Negatively charged excitons and photoluminescence in asymmetric quantum wells

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We study photoluminescence (PL) of charged excitons (X^-) in narrow asymmetric quantum wells in high magnetic fields B. The binding of all X^- states strongly depends on the separation δ of electron and hole layers. The most sensitive is the ''bright'' singlet, whose binding energy decreases quickly with increasing δ even at relatively small B. As a result, the value of B at which the singlet-triplet crossing occurs in the X^- spectrum also depends on δ, and decreases from 35 T in a symmetric 10 nm GaAs well to 16 T for $\delta = 0.5$ nm. Since the critical values of δ at which different X^- states unbind are surprisingly small compared to the well width, the observation of strongly bound X^- states in an experimental PL spectrum implies virtually no layer displacement in the sample. This casts doubt on the interpretation of PL spectra of heterojunctions in terms of X^- recombination.

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I. INTRODUCTION

The optical properties of a quasi-two-dimensional electron gas (2DEG) in a high magnetic field B have been widely studied both experimentally[1–15] and theoretically.[16–33] The 2DEG is usually realized in semiconductor quantum wells (QW's) or heterojunctions (HJ's). In QW's, where electrons (e) and valence holes (h) are confined in the same 2D layer, the photoluminescence (PL) spectrum shows emission from the radiative states of neutral ($X=e+h$) and charged ($X^- = 2e+h$) excitons interacting with one another and with free electrons.

The existence of a bound X^- complex was first predicted by Lampert[16] in bulk semiconductors; however, it could not be observed experimentally because of the small binding energy Δ. It was later shown by Stebe and Ainane[17] that the X^- binding is significantly enhanced in 2D systems. Indeed, an X^- state with a Δ of about 3 meV was detected by Kheng *et al.*[2] in a CdTe QW. Subsequent extensive experimental[3–12] and theoretical[20–25] studies established that X^- occurs in the form of a number of different bound states. The state observed by Kheng *et al.* was the singlet, X_s^-, whose total electron spin J is zero. This is the only bound X^- state in the absence of a magnetic field.

MacDonald and Rezayi[18] showed that the decoupling of optically active excitons from electrons in the lowest Landau level (LL) due to the ''hidden symmetry''[34,35] causes an unbinding of X_s^- (and other optically active complexes larger than X) for $B\to\infty$. However, a different bound X^- state exists in this limit. It is a triplet X_{td}^-, with $J=1$ and finite angular momentum $\mathcal{L}=-1$.[20] Since both the hidden symmetry[21,34,35] and the angular momentum conservation[25–28] independently forbid recombination of an isolated X_{td}^- in the lowest LL, its optical lifetime τ_{td} in high magnetic fields is expected to be long, and is determined by scattering and/or disorder.

The fact that the X_{td}^- binding energy Δ_{td} decreases with decreasing B implies a singlet-triplet crossing in the X^- spectrum at a certain B, estimated[23,25] at about 35 T for a 10-nm GaAs QW. Although PL experiments in high magnetic fields indeed showed emission from a pair of X^- states,[4–8] the crossing was not found,[6] nor was the intensity τ^{-1} of the peak assigned to the triplet seen to decrease with increasing B or decreasing electron density. This apparent discrepancy between theory and experiment was recently resolved by a numerical discovery[25] of yet another X^- state: a ''bright'' triplet X_{tb}^-. The X_{tb}^- state has $\mathcal{L}=0$ and $J=1$, a large oscillator strength τ_{tb}^{-1}, and a small binding energy Δ_{tb}, and occurs in high magnetic fields in QW's of finite width.

While the identification of the experimentally observed triplet as the X_{tb}^- state explains its small binding energy, the fact that the more strongly bound X_{td}^- state is not observed confirms its very long optical lifetime τ_{td}. The reason why τ_{td} remains large in the presence of surrounding electrons (although the e-X^- scattering breaks the $\mathcal{L}=0$ selection rule for an isolated X^-) is the short range of e-X^- repulsion, which causes Laughlin e-X^- correlations[36,37] and the effective isolation of all X^- states from the 2DEG.[25] These correlations are also responsible for the insensitivity of the PL spectra of QW's to the electron density, and for the success of its description in terms of the X^- quasiparticles and their single-particle properties such as binding energy Δ, PL energy ω, or oscillator strength τ^{-1}.

The major difficulty in comparing the numerical and experimental data is that most experiments are carried out in asymmetrically doped QW's[4–8] or HJ's,[10,11] in which an electric field perpendicular to the 2DEG modifies confinement and, among other effects, leads to a displacement of electron and hole layers. This displacement or separation between the electron and hole layers has been ignored in the existing realistic calculations (those which take into account

the finite widths of the electron and hole layers, LL mixing, etc.),[23–25] although from more idealized calculations (zero-layer widths and no LL mixing)[33] it can be expected to weaken the X^- binding, possibly in a different manner for different X^- states.

In this paper we incorporate the finite electron-hole layer displacement δ into the model used earlier[25] to study X^- states in narrow symmetric QW's. The displacement δ and a pair of widths w_e^* and w_h^* of electron and hole layers make three independent effective parameters of our model. Although a finite δ is intended to describe mainly the polarizing effect of an electric field in asymmetric QW's or HJ's, it must be kept in mind that in reality the electric field not only causes displacement of layers, but modifies their shape and width as well.[38] On the other hand, our effective parameter δ is not equivalent to the bare displacement of single-electron and hole wave functions caused by the electric field only. Rather, δ measures the actual average electron-hole separation in the direction perpendicular to the layers (along the z axis) *within a bound state*, and thus (indirectly) also depends on the correlations in the z direction, details of the confining potentials, etc. Despite the simplicity and the phenomenological character of this model, dictated by a limited knowledge of the exact single-electron and hole wave functions and their dependence on a particular sample, we were able to obtain results that, in connection with the experimental PL spectra of asymmetric QW's or HJ's, add to our general understanding of X^- states in these structures.

Using exact numerical diagonalization in Haldane's spherical geometry,[39] we examine the dependence of binding energies of all different bound X^- states on both the magnetic field and the displacement δ. In addition to the bright singlet X_s^- (denoted here by X_{sb}^-) and two triplets, X_{td}^- and X_{tb}^-, we identify a dark singlet X_{sd}^- with an angular momentum $\mathcal{L}=-2$ which occurs at $\delta>0$, in analogy to a known[40,41] D^- (charged donor) state at the same $\mathcal{L}$. We demonstrate that the binding energies of all X^- states depend strongly on δ. Most sensitive is the X_{sb}^- state, which unbinds when δ reaches merely 5–10 % of the QW width (depending on B).

Two major conclusions follow from this result: (i) In the presence of even small layer displacement, the singlet-triplet crossing in the X^- spectrum shifts to a considerably lower magnetic field (e.g., from 35 T in a symmetric 10-nm GaAs QW to 16 T for $\delta=0.5$ nm). We expect that this could stabilize the hypothetical two-component incompressible fluid states involving long-lived X_{td}^- quasiparticles,[26,27] and enable its detection in transport experiments. (ii) The observation of strongly bound X^- states in an experimental PL spectrum implies zero or very small layer displacement in the sample (compared to the QW width). While for asymmetrically doped QW's the displacement can be decreased due to electron-hole correlations in the direction perpendicular to the QW, the interpretation of PL spectra of HJ's in terms of X^- recombination is questionable.

II. MODEL

In order to preserve the 2D symmetry of a QW in a finite-size calculation, the electrons and holes are confined to a Haldane sphere[39] of radius R. The magnetic field B normal to the surface is due to a Dirac magnetic monopole in the center of the sphere. The monopole strength $2S$ is defined in the units of elementary flux, $\phi_0=hc/e$, so that $2S\phi_0=4\pi R^2 B$, and the magnetic length is $\lambda=R/\sqrt{S}$.

The single-particle orbitals (monopole harmonics) are eigenstates of angular momentum[42,43]:

$$L^2|S,l,m\rangle=\hbar^2 l(l+1)|S,l,m\rangle,$$

$$L_z|S,l,m\rangle=\hbar m|S,l,m\rangle. \tag{1}$$

Their energies

$$\varepsilon_{Slm}=\hbar\omega_c\left(n+\frac{1}{2}+\frac{n(n+1)}{2S}\right) \tag{2}$$

form $(2l+1)$-fold degenerate shells (LL's) labeled by $n=l-S=0,1,\ldots$, and (in the limit of large $2S$) separated by the cyclotron energy $\hbar\omega_c=\hbar eB/\mu c$ (where μ is the effective electron or hole cyclotron mass).

The parameters we used for calculation are appropriate for GaAs/Al_xGa_{1-x}As QW's of width $w=10$ nm and an Al concentration $x=0.33$. In such structures, mixing between light- and heavy-hole subbands in the valence band is not very strong[23] and both electrons and (heavy) holes can be described in the effective-mass approximation. The valence subband mixing enters the model through the dependence of the effective cyclotron mass of the hole μ_h on the magnetic field (after Cole *et al.*[44]). We omit the Zeeman splitting of electron and hole spin states $|\sigma\rangle$ and only discuss the Coulomb part of the binding energy. While the actual electron and hole g factors depend on the QW width[45] and magnetic field,[46] and on the wave vector k (and thus on a particular X or X^- wave function[7]), they mainly affect the stability of spin-unpolarized complexes[25] and affect much less the splitting of PL peaks for a given polarization of light. Let us note that although the Zeeman contribution to the X^- binding energy is fairly small in GaAs structures due to a small g factor in this material, it is much larger in other materials (CdTe, ZnSe, GaN, etc.), where it strongly favors the triplet X^- states over the singlet ones.[9,14] We also neglect mixing between different electron and hole QW subbands and (weak[23]) electron-hole correlations in the z direction. Instead, we use effective widths of electron and hole layers, w_e^* and w_h^*, and their effective displacement δ, which account both for actual widths and displacement of single-particle wave functions and for the effects of QW subband mixing and correlations.

Thus the single-particle states used in our calculation are labeled by a composite index $i=[n,m,\sigma]$, and describe an electron or a heavy hole with spin projection σ, whose in-plane quantum numbers are n and m, and the wave functions in the z direction are fixed and controlled by w_e^*, w_h^*, and δ. The electron-hole Hamiltonian can be generally written as

$$H=\sum_{i,\alpha} c_{i\alpha}^\dagger c_{i\alpha}\varepsilon_{i\alpha}+\sum_{ijkl,\alpha\beta} c_{i\alpha}^\dagger c_{j\beta}^\dagger c_{k\beta}c_{l\alpha}V_{ijkl}^{\alpha\beta}, \tag{3}$$

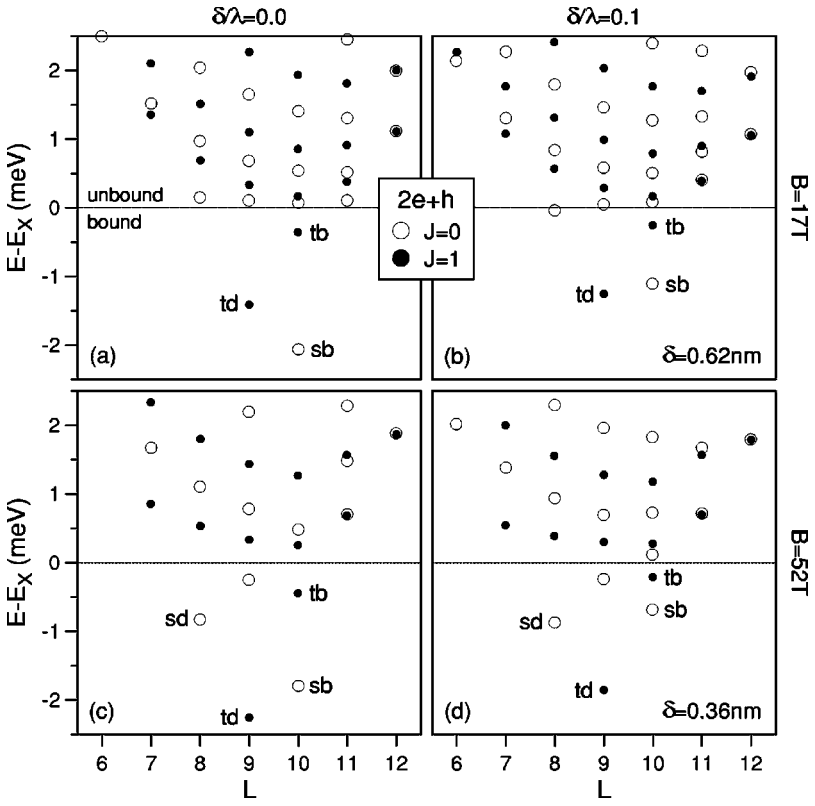

FIG. 1. The energy spectra (energy E vs angular momentum L) of the $2e+h$ system on a Haldane sphere with $2S=20$. Open and full circles mark singlet and triplet states, respectively. The magnetic field is $B=17$ T [(a) and (b)] and $B=52$ T [(c) and (d)]. The layer displacement is $\delta/\lambda=0$ [(a) and (c)] and $\delta/\lambda=0.1$ [(b) and (d)]. λ is the magnetic length. The angular momentum multiplets at $L=8$, 9, and 10 on a sphere correspond to those of angular momentum projection $\mathcal{M}\leqslant -2$, $\mathcal{M}\leqslant -1$, and $\mathcal{M}\leqslant 0$ on a plane, respectively.

where $c^{\dagger}_{i\alpha}$ and $c_{i\alpha}$ create and annihilate particle α (e or h) in state i, and $V^{\alpha\beta}_{ijkl}$ are the Coulomb matrix elements. While the 3D Coulomb matrix elements for an arbitrary electron and hole density profiles $\varrho(z)$ can be integrated numerically,[23,24] we make the following approximation.[25] For the density functions in the z direction we take $\varrho(z)\propto\cos^2(\pi z/w^*)$, that is, we replace the actual QW by one with infinite walls at the interface and a larger effective width w^*. For 10-nm GaAs QW's the best fits to the actual wave functions are obtained for $w_e^*=13.3$ nm and $w_h^*=11.5$ nm. The effective 2D interaction

$$V(r)=\pm\int dz\int dz'\frac{\varrho(z)\varrho(z')}{\sqrt{r^2+(z-z')^2}} \tag{4}$$

is approximated by $V_d(r)=\pm 1/\sqrt{r^2+d^2}$, where the parameter d accounts for the finite widths and displacement of the layers.[47] For the e-e repulsion we take $w^*=w_e^*$ and $d=w^*/5$, and for the e-h attraction $w^*=\frac{1}{2}(w_e^*+w_h^*)$ and $d=w^*/5+\delta$. The 2D matrix elements of $V_d(r)$ are close to the 3D ones, and can be evaluated analytically.

The Hamiltonian H is diagonalized numerically for a system of two electrons and one hole, in the basis including up to five LL's ($n\leqslant 4$), with up to $2S+1=21$ orbitals in the lowest LL. The eigenstates are labeled by two total angular momentum quantum numbers L and L_z, and the total spin of the pair of electrons $J=0$ or 1. The conservation of two orbital quantum numbers in a finite Hilbert space is the major advantage of using Haldane's spherical geometry to model an infinite planar system with 2D translational symmetry. The pair of numbers L and L_z corresponds directly to a pair of conserved quantities on a plane: total angular momentum projection $\mathcal{M}$ and an additional number $\mathcal{K}$ associated with the partial decoupling of the center-of-mass motion in a magnetic field.[28,48] On a sphere, the states within a LL have different L_z's and the same L, and on a plane they have different $\mathcal{K}$'s and the same $\mathcal{L}=\mathcal{M}+\mathcal{K}$.

The conservation of L (or $\mathcal{L}$) in the calculation is essential to identify of the X^- optical selection rules.[25] Since the optically active electron-hole pair has $L=0$ ($\mathcal{L}=0$), and the electron left behind after the X^- recombination has $l=S$ ($\mathcal{L}=0$), only those X^- states at $L=S$ ($\mathcal{L}=0$) are radiative (''bright''). Other (''dark'') states cannot recombine unless the 2D symmetry and the resulting angular momentum conservation are broken (e.g., in a collision with an impurity or another particle).

The spherical model obviously has some limitations, the most important of which is a modification of interactions due to the surface curvature. However, if the correlations modeled have a finite (short) range ξ that scales with λ (as for the electron-hole correlations that cause binding of the X^- states), ξ can be made small compared to R at large $2S$, and the finite-size effects are eliminated in the $2S\rightarrow\infty$ limit.

III. RESULTS AND DISCUSSION

The $2e+h$ low-energy spectra for two different values of $B=17$ and 52 T, and at $\delta/\lambda=0$ and 0.1, are shown in Fig. 1. The calculation was carried out for $2S=20$ and including five LL's ($n\leqslant 4$). We have checked[25] that these numbers are sufficient to obtain quantitatively meaningful results. The energy E is measured from the exciton energy E_X, so that for the bound X^- states below the dotted lines, the vertical axes

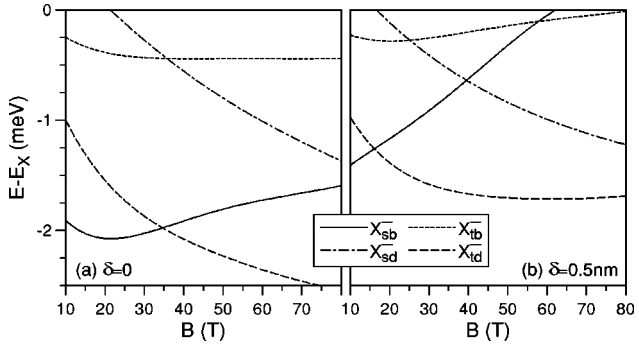

FIG. 2. The X^- binding energies E calculated on a Haldane sphere with the LL degeneracy $2S+1=21$, plotted as a function of the magnetic field B. The parameters are appropriate for a 10-nm GaAs quantum well. The layer displacement is $\delta=0$ (a) and $\delta=0.5$ nm (b).

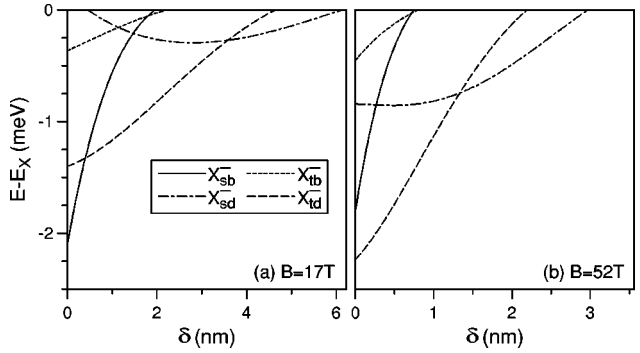

FIG. 3. The X^- binding energies vs the displacement of the electron and hole layers in a 10-nm asymmetric quantum well. The magnetic field is $B=17$ T (a) and $B=52$ T (b). E_X is the exciton energy.

show the negative of their binding energy: $-\Delta=E-E_X$. Singlet ($J=0$) and triplet ($J=1$) states are marked with open and full dots, respectively. The energy is plotted as a function of total angular momentum, and each data point represents a degenerate L multiplet.

The states of particular interest are bound states with the largest Δ and/or bright states at $L=S$. Depending on B and δ, we identify all or some of the following bound X^- states in the spectrum: bright singlet X^-_{sb} at $L=S$ ($\mathcal{L}=0$), dark singlet X^-_{sd} at $L=S-2$ ($\mathcal{L}=-2$), bright triplet X^-_{tb} at $L=S$ ($\mathcal{L}=0$), and dark triplet X^-_{td} at $L=S-1$ ($\mathcal{L}=-1$). As shown in Figs. 1(a) and 1(c), in the absence of layer displacement the X^-_{sb} is the ground state at the lower magnetic field of $B=17$ T, but at a higher magnetic field of $B=52$ T it is X^-_{td} that has the lowest energy. Another bright state X^-_{tb} occurs in the spectrum, but it has higher energy than X^-_{sb} or X^-_{td} at all fields. There is also a dark X^-_{sd} state that becomes bound at a sufficiently large B, but it is not expected to affect the PL spectrum because it is neither radiative nor strongly bound at any B. The situation is dramatically different when a finite layer displacement is included in Figs. 1(b) and 1(d). For $\delta=0.1\lambda$, the binding energies of all X^- states are significantly reduced. The most affected is the bright singlet X^-_{sb}, which is no longer the ground state even at a relatively low magnetic field of $B=17$ T. It is quite remarkable that a displacement as small as $\delta=0.62$ nm (at $B=17$ T) or $\delta=0.36$ nm (at $B=52$ T), that is only a few percent of the QW width of $w=10$ nm and certainly could be expected in asymmetric QW's, causes such a reconstruction of the X^- spectrum. The ground-state transition from a bright singlet to a dark triplet, induced at lower B, is similar to that caused by a magnetic field at $\delta=0$.[23,25]

The effect of the layer displacement on the dependence of the X^- binding energies on the magnetic field is shown in Fig. 2. At $\delta=0$, the binding energies of the two bright states remain almost constant over a wide range of B, in contrast to the two dark states, which quickly gain binding energy when B increases. As found in previous studies,[23,25] this different $\Delta(B)$ dependence results in a singlet-triplet ground-state transition at $B\approx35$ T. At a small displacement of $\delta=0.5$ nm, the binding energy of the bright singlet X^-_{sb} decreases rather quickly as a function of B, more so than the binding energies of other X^- states. As a result, the singlet-triplet transition occurs at a much lower magnetic field of $B\approx16$ T, and the bright singlet unbinds completely at a value of B larger than about 60 T. Actually, neither bright state is strongly bound at $B>60$ T, while the binding energies of both dark states remain fairly large (e.g., $\Delta_{sd}=1.0$ meV and $\Delta_{td}=1.7$ meV at $B=60$ T).

To illustrate the effect of the layer displacement on the X^- states most clearly, in Fig. 3 we plot the X^- binding energies as a function of δ for two values of the magnetic field. In both frames, δ goes from 0 to λ (where $\lambda=6.2$ and 3.6 nm at $B=17$ and 52 T, respectively). For $B=17$ T the ground-state transition from X^-_{sb} to X^-_{td} occurs at $\delta=0.4$ nm, and for $B=52$ T X^-_{td} is the ground state at all displacements. It is clear that the displacement has more effect on the binding energy of X^-_{sb} than on the binding energy of the next most strongly bound state, X^-_{td}. This can be understood by noting that the X^-_{sb} complex has smaller $|\mathcal{L}|$ and thus a smaller average electron-hole distance $\langle r_{eh}\rangle$, and that the effect of a finite δ in $V_d(r)$ decreases as r increases.

Let us point out that the binding energies obtained here are rather sensitive, not only to B or δ, but also to other details of our model, including some of its simplifications or approximations. For example, a slightly different approximation used here to calculate the e-h Coulomb matrix elements at $\delta=0$ resulted in smaller binding energies compared to Ref. 25 (although the difference in Δ appears to be similar for all X^- states, and the singlet-triplet crossing is obtained at the same B, which means that the difference between the models affects E_X rather than E_{X^-}). Whittaker and Shields[23] showed that even in narrow QW's the inclusion of higher QW subbands and electron-hole correlations in the z direction somewhat enhances the X^- binding, especially that of the X^-_{sb} state. Based on their calculation, one can expect that our values, obtained in the lowest subband approximation, are underestimated by up to 0.5 meV, depending on B and the particular X^- state. Despite the difficulty in obtaining definite values of Δ, two conclusions arising from our calculation seem important, and at the same time independent of the approximations made.

(i) Even a small displacement of electron and hole wave functions in the z direction shifts the singlet-triplet transition to a considerably lower value of the magnetic field. Therefore, the theoretical value of $B \approx 35$ T for the crossing in a 10-nm QW must be understood as the upper estimate, and in an experimental sample the crossing can occur at any smaller value. This effect broadens the range of magnetic fields in which the X_{td}^-'s, together with electrons, are both the most stable and long-lived quasiparticles in the electron-hole system. It thus seems that the proposed[26,27] incompressible fluid states of X_{td}^-'s and electrons could be observed more easily in slightly asymmetric QW's.

(ii) The binding energies of both bright X^- states are strongly sensitive to layer displacement. Therefore, the recombination from strongly bound X^- states observed in an experimental PL spectrum implies zero or very small displacement in the sample (compared to the QW width). The parameter δ used in our model describes the displacement of electron and hole wave functions in the z direction within a particular bound X or X^- state, and must be distinguished from the bare displacement δ_0 of single-electron and single-hole wave functions due to an external electric field (e.g., caused by a charged doped layer). It is therefore possible that, even in strongly asymmetric QW's, electron-hole correlations in the z direction (which favor small displacement) dominate the effect of external electric field (which causes displacement), and the resulting δ is much smaller than δ_0. If correct, this picture of symmetry (partially) restored by correlations would explain the success of ''symmetric models''[23–25] in describing a wide class of both symmetric and asymmetric QW's (and invalidate the use of the lowest subband approximation with δ_0 taken for unbound particles). However, it does not seem possible that any X^- states should form in HJ's where the electrons are confined in a narrow 2D layer and the holes remain outside of this layer. Consequently, the interpretation of multiplets in the PL spectra of HJ's in terms of X and X^- recombination seems questionable. A recent alternative interpretation[33] involves coupling a distant hole to (Laughlin) charge excitations of the 2DEG, and the formation of bound and radiative (fractionally charged) excitonic complexes of a different type.

IV. CONCLUSION

Using exact numerical diagonalization in Haldane's spherical geometry, we have studied the effect of the displacement δ of electron and hole layers on the binding energies of the X^- states formed in narrow asymmetric QW's in high magnetic fields B. Depending on B and δ, different bound X^- states were identified in the $2e+h$ spectrum: bright singlet X_{sb}^-, dark singlet X_{sd}^-, bright triplet X_{tb}^-, and dark triplet X_{td}^-. The binding energies of all X^- states quickly decrease as a function of δ. The most sensitive is the strongly bound X_{sb}^- state, and even at displacements very small compared to the QW width, the magnetic-field-induced transition from this bright ground state to the dark X_{td}^- ground state occurs at significantly lower values of B. The critical displacement for which the bright X^- states unbind is only 5–10 % of the QW width (depending on B). Therefore, detection of the X^- recombination in an experimental PL spectrum implies virtually no displacement of electron and hole layers (within the observed X^- states). While in asymmetric QW's small values of δ can result from electron-hole correlations, the interpretation of the PL spectra of HJ's in terms of X^-'s is questionable.

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