

Charged Magnetoexcitons in Two-Dimensions: Magnetic Translations and Families of Dark States

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We show that optical transitions of charged excitons in semiconductor heterostructures are governed in magnetic fields by a novel exact selection rule, a manifestation of magnetic translations. It is shown that the spin-triplet ground state of the quasi-two-dimensional charged exciton X^- —a bound state of two electrons and one hole—is optically inactive in photoluminescence at finite magnetic fields. Internal bound-to-bound X^- triplet transition has a specific spectral position, below the electron cyclotron resonance, and is strictly prohibited in a translationally invariant system. These results allow one to discriminate between free and disorder-affected charged excitons.

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In quasi-two-dimensional (quasi-2D) electron-hole ($e-h$) systems with low density of particles, a variety of hydrogenic few-particle complexes can be formed. Optical spectroscopy in magnetic fields is one of the basic tools for studying such complexes. Recently, much experimental [1–5] and theoretical [6–10] attention has been devoted to studying negatively X^- ($2e-h$) and positively X^+ ($2h-e$) charged excitons in magnetic fields B . These complexes are often considered to be semiconductor analogs of the hydrogen atomic H^- and molecular H_2^+ ions, respectively. In B , in addition to the spin-singlet, the higher-lying spin-triplet bound states of X^- and X^+ develop [1]. The question as to whether these complexes are mobile and free to move, or are localized—by single donor impurities [3,5,8], disorder due to long-range fluctuating potential of remote donors [4], etc.—is a matter of current controversy. To explore these issues, we theoretically address from first principles the following question: *Are there fundamental differences in optical transitions between mobile and localized charged $e-h$ systems in magnetic fields?*

For a one-component translationally invariant interacting electron system in B , the well-known Kohn theorem [11] states that intraband transitions can occur only at the bare electron cyclotron resonance (e -CR) energy $\hbar\omega_{ce} = \hbar eB/m_e c$. This is a consequence of the center-of-mass (CM) separation from internal degrees of freedom in B . For $e-h$ systems such separation is not possible and the CM and internal motions are coupled in B [12,13]. Nonetheless, for any system of charged particles in a uniform B an exact symmetry—magnetic translations—exists ([13], and references therein). It has been used to study the motion of atoms and ions [14] and neutral excitons [12] in constant magnetic and electric fields. In this paper, we introduce for charged semiconductor $e-h$ complexes in B an exact classification of states, which is based on magnetic translations. In this scheme, in addition to the total

orbital angular momentum projection M_z and spin of electrons S_e and holes S_h , an exact quantum number, the discrete oscillator quantum number k , appears. Surprisingly, only very general consideration of radiation processes in B using magnetic translations has been given [13]. To our knowledge, no selection rule associated with k has been established for dipole-allowed magneto-optical transitions of *charged* $e-h$ complexes. We show that k is conserved in the intraband and in interband magneto-optical transitions. This leads to striking spectroscopic consequences for charged excitons.

Consider a many-body Hamiltonian of interacting particles of charges e_i in a magnetic field $\mathbf{B} = (0, 0, B)$,

$$H = \sum_i \frac{\hat{\pi}_i^2}{2m_i} + \frac{1}{2} \sum_{i \neq j} U_{ij}(\mathbf{r}_i - \mathbf{r}_j). \quad (1)$$

Here $\hat{\pi}_i = -i\hbar\nabla_i - \frac{e_i}{c} \mathbf{A}(\mathbf{r}_i)$ and potentials of interparticle interactions U_{ij} can be rather arbitrary. In the symmetric gauge $\mathbf{A} = \frac{1}{2} \mathbf{B} \times \mathbf{r}$ the total angular momentum projection M_z , an eigenvalue of $\hat{L}_z = \sum_i (\mathbf{r}_i \times -i\hbar\nabla_i)_z$, is an exact quantum number. In a uniform \mathbf{B} the Hamiltonian (1) is also invariant under a group of magnetic translations whose generators are the components of the operator $\hat{\mathbf{K}} = \sum_i \hat{\mathbf{K}}_i$, where $\hat{\mathbf{K}}_i = \hat{\pi}_i - \frac{e_i}{c} \mathbf{r}_i \times \mathbf{B}$ and $[\hat{K}_{ip}, \hat{\pi}_{iq}] = 0$, $p, q = x, y$ [12–14]. $\hat{\mathbf{K}}$ is an exact integral of the motion $[H, \hat{\mathbf{K}}] = 0$. Its components commute as

$$[\hat{K}_x, \hat{K}_y] = -i \frac{\hbar B}{c} Q, \quad Q \equiv \sum_i e_i. \quad (2)$$

For neutral complexes (excitons, biexcitons) $Q = 0$ and the classification of states in \mathbf{B} is due to the two-component *continuous* vector—the 2D magnetic momentum $\mathbf{K} = (K_x, K_y)$ [12,13]. For charged systems $Q \neq 0$, and the components of $\hat{\mathbf{K}}$ do not commute. This determines

the macroscopic Landau degeneracy of eigenstates of (1). Using a dimensionless operator $\hat{\mathbf{K}} = \sqrt{c/\hbar B}|Q|\hat{\mathbf{K}}$ whose components are canonically conjugate, one obtains raising and lowering Bose ladder operators $\hat{k}_{\pm} = (\hat{k}_x \pm i\hat{k}_y)/\sqrt{2}$ such that $[\hat{k}_-, \hat{k}_+] = Q/|Q|$. Therefore, $\hat{\mathbf{K}}^2 = \hat{k}_+ \hat{k}_- + \hat{k}_- \hat{k}_+$ has the oscillator eigenvalues $2k + 1$, $k = 0, 1, \dots$. Since $[\hat{\mathbf{K}}^2, H] = 0$ and $[\hat{\mathbf{K}}^2, \hat{L}_z] = 0$, the exact charged eigenstates of (1) can be simultaneously labeled by the *discrete* quantum numbers k and M_z [13]. For charged e - h complexes in B the labeling therefore is $|kM_z S_e S_h \nu\rangle$, where ν is the ‘‘principal’’ quantum number, which can be discrete (bound states) or continuous (unbound states forming a continuum); concrete examples are given below. The $k = 0$ states are *parent states* (PS’s) within a degenerate manifold. All other daughter states, $k = 1, 2, \dots$, in each ν th family can be generated out of the PS: for, e.g., $Q < 0$,

$$|k, M_z - k, S_e S_h \nu\rangle = \frac{1}{\sqrt{k!}} \hat{k}_-^k |0, M_z, S_e S_h \nu\rangle, \quad (3)$$

where we have used $[\hat{L}_z, \hat{k}_{\pm}] = \pm \hat{k}_{\pm}$. The values of M_z that the PS’s have are determined by particulars of interactions and cannot be established *a priori* (cf. with 2D electron systems in strong B [15]).

In the dipole approximation the quantum number k is conserved in intraband and interband magneto-optical transitions. Indeed, for internal intraband transitions in the Faraday geometry (light propagating along \mathbf{B}) the Hamiltonian of the interaction with the radiation of polarization σ^{\pm} is of the form $\hat{V}^{\pm} = \sum_i (e_i \mathcal{F}_0 \hat{\pi}_{i\pm} / m_i \omega) e^{-i\omega t}$, where \mathcal{F}_0 is the radiation electric field, $\hat{\pi}_{i\pm} = \hat{\pi}_{ix} \pm i\hat{\pi}_{iy}$ (e.g., [11]). Conservation of k follows from the commutativity $[\hat{V}^{\pm}, \hat{\mathbf{K}}] = 0$ [16]. (In fact the perturbation $\hat{V} = F(\hat{\pi}_i, t)$ can be an arbitrary function of kinematic momentum operators $\hat{\pi}_i$ and time t , corresponding, e.g., to other geometries and polarization.) The other usual selection rules are conservation of spins S_e, S_h and $\Delta M_z = \pm 1$ for the envelope function in the σ^{\pm} polarization. This means that the PS’s should be connected by the dipole transition, i.e., have proper spins and $M'_z - M_z = \pm 1$. Indeed, for the transition dipole matrix element between the daughter states we have

$$\begin{aligned} \mathcal{D}_{\nu'\nu} &= \langle k', M'_z - k', S_e S_h \nu' | \hat{V}^{\pm} | k, M_z - k, S_e S_h \nu \rangle \\ &= \delta_{k',k} \delta_{M'_z, M_z \pm 1} \langle 0, M'_z, S_e S_h \nu' | \hat{V}^{\pm} | 0, M_z, S_e S_h \nu \rangle. \end{aligned} \quad (4)$$

Here we have used (3) and the operator algebra $[\hat{V}^{\pm}, \hat{k}_-] = [\hat{V}^{\pm}, \hat{k}_+] = 0$, $[\hat{k}_+, \hat{k}_-] = 1$. From (4) it is clear that $\mathcal{D}_{\nu'\nu}$ is the same in all generations and, thus, characterizes the two families of states. Similar considerations apply to interband transitions with e - h pair creation or annihilation: The interaction with the radiation field is described by the luminescence operator $\hat{\mathcal{L}}_{\text{PL}} = p_{\text{cv}} \int d\mathbf{r} \hat{\Psi}_e^{\dagger}(\mathbf{r}) \hat{\Psi}_h^{\dagger}(\mathbf{r}) + \text{H.c.}$, where p_{cv} is the

interband momentum matrix element (e.g., [17]). Here we have $[\hat{\mathcal{L}}_{\text{PL}}, \hat{\mathbf{K}}] = 0$, so that k is conserved. Because of the change of the Bloch parts in this case, the usual selection rule $\Delta M_z = 0$ holds for the envelope functions.

Conservation of k constitutes an exact selection rule for the dipole-allowed magneto-optical transitions in any charged e - h system in B . In the limiting case of low B , k can be related to the center of the cyclotron motion of the complex as a whole [13,14]. This gives some physical insight into its conservation. In the derivation above we used only translational invariance in the plane perpendicular to \mathbf{B} . Therefore, conservation of k holds for systems of *different dimensionality* and in *arbitrary* magnetic fields. At low magnetic fields the quasi-2D X^- states in higher Landau levels (LL’s) merge with the continuum corresponding to the motion of neutral excitons (see below) and become resonances. This situation is also typical for bulk 3D systems, where X^- and H^- [13] states in higher LL’s always merge with the continuum of the unbound internal z motion in lower LL’s. The established selection rule works also in this case and predicts which of the resonances are dark and which are not. Importantly, it is also applicable to semiconductors with a *complex* valence band [18]. The Hamiltonian describing the quasi-2D X^- states in a perpendicular field \mathbf{B} is of the form $\hat{H}_{X^-} = \hat{H}_h^0 + (H_{e1}^0 + H_{e2}^0 + H_{\text{int}}) \otimes \hat{I}_4$, where \hat{H}_h^0 is the 4×4 Luttinger Hamiltonian for the valence-band hole with $J_{\text{hz}} = \pm \frac{3}{2}, \pm \frac{1}{2}$ (see, e.g., [17]). The translational symmetry is always preserved $[\hat{H}_{X^-}, \hat{I}_4 \otimes \hat{\mathbf{K}}] = 0$, while the rotational symmetry is preserved in the usual axial approximation [19] $[\hat{H}_{X^-}, \hat{L}_z] = 0$, where $\hat{L}_z = \hat{L}_z \otimes \hat{I}_4 + \hat{J}_{\text{hz}}$. Therefore, the exact labeling is $|k \mathcal{M}_z P S_e \nu\rangle$, where \mathcal{M}_z is an eigenvalue of \hat{L}_z and P is the parity in the z direction. The above selection rules are formulated [18] in terms of the exact quantum numbers k and \mathcal{M}_z with P conserved for the normal incidence of light.

To demonstrate how the developed formalism works, we consider the strictly 2D e - h system with a simple valence band in the limit of high B [7,8], when $\hbar\omega_{\text{ce}}, \hbar\omega_{\text{ch}} \gg E_0 = \sqrt{\pi/2} e^2 / \epsilon l_B$ and mixing between LL’s can be neglected; $l_B = (\hbar c / eB)^{1/2}$. E_0 is the characteristic energy of Coulomb interactions, the only energy scale in the problem. The basis for the X^- states in the electron and hole LL’s ($N_e N_h$) is of the form $\phi_{n_1 m_1}^{(e)}(\mathbf{r}_e) \phi_{n_2 m_2}^{(e)}(\mathbf{R}_e) \phi_{N_h m_h}^{(h)}(\mathbf{r}_h)$ and includes different three-particle $2e$ - h states such that the total angular momentum projection, $M_z = n_1 + n_2 - m_1 - m_2 - N_h + m_h$, and LL’s $N_e = n_1 + n_2$, N_h are fixed [20]. Here $\phi_{nm}^{(e,h)}$ are the e and h single-particle factored wave functions in B (e.g., [13,14]); n is the LL quantum number and m is the single-particle oscillator quantum number ($m_{ze} = -m_{zh} = n - m$). We use the electron relative $\mathbf{r}_e = (\mathbf{r}_{e1} - \mathbf{r}_{e2})/\sqrt{2}$ and CM $\mathbf{R}_e = (\mathbf{r}_{e1} + \mathbf{r}_{e2})/\sqrt{2}$ coordinates. The electron relative motion angular momentum $n_1 - m_1$ should be even (odd) in the electron spin-singlet ($S_e = 0$) (triplet $S_e = 1$) state. An

additional canonical transformation diagonalizing $\hat{\mathbf{k}}^2$ and fixing k is performed; details will be given elsewhere [18].

The calculated three-particle $2e-h$ eigenspectra with electrons in the triplet $S_e = 1$ state in two lowest LL's are shown in Fig. 1. Generally, the eigenspectra associated with each LL consist of continuous bands of finite widths $\sim E_0$, corresponding to the extended motion of a *neutral* magnetoexciton (MX) as a whole with the second electron in a scattering state. The continuum in the, e.g., $(N_e N_h) = (10)$, LL consists of the MX band of width E_0 extending down in energy from the free (10) LL. This corresponds to the $1s$ MX in zero LL's [21] plus a scattered electron in the first LL, labeled $X_{00} + e_1$. (A similar continuum exists in zero LL's.) In addition, there is another MX band of width $0.574E_0$ also extending down in energy from the free (10) LL. This corresponds to the $2p^+$ MX [21] plus a scattered electron in the zero LL, labeled $X_{10} + e_0$. Moreover, there is a quasicontinuum above each free LL (labeled $2e + h_0$ in Fig. 1) originating from the bound internal motion of two 2D electrons in B (cf. [15]). Bound X^- states lie outside the continua. In the strictly 2D high- B limit the only family of bound X^- states in zero LL's is the triplet X_{t00}^- . There are no bound singlet X_s^- states [7,8], in contrast to the $B = 0$ case. The obtained X_{t00}^- binding energy $0.043E_0$ is in agreement with Refs. [7,8]. In the next electron LL there are no bound singlet X_s^- states, and only one family of bound triplet states X_{t10}^- . The X_{t10}^- binding energy is

$0.086E_0$, twice that of the X_{t00}^- . This is due to the fact that the two electrons in the triplet X_{t10}^- state can occupy the single-particle states with zero $e-h$ relative angular momenta $1s$ (zero LL) and $2s$ (first LL). This enhances the $e-h$ attraction relative to the ground X_{t00}^- state in which electrons can occupy an antisymmetric combination of the $1s$ and $2p^-$ single-particle states in zero LL.

We first discuss internal X^- triplet transitions. In the σ^+ polarization the inter-LL $\Delta N_e = 1$ transitions are strong and gain strength with B . Both bound-to-bound $X_{t00}^- \rightarrow X_{t10}^-$ and photoionizing bound-to-continuum transitions are possible. For the latter, due to the rich structure of the continuum, two exact selection rules (4) are easily simultaneously satisfied. As a result, the photoionizing absorption spectra have intrinsic linewidth $\sim E_0$ with two prominent peaks above the e -CR (Fig. 2). These peaks are associated with high densities of states at the edges of the two MX bands indicated in Fig. 1. Most of these qualitative features of photoionizing transitions are preserved at finite fields and confinement, where both the triplet and singlet bound X^- states exist. This has been shown by high-accuracy calculations for realistic GaAs/GaAlAs quantum wells at $B > 8$ T, which are confirmed in recent experiments and will be reported elsewhere [22]. Here we are interested in the bound-to-bound $X_{t00}^- \rightarrow X_{t10}^-$ transition. Note first that, since the final state is more strongly bound, this transition has a specific spectral position—it lies below the e -CR energy $\hbar\omega_{ce}$. However, in a translationally invariant system it is *strictly prohibited*. Indeed, the X_{t00}^- PS (with $k = 0$) has $M_z = -1$, while the X_{t10}^- PS has $M_z = 1$ (Fig. 1). It follows then that the selection rules (4) $\Delta k = 0$ and $\Delta M_z = 1$ cannot be simultaneously satisfied. This also holds at finite $B > 8$ T and in quasi-2D quantum wells

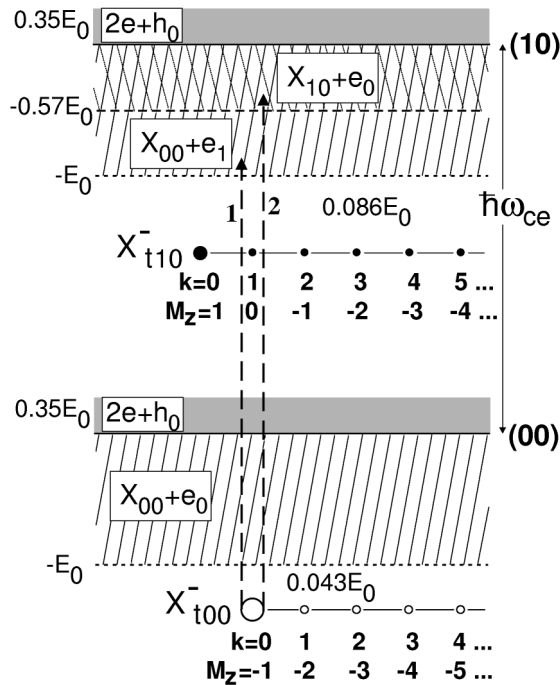


FIG. 1. Schematic drawing of bound and scattering electron triplet $2e-h$ states in the lowest LL's $(N_e N_h) = (00), (10)$. Large (small) dots correspond to the bound parent $k = 0$ (daughter $k = 1, 2, \dots$) X^- states. Allowed internal transitions must satisfy $\Delta M_z = 1$ and $\Delta k = 0$.

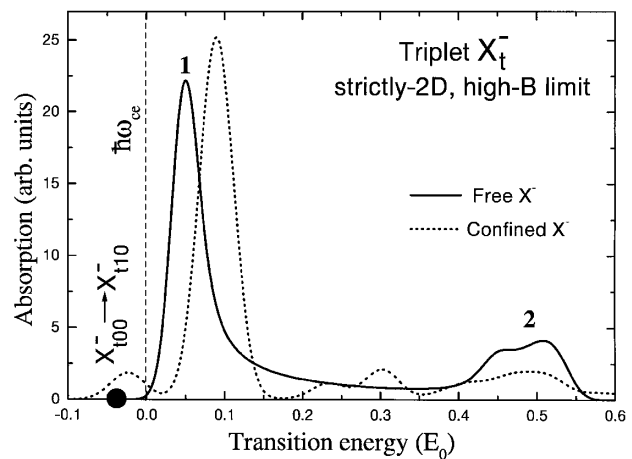


FIG. 2. Energies and dipole matrix elements of the X^- internal transitions corresponding to Fig. 1 (solid line). The solid dot shows the position of the $X_{t00}^- \rightarrow X_{t10}^-$ transition, which is forbidden for free X^- . Dashed line: the same for the X^- confined in a parabolic potential with the oscillator length $L = (\hbar/2m_e\omega_e)^{1/2} = (\hbar/2m_h\omega_h)^{1/2} = 0.4l_B$.

[22]. Localization of charged excitons, which we model by a parabolic lateral confinement $V_e = \frac{1}{2}\omega_e^2 r_e^2$ and $V_h = \frac{1}{2}m_h\omega_h^2 r_h^2$, breaks translational invariance and relaxes the selection rule. As a result, the bound-to-bound $X_{100}^- \rightarrow X_{110}^-$ transition develops *below* the e -CR (Fig. 2). Such a peak is a telltale mark of localization of triplet excitons X_t^- . The strong triplet T^- transition [20,23] of the D^- center (two electrons bound by a donor ion) can be thought of as another possible limiting case, namely, when the hole is completely localized.

Consider now photoluminescence (PL) from the triplet ground state $X_{100}^- \rightarrow \text{photon} + e_n^-$ with the electron in the n th LL in the final state; $n = 1, 2, \dots$ correspond to shake-up processes in the PL [2,3]. The PL selection rules are $\Delta k = 0$ and $\Delta M_z = 0$. The triplet X_{100}^- ground PS with $k = 0$ has $M_z = -1$ (also at finite $B > 8$ T and in quasi-2D quantum wells [8,22]), while the electron in the n th LL, with the factored wave function $\phi_{nm}^{(e)}$, has $m_z = n - m$. The corresponding optical matrix element for transition to any LL $n \geq 0$ is zero: $\langle \phi_{nm}^{(e)} | \hat{L}_{\text{PL}} | X_{100}^-(M_z = -1, k = 0) \rangle \sim \delta_{m,k=0} \delta_{n-m,-1}$. This means that the ground triplet state of an isolated X_t^- is optically inactive—*dark in PL*. In the strictly 2D high- B limit this also follows [7] from the “hidden symmetry” in e - h systems. Our result is much more general. Indeed, as discussed above, quasi-2D effects, e - h asymmetry, admixture of higher LL’s, and the complex character of the valence band [19] break neither axial nor translational symmetry. Therefore, even in the presence of these effects, the triplet stays dark—as long as the ground X_t^- PS has $M_z < 0$. Note that the quasi-2D X_s^- singlet ground PS has $M_z = 0$ [8,9,22] and is optically active in PL: $\langle \phi_{nm}^{(e)} | \hat{L}_{\text{PL}} | X_{s00}^-(M_z = 0, k = 0) \rangle \sim \delta_{m,0} \delta_{n,0}$. We see, however, that the shake-up processes are *prohibited* in PL from the isolated singlet ground state X_s^- . The question now remains why in fact the X_t^- ground state is visible in experiment in B [1–5] and the singlet X_s^- shake-up processes are commonly observed in PL—even at very low densities of excess free carriers [2,3]. Our results show that there should be mechanisms breaking the underlying exact translational and rotational symmetries. We interpret this as an indication toward a significant effect of scattering in the disorder potential. Note that as rather heavy particles, the charged excitons are very likely to be localized in real samples, especially under the confining effect of B .

In conclusion, we have shown that due to magnetic translations, dipole-allowed transitions of charged mobile semiconductor complexes are governed in magnetic fields B by a novel exact selection rule. Some experimentally observed features in interband photoluminescence of 2D charged excitons X^- in B cannot be explained without accounting for symmetry-breaking effects, an indication toward the essential role of the disorder. The appearance of the peak below the electron cyclotron resonance, corre-

sponding to the internal bound-to-bound X^- triplet transition, is a characteristic mark associated with breaking of translational invariance. We propose using this as a tool for studying the extent of X^- localization.

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