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ISOLATED AND "SCREENED" D^- CENTERS IN QUANTUM WELLS IN HIGH MAGNETIC FIELDS

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The spectra of magneto-optical transitions of D^- centers in GaAs/GaAlAs quantum wells (QWs) in high magnetic fields are considered. Excellent agreement with the experimental data of Huant et al. for the singlet D^- transitions is found; our predictions for the triplet D^- transitions can be directly experimentally tested. We discuss changes in the spectra which arise with increasing concentration of free electrons in QWs.

1. The observation of D^- centers (i.e. neutral shallow donors D^0 trapped a second electron) in modulation-doped GaAlAs/GaAs QWs in magnetic fields $B > 4$T by Huant et al. [1] has motivated much interest in these two-electron impurity-bound states (see Refs.[2--8]). In our previous work [6] we have presented the non-variational approach for calculation of D^- states and transition energies which incorporates consistently effects of GaAs conduction band non-parabolicity (NP) and magneto-polaron effects (the latter become increasingly important for fields $B > 10$T). In this Letter we present the relevant details of our scheme and report on (i) the binding energies of D^- singlet and triplet ground states; (ii) transition energies of D^- in wide QWs with mixing of different electric subbands taken into account. We shall also discuss the features observed in the recent experiments of Cheng *et al.* [8] on the effect of excess free electrons on magneto-optical tansitions associated with impurities.

2. In the effective mass approximation, the Hamiltonian of the problem has the form

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + \delta\hat{\mathcal{H}}_{\rm NP} - \frac{e^2}{\varepsilon r_1} - \frac{e^2}{\varepsilon r_2} + \frac{e^2}{\varepsilon|\mathbf{r}_1 - \mathbf{r}_2|} \qquad (1)$$

where the position of an electron is denoted by $\mathbf{r}_i = (\rho_i, z_i)$ and an impurity is assumed to be at the center of a QW. The Hamiltonian of free electrons in a QW in a perpendicular magnetic field B is given by

$$\hat{\mathcal{H}}_0 = \sum_{i=1,2} \hat{\mathcal{H}}_0^{(i)} = \sum_{i=1,2} \left\{ \frac{1}{2m^*} \left(\hat{\mathbf{p}}_i + \frac{e}{c}\mathbf{A}_i\right)^2 + V(z_i) + \frac{1}{2} g^* \mu_{\rm B} B \sigma_{zi} \right\} \qquad (2)$$

where the confining potential is taken to be $V(z) = V_0$ for $|z| > d/2$ and $V(z) = 0$ for $|z| < d/2$; we found that different existing approximations for dependence of V_0 on x in $\mathrm{Ga}_{1-x}\mathrm{Al}_x\mathrm{As/GaAs}$ QWs give practically the same results for the D^- interaction energies. The term $\delta\hat{\mathcal{H}}_{\rm NP} = \sum_{i=1,2} \delta\hat{\mathcal{H}}_{\rm NP}^{(i)}$ describes GaAs conduction

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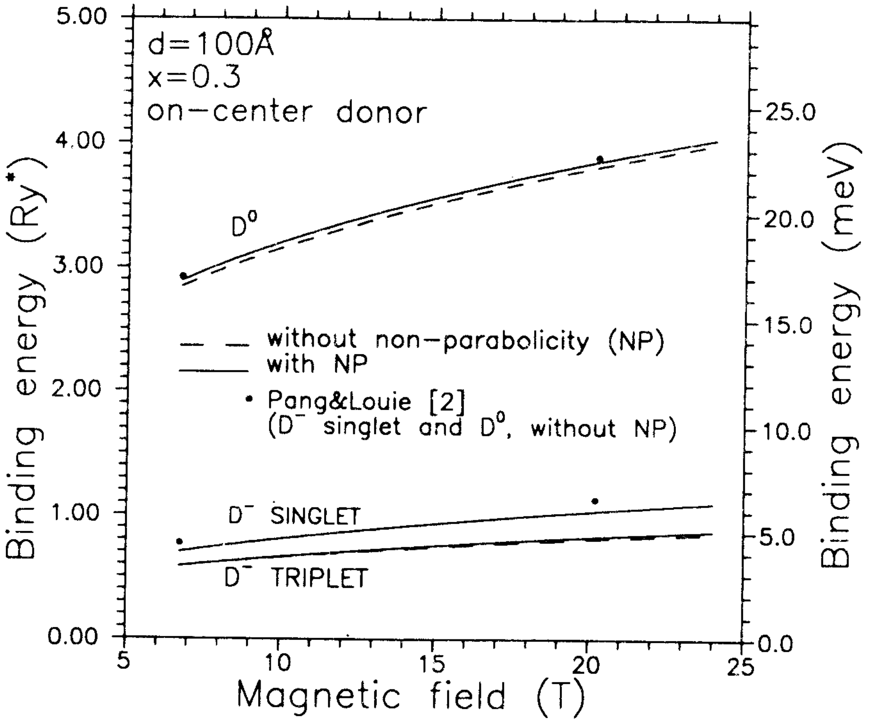

Fig.1. The binding energies (in units of the effective Rydberg Ry* = 5.83meV) of the neutral donor D^0 and the D^- center in Ga$_{0.7}$Al$_{0.3}$As/GaAs QW of the width $d = 100$ Å with (solid curves) and without (dashed curves) corrections owing to band non-parabolicity (NP); for the singlet D^- state the corrections are negligible.

T± (and the corresponding final states as $|1,1;\mathrm{T}\pm\rangle$). In available magnetic fields $B < 80$T, due to the low values of the g^*-factor in GaAs/GaAlAs QWs, the D^- singlet $|0,0;\mathrm{S}\rangle$ is still the ground state, hence the triplet D^- p-ground state is depopulated at low temperatures. Besides, the transition matrix elements for the triplet are by nearly two times smaller than that of the singlet (Fig.2). Nevertheless, the calculated relative intensities $R_\pm = I_T^\pm/I_S$ of the two strong triplet T± transitions to that of the singlet at elevated temperatures T≃ 10K turn out to be ~ 0.5 (while at T=4K $R_\pm \sim 0.1$) [6]. Hence, these transitions can be experimentally tested; note, however, that the T+ transition is masked by the strong $1s \longrightarrow 2p_+$ transition of D^0 in a QW which occurs in the same spectral region (see Fig.2).

5. Magneto-polaron effects lead to a shift of energies of donors in intermediate magnetic fields and give rise to a resonant splitting in high magnetic fields $B > 15$T (e.g. [12,13]). Here we present the results for the polaron corrections to the energies of quasi-2D D^- centers, i.e. the bound magneto-bipolarons. Because GaAs is a weak polar material (the electron-phonon coupling $\alpha = 0.068$), we shall use the two second-order perturbation theory approaches: (i) Rayleigh-Shrödinger (RS) for the ground states and Wigner-Brillouin (WB) for the excited states (RS-WB), and (ii) the improved Wigner-Brillouin (IWB) which ensures pinning behavior at $\hbar\omega_{LO}$

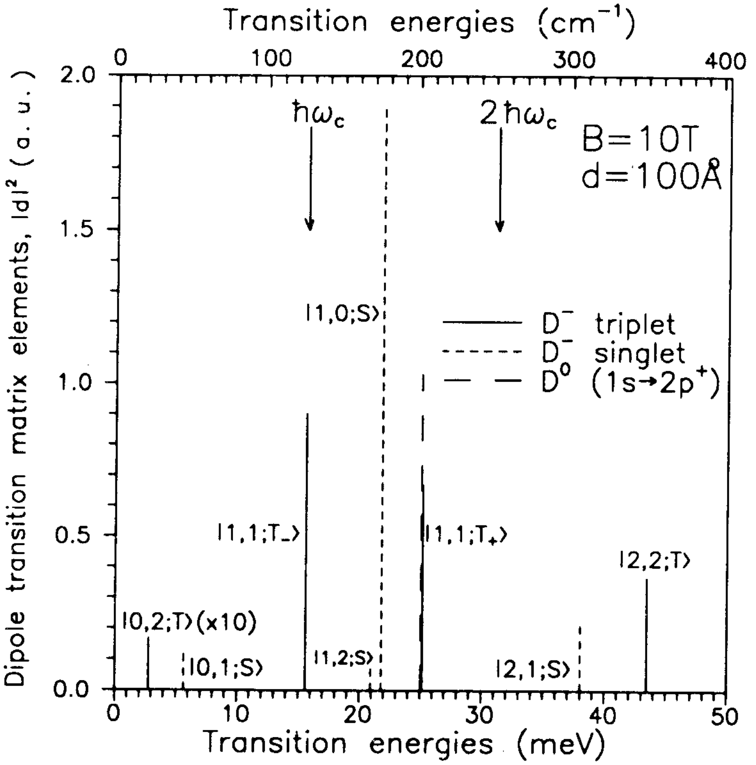

Fig.2. Dipole transition matrix elements $|d|^2$ and the energies corresponding to the $1s \rightarrow 2p_+$ $[n = m = 0 \rightarrow n = 1,\ m = 0]$ D^0 transition and to the transitions from the singlet $|0,0;\mathrm{S}\rangle$ and the triplet $|0,1;\mathrm{T}\rangle$ D^- ground states; quantum numbers of the final D^- states are explicitly shown. Transitions with $\Delta M = 0$ are strong and correspond to the left circular polarization σ^-; transitions with $\Delta M \neq 0$ are weak in a strong magnetic field as $[(e^2/\epsilon r_H)/\hbar\omega_C]^2 \sim B^{-1}$. The positions of the cyclotron resonance $\hbar\omega_C$ and its multiple $2\hbar\omega_C$ are shown by arrows.

(see [13] and references therein). For the polaron correction to the energy of the ith D^- state, ΔE_i, one has

$$\Delta E_i = \sum_{\mathrm{q}} \sum_{j} \frac{|\langle j;\mathrm{q}|H_{e-ph}|i;0\rangle|^2}{E_i^0 - E_j^0 - \hbar\omega_{LO} + \Delta E_i'}, \tag{4}$$

where

$$H_{\text{e-ph}} = \sum_{i=1,2} \frac{1}{\mathrm{V}} \sum_{\mathrm{q}} \left(V_{\mathrm{q}} \exp(i\mathrm{q}\mathrm{r}_i)\, b_{\mathrm{q}} + \mathrm{H.c.}\right), \tag{5}$$

$$|V_{\mathrm{q}}|^2 = 4\pi\alpha \left(\frac{\hbar}{2m^*\omega_{LO}}\right)^{1/2} \left(\frac{\hbar\omega_{LO}}{q}\right)^2 \tag{6}$$

is the Hamiltonian of the Fröhlich interactions with bulk dispersionless LO phonons and $|j;\mathrm{q}\rangle$ denotes a D^- in the jth state with the unperturbed energy E_j^0 plus LO phonon of momentum q and the energy $\hbar\omega_{LO} = 36.25\,\mathrm{meV}$. The form of $\Delta E_i'$ in Eq.4 depends on the choice of a perturbation approach. For, e.g. RS (WB) perturbation theory $\Delta E_i' = 0(\Delta E_i)$. Here we consider the corrections to the ground singlet and triplet D^- states ($|i\rangle = |0,0;\mathrm{S}\rangle$, $|0,1;\mathrm{T}\rangle$) and to the states which are the

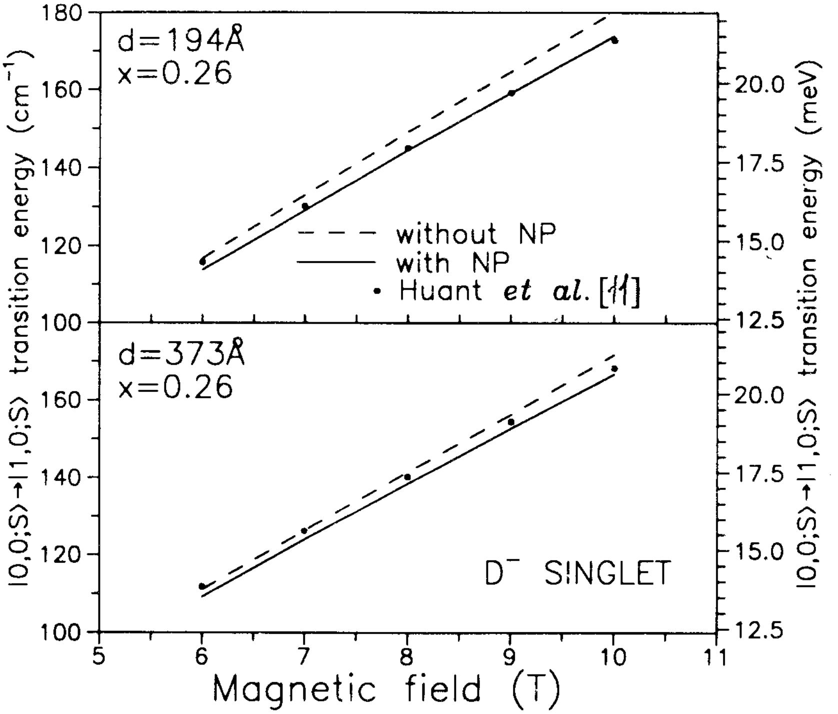

Fig.3. Transition energies from the ground singlet D^- state for two QWs with $d = 194$ Å and $d = 373$ Å versus B. Dots are the experimental data of Huant et al. [11].

final states of the strong magneto-optical transitions: $|i\rangle = |1,0;\mathrm{S}\rangle$ for the singlet and $|i\rangle = |1,1;\mathrm{T}\pm\rangle$ for the triplet (see Fig.2). To achieve reasonable accuracy, we include in the sum of Eq.4 from 5 to 7 relevant states of D^- which were obtained in the previous step of our calculations. Polaron corrections to the energies of D^- states turn out to be large ($\sim 4\alpha$). For the D^- singlet ground state at $B = 20\mathrm{T}$ $\Delta E_{00S} = -5.28$ meV which exceeds the correction to the D^0 ground state ($\sim \alpha$) by nearly four times, as it should be for a bound bipolaron [6]. It turns out that at fields $B < 10\mathrm{T}$ corrections to different D^- states strongly compensate each other, and transition energies are altered modestly (Fig.4). For the $\Delta N = 1$ singlet transition energy we find that in the high field region of resonant splittings the IWB perturbation theory gives the results which are below the experimental values (at $B = 16.6\mathrm{T}$ by $\simeq 3\%$), i.e. it overestimates polaron corrections, while the RS-WB approach (not shown) underestimates them (at $B = 16.6\mathrm{T}$ by $\simeq 4\%$). Our predictions for the magneto-polaron effects on the triplet D^- transitions need experimental verification.

6. The essential point in our consideration is that due to a complete discretness of quasi-2D D^- spectra in quantizing magnetic fields, the binding and transition energies differ considerably [3,4] and should not be confused. Our results differ much in this point from the work of Shi *et al.* [7]. These authors also incorporated into their variational calculations magneto-polaron corrections. It should be pointed out here that (i) variational calculations [7] provide rather poor accuracy; in comparison

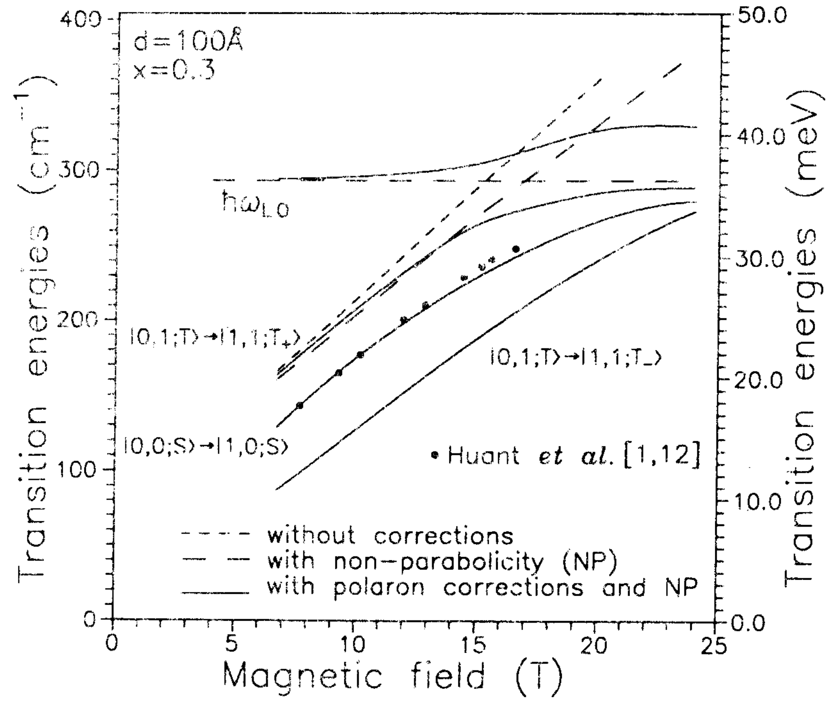

Fig.4. Transition energies from the ground singlet and triplet D^- states in the QW with $d = 100$ Å and $x = 0.3$. For the triplet T_+ transition the energies without corrections (dashed line), with NP but without polaron effects (long-dashed line) and one upper polaron branch (solid line) are also shown.

with [2] the value for the singlet D^- ground state E_b is underestimated by more than 30% (see Fig.3 of Ref. [7]); (ii) taking the final state of a magneto-optical transition as a neutral donor and a free electron, in [7] the polaron correction to its energy turns out to be $\sim 2\alpha$ and its value is underestimated by a factor of two. Thus, the quantitative agreement with the experiments [1] claimed in [7] appears to be fortuitous due to the cancellations following from (i), (ii) and a misinterpretation of D^- transitions.

7. Let us discuss the features observed by Cheng *et al.* [8] in the magneto-optical spectra associated with the impurity transitions when the concentartion of excess free electrons n_{ex} in QWs is varied from $2\times10^{10}\,\mathrm{cm}^{-2}$ to $2.8\times10^{11}\,\mathrm{cm}^{-2}$. First, a broad shoulder appearing at $n_{ex} > 8\times10^{10}\,\mathrm{cm}^{-2}$ ($\nu = n_{ex}hc/eB > 0.4$ at $B = 9\mathrm{T}$) on the low-energy side of the cyclotron resonance near $\hbar\omega_C$ we assign (see Fig. 2) to the transition evolving from the triplet T− transition (or, in a single-particle picture, from the $n = 0$, $m = 1 \rightarrow n = m = 1$ $[2p_- \rightarrow 2s]$ impurity transition).

Another feature observed in [8] is that with increasing n_{ex} the transition associated with the singlet D^- is shifted to *higher* energies. This shift is considerable only for $\nu > 0.3$ [8], when the spatial extent of the D^- wavefunctions is comaparable with the mean distance between free electrons. Hence, one should abandon a picture of D^- states and rather speak in terms of collective magnetoplasma excitations localized at (essentially isolated) Coulomb impurity. As is shown by Dzyubenko and Lozovik [14], who considered for $\nu = 1, 2$ a strictly 2D limit neglecting virtual transitions between LLs, the two infrared-active impurity-localized magnetoplasma modes exist in the spectrum. One is lying below $\hbar\omega_C$ and the other above $\hbar\omega_C$ near the upper free magnetoplasma band edge. Due to the exchange effects the energy positions of the impurity-localized collective modes are shifted to higher energies relative to the transitions of D^- from which they are evolved with increasing ν. At $\nu = 1$ the shifts for the lower (upper) mode $\delta_\nu^\mp$ in units of $e^2/\epsilon r_H$ are given by $\delta^-_{\nu=1} = 0.20$, and $\delta^+_{\nu=1} = 0.19$. At $\nu = 2$, owing to the increased exchange effects, the shifts are larger: $\delta^-_{\nu=2} = 0.29$, and $\delta^+_{\nu=2} = 0.42$. The experimental value for the shift at $B = 9\,\mathrm{T}$ and $n_{ex} = 2.8 \times 10^{11}\,\mathrm{cm}^{-2}$ ($\nu = 1.3$) of $13\,\mathrm{cm}^{-1}$ [8] in units of $e^2/\epsilon r_H$ gives $\delta^+_{\nu=1.3} = 0.10$. Agreement between the strictly-2D theory and the experiment can be improved, at least partially, by taking into account mixing between LLs and quasi-2D effects (which lower interaction energies). Besides, due to the presence in the same spectral region of a branch of free magnetoplasma excitations (giving zeros of the dynamical dielectric function $\varepsilon(\mathrm{k}, \omega)$), the effects of dynamical screening on the energies of impurity optical transitions should be very important.

We conclude by noting that: (i) the picture of impurity-localized magnetoplasma modes is consistent with the abrupt slope changes at integer ν observed in [8]. It also predicts the discontinuities in the transition energies at integer ν; (ii) since in the singlet and triplet T+ transitions the final D^- states are unbound and more extended than the initial ones [4], a simple picture of screened D^- states (valid when $\nu \ll 1$) also predicts a shift to *higher* transition energies.

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