Well-width dependence of D^- cyclotron resonance in quantum wells

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Negative-donor (D^- center) cyclotron resonance is studied in GaAs quantum wells of different widths to obtain information about the well-width dependence of the D^- ground-state binding energy and optical transition energies in the range of 58–373 Å. The experimental magneto-optical transition energy is shown to increase with decreasing well width, in good agreement with variational quantum Monte Carlo results.

I. INTRODUCTION

Electron correlation is crucial for the binding of a second electron to neutral shallow donors D^0 in semiconductors, i.e., the formation of D^- centers.[1] D^- centers are one of the simplest "many-body" electronic systems which may be used as a test for theoretical descriptions of electron correlation. In this respect, the diffusion quantum Monte Carlo approach used in Ref. 2 to compute the D^- binding energy in a bulk semiconductor in a high magnetic field has proven superior to previous variational calculations.[3] Since correlation effects are expected to be enhanced with reduced dimension, there is interest in studying D^- centers in quantum wells (QW's) with decreasing well widths. Such a study is made experimentally feasible because a stable D^- population can be engineered in QW's by appropriate doping of a multiple-quantum-well (MQW) structure.[4–6]

In the present paper, we focus on the well-width dependence of D^- cyclotron resonance (CR) in the range from 58 to 373 Å. Apart from studying correlation effects with increasing quantum confinement, this study is also motivated by the observation that the energy of the characteristic D^- peak (the D^- CR) in far-infrared (FIR) magneto-optical spectra is not very sensitive to the well width L_W, while D^0 transitions are clearly L_W sensitive. This apparent inconsistency is shown to be due to band nonparabolicity. The analysis of the experimental spectra reveals a clear enhancement of the excitation energy with decreasing well width, in agreement with quantum Monte Carlo (QMC) simulations.

II. SAMPLES, EXPERIMENTAL AND THEORETICAL METHODS

A series of molecular-beam-epitaxy grown GaAs/(Ga,Al)As MQW samples intentionally doped with silicon has been studied by means of FIR magneto-optics. A list of the samples is given in Table I. The samples were double-planar-doped (DPD) with growth interruption during doping at the middle of the wells and middle of the barriers. This double-planar-doping technique produces a controlled electron surplus in QW's which favors the formation of D^- centers.[4–6] The donor concentrations were obtained from Hall measurements at room temperature and are given in Table I. For a comparison with calculations, we have determined L_W, the Al content in the (Ga,Al)As barriers (and thus the barrier

TABLE I. List of the samples with relevant parameters. The number of periods is 100.

Sample	L_W (Å)	L_b (Å)	Al content (%)	N_W per well ($\times 10^{10}$ cm^{-2})	N_b per barrier ($\times 10^{10}$ cm^{-2})
*FC*22	58	191	25	0.85	0.85
*F*606	95	194	26	0.84	0.84
*FC*14	144	187	23	1.1	1.1
*FC*18	194	196	26	0.5	0.5
*G*312	373	191	26	0.44	0.44

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width L_b) by x-ray-diffraction measurements.[7] FIR magnetotransmission spectra were recorded at a fixed magnetic field up to 13 T in the Faraday geometry using a BRUKER IFS 113-v fast-scan Fourier-transform spectrometer connected to a superconducting magnet. The substrates were wedged by 4° to avoid line-shape distortion by multireflections.

Our diffusion QMC calculations for the ground-state energy of D^0 and D^- centers have been discussed in some detail in Ref. 2. For the variational calculations of the excited-state energies, we adopt a wave function of the following form for the D^- center:

$$\Psi(\mathbf{r}_1,\mathbf{r}_2)=f(\mathbf{r}_{12})[g_1(\mathbf{r}_1)g_2(\mathbf{r}_2)+g_1(\mathbf{r}_2)g_2(\mathbf{r}_1)]\prod_{i=1}^{2}\varphi(z_i)\ , \tag{1}$$

where z is along the magnetic-field direction, and $\varphi(z)=\cos(kz)$ in the well and $\cos(kL_W/2)\exp[\lambda(L_W/2-|z|)]$ outside the well (k and λ are determined by solving for the solution of a single electron in such a quantum well), $f(\mathbf{r})=\exp\{ar/[1+(c_\rho^2\rho^2+c_z^2z^2)^{1/2}]\}$, where a is chosen to be 0.5 to satisfy the cusp condition at small distance between electrons, and the single-particle orbital g_i in the form as that used by Larsen,[8] which is

$$g_i(\mathbf{r})=\rho^{|M|}\exp(-\mu_i\rho^2-\kappa_i\sqrt{\rho^2+\alpha_iz^2})\ , \tag{2}$$

with M the z-component angular moment of the orbital. There are eight variational parameters (c_ρ, c_z, μ_1, μ_2, κ_1, κ_2, α_1, and α_2). Total energies are minimized with respect to these parameters to obtain the variational eigenvalues of the Hamiltonian. 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 the derivatives are less than a certain value (typically 10^{-3} a.u.). A set of configurations is regenerated 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 $g(\mathbf{r})\varphi(z)$ in Eqs. (1) and (2).

III. RESULTS AND DISCUSSION

Typical transmission spectra taken at 10 T and normalized to zero-field spectra are shown in Fig. 1. The three peaks are labeled $A-C$ in order of decreasing energy to agree with previous nomenclature.[4,5] Peaks A and C are the $1s\rightarrow2p^+$ transitions for well-center donors $D^0(W)$ and barrier-center donors $D^0(b)$, respectively.[4,9,10] They are the only observable peaks in samples which are doped at the middle of the wells and the middle of the barriers, respectively. Peak B is the characteristic D^- feature which is seen in DPD samples such as those of Fig. 1. The formation of stable D^- centers in the wells in DPD samples is due to donors in the wells trapping a second electron given by barrier donors.[4–6] An energy-level diagram is given in Fig. 2 which shows the optical transitions of interest in this study.

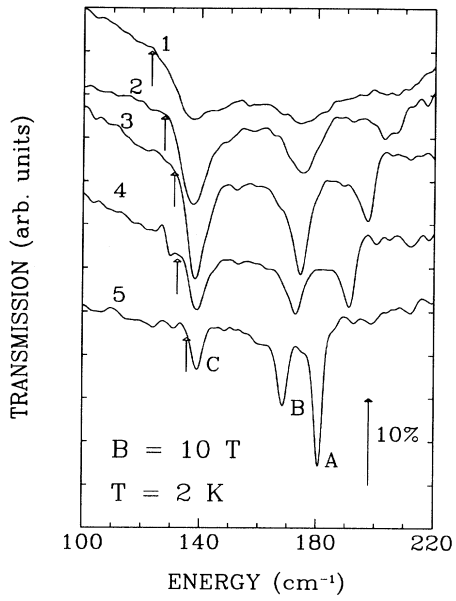

FIG. 1. Typical magnetotransmission spectra (resolution 2 cm^{-1}) of the samples of Table I. Curves 1–5 are for sample $FC22$, $F606$, $FC14$, $FC18$, and $G312$, respectively. The vertical arrows point out the electron CR energies measured at higher temperature (Ref. 11) at the same field of 10 T. Peaks A and C are the $1s\rightarrow2p^+$ transitions for D^0 donors at the middle of the wells and middle of the barriers, respectively. Peak B is the characteristic D^- CR peak.

At low temperature, the impurity transitions $A-C$ are the only observable features. As temperature increases, the impurities are thermally ionized and the impurity transitions are replaced by an electron CR whose energy is indicated by the arrows in Fig. 1. It is seen in Fig. 1 that the CR energy decreases with decreasing well width. This behavior is entirely due to band nonparabolicity as discussed in Ref. 11, where it was shown that the cyclotron mass in a QW can be fairly well understood in terms of the parallel effective mass introduced by Ekenberg[12] (see the Appendix). Band nonparabolicity has important consequences on the D^- CR energy, as we will see, and on the D^0 transition energies as we shall first discuss.

A. Neutral donor (D^0) transitions

As seen in Fig. 1, the energy of peak A increases by 27.5 ± 1 cm^{-1} from the widest well (sample $G312$) to the narrowest one (sample $FC22$). This is a consequence of increasing confinement as the electron wave function is pushed closer to the impurity ion for decreasing well width.[13] We have carried out variational calculations[14,15] of the transition energies A and C for samples $G312$ and $FC22$, using the sample characteristics of Table I. The potential depth is taken as 67% (Ref. 16) of the gap difference between GaAs and $Ga_{1-x}Al_xAs$: $\Delta E_g=1.45x$ (eV).[7] The electron-phonon interaction is neglected: This is justified for fields not greater than 10 T, for which mixing of the $2p^+$ state with the state formed by the $1s$ state plus one longitudinal-optical phonon is small.[14] Neglect-

ing the band nonparabolicity, we calculate that the energy of peak A increases by 37.5 cm^{-1} from the wide to the narrow well at a field of 10 T. However, this enhancement is reduced by band nonparabolicity similar to the CR energy because the electron CR and impurity CR have similar field slopes, which is different in different samples. Computing the transition energies with the effective mass corrected for band nonparabolicity (see the Appendix), we indeed find this enhancement to be 28.8 cm^{-1}, in good agreement with the measured value. It is also seen in Fig. 1 that the energy of peak C is approximately independent of the sample. The energy structure of a $D^0(b)$ donor in a thick barrier is primarily determined by L_b,[14] which is approximately the same in the samples investigated. For decreasing L_W, however, the electron in the well is pushed closer to its parent donor at the middle of the barrier and should therefore feel more binding. The energy of peak C should increase by 3.9 cm^{-1} according to our variational estimate without band nonparabolicity, while a slight decrease ($\approx 3\pm 1$ cm^{-1}) is observed. Again, this is due to band nonparabolicity. Our variational estimate after correction for band nonparabolicity gives a 8.9-cm^{-1} decrease from the widest to the narrowest well.

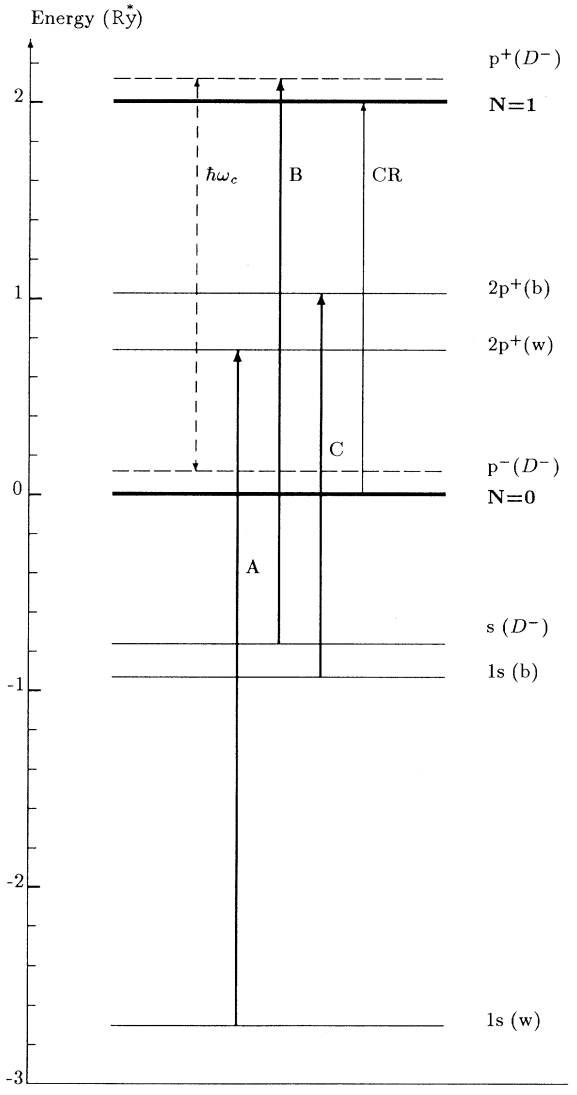

FIG. 2. An energy-level diagram showing the D^0 and D^- levels and transitions in sample $F606$ in a reduced magnetic field of $\gamma = 1$ (around 6.75 T in GaAs). Band nonparabolicity is neglected. The energies are expressed in effective Ry. The D^0 levels have been computed using the variational approach of Ref. 14. The D^- levels have been computed for a 95-Å single well using the variational QMC method described in the text. The $p^-(p^+)$ like excited states of D^- are lying above the $N=0$ ($N=1$) Landau levels, and are therefore not bound.

B. Negative-donor (D^-) transitions

We now turn to the main point of the paper, namely the behavior of the D^--related peak B. Since the energy of this peak increases almost linearly with magnetic field at a slope close to that of the cyclotron energy $\hbar\omega_c$, the final state of this transition is connected to the $N=1$ Landau level (see Fig. 2). In our previous work, we proposed that this final state is the $N=1$ Landau level itself.[4,5] This assignment was made plausible by analogy to what is experimentally[17] and theoretically[3] known in bulk material: that D^- centers are observed in photoionization transition due to spin conservation. Recently, Larsen and McCann[18] and Dzyubenko[19] have shown that, in QW's, the final state of the D^- peak B is in fact not the $N=1$ Landau level, but an unbound localized p^+-like D^- singlet state connected to it and lying slightly higher in energy (Fig. 2). This is because in a magnetic field, all D^- states formed from a donor and a second electron confined to a common QW are discrete due to complete quantization of the electron motion. This is very similar to a problem that was previously debated for neutral donors[20] and excitons[21] in QW's. The variational calculations of Mueller *et al.*[8] found that the negative binding energy of this excited unbound singlet state relative to the $N=1$ Landau level (see Fig. 2) is very small, of the order of -1 cm^{-1} for a 500±50-Å-thick QW at 5 T. We have extended the calculation to a series of well widths using the variational QMC method (see below).

To avoid ambiguous or cumbersome labeling of the $\Delta M=+1$ transition B,[22] we refer to it as the "D^- CR" in the rest of this paper.[23] This is similar to calling the $1s\rightarrow 2p^+$ transition for shallow donors the impurity CR. Similarly, the fundamental D^- transition of $\Delta M=-1$ that we reported in Refs. 4 and 5 is the transition from the D^- ground state to the p^--like unbound singlet excited state which lies slightly above the $N=0$ Landau level in energy (see Fig. 2). Not that the p^+- and p^--like states are replicas of each other, with $M=+1$ and $M=-1$, respectively, and that they are separated by the cyclotron energy $\hbar\omega_c$.[18,19] Thus a measurement of the energy of peak B at some field minus the electron CR energy measured at the same field gives the $\Delta M=-1$ transition energy at that field. The validity of this procedure was checked in Refs. 4 and 5 in a 100-Å QW in a magnetic field up to 21 T.[24]

In Fig. 1, it is striking at first sight that peak B is al-

most independent of L_W up to 144 Å, and tends to decrease only for the widest well (7 cm^{-1} from the 144-Å well to the 373-Å well). This is elucidated by subtracting the electron CR energy from peak B, which gives the transition energy from the D^- ground state to the p^--like unbound singlet state (see Fig. 2). The results are shown in Fig. 3. It is found that this transition energy indeed increases for decreasing L_W. This increase is offset by the band nonparabolicity which softens the CR energy for decreasing L_W at a given field in such a way that the energy of transition B remains approximately constant over a wide L_W range. Thus it is convenient to discuss the optical transition from the D^- ground state to the p^--like state to avoid complications with band nonparabolicity, which masks part of confinement effects.

In Fig. 4, we compare the transition energies of Fig. 3 with the D^- binding energies in the 95- and 194-Å single wells calculated using the diffusion QMC method of Ref. 2. We have corrected the computed binding energies for band nonparabolicity, which deepens slightly shallow impurity states.[14,25] This is done by rescaling energies and magnetic fields as described in the Appendix. As can be seen in Fig. 4, agreement between the theoretical binding energies and experimental results is achieved within 8% for the two well widths investigated, with the theoretical values being low. Without nonparabolic corrections, agreement is achieved within 12%.

When the final state of the transition in Fig. 3 is attributed to the p^--like excited state of the QW D^- center (Fig. 2), the agreement is improved systematically, due to the negative binding energy of the p^--like excited state. The variational QMC method was first used to calculate the binding energy of the D^- ground state, which compared well to the corresponding diffusion QMC results in both bulk and quantum wells. Then it was used to calculate the transition energy, of which the final state is the p^--like excited state of D^-. The variational QMC results are presented in Fig. 5 together with the corresponding experimental results for three quantum-well sizes (L_W=95, 194, and 373 Å). Open and solid squares are theoretical and experimental results for L_W=95 Å, whereas the circles and diamonds are for L_W=194 and 373 Å wells, respectively. In the calculation, the experi-

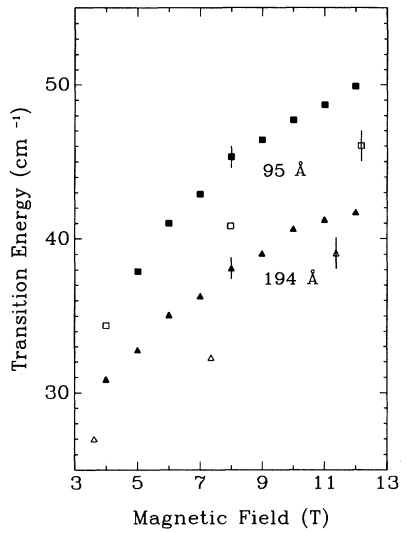

FIG. 4. The same as Fig. 3, but for the 95-Å and the 194-Å QW's only. The open symbols are the corresponding results of the diffusion QMC simulation (with statistical noise) after correction for band nonparabolicity.

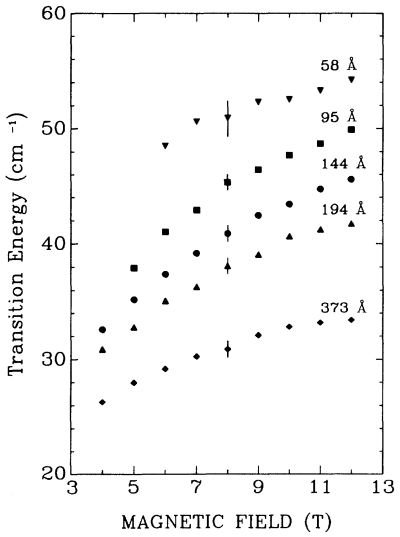

FIG. 3. The field dependence of the transition energy from the D^- ground state to the first excited p^--like singlet state for the five well widths indicated on the curves. This transition energy is obtained from the D^- CR energy minus the actual electron CR energy (Fig. 1). The small vertical bars are typical error bars.

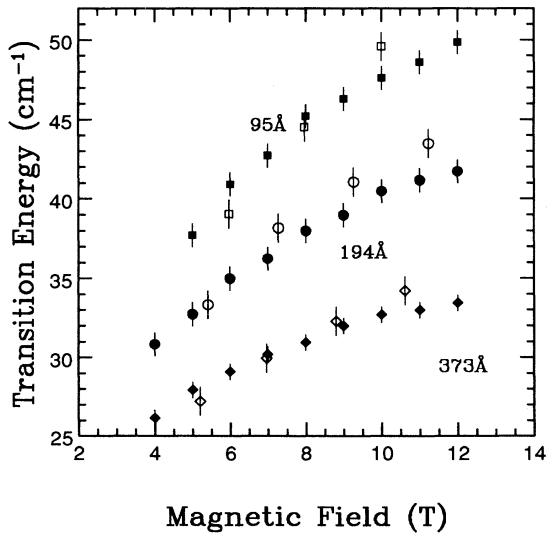

FIG. 5. The same as Fig. 3, but for the 95-Å, 194-Å, and 373-Å QW's only. The open symbols are the corresponding transition energies of the variational QMC calculation (with statistical noise). The final state is the p^--like excited state.

mentally determined cyclotron mass was used to include the band-nonparabolicity effects (see the Appendix). A good agreement is now found between the theoretical values and experimental results, although there are still noticeable deviations at very high fields. We attribute the deviations partly to polaron effects. In the strong-field regime, due to the electron-phonon (Fröhlich) interaction, the p^+ excited state begins to mix with the state corresponding to the D^- ground state plus a longitudinal optical phonon, lowering the D^- CR energy.[26] Our theoretical values are therefore above the experimental data in the high-field regime. An estimate of the energy shift due to the polaron effect gives values comparable to the observed energy differences between variational QMC results and experiment.[27] Thus, in the strong magnetic-field regime, polaron effects are important at the level of discrepancies shown in Fig. 5 and need to be taken into account for further improvement in agreement between theory and experiment.

IV. CONCLUSION

We have reported a study of D^- CR—i.e., the spin-singlet magneto-optical transition from the D^- ground state to the p^+-like excited state—in a series of well-characterized quantum wells with width ranging from 58 to 373 Å. The D^- CR energy is shown to depend weakly on the well width because band-nonparabolicity effects become important for narrow wells, masking part of the confinement effects. Complications due to band nonparabolicity are removed by subtracting the actual electron CR energy from the measured D^- CR energy, giving the transition energy from the D^- ground state to the p^--like first excited state. The latter transition energy is found to be significantly above the diffusion QMC predictions for the D^- binding energy. It is, however, found to be in good agreement with variational QMC predictions for this excitation energy. This shows unambiguously that the final states of spin-singlet magneto-optical transitions of D^- centers in QW's are unbound.

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APPENDIX: BAND-NONPARABOLICITY EFFECTS

A simple way to describe band-nonparabolicity effects on the D^0 energies is to rescale the calculated energies with $(m_\parallel^*/m_b^*)$ (from the mass dependence of the effective rydberg Ry*) and magnetic fields with $(m_\parallel^*/m_b^*)^2$ (from the mass dependence of the reduced magnetic field $\gamma=\hbar\omega_c/2$ Ry*). Here, m_b^* is the band-edge mass in GaAs and $m_\parallel^*$ is the parallel mass as defined by Ekenberg:[12]

$$m_\parallel^*(E_1)=m_b^*[1+(2\alpha+\beta)E_1]\ . \tag{A1}$$

In Eq. (A1), $\alpha=0.642$ eV^{-1} and $\beta=0.697$ eV^{-1} are the nonparabolic and anisotropic terms in the conduction band of GaAs. E_1 is the energy of the ground electronic level in the QW. The approach of Ekenberg has been shown to give a very good description of nonparabolicity effects on cyclotron mass in GaAs QW's.[11] Thus the above corrections should give a fairly good parametrization of nonparabolicity effects on the D^0 binding energy. In the above corrections, the weak magnetic-field dependence of the mass is neglected.

This weak magnetic-field dependence can be accounted for by replacing $m_\parallel^*$ by the measured cyclotron mass $m_{\rm CR}^*$. This was done to correct the D^- energies for band nonparabolicity. This is consistent with our determination of the transition energy in Figs. 3 from the energy of the D^- CR energy minus the electron CR energy. The differences between the use of either $m_\parallel^*$ or $m_{\rm CR}^*$ in the above corrections for the D^- energies are of the order of only 2% of the computed binding energies.

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$$\langle \psi_{2p^\pm}|H_e|\psi_{2p^\pm}\rangle=\frac{\pi m_w}{\alpha_{2p^\pm}}\int_{-w/2}^{w/2+b}dz\frac{f^2(z)}{m_e^*(z)}\sum_{n=-\infty}^{\infty}e^{-2\beta_{2p^\pm}(z-z_I+nl)^2}\left\{1+\left[\frac{\gamma}{4\alpha_{2p^\pm}}\right]^2\pm\frac{\gamma}{4\alpha_{2p^\pm}}+\tfrac{1}{2}A_{2p^\pm}^{(n)}(z)\right.$$
$$\left.+\tfrac{1}{2}B_{2p^\pm}^{(n)}(z)[\tfrac{1}{2}-2\alpha_{2p^\pm}(z-z_I+nl)^2]-\frac{m_e^*(z)}{m_W}|z-z_I+nl|\right\}.$$

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[22]In addition to spin conservation, the dipole-excited D^- transitions in Faraday geometry must obey the $\Delta M=\pm 1$ selection rule, where M is the D^- angular momentum along the field axis. The $\Delta M=+1$ and -1 D^- transitions are analogous to the $1s\rightarrow 2p^+$ and $1s\rightarrow 2p^-$ transitions for a neutral donor, respectively.
[23]A natural alternative labeling for the D^- transition B could refer to the p^+-like nature of the outer orbital in the final state. In our opinion, this is not really satisfactory since sharing a D^- orbital in an inner s-like and outer (s-like or p-like, depending on symmetry) is an artifact of variational calculations. Indeed, there is constant exchange between the two D^- electrons, so that it is not possible to distinguish between outer and inner orbitals. This is easily seen by looking at the D^- charge density defined by $\rho(r_1)=\int d\mathbf{r}_2|\Psi(\mathbf{r}_1,\mathbf{r}_2)|^2$, where $\Psi(\mathbf{r}_1,\mathbf{r}_2)$ is the two-electron D^- wave function. It is obvious that $\rho(r_1)=\rho(r_2)=\int d\mathbf{r}_1|\Psi(\mathbf{r}_2,\mathbf{r}_1)|^2$.
[24]The p^-- and p^+-like states are exactly separated by the cyclotron energy only for parabolic conduction bands. The analysis of our previous work shows that in a 100-Å GaAs QW, they are separated by the actual CR energy to a good approximation after correction for polaron effects in a high field (>11 T).
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