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 $\delta$ of electron and hole layers. The most sensitive is the “bright” singlet, whose binding energy decreases quickly with increasing $\delta$ 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 $\delta$ 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 $\delta$ 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.

1. INTRODUCTION

The optical properties of a quasi-two-dimensional electron gas (2DEG) in a high magnetic field $B$ have been widely studied both experimentally and theoretically. The 2DEG is usually realized in semiconductor quantum wells (QW) or heterojunctions (HJ). In the 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 in bulk semiconductors; however, it could not be observed experimentally because of the small binding energy $\Delta$. It was later shown by Stebe and Aina that the $X^-$ binding is significantly enhanced in 2D systems. Indeed, the $X^-$ state with $\Delta$ of about 3 meV has been detected by Kheng et al. in CdTe QW. The subsequent extensive experimental and theoretical studies established that the $X^-$ occurs in 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 showed that the decoupling of optically active excitons from electrons in the lowest Landau level (LL) due to the “hidden symmetry” breaks unbinding of the $X^-$ (and other optically active complexes larger than $X$) for $B \rightarrow \infty$. However, a different bound $X^-$ state exists in this limit. It is a triplet $X^-_{T_{\delta}}$ with $J = 1$ and finite angular momentum $\mathcal{L} = -1$. Since both the hidden symmetry and the angular momentum conservation independently forbid recombination of an isolated $X^-_{T_{\delta}}$ in the lowest LL, its optical lifetime $\tau_{b}$ in high magnetic fields is expected to be long and determined by scattering and/or disorder.

The fact that the $X^-_{T_{\delta}}$ binding energy $\Delta_{T_{\delta}}$ decreases with increasing $B$ implies a singlet–triplet crossing in the $X^-$ spectrum at a certain $B$, estimated as about 35 T for a 10 nm GaAs QW. Although the PL experiments in high magnetic fields indeed show emission from a pair of $X^-$ states, neither the crossing has been found nor the intensity $\tau^{-1}$ of the peak assigned to the triplet state increased with increasing $B$ or decreasing electron density. This apparent discrepancy between theory and experiment has been recently resolved by a numerical discovery of yet another $X^-$ state, a “bright” triplet $X^-_{T_{b}}$. $X^-_{T_{b}}$ state has $\mathcal{L} = 0$, $J = 1$, large oscillator strength $\tau_{b}^{-1}$, and small binding energy $\Delta_{b}$, and occurs in high magnetic fields in QW’s of finite width.

While the identification of the experimentally observed triplet as the $X^-_{T_{b}}$ state explains its small binding energy, the fact that the more strongly bound $X^-_{T_{\delta}}$ state is not observed confirms its very long optical lifetime $\tau_{b}$. The reason why $\tau_{b}$ 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, and the effective isolation of all $X^-$ states from the 2DEG. 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 $\Delta$, PL energy $\omega$, or oscillator strength $\tau^{-1}$.

The major difficulty in comparing the numerical and experimental data is that most experiments are carried out in asymmetrically doped QW’s or HJ’s, in which an electric field perpendicular to the 2DEG modifies confinement and leads to displacement of electron and hole layers. This displacement or separation between the electron and hole layers, has been ignored in the existing realistic calculations (which take into account the finite widths of the electron and hole layers, LL mixing, etc.) although from more idealized calculations (zero layer widths and no LL mixing) 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 $\delta$ into the model used earlier to study the $X^-$ states in narrow symmetric QW’s. Using exact numerical diagonalization in Haldane’s spherical geometry we examine the dependence of binding energies of all different bound $X^-$ states on both magnetic field and the displacement. In addition to the bright singlet $X^s_0$ (denoted here by $X^s_0$) and two triplets, $X^t_{ld}$ and $X^t_{ld'}$, we identify a dark singlet $X^s_{sd}$ with angular momentum $J = 2$ which occurs at $\delta > 0$, in analogy to a known $D^-$ (charged donor) state at the same $L$. We demonstrate that the binding energies of all $X^-$ states strongly depend on $\delta$. Most sensitive is the $X^s_{sd}$ state which unbinds when $\delta$ 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 $X^s_{id}$ quasiparticles 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 a 2D symmetry of a QW in a finite-size calculation, the electrons and the holes are confined to a Haldane sphere 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 are called monopole harmonics. They are the eigenstates of angular momentum:

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

and their energies,

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

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 $\mu$ is the effective electron or hole cyclotron mass).

The parameters we used for calculation are appropriate for GaAs/AlGaAs QW’s of width $w = 10$ nm and Al concentration $x = 0.33$. In such structures, mixing between the light- and heavy-hole subbands in the valence band is not very strong, 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 $\mu_h$ on the magnetic field (after Cole et al.). 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 factors depend on the QW width and magnetic field and on the wavevector $k$ (and thus on a particular $X$ or $X^-$ wavefunction), they mainly affect the stability of spin-unpolarized complexes and much less the splitting of PL peaks for a given polarization of light. We also neglect mixing between different electron and hole QW subbands and the (weak) electron–hole correlations in the direction perpendicular to the QW (along the $z$ axis). Instead, we use effective widths of electron and hole layers, $w^e_\sigma$ and $w^h_\sigma$, and their effective displacement $\delta$, which account both for actual widths and displacement of single-particle wavefunctions 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 $\sigma$, whose in-plane quantum numbers are $n$ and $m$, and the wavefunctions in the $z$-direction are fixed and controlled by $w^e_\sigma$, $w^h_\sigma$, and $\delta$. The electron–hole Hamiltonian can be generally written as

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

where $c_{i\alpha}$ and $c_{i\alpha}$ create and annihilate particle $\alpha$ ($e$ or $h$) in state $i$, and $V_{ijkl}^{\alpha\beta}$ are the Coulomb matrix elements. While the 3D Coulomb matrix elements for an arbitrary electron and hole density profiles $g(z)$ can be integrated numerically, we make the following approximation
For the density functions in the \( z \)-direction we take \( \rho(z) \propto \cos^2(\pi z/\omega^*) \), that is, we replace the actual QW by one with infinite walls at the interface and a larger effective width \( \omega^* \). For a 10 nm GaAs QW’s the best fits to the actual wavefunctions are obtained for \( \omega^*_y = 13.3 \) nm and \( \omega^*_h = 11.5 \) nm. The effective 2D interaction

\[
V(r) = \pm \int dz \int dz' \frac{\rho(z)\rho(z')}{\sqrt{r^2 + (z - z')^2}},
\]

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. 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 the system of two electrons and one hole, in the basis including up to five LL’s \((n \leq 4)\) with up to \( 2S + 1 = 21 \) orbitals in the lowest LL. The eigenstates are labeled by \( L \) 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 the 2D translational symmetry. The pair of numbers, \( L \) and \( L_z \), correspond directly to a pair of conserved quantities on a plane: total angular momentum \( \mathcal{M} \) and an additional number \( \mathcal{K} \) associated with the partial decoupling of the center-of-mass motion in a magnetic field. On a sphere, the states within a LL have different \( L \) and the same \( L_z \), and on a plane, they have different \( \mathcal{K} \) 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. 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 and the most important one is modification of interactions due to the surface curvature. However, if the correlations modeled have a finite (short) range \( \xi \) that scales with \( \lambda \) (as it is for the electron–hole correlations that cause binding of the \( X^- \) states), \( \xi \) can be made small compared to \( R \) at large \( 2S \) and the finite-size effects are eliminated in the \( 2S \to \infty \) 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 \((n \leq 4)\). We have checked 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 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 the bound states with the largest \( \Delta \) and/or the bright states at \( L = S \). Depending on \( B \) and \( \delta \), 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 Fig. 1ac), 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 \( X_{sd}^- \) 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 Fig. 1bd). 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 \( \omega = 10 \) nm and certainly could be expected in asymmetric QW’s, causes such reconstruction of the \( X^- \) spectrum. The ground state transition from the bright singlet to the dark triplet induced at lower \( B \) is similar to that caused by a magnetic field at \( \delta = 0.2 \) nm.

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 the previous studies, 23-25 this different \( \Delta(B) \) dependence results in a singlet–triplet ground state transition at \( B \approx 35 \) 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 \( B \approx 16 \) T and the bright singlet unbinds completely at \( B \) 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 \( \delta \) for two values of the magnetic field. In both frames, \( \delta \) goes from 0 to 0.1 \( \lambda \) (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 \( X_{st} \) occurs at \( \delta = 0.4 \) nm and for \( B = 52 \) T the \( X_{td} \) 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 \( |L| \) and thus smaller average electron–hole distance \( \langle r_{eh} \rangle \), and that the effect of a finite \( \delta \) in \( V(r) \) decreases as \( r \) increases.

Let us point out that the binding energies obtained here are rather sensitive not only to \( B \) or \( \delta \), 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 \( e-\hbar \) Coulomb matrix elements at \( \delta = 0 \) resulted in smaller binding energies compared to Ref. 24 (although the difference in \( \Delta \) 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 25 showed that even in narrow QW’s the inclusion of higher QW subbands and electron–hole correlations in the \( z \)-direction enhances somewhat the \( X^- \) binding, specially 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 with obtaining definite values of \( \Delta \), two conclusions arising from our calculation seem quite important and at the same time independent of the approximations made.

(i) Even a small displacement of electron and hole wavefunctions 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 well 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 most stable and long-lived quasiparticles in the electron–hole system. It thus seems that the proposed 23-25 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 the layer displacement. There-
fore, 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 $\delta$ used in our model describes displacement of electron and hole wavefunctions in the $z$-direction within a particular bound $X$ or $X^-$ state and must be distinguished from the bare displacement $\delta_0$ of single-electron and single-hole wavefunction 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, the electron–hole correlations in the $z$-direction (which favor small displacement) dominate the effect of external electric field (which causes displacement), and the resulting $\delta$ is much smaller than $\delta_0$. If correct, this picture of symmetry (partially) restored by correlations explains the success of “symmetric models” to describe a wide class of symmetric and asymmetric QW’s (and invalidates the use of the lowest subband approximation with $\delta_0$ taken for unbound particles). However, it does not seems 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 involves coupling of a distant hole to (Laughlin) charge excitations of the 2DEG and 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 $\delta$ 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 $\delta$, different bound $X^-$ states were identified in the $2e + h$ spectrum: bright singlet $X_{ab}^-$, dark singlet $X_{ad}^-$, bright triplet $X_{tb}^-$, and dark triplet $X_{td}^-$. The binding energies of all $X^-$ states quickly decrease as a function of $\delta$. The most sensitive is the strongly bound $X_{ab}^-$ 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 $X_{id}^-$ 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 $\delta$ 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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