Optics of Charged Excitons in Quantum Wells: Free versus Donor-bound Complexes

A. B. Dzyubenko1,2, D. A. Cosma3, T. D. Kelly II4, A. R. Todd1, and A. Yu. Sivachenko5
1Department of Physics, California State University at Bakersfield, Bakersfield, CA 93311, USA
2General Physics Institute, Russian Academy of Sciences, Vavilova 38, Moscow 119991, Russia
3Ariadne Genomics Inc., 3000 Great Seneca Highway, Rockville, MD 20850, USA

(Dated: March 23, 2006)

We theoretically study localization of quasi-two-dimensional negatively charged excitons X− on isolated charged donors in magnetic fields. We consider donors located in a barrier at various distances L from the heteroboundary as well as donors in the quantum well. We establish how many different singlet X1− and triplet X2− bound states a donor ion D+ can support in magnetic fields B > 6 T. We find several new bound states, some of which have surprisingly large oscillator strengths.

PACS numbers: 71.35.Cc, 71.35.Ji, 73.21.Fg

INTRODUCTION

Optical signatures of spin-singlet X1− and spin-triplet X2− charged excitons are commonly observed in semiconductor nanostructures in magnetic fields. Despite the status of X− as one of the simplest few-body systems with Coulomb interactions and a large amount of experimental and theoretical work, some important issues remain unresolved. One of these issues is the degree of localization of charged excitons in realistic quantum wells (QW’s) and how localization of X− manifests itself in optics [1,2].

In this work, we discuss exact selection rules that govern transitions of charged excitons in magnetic fields. We also demonstrate, on a quantitative level, how these selection rules work when applied to free X− and donor-bound (D+, X−) charged excitons; the latter can also be considered as excitons bound to a neutral donor (D0, X).

CLASSIFICATION OF STATES AND SELECTION RULES

Classification of states of free charged electron-hole complexes in magnetic fields is based on magnetic translations and the axial symmetry about the magnetic field axis [2]. The corresponding orbital quantum numbers are the oscillator quantum number k = 0, 1, 2, . . . and the total angular momentum projection on the z-axis, M. The former has the meaning of the mean squared distance to the orbit guiding center; there is an infinite-fold Landau degeneracy in k. Each family of degenerate X− states starts with its parent k = 0 state that has some specific value of M that follows from the solution of the Schrödinger equation. Degenerate daughter states k = 1, 2, . . . have values M − 1, M − 2, . . . for the total angular momentum projection. Selection rules for interband transitions are ∆M = 0 and ∆k = 0 and lead to the following results: Photoluminescence (PL) of a free X− must leave an electron in a LL with the number n equal to the angular momentum M of the parent state, X− → nωX− + eωX−. Therefore, (i) families of X− states that start with M < 0 are dark in PL and (ii) shake-ups to multiple Landau levels (LL’s) are strictly prohibited [2].

The presence of a donor ion D+ breaks the translational symmetry, lifts the degeneracy in k, and makes many of the previously prohibited transitions allowed. Let us discuss spectroscopic consequences of the remaining axial symmetry for a donor-bound state (D+, X−) with angular momentum M and wavefunction ΨM(r1, r2; rh). The dipole matrix element for interband transition to a final electron state φm/(r) with angular momentum projection m/f is

\[ f \sim \left| p_{cv} \int dr \int dr' \phi_{m/f}(r) \Psi_M(r, r', r') \right|^2 \sim \delta_{M,m/f}. \] (1)

Conservation of angular momentum M = m/f can be satisfied for a number of final states φm/(r) belonging to different LL’s. Therefore, shake-up processes become allowed in PL. More than that, PL of (D+, X−) states with M > 0 must proceed via shake-ups to higher LL’s [3]. This is because electron states with angular momenta m/f = M > 0 are only available in n = M or higher LL’s. Note that the shake-up processes are due to the Coulomb induced admixture of LL’s and are suppressed in strong fields as B−2.

NUMERICAL APPROACH

We obtain the energies and wavefunctions of the (D+, X−) complexes and of free X− excitons by diagonalization of the interaction Hamiltonian using a complete basis of states compatible with both axial and electron permutational symmetries. The basis states are constructed out of the in-plane wavefunctions in LL’s.
and size quantization levels in a QW with proper symmetrization for triplet and singlet states. We consider up to $2 \times 10^5$ basis states and, by applying an adaptive scheme, we choose out of these about $6 \times 10^3$ states to be diagonalized in the Hamiltonian matrix. A stability for the donor-bound charged exciton is determined with respect to its dissociation to a neutral donor and a free exciton, $(D^+, X^-) \rightarrow D^0 + X$. Accordingly, a binding energy of a stable $(D^+, X^-)$ complex is defined as the energy difference between the total Coulomb energies

$$E_{(D^+, X^-)}^b = E_{D^0}^{\text{Coul}} + E_X^{\text{Coul}} - E_{(D^+, X^-)}^{\text{Coul}} > 0,$$

with $D^0$ and $X$ being in their ground states. Binding energy 2 determines the energy difference between the positions of a neutral exciton and a donor-bound charged exciton PL emission lines, $\hbar \omega_X - \hbar \omega_{(D^+, X^-)} = E_{(D^+, X^-)}^b$.

RESULTS AND DISCUSSION

The calculated binding energies of the various charged exciton states in a 100 Å GaAs QW as functions of the distance $L$ to the donor ion $D^+$ are shown in Fig. 1. We estimate the accuracy in binding energies to be of the order of $\pm 0.1$ meV.

The limiting case $L = \infty$ corresponds to free charged excitons $X^-$. There are three documented bound states in this limit: the bright singlet $X_s^-$ with $M = 0$, the dark triplet $X_{td}^-$ with $M = -1$, and the bright triplet $X_{tb}^-$ with $M = 0$ (see 2, 4, 5 and references therein). We found two new bound states: the second dark triplet state with $M = -1$, labeled $X_{td2}^-$ in Fig. 1, and the second bright singlet state with $M = 0$, labeled $X_{s2}^-$. These states are very weakly bound and will be discussed in more detail elsewhere.

Our results show that the parent bright singlet state $X_s^-$ with $M = 0$ remains always bound. Its binding energy initially decreases with decreasing $L$, reaches its minimum when the donor $D^+$ is very close to the heteroboundary, and then increases again. We interpret this as an indication toward a rearrangement of the type of binding in the singlet $(D^+, X_s^-)$ state: At very large distances $L$, the donor ion binds $X_s^-$ as a whole, barely affecting its internal structure. In the opposite limit of an in-well donor, the interaction of electrons with the $D^+$ is stronger than that with the hole. The donor-bound complex formed in this case is better described as an exciton bound to a neutral donor $(D^0, X)$.

Notice a systematic change in the dipole transition matrix elements $f$ in Fig. 1: as the binding energy of a complex decreases, its spatial extent increases leading to the increase in $f$. This is consistent with the notion of “giant oscillator strengths” 3.

We found just one state that only exists in the presence of the $D^+$ and does not have its free $L = \infty$ counterpart: the dark singlet state $(D^+, X_{sd}^-)$ with $M = 1$. It only becomes bound when the $D^+$ is located in a QW or very near to it. This is also the only donor-bound state that remains bound in the strictly 2D high-field limit in symmetric electron-hole systems 4. According to 1, the PL from this state goes mostly via shake-ups to $n = 1$ electron LL. As a result, the dipole transition matrix elements $f$ shown in Fig. 1 are very small.

FIG. 1: Binding energies of charged excitons $X^-$ in a 100 Å GaAs/Al$_{0.3}$Ga$_{0.7}$As QW at $B = 10$ T. Sizes of the dots are proportional to the interband dipole transition matrix elements $f$. The solid diamonds designate dark $X^-$ states.

 FIG. 2: Lifting of the Landau degeneracy in the family of bright singlet states $X_s^-$. The states are characterized by different total angular momentum projections $M = 0, -1, -2, \ldots$ and all are optically active.

In contrast to singlet states, the dark $X_{td}^-$ and bright $X_{tb}^-$ triplet states survive only for sufficiently large distances $L$ to the donor ion $D^+$ (Fig. 1). This is because
electrons in triplet states cannot simultaneously occupy the s-state in the lowest LL and, therefore, it is difficult to find a configuration with optimized electron-donor interactions. Notice the finite oscillator strengths for the PL from the donor-bound complex \((D^+, X_{td}^-)\) originating from the dark triplet state.

We stress that each free \(X^-\) state gives rise to a family of degenerate states; only the evolution of the parent \(X^-\) states is shown in Fig. 1. The degeneracy in the in-plane position of the guiding center (quantum number \(k\)) is lifted in the presence of the donor ion \(D^+\). Figure 2 demonstrates this for the family of singlet bright states \(X^-_s\). When the distance to the donor \(L\) decreases, all but one state (with \(M = 0\)) become one by one unbound. This leads to a number of optically active states with large oscillator strengths.

In conclusion, we have shown there is a multitude of donor-bound \(X^-\) states that may exhibit relatively weak dependencies of binding energies and oscillator strengths on positions of remote donors. Our results may be relevant for explanation of the PL from the dark triplet state \(X_{td}^-\) of the multiple PL peaks observed in different experiments, and of the \(X^-\) shake-ups in PL.

This work is supported in part by NSF grants DMR-0203560 and DMR-0224225, and by a College Award of Cottrell Research Corporation.

* Electronic address: adzyubenko@csub.edu

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