7] to classify D^- states (as well as states of any axially symmetric one-component electron system) by the quantum numbers $\{N, M\}$; here $N = n_1 + n_2$ is the total Landau level quantum number and $M = m_1 + m_2 = N - M_z$ is the total oscillator quantum number. The scheme of the calculated set of the low-lying D^- eigenstates is shown in Fig. 1. In the absence of NP the states from the groups $\{N, M\}$ and $\{M, N\}$, which are $t \to -t$ counterparts, differ in energies exactly[15] by $(N-M)\hbar\omega_c = M_z\hbar\omega_c$. Note that this $t \to -t$ symmetry becomes especially transparent in the quantum numbers N, M and the eigenstates in Fig. 1 are symmetrical about the diagonal passing through the "self-dual" $\{N, N\}$ s states.[7,12] D^- states with the energies below the $1s$ D^0 ground state have positive binding energies, i.e., they are bound (see Fig. 1). The results for the calculated energies of the singlet s ground state $|0,0;S\rangle$, of the triplet p ground state $|0,1;T\rangle$, and of the final states of the strong magneto-optical transitions (see below) are presented in Table I. Note that at $B = 20.2$ T, the singlet ground state binding energy (calculated without NP) is by $3.5\,\mathrm{cm}^{-1}$ (1.5% of the D^- singlet Coulomb interaction energy) lower than that obtained by Pang and Louie.[5] This underestimation of the interaction energies is systematic in our approach and, due to the variational principle for the first $\mathcal{N}$ levels (see Ref. 16), both the ground *and* excited states will deepen with the inclusion of higher Landau levels and higher subbands. Therefore, being systematic, the errors compensate each other and the accuracy for *transition* energies should be higher than the accuracy of the ground state energy; we expect that for $\Delta N = 1$ transition energies we obtain the accuracy of 1–$2\,\mathrm{cm}^{-1}$. It should also be stressed that we find that in the fields up to 25 T the effect of NP on the *binding* energies of both the D^0 and D^- states is small and for the singlet D^- it is practically absent. These results are in disagreement with the calculations of Pang and Louie.[5]

Consider now magneto-optical transitions with $\Delta M_z = \pm 1$ allowed in the Faraday geometry (Figs. 1 and 2). Only transitions with $\Delta M = 0$, i.e., conserving total oscillator quantum number, are strong in high magnetic fields. Indeed, all other transitions become allowed only due to the admixture of higher Landau levels (induced by the Coulomb interactions) and, hence, they are weak as $[(e^2/\varepsilon r_H)/\hbar\omega_c]^2 \sim B^{-1}$.[7] From the singlet ground state

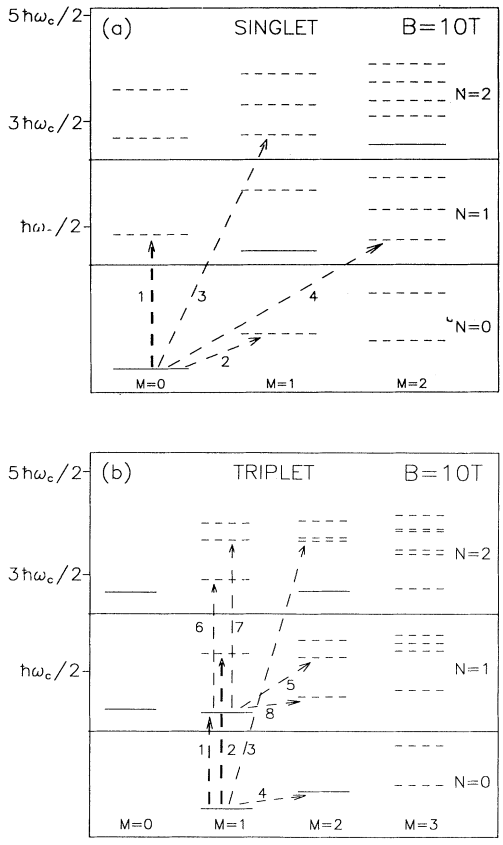

FIG. 1. Scheme of the calculated (a) singlet and (b) triplet levels of D^- at $B = 10$ T (energies are shown in scale). The states denoted by solid (dashed) lines are bound (unbound). Horizontal solid lines are guides to the eye separating the states with different Landau quantum numbers N; the actual energy positions of Landau levels are also indicated. The arrows (1–4) show the strongest allowed optical transitions from the ground D^- states; transitions (5–8) in (b) are from the strongly bound $|1,1;T-\rangle$ state.

TABLE I. Energies [in Ry* $= m^*e^4/2\varepsilon^2\hbar^2 = 5.83$ meV, measured relative to the energy of one or two free electrons (for D^0 and D^-, respectively) in their zero Landau level in the lowest subband without spin energy and NP energy corrections] of the D^0 $1s$ and $2p^+$ and the D^- singlet and triplet states involved in the strong magneto-optical transitions.

Magnetic field B (T)	6.75	10	15	20.2
$\gamma = \hbar\omega_c/2$ Ry*	1.0	1.5	2.2	3.0
$1s$	−2.86	−3.13	−3.52	−3.83
$2p^+$	0.64	1.36	2.53	3.82
$\|0,0;S\rangle$	−3.55	−3.95	−4.48	−4.88
$\|1,0;S\rangle$	−0.72	0.01	1.13	2.41
$\|0,1;T\rangle$	−3.42	−3.79	−4.27	−4.65
$\|1,1;T-\rangle$	−1.55	−0.92	0.06	1.24
$\|1,1;T+\rangle$	0.12	0.78	1.87	3.09

$|0,0;S\rangle$ there are the pairs of transitions separated in energy, not exactly in the presence of NP, by $\hbar\omega_c$.[6,7] As a consequence of the $t \to -t$ symmetry, the same holds true for transitions from all $\{N,N\}$ s states of arbitrary one-component electron systems[12,15] (see also Ref. 9). In each pair the transition with $\Delta M_z = -1$ (corresponding to the right circular polarization σ^+) is weaker in high fields. We find very good agreement with the experimental data of Huant *et al.* for the transitions from the singlet ground state [see Fig. 3(a)]. From the triplet ground state $|0,1;T\rangle$ there are *two* strong $\Delta M = 0$ transitions[7] [shown by bold arrows 1 and 2 in Fig. 1(b)] to the final states which we denote as $|1,1;T\pm\rangle$. The transition 1 is to the *strongly bound* state $|1,1;T-\rangle$ and the transition energy is very close to $\hbar\omega_c$ (cf. Ref. 7). Note that the $\Delta N = 0$ triplet transition 4 in Fig. 1(b) is to the final bound state $|0,2;T\rangle$ and, hence, its energy is less than the triplet ground state binding energy; this transition, however, turns out to be very weak [Fig. 2(b)]. In available magnetic fields $B < 80$ T, due to the low values of the g factor in GaAs QW's, the D^- singlet is still the ground state (see also Ref. 10), hence the triplet is depopulated at low temperatures. Besides, the transition matrix elements for the triplet are nearly two times

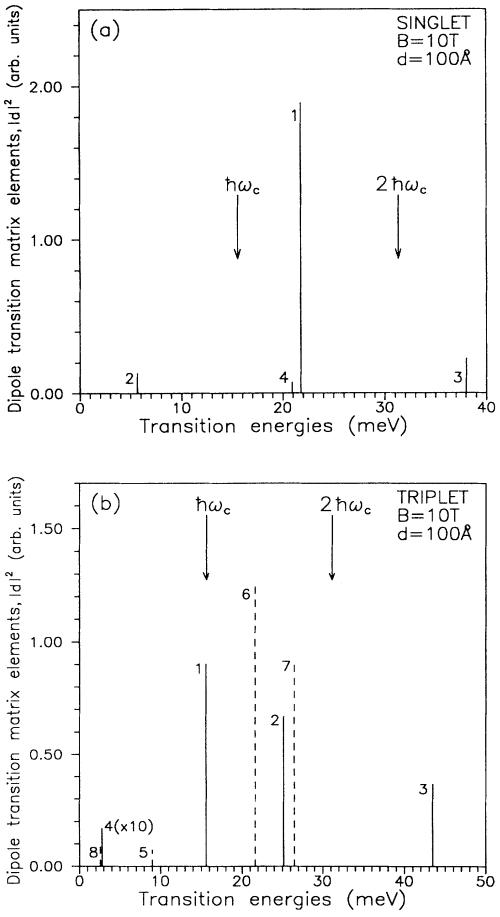

FIG. 2. Dipole transition matrix elements $|d|^2$ corresponding to Fig. 1. Transitions from the triplet state $|1,1;T-\rangle$ in (b) are shown by dashed lines. The effect of band nonparabolicity is taken into account.

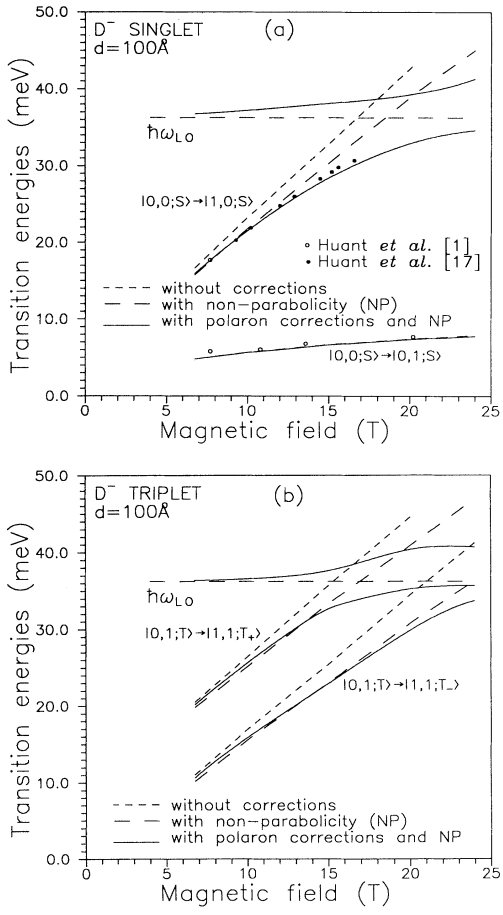

FIG. 3. Transition energies from the ground (a) singlet and (b) triplet D^- states vs B. Polaron corrections are shown for the improved Wigner-Brillouin (IWB) perturbation theory; only one upper polaron branch is depicted. The very weak transition of the triplet in the $N = 0$ Landau level is not shown.

smaller (see Fig. 2). The calculated ratios of intensities of the two strong triplet transitions to that of the singlet $R_{\pm} = I_{\mathrm{T}}^{\pm}/I_{\mathrm{S}}$ at the temperature $T = 4$ K and $B = 10$ T are given by $R_{-} = 0.15$, $R_{+} = 0.12$, while at $T = 10$ K $R_{-} = 0.58$, $R_{+} = 0.43$, i.e., they are not small. However, these two strong triplet transitions turn out to be close in energy, correspondingly, to the cyclotron resonance of free electrons $\hbar\omega_c$ and the $1s \rightarrow 2p^{+}$ ($\Delta M = 0$, $\Delta N = 1$) transition of D^0 within a QW. Note that the latter transition may in principle be suppressed when all D^0 centers are converted[2,3] to D^- and temperature is not too high so that D^- centers are not ionized. Our predictions for the triplet D^- transitions need experimental verification.

Magnetopolaron effects lead to a shift of shallow donor energies in QW's in intermediate magnetic fields and give rise to resonant splittings in high fields $B > 15$ T (see, e.g., Refs. 17 and 4 and references therein). To describe this effect for D^-, we have taken account of the Fröhlich interactions of electrons with bulk GaAs LO phonons ($\hbar\omega_{\mathrm{LO}} = 36.25$ meV, the electron-phonon coupling constant $\alpha = 0.068$) within the two second order perturbation theory schemes: (i) Rayleigh-Schrödinger for the ground and Wigner-Brillouin for excited D^- states (RS-WB) and (ii) the improved Wigner-Brillouin (IWB) (see Ref. 4 and references therein) which ensures the pinning behavior at $\hbar\omega_{\mathrm{LO}}$. After performing an infinite summation over momentum of LO phonon, we include in the perturabtion theories from five to seven relevant D^- states (compare with Ref. 4). All other discarded D^- states are localized far away from the states of interest, so that the matrix elements become very small and exponentially decreasing. The magnetopolaron corrections to the energies of D^- states turn out to be large ($\sim 4\alpha$), as it should be for a bound magnetobipolaron (when α is small) for which a polarization cloud of LO phonons is enhanced by a factor of 2 compared with that for D^0; to some extent this resembles the giant binding of D^- centers in bulk strongly polar semiconductors.[18] For, e.g., the D^- singlet s ground state containing two equivalent electrons, in the field $B = 20$ T the correction is -5.28 meV, which exceeds that to the D^0 ground state by nearly four times. Nevertheless, the $\Delta N = 0$ transition energies [Fig. 3(a)] are altered very modestly (the corrections to the initial and final D^- states compensate each other). The same conclusion for the $\Delta N = 0$ singlet D^- transition has been recently reached by Peeters.[19] In the high-field region of resonant splittings, the available published experimental data for $\Delta N = 1$ D^- transition energies (not diminished by a high-field $\hbar\omega_c$ as in Ref. 1) are from Ref. 17, where the peak B should be assigned to D^-. We find that IWB gives the results below the experiment (at $B = 16.6$ T by $\sim 3\%$), i.e., overestimates polaron corrections [see Fig. 3(a)], while RS-WB underestimates them by $\sim 4\%$ (not shown). We believe that for D^- the high-field region of resonant splittings needs further experimental and theoretical studies.

Note added. In a recently published paper[20] an additional feature in the magneto-optical absorption spectra of double-planar-doped GaAs/$Ga_{1-x}Al_xAs$ QW's has been observed on the low-energy side of the cyclotron resonance when the concentration of excess free electrons n_{ex} was increased. According to our calculations, we assign this feature to the transition evolving with increasing n_{ex} from the triplet D^- transition $|0,1;T\rangle \rightarrow |1,1;T-\rangle$.

It is a pleasure to thank S. G. Tikhodeev for helpful discussions and encouragement. We are also most grateful to D. M. Larsen for sending a copy of Ref. 10 prior to publication and to S. Huant for communicating to us their recent results prior to publication. A.B.D. wishes to thank F. M. Peeters for a fruitful discussion of magnetopolaron effects and to express gratitude to G. E. W. Bauer and the Department of Theoretical Physics, Delft University of Technology for hospitality.

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