

CHARGED MOBILE COMPLEXES IN MAGNETIC FIELDS: A NOVEL SELECTION RULE FOR MAGNETO-OPTICAL TRANSITIONS

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We discuss implications of magnetic translations for internal optical transitions of charged mobile electron-hole ($e-h$) complexes and ions in a uniform magnetic field B . It is shown that transitions of such complexes are governed by a novel exact selection rule. Internal intraband transitions of two-dimensional (2D) charged excitons X^- in strong magnetic fields are considered as an illustrative example.

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Recently, there has been considerable experimental and theoretical interest in the behavior of 2D semiconductor complexes — negatively X^- ($2e-h$) and positively X^+ ($e-2h$) charged excitons in magnetic fields (e.g., [1–5] and references therein). These three-particle bound states can be considered as analogs of the hydrogen atomic H^- and molecular H_2^+ ions, respectively. The application of magnetic field B changes the hydrogenic spectra drastically (e.g., [6–8]). As an example, while at $B = 0$ the H^- supports in 3D only one bound singlet state, at any finite B there appear [7] bound triplet H^- states and infinite number of quasi-bound states (resonances) associated with higher Landau levels (LL's). In 2D systems, it is the X^- triplet that becomes the ground state in high magnetic fields [2, 3]. Singlet and triplet X^- and X^+ states are observed in 2D systems by means of interband optical magneto-spectroscopy [1]. Intraband magneto-spectroscopy, in which internal transitions from populated (ground) to excited states are induced by a photon, can provide additional information about binding of hydrogen-like complexes. In the σ^+ polarization internal transitions in high B are predominantly induced to the next electron LL. Such photoionizing bound-to-continuum X^- singlet and triplet transitions in the far-infrared (FIR) have been predicted theoretically and recently observed experimentally in GaAs quantum wells [4]. In this work we describe some general implications of the existing exact symmetry — magnetic translations — for internal transitions of charged mobile $e-h$ complexes in B . We also present theory predictions for internal transitions from the X^- ground triplet state in the σ^- polarization in high B . A preliminary account of some of these results and implications for interband magneto-optics of X^- have been reported in [5].

Consider Hamiltonian of interacting particles of charges e_i in a magnetic field 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 $\mathbf{B} = (0, 0, B)$ the Hamiltonian (1) is also invariant under a group of magnetic translations whose generators are the components of the operator $\hat{\mathbf{K}} = \sum_j \hat{\mathbf{K}}_j$, $\hat{\mathbf{K}}_j = \hat{\pi}_j - \frac{e_j}{c}\mathbf{r}_j \times \mathbf{B}$ (e.g., [6, 7]). $\hat{\mathbf{K}}$ is an exact integral of the motion $[H, \hat{\mathbf{K}}] = 0$ whose components commute in B as

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

while $[\hat{K}_{ip}, \hat{\pi}_{jq}] = 0$, $p, q = x, y$. For neutral complexes (atoms, excitons, biexcitons) $Q = 0$, and classification of states in \mathbf{B} is due to the two-component continuous vector — the 2D magnetic momentum $\mathbf{K} = (K_x, K_y)$ [6, 7]. For charged systems $Q \neq 0$ and the components of $\hat{\mathbf{K}}$ do not commute, which determines the macroscopic Landau degeneracy of exact 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 $\hat{k}_{\pm} = (\hat{k}_x \pm i\hat{k}_y)/\sqrt{2}$: $[\hat{k}_-, \hat{k}_+] = Q/|Q|$ (see (5) below). 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 [7].

The usual optical selection rules for the dipole-allowed transitions in the Faraday geometry (the light propagates along \mathbf{B}) is conservation of spin and $\Delta M_z = \pm 1$ for the left- and right-circularly polarized light σ^{\pm} . There is an additional selection rule: the quantum number k is conserved. Indeed, the Hamiltonian $\hat{V}^{\pm} = \sum_i (e_i \mathcal{F}_0 \hat{\pi}_i^{\pm} / m_i \omega) e^{-i\omega t}$ describing interaction with the light of polarization σ^{\pm} (\mathcal{F}_0 is the radiation electric field, $\hat{\pi}_i^{\pm} = \hat{\pi}_{ix} \pm i\hat{\pi}_{iy}$) commutes with $\hat{\mathbf{K}}_i$ and, therefore,

$$[\hat{V}^{\pm}, \hat{\mathbf{k}}^2] = 0 \Rightarrow k \text{ is conserved.} \quad (3)$$

In fact perturbation $\hat{V} = F(\hat{\pi}_i, t)$ can be an arbitrary function of kinematic momentum operators $\hat{\pi}_i$ and time t . In some limiting cases (e.g., at low fields B) k can be associated [7] with the center of the cyclotron motion of a charged system as a whole. This gives some physical insight into its conservation. This selection rule is applicable to any charged e - \hbar system in B . In particular, to mobile complexes — charged excitons X^- , X^+ , multiply-charged excitons X^{-n} (i.e., bound complexes $(n+1)e$ - \hbar with $n > 1$, which can exist in special quasi-2D geometries [9]), and to charged multiple-excitons X_N^- (X_N - e), which exist in 2D systems in high B [2]. In deriving (3) we only used translational invariance in the plane perpendicular to \mathbf{B} . Therefore, (3) holds in arbitrary magnetic fields and for systems of different dimensionality, including semiconductors with complex valence band described by the Luttinger Hamiltonian [10]. The selection rule (3) is also valid for internal transitions in electron systems. Note that for a translationally invariant one-component (e.g., electron) system with constant charge-to-mass ratio $e_i/m_i = \text{const}$, the well-known Kohn theorem [11] states that internal transitions can occur only at the bare electron cyclotron (e -CR) energy $\hbar\omega_{ce} = \hbar eB/m_e c$. This is a consequence of the operator algebra $[H, \hat{V}^{\pm}] = \pm \hbar\omega_{ce} \hat{V}^{\pm}$ involving the center-of-mass *inter*-LL ladder operators. On the other hand, (3) is based on the algebra of *intra*-LL ladder operators. However, for one-component systems the center-of-mass decouples from internal degrees of freedom in

B and both theorems – though based on different operator algebras – give in this case equivalent predictions.

To make further discussion concrete, we shall consider transitions in the σ^- polarization in a 2D three-particle $2e-h$ system with the Coulomb interactions, simple valence band, and in the limit of high magnetic fields:

$$\hbar\omega_{ce}, \hbar\omega_{ch}, |\hbar\omega_{ce} - \hbar\omega_{ch}| \gg E_0 = \sqrt{\frac{\pi}{2}} \frac{e^2}{\epsilon l_B}, \quad l_B = \left(\frac{\hbar c}{eB}\right)^{1/2}. \quad (4)$$

Then mixing between different LL's can be neglected and the X^- states can be classified by total electron and hole LL numbers, $(N_e N_h)$. The corresponding basis for X^- is of the form [12] $\phi_{n_1 m_1}^{(e)}(\mathbf{r}) \phi_{n_2 m_2}^{(e)}(\mathbf{R}) \phi_{N_h M_h}^{(h)}(\mathbf{r}_h)$, and includes different three-particle $2e-h$ states such that the total angular momentum projection $M_z = N_e - N_h - m_1 - m_2 + M_h$, and LL's $N_e = n_1 + n_2$, N_h are fixed. Here $\phi_{nm}^{(e,h)}$ are the e - and h - single-particle factored wave functions in B ; n is the LL quantum number and m is the oscillator quantum number [$m_{ze(h)} = \begin{smallmatrix} + \\ - \end{smallmatrix} (n - m)$]. We use the electron relative and center-of-mass coordinates: $\mathbf{r} = (\mathbf{r}_{e1} - \mathbf{r}_{e2})/\sqrt{2}$ and $\mathbf{R} = (\mathbf{r}_{e1} + \mathbf{r}_{e2})/\sqrt{2}$. Permutational symmetry requires that for electrons in the spin-singlet s (triplet t) state the relative motion angular momentum $n_1 - m_1$ should be even (odd). To make this basis compatible with magnetic translations, i.e., to fix k , an additional Bogoliubov canonical transformation should be performed [5].

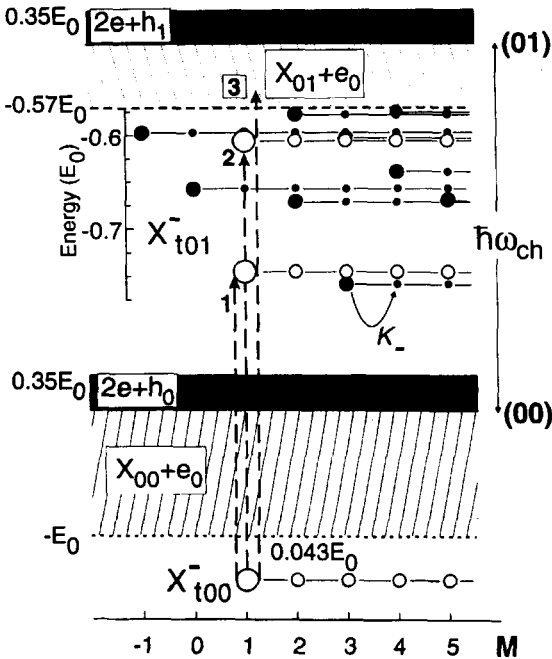


Fig.1. Schematic drawing of bound and scattering electron triplet $2e-h$ states in the lowest LL's $(N_e N_h)=(00), (01)$. The quantum number $M = -M_z$ for the $(N_e N_h)=(00)$ states and $M = -M_z - 1$ for the $(N_e N_h)=(01)$ states. Large (small) dots correspond to the bound parent $k = 0$ (daughter $k = 1, 2, \dots$) X^- states. Allowed strong transitions in the σ^- polarization must satisfy $\Delta N_h = 1$, $\Delta M_z = -1$, and $\Delta k = 0$. Filled dots in the (01) LL correspond to families of dark X_{t01}^- states (see text)

The calculated eigenspectra of the three-particle $2e-h$ states with two electrons in the spin-triplet state t ($S_e = 1$) are shown for two lowest $(N_e N_h)=(00), (01)$ LL's in Fig.1. The spectra consist of continua, which correspond to the motion of a neutral magnetoexciton (MX) [4, 8]. In addition, there are discrete bound X^- states lying outside the continua in which internal motions of all particles are finite. The continuum in the

$(N_e N_h) = (00)$ LL consists of a MX band of the width E_0 extending down in energy from the free (00) LL and corresponding to the $1s$ MX ($N_e = N_h = 0$) [8] plus a scattered electron in the zero LL, labeled $X_{00} + e_0$. In the next hole LL $(N_e N_h) = (01)$, the MX band is of the width $0.574E_0$ [8] and corresponds to the $2p^-$ MX ($N_e = 0, N_h = 1$) plus a scattered electron in $N_e = 0$ LL, labeled $X_{01} + e_0$. Moreover, there is a band above each free LL originating from the bound internal motion of two 2D electrons in B (labeled $2e + h_{N_h}$) [4]. The spectra of the discrete bound X^- states are the following. In the lowest $(N_e N_h) = (00)$ LL there exists only one bound X_{i00}^- triplet state, lying below the lower edge of the $1s$ MX band, whose binding energy is $0.043E_0$ [2, 3]. In the next hole LL $(N_e N_h) = (01)$ the $2p^-$ MX band is narrow, and there appear *many* bound X_{i01}^- states lying below the continuum edge (Fig.1). This should be contrasted with the situation in the next electron LL $(N_e N_h) = (10)$, where only one bound triplet state X_{i10}^- exists [4].

In the σ^- polarization, the h -CR-like inter-LL $\Delta N_h = 1$ transitions are strong and gain strength in B . These are allowed by the usual selection rules: spin conserved, $\Delta M_z = -1$. Consider first photoionizing X^- transitions in which final three-particle states belong to the $X_{01} + e_0$ continuum. There is an onset at the edge indicated in Fig.1 by transition 3. It occurs at an energy $\hbar\omega_{ch} + 0.469E_0$, i.e. above the h -CR at an energy that equals the difference in the $1s$ and $2p^-$ MX binding energies, plus the X_{i00}^- binding energy. This transition may be thought of as the $1s \rightarrow 2p^-$ internal transition of the MX [13], which is shifted and broadened by the presence of the second electron. Photoionizing transitions to the $2e + h_1$ band have extremely small oscillator strengths and can be neglected.

In order to understand bound-to-bound transitions $X_{i00}^- \rightarrow X_{i01}^-$ we shall describe the structure of X^- states in more detail. Generally, there exist *families* of macroscopically degenerate X^- states in B . Each i -th family starts with its *Parent State* (PS) $|\Psi_{M_z}^{(P_i)}\rangle$, which has $k = 0$ and the maximal possible in the family value of M_z (cf. with translationally invariant states in 2D electron systems in strong B [14]). The normalized daughter states with $k = 1, 2, \dots$ in the i -th family, $|\Psi_{M_z'}^{(D_i)}\rangle$, are constructed out of the PS with the help of the ladder operators:

$$|\Psi_{M_z-l}^{(D_i)}\rangle = \frac{1}{\sqrt{l!}} \hat{k}_-^l |\Psi_{M_z}^{(P_i)}\rangle, \quad (5)$$

where we used $[\hat{L}_z, \hat{k}_-] = -\hat{k}_-$. Conservation of k implies therefore that internal transitions in the σ^\pm polarization, satisfying the usual selection rule $\Delta M_z = \pm 1$, spin conserved, are allowed only between the states from families such that their PS's are connected by the dipole transition $|\Psi_{M_z'}^{(P_j)}\rangle \rightarrow |\Psi_{M_z}^{(P_i)}\rangle$, i.e. have $M_z' = M_z \pm 1$. Indeed, for the transition dipole matrix element between the daughter states in the m -th and n -th generations we have from (5):

$$\mathcal{D}_{ij} = \langle \Psi_{M_z'-m}^{(D_i)} | \hat{V}^\pm | \Psi_{M_z-n}^{(D_j)} \rangle = \frac{1}{\sqrt{n!m!}} \langle \Psi_{M_z'}^{(P_i)} | \hat{k}_+^m \hat{V}^\pm \hat{k}_-^n | \Psi_{M_z}^{(P_j)} \rangle. \quad (6)$$

From the commutativity $[\hat{V}^\pm, \hat{k}_-] = [\hat{V}^\pm, \hat{k}_+] = 0$ we see that either \hat{k}_- annihilates the left-PS ($n > m$) or \hat{k}_+ annihilates the right-PS ($n < m$) in (6). Therefore, $\mathcal{D}_{ij} = 0$, unless $n = m$ and $M_z' - M_z = \pm 1$. From the operator algebra it is clear that \mathcal{D}_{ij} is the same in all generations and, thus, characterizes the two families of states.

This selection rule, due to a rich structure of the continuum, is easily satisfied for bound-to-continuum transitions. However, considering many families of the bound X_{i01}^-

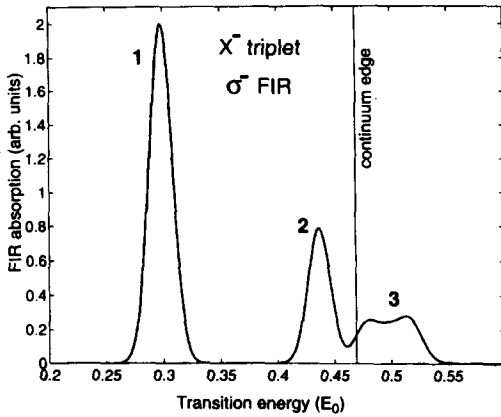


Fig.2. Energies (counted from $\hbar\omega_{ch}$, in units of $E_0 = \sqrt{\pi/2} e^2/\epsilon l_B$) and dipole matrix elements of the inter-LL $\Delta N_h = 1$ transitions (Fig. 1) from the ground triplet X_{t00}^- state in the high-field limit. Spectra have been convoluted with the Gaussian of the $0.02E_0$ width

states in the next hole LL ($N_e N_h$)=(01), we see that there are only two PS's that are connected by the FIR transition (Figs.1,2). Therefore all families except these two are dark, i.e., are not accessible by internal transitions from the ground X_{t00}^- bound states: There exist only two strong bound-to-bound $X_{t00}^- \rightarrow X_{t01}^-$ transitions in the σ^- polarization, both lying above the \hbar -CR (cf. [4]). Breaking of translational invariance (e.g., by impurities) would make transitions to many dark states possible and lead to drastic changes in the absorption spectra. We note also that in 2D systems the X^- states belonging to higher LL's are genuinely discrete only in sufficiently strong B [cf. (4)]. With decreasing B , the discrete states merge with the MX continuum of lower LL's and become resonances. Such situation is also typical for bulk 3D systems, where X^- (and H^-) states in higher LL's always merge with the continuum of the unbound internal z -motion in lower LL's [7]. Since the quantum number k is still rigorously conserved, the selection rule predicts which of the resonances are dark and which are not. The absorption spectra in such situation can have an asymmetric form typical for Fano-resonances.

In conclusion, we demonstrated that internal optical transitions of charged mobile complexes and ions in magnetic fields are governed by a novel exact selection rule, a manifestation of magnetic translational invariance. Internal bound-to-bound transitions of charged excitons X^- in 2D systems in B should be very sensitive to breaking of translational invariance (by impurities, disorder etc.). This can be used for studying the extent of X^- localization in quantum wells.

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