Triplet transitions of D^- centers in quantum wells in high magnetic fields

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The spin-singlet and spin-triplet optical transitions of negative donor centers D^- in GaAs/(Al,Ga)As quantum wells in high magnetic fields are considered both theoretically and experimentally. The measured and calculated transition energies are shown to be in very good agreement for the D^- singlet transition (1%). The two strong triplet transitions that are predicted by theory are not observed in the experiment. This is thought to be due to accidental energy coincidences with one-particle excitations such as the electron cyclotron resonance and the $1s \rightarrow 2p^+$ transition of neutral donors.

INTRODUCTION

D^- centers are simple two-electron systems formed by neutral donors D^0 trapping a second electron. They are interesting to study in view of interparticle correlations. In a high magnetic field and in a quantum well (QW), correlation effects become very pronounced and a strong enhancement of the binding energy is observed.[1,2] Apart from the confinement effects, the QW geometry has the interesting property that a stable population of D^- centers can be created using the double-planar doping technique.[1,3] This makes the study of the energy spectrum of D^- centers considerably easier than in bulk material where sample and experimental conditions for populating D^- states are very restrictive.[4]

The energy spectrum of confined D^- centers in a magnetic field has been the focus of several recent studies.[5–11] In QW's at low temperatures, only the spin-singlet s ground state of D^- is populated and gives rise to an intense spin-singlet transition. This transition has already been measured over a large range of well widths and magnetic fields and a very good agreement between experiment and various theoretical approaches has been found.[8,9] In high fields, in addition to this singlet D^- transition, theory predicts[6,7] two intense transitions from the triplet p ground state to the excited triplet states which have found no experimental confirmation so far. This seems paradoxical in view of the common observation of the singlet transition.[9,12]

In this paper, we describe our search for the D^- triplet transitions which should be observed at a higher temperature when the lowest triplet state starts to be thermally occupied.[8,10] We find evidence for an interexcited state transition of D^0 donors. However, a careful analysis of the optical spectra supplemented by calculations of various transition energies reveals that the triplet transitions are in fact not seen because they are not energetically discriminated from either the electron cyclotron resonance (CR) or the D^0 $1s \rightarrow 2p^+$ transitions in the samples studied so far, as revealed by our calculations.

SAMPLES AND EXPERIMENTAL DETAILS

Three $GaAs/Al_{0.25}Ga_{0.75}As$ multiple-quantum-well structures grown by molecular-beam epitaxy have been studied by means of far-infrared (FIR) magnetotransmission measurements in magnetic fields B up to 13 T. The barrier width and the well width are approximately 200 Å. Sample 1 (sample 3) is planar doped with silicon donors at the middle of the wells (barriers) only. Sample 2 is double-planar doped, that is, planar doped at the middle of the wells and middle of the barriers (concentration of 5×10^9 cm^{-2} per doping plane). This is the only sample which reveals D^- centers as confirmed by the strong singlet transition in the magneto-optical spectra.[3] For a comparison with calculations, we have determined the well width d, barrier width, and Al content in the barrier of sample 2 by x-ray diffraction measurements:[13] these parameters are, respectively, 194 Å, 196 Å, and 0.26. FIR transmission measurements are performed without visible light illumination, at various

temperatures between 2 K and 30 K, using a fast-scan Fourier-transform spectrometer (Bruker IFS 113 v). The instrumental resolution is taken at 2 cm^{-1}.

THEORETICAL MODEL

In the effective mass approximation, the Hamiltonian of the problem has 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 positions of electrons are denoted by $\mathbf{r}_i = (\boldsymbol{\rho}_i, z_i)$, and the impurity is assumed to be at the center of a QW. The Hamiltonian H_0 of free electrons in a perpendicular magnetic field B is given by

$$H_0 = \sum_{\sigma,j,n,m} \left(E_{jn} + \tfrac{1}{2} g^* \mu_B B \sigma_z\right) a^\dagger_{jnm\sigma} a_{jnm\sigma}\,, \tag{2}$$

$$E_{jn} = \epsilon_j + \hbar\omega_c (n + \tfrac{1}{2})\,. \tag{3}$$

The operator $a^\dagger_{jnm\sigma}$ creates an electron with the spin projection $\sigma = \uparrow, \downarrow$ ($\sigma_z = \pm 1$) and with the coordinate wave function $\zeta_j(z)\phi_{nm}(\boldsymbol{\rho})$; here j is the subband index, n is the Landau level number, and m the oscillator quantum number, $m_z = n - m$ (see, e.g., Ref. 6 and references therein), and $\hbar\omega_c$ is the cyclotron energy. The wave functions of electric subbands $\zeta_j(z)$ and the corresponding energies ϵ_j are determined in a standard procedure in which we neglect the difference between the electron effective masses in GaAs and in $Ga_{1-x}Al_xAs$ and take $m^* = 0.067 m_0$. The confining potential $V(z)$ is taken to be $V(z) = V_0$ for $|z| > d/2$ and $V(z) = 0$ for $|z| < d/2$ and $V_0 = 0.67\Delta E_g(x)$,[14] $\Delta E_g = 1.45x$ (eV).[13] To take into account the effect of GaAs conduction band nonparabolicity (NP), we use the analytical expression of Ekenberg[15] for the Landau level energies in QW's; the relevant details have been presented elsewhere.[8,16] Magnetopolaron effects on D^- transition energies in fields up to 10 T are small,[8,11] and we neglect them here.

We construct wave functions of D^- with the total angular momentum projection M_z, out of the noninteracting wave functions $\zeta_i(z)\phi_{nm}(\boldsymbol{\rho})$ as symmetrized (for the singlet, S) or antisymmetrized (for the triplet, T) combinations

$$\Psi_{M_z S(T)}(\mathbf{r}_1, \mathbf{r}_2) = \sum_{i,j} \sum_{n_1,m_1,n_2,m_2} A_{M_z S(T)}(n_1, m_1, n_2, m_2) \tfrac{1}{\sqrt{2}} (1 \pm \hat{P}_{12})\, \zeta_i(z_1)\phi_{n_1 m_1}(\boldsymbol{\rho}_1)\zeta_j(z_2)\phi_{n_2 m_2}(\boldsymbol{\rho}_2)\,, \tag{4}$$

involving the states with $n_1 + n_2 - m_1 - m_2 = M_z$; $\hat{P}_{12}$ is the operator of permutations of coordinates $\mathbf{r}_1 \leftrightarrow \mathbf{r}_2$; for D^- singlet, orbitals with coinciding quantum numbers $i = j$, $n_1 = n_2$, $m_1 = m_2$ are counted only once. We obtain the D^- eigenstates (i.e., the expansion coefficients $A_{M_z S(T)}$) and the corresponding eigenenergies by diagonalization of the matrix of Hamiltonian (1) calculated in the orthonormal basis set of two-electron noninteracting states in the QW and in the magnetic field. In the considered regime of high magnetic fields $r_H = (\hbar c/eB)^{1/2} < a_B = \varepsilon\hbar^2/m^* e^2$ ($B > 6$ T), a reasonable accuracy is obtained when the basis set is truncated by including only states with noninteracting energies $E = E_{in_1} + E_{jn_2} \le E_{\max} \simeq 9e^2/\varepsilon r_H$; thus only the three to seven lowest Landau levels $n_1 + n_2 \le 3$–7 (depending on B) and the three to five lowest subbands $i + j \le 3$–5 (depending on the well width d) are taken into account. We estimate the achieved relative accuracy in determining the D^- interaction energies [i.e., the total contribution of the Coulomb e-e and electron-impurity interactions from (1)] to be $\sim 4\%$ (which corresponds to absolute accuracy ~ 6 cm^{-1} at $B = 10$ T). We believe that this accuracy is mainly limited by the neglect of single-particle electronic states from the continuum above the well (see, e.g., calculations of the magnetoexciton spectra in QW's[17]). It is important to stress that underestimations in interaction energies are systematic and are similar for both the ground and excited states in our approach. Hence, due to a compensation of these underestimations, the accuracy for D^- *transition* energies is much higher. D^- states are classified by the high-field quantum numbers $|N, M; S(T)\rangle$ (and, when necessary, by an additional index distinguishing between the states within the group $\{N, M\}$);[6,10] here $N = n_1 + n_2$ and $M = m_1 + m_2 = N - M_z$ is the total oscillator quantum number.

A similar approach is used for calculations of the D^0 eigenstates whose wave functions are in the form

$$\Phi_{M_z}(\mathbf{r}) = \sum_i \sum_{n,m} A_{M_z}(n, m)\, \zeta_i(z)\phi_{nm}(\boldsymbol{\rho}) \tag{5}$$

with $n - m = M_z$. The expansion coefficients $A_{M_z}(n, m)$ and the D^0 eigenenergies are obtained by numerically solving the corresponding secular equation which involves the one-electron part of Hamiltonian (1).

RESULTS AND DISCUSSION

Figure 1 shows a low-temperature transmission spectrum for the double-planar doped sample 2. Three peaks are seen in this spectrum. Peaks A and C are the $1s \rightarrow 2p^+$ transitions for neutral donors in the wells and in the barriers, respectively: they are the only features seen in samples 1 and 3, respectively. Peak B is the strong transition from the D^- singlet s ground state $|0, 0; S\rangle$ to the excited state $|1, 0; S\rangle$ associated with the first Landau level. The energy of this transition has been shown to be in very good agreement with the results of the direct diagonalization of the interaction Hamiltonian[8] and with variational Monte Carlo calculations[9] over a

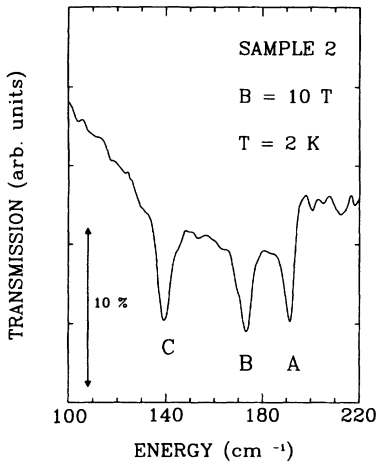

FIG. 1. Magnetotransmission spectrum of the double-planar doped sample 2 at $B = 10$ T and $T = 2$ K, showing the strong D^- singlet transition in this sample (peak B).

large well width and magnetic field range.

Figure 2 shows transmission spectra for all three samples at higher temperatures. At these temperatures, some donors are thermally ionized and the cyclotron resonance is seen. The interesting behavior to stress here is that a peak develops at approximately 10 cm^{-1} below the CR line in *both samples 1 and 2*. The experimental finding that this peak is also seen in sample 1, *which has no D^- centers*, demonstrates that *it is not due to a D^- transition* — such as, e.g., a spin-triplet transition—as believed by some other authors.[18] We assign this peak to the $2p^- \rightarrow 2s$ transition of neutral donors D^0 in the wells. This assignment is supported by the good agreement with calculations (see below) and by the relative occupancies of the $1s$ and $2p^-$ states at 27 K as compared to the relative intensities of this additional peak and of peak A. Careful experiments carried out at higher resolution (up to 0.5 cm^{-1}) and various temperatures between 4 K and 30 K failed to reveal any additional peak near the CR line and near peak A where D^- triplet transitions are expected (see below).

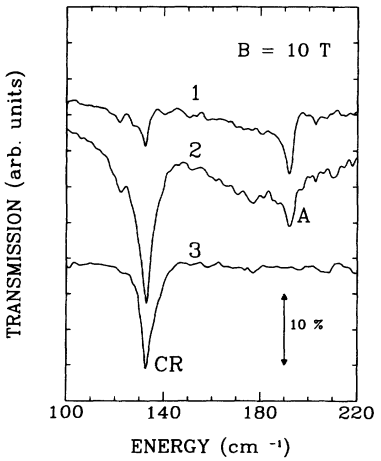

FIG. 2. Magnetotransmission spectra at 10 T of the on-well doped sample 1 at 27 ± 2 K, the double-planar doped sample 2 at 22 ± 2 K, and the on-barrier doped sample 3 at 20 ± 2 K. The spectra have been vertically displaced for the sake of clarity.

The calculated energies of the strong D^- magneto-optical transitions are shown for sample 2 in Fig. 3. Also shown for comparison are the experimental energies of the singlet transition. These strong transitions are the transition between the singlet states denoted S and the *two* strong transitions with $\Delta N = 1$, $\Delta M = 0$ ($\Delta M_z = +1$) from the triplet p ground state $|0,1;T\rangle$. The latter are in different spectral regions; we denote them by $T\pm$ (and the corresponding final states as $|1,1;T\pm\rangle$).[19]

For magnetic fields less than 80 T, due to the low values of the g^* factor in GaAs, the D^- singlet $|0,0;S\rangle$ is still the ground state, hence the triplet D^- p ground state is depopulated at low temperatures. Also, the transition matrix elements for the triplet are nearly two times smaller than that of the singlet.[8,10] Thus, the triplet $T\pm$ transitions can be experimentally tested only at elevated temperatures. Indeed, the calculated relative intensities $R_\pm = I_T^\pm / I_S$ of the two strong triplet $T\pm$ transitions to that of the singlet at $T \simeq 10$ K turn out to be ~ 0.5 (while at $T = 4$K $R_\pm \sim 0.1$). This intensity should be sufficient to detect triplet transitions in the experiment.

The question remains why the triplet transitions $T+$ and $T-$ are not in fact seen in the experiment? We think that in the presence of broadening they are masked by the strong $1s \rightarrow 2p^+$ transition of D^0 within a QW and by the CR, which occur in the same spectral region, respectively.[8] This is shown in Table I which summarizes all relevant transition energies in sample 2 at three field strengths. Table I gives support to our interpretation of the additional peak of Fig. 1 as due to the $2p^- \rightarrow 2s$ transition of D^0 donors in the wells. It can be seen that experiment and theory agree within typically 1%. For instance, the results of theoretical calculations of the singlet D^- transition energy at $B = 10$ T coincide with

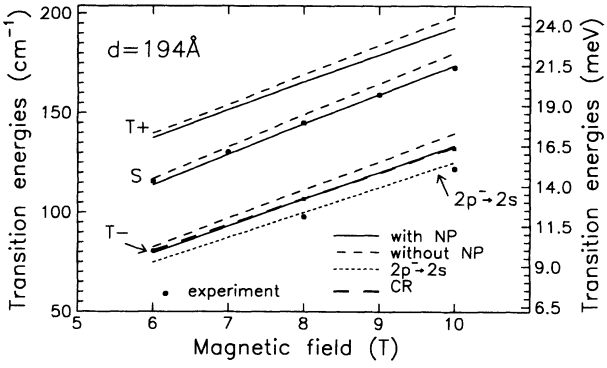

FIG. 3. Energies of the strong magneto-optical transitions from the singlet D^- s ground state (S) and from the triplet D^- p ground state ($T\pm$) for the QW with $d = 194$ Å. Solid (dashed) lines are the calculated values with (without) allowance for the effects of nonparabolicity (NP). The full symbols are the experimental data for the singlet D^- transition, CR, and $2p^- \rightarrow 2s$ transition. The calculated CR (large-dashed line) and the calculated $2p^- \rightarrow 2s$ transition energy (small-dashed line) are also shown.

TABLE I. Transition energies in cm^{-1} for sample 2 (d = 194 Å) at three magnetic field strengths. For each transition, the first row is the theoretical prediction and the second row is the experimental value (when available). For the D^- triplet transitions $T-$ and $T+$, only theoretical predictions are given to show the proximity of their energies to the CR $\hbar\omega_c$ and to the D^0 $1s \rightarrow 2p^+$ transition energy, respectively.

B (T)	$2p^- \rightarrow 2s$	$T-$	$\hbar\omega_c$	S	$T+$	$1s \rightarrow 2p^+$
6	74.7	79.4	80.6	113.8	137.4	136.0
			80.6	115.7		138.1
8	100.0	106.8	106.9	144.5	165.6	163.8
	97.8		106.9	145.0		164.8
10	125.2	133.4	132.9	173.8	192.7	190.9
	121.9		132.1	172.7		191.2

the experiment within 0.6% (1.1 cm^{-1}). This remarkable agreement gives us confidence in the calculated proximity between the $T+$ ($T-$) triplet transition energies and the $1s \rightarrow 2p^+$ (CR) transition energies as shown in Table I.

CONCLUSION

In conclusion, we have considered the spin-singlet and spin-triplet transitions of D^- centers in quantum wells in high magnetic fields. The calculation of the singlet transition agrees remarkably well with the experiment. Our analysis reveals that the two strong triplet transitions $T\pm$ which are expected from theory are not seen in the experimental spectra. This is thought to be due to the fact that they are not distinguished from other one-particle strong magneto-optical transitions due to the accidental energy coincidences. We have shown that upon increasing temperature, a peak develops on the low-energy side of the CR line in well-center doped and double-planar doped samples. Its energy position makes it tempting to interpret as the $T-$ triplet transition.[18] We have demonstrated, however, that this peak is in fact due to the $2p^- \rightarrow 2s$ transition of D^0 donors at the middle of the quantum wells.

Finally we mention an interesting possibility to find traces of D^- triplet transitions in the optical spectra of D^- doped QW samples.[3] This is to study the evolution of magneto-optical spectra associated with impurities with increasing concentration of excess free electrons n_{ex} in a QW.[20] For Landau level filling factors $\nu \equiv 2\pi r_H^2 n_{ex} \sim 1$, the picture of the transitions S and $T\pm$ between the two-particle D^- states should be changed to a picture of collective magnetoplasma excitations localized at the Coulomb impurity.[8,21] There are two such localized optically active collective modes lying in different spectral regions: one develops from the $T+$ and S transitions of D^- (and, in agreement with the experiment of Ref. 20, is shifted to higher energies due to the exchange effects), and the other develops from the $T-$ transition and lies below $\hbar\omega_c$. The latter mode could explain the low-energy feature of the CR in Ref. 20 at $B = 9$ T and $\nu \sim 1.3$.

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[19] The D^- singlet S and the triplet $T+$ transitions have their origin in the single-particle $1s \rightarrow 2p^+$ D^0 transition which is shifted due to the e-e interaction. The shifts, obviously, are different for the singlet and triplet D^- states. Similarly, the origin of the triplet $T-$ transition is the $2p^- \rightarrow 2s$ transition of D^0 which is also shifted due to the e-e interaction. In the high-magnetic-field limit and strictly two-dimensional consideration of Refs. 6 and 7, these shifts turn out to be zero.

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