Stability of optically-active charged excitons in quasi-two dimensional systems

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Abstract

A negatively charged quasi-two dimensional exciton ($X^-$) is solved exactly numerically in the presence of a uniform perpendicular B-field. Various quasi-two dimensional geometries are studied. The charge distribution of the $X^-$ parallel to the B-field is found to be crucial in determining the stability of the optically-active $X^-$ and hence its photoluminescence (PL) signature. The theory provides a quantitative explanation of recent experimental results obtained for a GaAs quantum well. Effects are found which cannot be described within a lowest Landau level approximation.

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Excitonic effects are used to explain the majority of discrete lines appearing in the interband optical spectra obtained from semiconductors. However new lines have been seen recently which cannot be explained by excitons, either intrinsic or impurity-bound (see eg Refs. [1] [2]). It has been proposed that these discrete lines are associated with the formation of a charged electron-hole complex. The stability of charged excitons was first predicted for three-dimensional (3D) systems by Lampert [3]. This was subsequently studied theoretically at zero B-field in both 3D [4] and two-dimensions (2D) [5]. In both cases the charged exciton was found to be stable. The 2D charged exciton binding energies are about ten times larger than those in 3D due to the extra confinement [5] thereby facilitating their experimental observation. Negatively charged excitons, $X^- = 2e + 1h$, were first observed in CdTe/CdZnTe quantum wells [6] [7] and were later seen in GaAs/GaAlAs quantum wells [8] [1] [9] [2] [10]. Positively charged excitons, $X^+ = 1e + 2h$, have also been seen [1]. The charged excitons have been observed experimentally using polarized spectroscopy, in particular photoluminescence (PL) with a uniform magnetic field B applied perpendicular to the quantum well plane. $X^-$ stability requires a two-dimensional electron gas (2DEG) of intermediate electron density which is typically near the metal-insulator transition, the reduced screening effect of the other electrons helping to stabilise the $X^-$ [8]. In the low density limit the exciton ($X$) dominates the PL spectrum as expected. At high densities and low B-fields, a Fermi-edge-singularity (FES) is seen. The FES moves smoothly into the $X^-$ line as the electron density is decreased [1] [1] and develops into sharp $X$ and $X^-$ peaks with increasing B-field [7]. These results suggest that a study of the $X^-$ is crucial for an understanding of the optical properties of 2DEGs at finite B-fields, as was claimed recently in Ref. [1].

In this paper an accurate numerical solution is presented for a quasi-2D $X^-$ consisting of two electrons and a hole situated in a weak in-plane confinement potential and a perpendicular uniform B-field. The $X^-$ properties are studied over a wide range of experimentally applicable B-fields. Various relevant quasi-2D geometries are considered, each having a different charge distribution parallel to the B-field, i.e. perpendicular to the 2DEG. These
geometries are as follows:

(i) electrons and hole are strictly 2D but move on separate parallel planes (biplanar geometry),

(ii) electrons and hole occupy the same plane but have their charge smeared in a rod-like distribution along the perpendicular direction (rod geometry),

(iii) electrons and hole are strictly 2D and occupy the same plane (coplanar geometry).

The stability of the optically-active $X^-$ and hence its PL spectrum are found to vary drastically depending on the geometry used. It is shown that recent experimental PL spectra obtained from quantum wells can only be explained using the rod geometry and higher Landau level mixing. A previous theoretical study only considered a coplanar geometry at a fixed B-field.

The Hamiltonian for the $X^-$ problem is:

$$
\mathcal{H} = \frac{1}{2m_h} \left[ \mathbf{p}_h - \frac{e}{c} \mathbf{A}(\mathbf{r}_h) \right]^2 + \frac{1}{2} m_h \omega_h^2 \mathbf{r}_h^2 + g_h \mu S_h^z \\
+ \sum_{i=1}^{2} \left\{ \frac{1}{2m_e} \left[ \mathbf{p}_{e,i} + \frac{e}{c} \mathbf{A}(\mathbf{r}_{e,i}) \right]^2 + \frac{1}{2} m_e \omega_e^2 \mathbf{r}_{e,i}^2 + g_e \mu \mathbf{S}_{e,i}^z - V_{eh}(|\mathbf{r}_{e,i} - \mathbf{r}_h|) \right\} \\
+ V_{ee}(|\mathbf{r}_{e,1} - \mathbf{r}_{e,2}|)
$$

(1)

where $m_e = 0.067m_0$ and $m_h = 0.475m_0$ are the masses of the conduction electrons and heavy holes respectively for GaAs. In order to facilitate comparison with a quasi-2D electron gas the parabolic confinement potentials, $\hbar \omega_e = 1.75\text{meV}$ and $\hbar \omega_h = 0.25\text{meV}$, are chosen to be weak compared with the cyclotron energies ($\hbar(\omega_e)_e = 17.2\text{meV}$ and $\hbar(\omega_e)_h = 2.4\text{meV}$ at $B=10T$). Also $\omega_e/\omega_h = m_h/m_e$ ensuring that the electron and hole wavefunctions have equal characteristic lengths, $l_{e,h}^2 = \hbar/(m_{e,h}\tilde{\omega}_{e,h}) = l^2$ where $\tilde{\omega}_{e,h}^2 = (\omega_{c,e,h})^2 + 4\omega_{e,h}^2$, as is the case in the limit of zero confinement.

The interaction potentials are Coulombic. In order to obtain a tractable model it is assumed that the single particle wavefunctions separate: $\Psi_e = \phi_e(z_e)\psi_e(\mathbf{r}_e)$ and $\Psi_h = \phi_h(z_h)\psi_h(\mathbf{r}_h)$. A specific form for $\phi_{e,h}$ is chosen, thereby freezing out the z-motion and
yielding a quasi-two dimensional model. For the biplanar geometry:

\[ |\phi_e(z_e)|^2 = \delta(z_e - 0) ; \quad |\phi_h(z_h)|^2 = \delta(z_h - d) \]  

(2)

while for the rod geometry:

\[ |\phi_{e,h}(z_{e,h})|^2 = \begin{cases} \frac{1}{L} & \text{if } -\frac{L}{2} < z_{e,h} < \frac{L}{2} \\ 0 & \text{otherwise} \end{cases} \]  

(3)

The coplanar geometry is the limit of the previous two where \( d \) and \( L \) tend to zero. The coplanar and rod models retain a singularity in \( V_{e,h} \) at \( r_e = r_h \) which the biplanar model loses. The differences in the forms of \( |\phi(z)|^2 \) lead to significantly different PL spectra.

The effects of the B-field and the Coulomb interaction compete: the expansion parameters are given by \( a_{e,h}/l \) where \( a_{e,h} = \epsilon \hbar^2 / m_{e,h} e^2 \) is the Bohr radius for the electron and hole respectively. Taking \( B \sim 8T \) and \( \epsilon_r = 12.53 \) yields (GaAs) \( a_h/l = 0.2 \) and \( a_e/l = 1.1 \), thereby rendering any perturbation approach unreliable. For this reason the \( X^- \) Hamiltonian is diagonalized exactly numerically. Rotational symmetry about the B-field leads to conservation of total angular momentum, \( M \). The symmetric gauge is hence chosen, \( \mathbf{A}(\mathbf{r}) = (\mathbf{B} \times \mathbf{r})/2 \), giving single particle states characterised by the Landau level \( (n) \) and azimuthal angular momentum \( (m) \). Angular momentum conservation makes the Hamiltonian block diagonal in this basis, each block being characterised by \( M \). The spin and spatial components of the wavefunction factorise, the spin states of the electrons being either singlet or triplet. In order to preserve the correct electron antisymmetry the triplet (singlet) states have a spatial component which contains antisymmetric (symmetric) combinations of the single particle electron wavefunctions.

The Hamiltonian is diagonalized in the basis of definite \( M \), \( S \) and \( S_z \). Since \( (a_{e,h}/l) > 1 \) higher Landau levels must be included in the basis. The actual basis used has 6 hole Landau levels, 2 electron Landau levels and 20 angular momentum states per Landau level (cf. [11]).
A larger number of hole Landau levels are used because of the small heavy-hole cyclotron energy. A finite number of angular momentum states are used due to the weak confinement which removes the degeneracy of the Landau level. From comparison with experiment (discussed later in this paper) the authors believe that this basis captures the essential physics of the $X^-$. The matrix elements are evaluated following a method outlined in [12]. In the coplanar limit ($L, d \rightarrow 0$) analytic results are obtained for the matrix elements [13]. Away from this limit, numerical integration is employed. Two completely different methods for evaluating the matrix elements have been used to check for numerical error in the integration routines. For comparison purposes the exciton lines are also calculated in a manner identical to that used for the $X^-$. 

Both excitons ($X$) and $X^-$ undergo recombination giving rise to a PL spectrum. It is assumed here, as in [2], that recombination is with the $\pm 3/2$ heavy holes. The oscillator strengths for the $X$ and $X^-$ PL lines are $\langle 0|\hat{L}|X \rangle$ and $\langle e|\hat{L}|X^- \rangle$ respectively, where $\hat{L} = \int \hat{\psi}_e(r)\hat{\psi}_h(r)d^2r$. This means that only excitons with orbital angular momentum $M=0$ will recombine. For both $X$ and $X^-$ the necessary change in the system’s angular momentum is taken up by the atomic part of the wavefunction giving rise to two lines, $\sigma^+$ and $\sigma^-$, which are Zeeman split. In the limit that the basis is restricted to the lowest Landau level and setting $V_{eh} = V_{ee}$, there is a well-documented hidden symmetry [14] [15]. This forces the triplet $X^-$ ($X^-t$) PL energy to be identical with that of the lowest exciton ($X$) line. In this paper the symmetry is broken by the weak confinement and more importantly by the inclusion of higher Landau levels. For the case of a biplanar $z$-geometry the interactions $V_{ee}$ and $V_{eh}$ are no longer equal, thus further breaking the hidden symmetry. The reliability of the calculation has been verified by reproducing known results as follows: (a) in the limit of zero confinement and using the rod geometry ($V_{ee} = V_{eh}$) the hidden symmetry result is recovered, (b) the B=0 confined 2-electron results of [16] are reproduced, (c) results found in a previous $X^-$ study [11] are obtained.

The theoretical PL spectra resulting from the coplanar, biplanar and rod geometries are now discussed. When a strictly coplanar geometry is used both the optically-active singlet
\((X^-)\) and triplet \((X^-)\) states are stable with respect to the decomposition \(X^- \rightarrow X + e\). This stability persists over a calculated range in B-field of at least \(6 \rightarrow 14\)T. However, the agreement with recent experimental quantum well data (see Fig 1a) is not good since the calculated \(X^- - X^-\) splitting is too large (3.8meV at B=8T) and there is no strong increase of the PL energies with B. Using the biplanar geometry the PL spectrum is found to be very sensitive to changes in the plane separation, \(d\). For an \(X^-\) with zero total angular momentum, \(M=0\), the optically-active singlet and triplet species are stable when \(d = 0\). Increasing \(d\) from zero causes the singlet \((X^-)\) to unbind \((d \sim 0.5l)\) with the triplet \((X^-)\) unbinding soon after \((d \sim l)\). Similar unbinding occurs for the optically-active \(M\neq0\ \ X^-\) studied. Thus the biplanar geometry PL spectra differ strongly from the experimental quantum well PL spectra (Fig 1a). These results demonstrate that quasi-2D systems which approximate to a biplanar system (eg heterojunctions) with \(d > l\) are most unlikely to exhibit PL effects due to charged excitons.

The experimental PL spectrum of Shields et al. for a GaAs quantum well [2] shown in Fig 1a is typical of those found in the literature. The exciton \((X^-)\), singlet \((X^-)\) \((X^-)\), and triplet \((X^-)\) \((X^-)\) are marked in accordance with the experimental assignments of Ref. [2] (for clarity, only the \(\sigma^-\) lines are shown in all figures in this paper). The theoretical lines shown in Fig 1b are calculated using the rod geometry with a rod length \(L = 225\)Å and total angular momentum \(M=0\). For the range of B-fields shown in Fig 1, \(L\) is less than the width of the \(X^-\) in the plane thus validating the assumption that the \(X^-\) is quasi-2D. The \(L\) value is also consistent with the width of the envelope function in the experimental quantum well used by Shields et al. [2]. Figure 1 shows that the theoretical lines exhibit many of the quantitative features of the experimental spectrum. The energy of the calculated PL lines increases with B-field in a manner closely matching that of the experimental lines. This is a non-trivial agreement since the slope is not the result of simply adding the single-particle cyclotron energies. The \(X^- - X^-\) splitting \((\sim 1.4\)meV at 8T) is close to the experimental value \((\sim 1.7\)meV at 8T). With increasing B-field the triplet line \((X^-)\) becomes more separated from the exciton line \((X^-)\) hence \((X^-)\) becomes increasingly
stable. However, $X_t^{-}$ only becomes stable at a finite B-field (5T theoretically compared with 2.5T experimentally). The ordering of the PL lines in energy is very stable to changes in $L$. The quantitative theory discussed here thus allows a definite assignment of the PL lines which is in agreement with that proposed for the original experimental spectra [2]. The slight differences between the theory and experiment in Fig 1 are to be expected since the model is necessarily a simple, quasi-2D representation of a complex 3D system.

Recombination from charged excitons in the rod geometry with total angular momentum $M<0$ should also be considered as they give PL lines of similar energies to $M=0 X^{-}$. The PL spectra for a $M=-2 X^{-}$ at $B=10T$ are calculated both with and without an in-plane confinement and are shown in Figs 2a and 2b respectively. As the in-plane confinement is removed the lowest triplet state has its oscillator strength drastically lowered to the benefit of the first excited triplet state (cf Figs 2a and 2b). The PL spectra in the zero confinement limit agree well with the experimental PL spectra in Ref. [2]. The samples used in Ref. [2] are reported to have a thick undoped spacer layer between the donor atoms and the well. This minimizes the spatial confinement in the plane of the well, giving mobilities which demonstrate a high degree of homogeneity and allowing comparison with calculations in the zero confinement limit. Fig 2c shows the PL spectrum of the $M=0 X^{-}$ for $B=10T$ with the weak in-plane confinement. As the in-plane confinement is removed (Fig 2d) the PL spectrum is almost unaffected. If the basis is restricted to the lowest Landau level, the hidden symmetry result (triplet and exciton PL lines coincident) is obtained when the in-plane confinement is removed (see inset in Fig 2). The fact that the splitting is retained in Figs 2b and d implies that higher Landau levels are crucially important in allowing the optically-active $X_t^{-}$ wavefunctions to form configurations stable against the decomposition $X_t^{-} \rightarrow X + e^{-}$: this breaks the hidden symmetry and causes the $X_t^{-}$ PL line to be seen. In fact the energy levels of the $X^{-}$ and exciton all change significantly when the basis is restricted to the lowest Landau level.

To summarize, a quasi-two dimensional $X^{-}$ has been solved exactly numerically in the presence of a uniform perpendicular magnetic field and a weak in-plane parabolic confine-
ment. The diagonalization basis contained higher Landau levels giving rise to results which differ significantly from those obtained within a lowest Landau level approximation. The quasi-2D coplanar, biplanar and rod geometries studied were found to yield significantly different PL spectra. Of all the geometries considered, the rod model approximates closest to a quantum well and was able to produce good quantitative agreement with recent experimental quantum well spectra. The biplanar model predicts an instability of optically-active $X^-$ with increasing $d$: this is consistent with the apparent failure to observe charged excitons in experimental quasi-2D systems other than quantum wells.

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Figure Captions

Figure 1: Variation of $X^-$ and exciton X PL spectra ($\sigma^-$) with B-field. (a) Experimental; adapted from for a 300 Å GaAs quantum well. (b) Theory; zero of energy shifted to include the band gap. Ranges are chosen to highlight the agreement of trends in the experimental and calculated spectra. Parameters given in text. Lines are guides to the eye.

Figure 2: PL spectrum ($\sigma^-$) at B=10T, $L=225\,\text{Å}$ for (a) $M=-2$, weak in-plane confinement (b) $M=-2$, zero in-plane confinement limit (c) $M=0$, weak in-plane confinement (d) $M=0$, zero in-plane confinement limit. Parameters given in text. Inset shows $X^+_r$ and $X$ lines calculated in the lowest Landau level (LLL) with zero in-plane confinement. A gaussian line-broadening has been introduced throughout.
Exciton X
Triplet X¹⁻
Singlet X¹⁻

Photoluminescence Energy (meV)

Magnetic Field (T)

(a)

(b)
Singlet X
− Triplet X
− Exciton X

(a)
(b)
(c)
(d)

LLL

Photoluminescence Strength (arb. units)

Photoluminescence Energy

2 meV

2 meV

LLL

(d)

Photoluminescence Energy

2 meV