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 tested directly experimentally. The changes in the spectra which arise with increasing concentration of free electrons in QWs are discussed.

1. The observation of D^- centers (i.e., neutral shallow donors D^0 trapped by a second electron) in modulation-doped GaAlAs/GaAs QWs in magnetic fields, $B>4$ T, by Huant *et al.*[1] has motivated considerable interest in these two-electron impurity-bound states (see Refs. 2–8). In our previous work,[6] we have considered a nonvariational approach for calculation of D^- states and transition energies, which incorporates consistently the effects of GaAs conduction band nonparabolicity (NP) and magneto-polaron effects (the latter become increasingly important in 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 allowance for the mixing of different electric subbands. 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 transitions associated with impurities.

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

$$\hat{\mathscr{H}}=\hat{\mathscr{H}}_0+\delta\hat{\mathscr{H}}_{\rm NP}-\frac{e^2}{\varepsilon r_1}-\frac{e^2}{\varepsilon r_2}+\frac{e^2}{\varepsilon|\mathbf{r}_1-\mathbf{r}_2|}, \tag{1}$$

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

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

where the confining potential is assumed 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 the dependence of V_0 on x in $Ga_{1-x}Al_xAs/GaAs$ QWs give nearly the same results for the D^- interac-

tion energies. The term $\delta\hat{\mathscr{H}}_{\rm NP}=\Sigma_{i=1,2}\delta\hat{\mathscr{H}}_{\rm NP}^{(i)}$ describes GaAs conduction band NP; for $\delta\hat{\mathscr{H}}_{\rm NP}^{(i)}$ we use Ekenberg's[9] analytical expression for the Landau level (LL) energies in QWs.

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

where n is a LL number, $r_H=(\hbar c/eB)^{1/2}$ [cf. Eq. (35) of Ref. 9, which we have slightly corrected by the proper symmetrization of kinematic momentum operators in the initial Hamiltonian]; in $\delta\hat{\mathscr{H}}_{\rm NP}$ the terms describing the spin–orbit splitting and the terms breaking the angular symmetry are small and have therefore been eliminated in (3). The calculated corrections to the cyclotron energy $\hbar\omega_c$ (e.g., -1.56 meV for 100 Å QW at $B=10$ T) which follow from Ekenberg's approach are in very good agreement with the experimentally determined values[1] (see also Refs. 6 and 10).

We construct the wave functions of D^- with the total angular momentum projection M_z, $\Psi_{M_z}(\mathbf{r}_1,\mathbf{r}_2)$, from the noninteracting eigenfunctions of the free-electron Hamiltonian $\hat{\mathscr{H}}_0^{(i)}(2)\zeta_j(z_i)\phi_{n,m}(\rho_i)$ (see Refs. 4 and 6 for details) as symmetrized (for the singlet, S) or antisymmetrized (for the triplet, T) combinations involving the states with $n_1+n_2-m_1-m_2=M_z$. We classify D^- states by the high-field quantum numbers[4] $|N,M;\mathrm{S(T)}\rangle$ (and, when necessary, by an additional index which distinguishes the states within the group $\{N,M\}$); here $N=n_1+n_2$ and $M=m_1+m_2=N-M_z$ is the total oscillator quantum number. The states from the groups $\{N,M\}$ and $\{M,N\}$ are $t\to -t$ counterparts and, in the absence of NP, they differ in energies exactly by[4,6] $(N-M)\hbar\omega_c=M_z\hbar\omega_c$.

3. We consider the high magnetic field regime $r_H\ll a_B=\varepsilon\hbar^2/m^*e^2$ ($B>6$ T) and truncate the basis set by including only several lowest LLs ($n_1+n_2<3$–7, depending on B) and several lowest subbands $\zeta_j(z)$ (depending on the well width d). To check the accuracy of our approach, we calculated the binding energy of a more simple system, D^0, and compared our results with high-accuracy quantum-diffusion Monte Carlo results of Pang and Louie[2] for the 100 Å QW; the relative accuracy turns out to be $\sim 1.5\%$ (see Fig. 1). The calculated binding energies E_b of the singlet s-ground state $|0,0;\mathrm{S}\rangle$ and of the triplet p-ground state $|0,1;\mathrm{T}\rangle$ are also shown in Fig. 1. At $B=20$ T, for the singlet s-ground state the calculated E_b is 8% lower than that obtained by Pang and Louie.[2] It is important to stress that errors in the interaction energies are systematic in our approach, and that they cancel each other, so that the accuracy of D^- *transition* energies is much higher.[6] It should be also noted that the effect of NP on the *binding* energies of the D^0 and D^- states was found to be small within this consistent approach (cf. Ref. 2; see also Ref. 10).

4. The calculated energies and dipole matrix elements of the D^0 and D^- magneto-optical transitions with $\Delta M_z=\pm 1$ which are allowed in the Faraday geometry, are shown in Fig. 2 for the 100 Å $Ga_{0.7}Al_{0.3}As/GaAs$ QW (see also Fig. 4) and the energies of the singlet D^- transitions for two wider 194 Å and 373 Å $Ga_{0.74}Al_{0.26}As/GaAs$ QWs are shown in Fig. 3. As a result of the $t\to -t$ symmetry, from the singlet ground state $|0,0;\mathrm{S}\rangle$ (as well as from all s-states of D^0 and $|N,N;\mathrm{S(T)}\rangle$ s-states of D^-) there is a *pair* of transitions separated in energy by $\hbar\omega_c$ (see also Refs. 3, 4, and

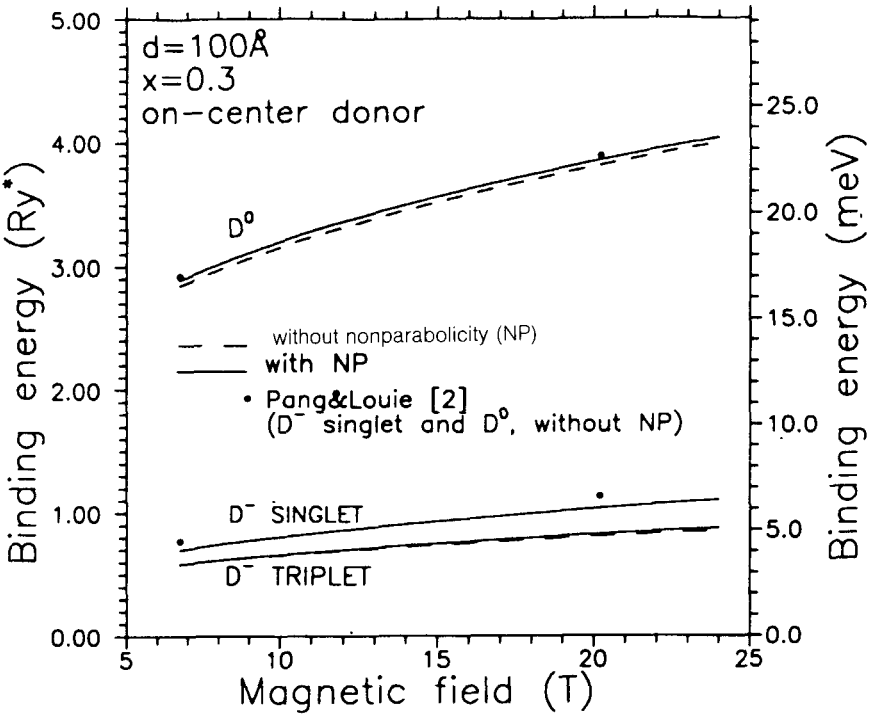

FIG. 1. The binding energies (in units of the effective Rydberg Ry* = 5.83 meV) of the neutral donor D^0 and the D^- center in $Ga_{0.7}Al_{0.3}As/GaAs$ QW of width $d = 100$ Å with corrections (solid curves) and without corrections (dashed curves) due to the band nonparabolicity (NP); for the singlet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

6). In each pair the transition with $\Delta M_z = 1$ induced by the radiation with the left circular polarization σ^- is stronger. Our results for the singlet D^- transitions are in excellent agreement with the experiments.[1,11]

From the triplet p-ground state $|0,1;T\rangle$ there are *two* strong transitions which turn out to be in different spectral regions.[4] We denote these transitions by T± (and the corresponding final states as $|1,1;T\pm\rangle$). In available magnetic fields $B < 80$ T, because of the low values of the g^*-factor in GaAs/GaAlAs QWs, the D^- singlet $|0,0;S\rangle$ is still the ground state; hence the triplet D^- p-ground state is depopulated at low temperatures. Furthermore, the transition matrix elements for the triplet are nearly twice as small as those 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 the singlet at elevated temperatures, $T \simeq 10$ K, turn out to be ~ 0.5 (while at $T = 4$ K we have $R_\pm \sim 0.1$).[6] Hence, these transitions can be experimentally tested; note, however, that the T+ transition is masked by the strong $1s \to 2p_+$ 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 in the energies of donors in intermediate

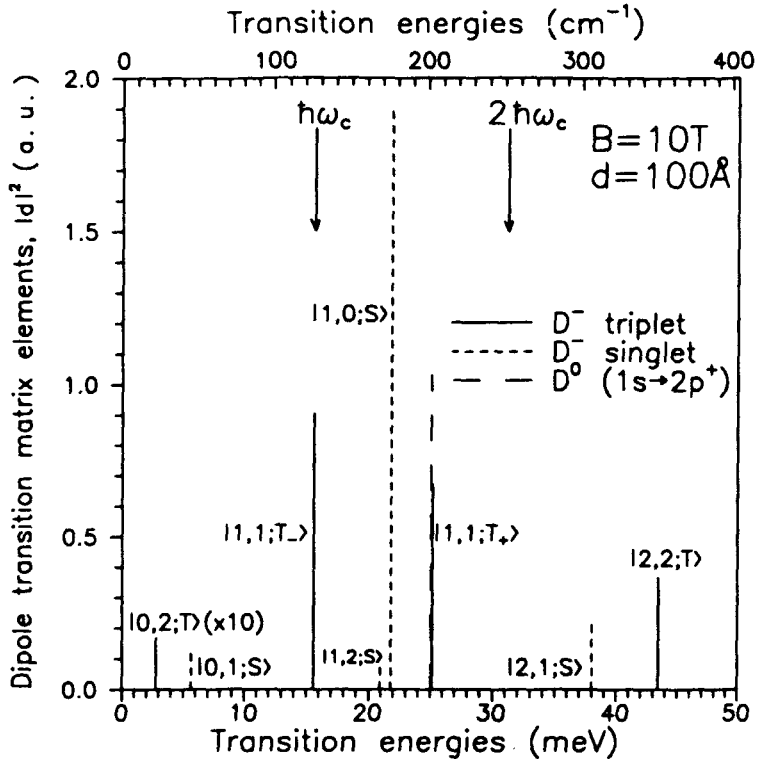

FIG. 2. Dipole transition matrix elements $|d|^2$ and the energies corresponding to the $1s\rightarrow 2p+$ $[n=m=0\rightarrow n=1,\ m=0]$ D^0 transition and to the transitions from the singlet $|0,0;S\rangle$ and the triplet $|0,1;T\rangle$ D^- ground states; quantum numbers of the final D^- states 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 since $[(e^2/\varepsilon\tau_H)/\hbar\omega_c]^2\sim B^{-1}$. The positions of the cyclotron resonance $\hbar\omega_c$ and its multiple $2\hbar\omega_c$ are indicated by the arrows.

magnetic fields and give rise to a resonant splitting in high magnetic fields, $B>15$ T (e.g., Refs. 12 and 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) the Rayleigh–Schrödinger (RS) approach for the ground states and the Wigner–Brillouin (WB) approach for the excited states (RS–WB), and (ii) the improved Wigner–Brillouin (IWB) approach, which ensures a pinning behavior at $\hbar\omega_{LO}$ (see Ref. 13 and the references cited there). For the polaron correction to the energy of the ith D^- state, ΔE_i,

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

where

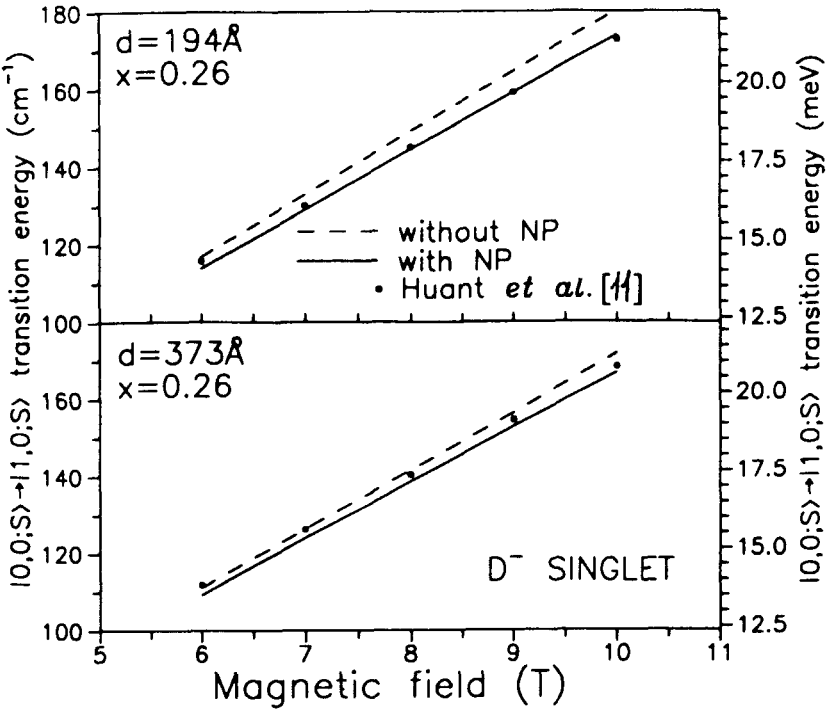

FIG. 3. Transition energies from the ground singlet D^- state for two QWs with $d=194$ Å and $d=373$ Å versus B. Dots are the experimental data of Huant *et al.*[11]

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

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

is the Hamiltonian of the Fröhlich interactions with the bulk dispersionless LO phonons, and $|j;\mathbf{q}\rangle$ denotes a D^- in the jth state with the unperturbed energy E_j^0 plus a LO phonon of momentum $\mathbf{q}$, and the energy $\hbar\omega_{LO}=36.25$ meV. The form of $\Delta E_i'$ in Eq. 4 depends on the choice of a perturbation approach. For the RS (WB) perturbation theory, for example, $\Delta E_i'=0(\Delta E_i)$. Here we consider the corrections to the ground singlet and triplet D^- states ($|i\rangle=|0,0;S\rangle$, $|0,1;T\rangle$) and to the states which are the final states of the strong magneto-optical transitions: $|i\rangle=|1,0;S\rangle$ for the singlet state and $|i\rangle=|1,1;T\pm\rangle$ for the triplet state (see Fig. 2). To achieve reasonable accuracy, we include in the sum of Eq. 4 from five to seven relevant states of D^- which were obtained in the previous step of the calculations. The polaron corrections to the energies of D^- states turn out to be large ($\sim 4\alpha$). For the D^- singlet ground state at $B=20$ T $\Delta E_{00S}=-5.28$ meV, which exceeds the correction to the D^0 ground state ($\sim\alpha$) by nearly a factor of four, as it should be for a bound bipolaron.[6] It turns

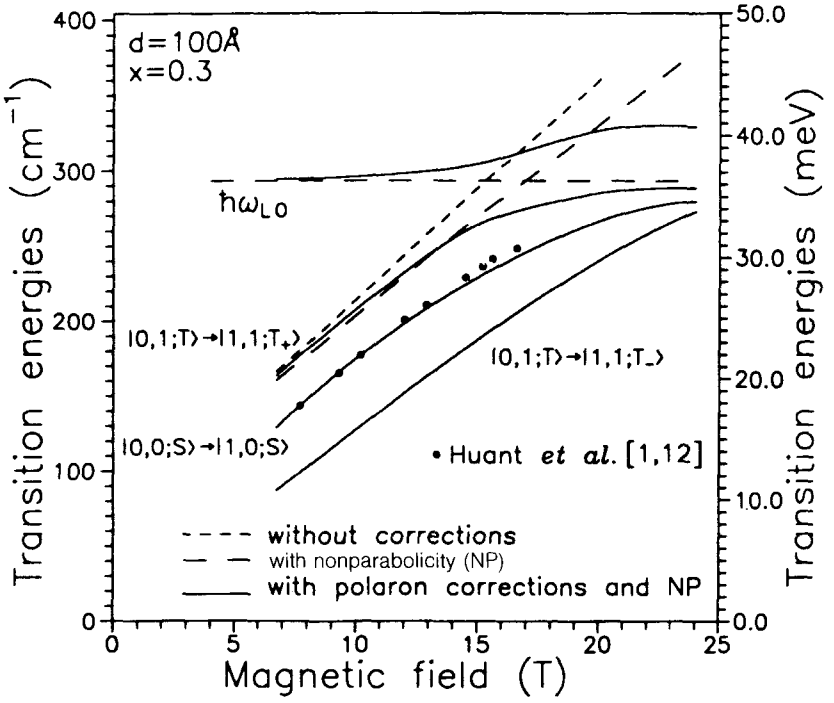

FIG. 4. Transition energies from the ground singlet and triplet D^- states in the QW with $d=100$ Å and $x=0.3$. For the triplet T_+ transition the energies without corrections (dashed line), with NP but without polaron effects (long-dashed line) and one upper polaron branch (solid line) are also shown.

out that at fields $B<10$ T the corrections to different D^- states fully cancel each other, and the transition energies are altered modestly (Fig. 4). For the $\Delta N=1$ single transition energy we find that in the high-field region of resonant splittings the IWB perturbation theory gives results which are below experimental values (at $B=16.6$ T by $\simeq 3\%$); i.e., it overestimates the polaron corrections, while the RS–WB approach (not shown) underestimates them (at $B=16.6$ 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 because of the complete discreteness 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 considerably in this respect from the work of Shi *et al.*[7] Those authors incorporated the magneto-polaron corrections into their variational calculations. It should be pointed out that (i) variational calculations[7] provide rather poor accuracy; in comparison with Ref. 2, the value for the singlet D^- ground state E_b is underestimated by more than 30% (see Fig. 3 in Ref. 7); (ii) assuming the final state of a magneto-optical transition to be a neutral donor and a free electron, the polaron correction to its energy in Ref. 7 turns out to be $\sim 2\alpha$ and its value is underestimated by a factor of 2. Thus, the quantitative agreement with the experiments[1] claimed in Ref. 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 concentration of excess free electrons n_{ex} in QWs is varied from 2×10^{10} cm^{-2} to 2.8×10^{11} cm^{-2}. First, a broad shoulder appearing at $n_{ex}>8\times10^{10}$ cm^{-2} ($\upsilon=n_{ex}hc/eB>0.4$ at $B=9$ T) on the low-energy side of the cyclotron resonance near $\hbar\omega_c$ can be assigned (see Fig. 2) to the transition evolving from the triplet T transition (or, in a single-particle picture, from the $n=0$, $m=1\to n=m=1$ [$2p_-\to 2s$] impurity transition).

Another feature observed in Ref. 8 is that, with an increase in n_{ex}, the transition associated with the singlet D^- shifts toward *higher* energies. This shift is considerable only for $\upsilon>0.3$ (Ref. 8), when the spatial extent of the D^- wave functions is comparable with the mean distance between free electrons. Hence, one should abandon a picture of D^- states and consider the problem instead in terms of collective magnetoplasma excitations localized at a (essentially isolated) Coulomb impurity. As was shown by Dzyubenko and Lozovik,[14] who considered for $\upsilon=1$, 2 a strictly 2D limit, ignoring virtual transitions between LLs, the two infrared-active impurity-localized magnetoplasma modes exist in the spectrum. One lies below $\hbar\omega_c$ and the other above $\hbar\omega_c$ near the upper free magnetoplasma band edge. Because of 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 evolve with an increase in υ. At $\upsilon=1$, the shifts for the lower (upper) mode, $\delta_\upsilon^\mp$ in units of $e^2/\varepsilon r_H$, are given by $\delta_{\upsilon=1}^-=0.20$, and $\delta_{\upsilon=1}^+=0.19$. At $\upsilon=2$, because of the increase in the exchange effects, the shifts are larger: $\delta_{\upsilon=2}^-=0.29$, and $\delta_{\upsilon=2}^+=0.42$. The experimental value for the shift at $B=9$ T and $n_{ex}=2.8\times10^{11}$ cm^{-2} ($\upsilon=1.3$) of 13 cm^{-1} (Ref. 8) in units of $e^2/\varepsilon r_H$, gives $\delta_{\upsilon=1,3}^+=0.10$. Agreement between the strictly 2D theory and the experiment can be improved, at least partially, by taking into account the mixing between LLs and quasi-2D effects (which lowers the interaction energies). Furthermore, the presence in the same spectral region of a branch of free magnetoplasma excitations [which accounts for the zeros of the dynamic dielectric function $\varepsilon(\mathbf{k},\omega)$], should make the effects of dynamic screening on the energies of impurity optical transitions very important.

We conclude by noting that: (i) the picture of impurity-localized magnetoplasma modes is consistent with the abrupt slope changes at integer υ observed in Ref. 8. It also predicts the discontinuities in the transition energies at integer υ; (ii) since in the singlet and triplet T+ transitions the final D^- states are not bound and are extended more than the initial states,[4] a simple picture of screened D^- states (valid when $\upsilon\ll1$) also predicts a shift to higher transition energies.

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