Charged mobile complexes in magnetic fields: a novel selection rule for magnetooptical transitions

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The implications of magnetic translations for internal optical transitions of charged mobile electron–hole ($e-h$) complexes and ions in a uniform magnetic field B are discussed. 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. © *1999 American Institute of Physics.* [S0021-3640(99)00520-4]

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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 (see, e.g., Refs. 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 a magnetic field B changes the hydrogenic spectra drastically (see, e.g., Refs. 6–8). As an example, whereas at $B=0$ the H^- ion supports in 3D only one bound singlet state, at any finite B there appear[7] bound triplet H^- states and an infinite number of quasi-bound states (resonances) associated with higher Landau levels (LLs). 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 magnetospectroscopy.[1] Intraband magnetospectroscopy, in which internal transitions from populated (ground) to excited states are induced by a photon, can provide additional information about binding of hydrogenlike 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 theoretical 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 magnetooptics of X^- have been reported in Ref. 5.

Consider the Hamiltonian of interacting particles of charges e_i in a magnetic field $\mathbf{B}$

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

here $\hat{\pi}_i = -i\hbar\nabla_i - \frac{e_i}{c}\mathbf{A}(\mathbf{r}_i)$, and the interparticle interaction potentials 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 $\hat{L}_z = \Sigma_i(\mathbf{r}_i \times -i\hbar\nabla_i)_z$, 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 $\hat{\mathbf{K}} = \Sigma_j \hat{\mathbf{K}}_j$, $\hat{\mathbf{K}}_j = \hat{\pi}_j - (e_j/c)\mathbf{r}_j \times \mathbf{B}$ (see, e.g., Refs. 6,7). The operator $\hat{\mathbf{K}}$ is an exact integral of the motion, $[H,\hat{\mathbf{K}}] = 0$, whose components commute in B as

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

while $[\hat{K}_{ip}, \hat{\pi}_{jq}] = 0$, $p,q = x,y$. For neutral complexes (atoms, excitons, biexcitons) $Q = 0$, and the states in $\mathbf{B}$ are classified according to the two-component continuous vector — the 2D magnetic momentum $\mathbf{K} = (K_x, K_y)$ (Refs. 6,7). For charged systems $Q \neq 0$, and the components of $\hat{\mathbf{K}}$ do not commute; this determines the macroscopic Landau degeneracy of the exact eigenstates of (1). Using the dimensionless operator $\hat{\mathbf{k}} = \sqrt{c/\hbar B|Q|}\,\hat{\mathbf{K}}$ whose components are canonically conjugate, one obtains the 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 $2k+1$, $k = 0,1,\ldots$. 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 (Ref. 7).

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

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

In fact the perturbation $\hat{V} = F(\hat{\pi}_i, t)$ can be an arbitrary function of the 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 $e-h$ system in B. In particular, it applies to mobile complexes — charged excitons X^-, X^+, multiply charged excitons X^{-n} (i.e., bound complexes $(n+1)e-h$ 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, relation (3) holds in arbitrary magnetic fields and for systems of different dimensionality, including semicon-

ductors with a 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 $e_i/m_i =$ 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, relation (3) is based on the algebra of *intra*-LL ladder operators. However, for one-component systems the center-of-mass motion decouples from the internal degrees of freedom in B, and the theorems — though based on different operator algebras — give equivalent predictions in this case.

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

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

Then mixing between different LLs can be neglected and the X^- states can be classified according to the total electron and hole LL numbers, $(N_e N_h)$. The corresponding basis for X^- is of the form[12] $\phi^{(e)}_{n_1 m_1}(\mathbf{r})\,\phi^{(e)}_{n_2 m_2}(\mathbf{R})\,\phi^{(h)}_{N_h M_h}(\mathbf{r}_h)$, 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 LLs $N_e = n_1 + n_2$, N_h are fixed. Here $\phi^{(e,h)}_{nm}$ 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 $[m_{ze(h)} = {}^{+}_{(-)}(n-m)]$. 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]

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) LLs 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 the internal motions of all the particles are finite. The continuum in the $(N_e N_h) = (00)$ LL consists of a MX band of 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 zeroth LL, labeled $X_{00} + e_0$. In the next hole LL $(N_e N_h) = (01)$, the MX band is of width $0.574 E_0$ (Ref. 8) and corresponds to the $2p^-$ MX ($N_e = 0$, $N_h = 1$) plus a scattered electron in the $N_e = 0$ 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^-_{t00} triplet state, lying below the lower edge of the $1s$ MX band, the binding energy of which is $0.043 E_0$ (Refs. 2,3). In the next hole LL $(N_e N_h) = (01)$ the $2p^-$ MX band is narrow, and there appear *many* bound X^-_{t01} states lying below the continuum edge (Fig. 1). This should be contrasted with the
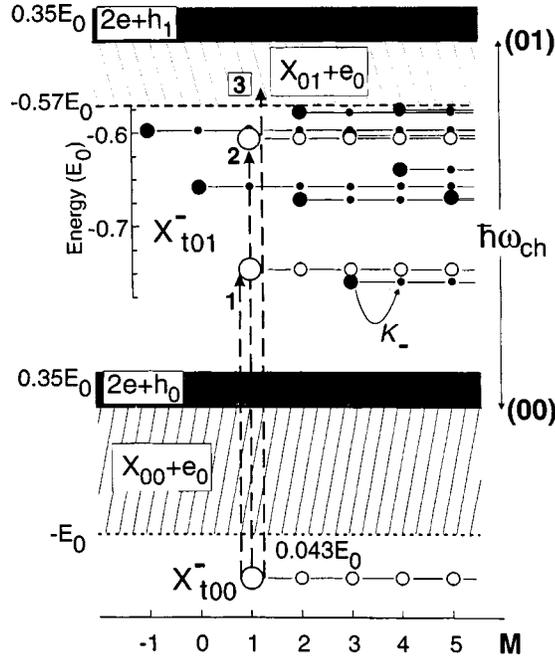
FIG. 1. Schematic drawing of bound and scattering electron triplet $2e-h$ states in the lowest LLs $(N_e N_h)$ $=(00)$, (01). The quantum number $M=-M_z$ for the $(N_e N_h)=(00)$ states and $M=-M_z-1$ for the $(N_e N_h)$ $=(01)$ states. Large (small) dots correspond to the bound parent $k=0$ (daughter $k=1,2,\ldots$) 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).

situation in the next electron LL $(N_e N_h)=(10)$, where only one bound triplet state X^-_{t10} 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 the photoionizing X^- transitions in which the 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^-_{t00} binding energy. This transition may be thought of as the $1s\rightarrow2p^-$ 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 $X^-_{t00}\rightarrow X^-_{t01}$, let us describe the structure of the X^- states in more detail. Generally, there exist *families* of macroscopically degenerate X^- states in B. Each ith family starts with its *parent state* (PS) $\left|\Psi^{(P_i)}_{M_z}\right\rangle$, for which $k=0$ and M_z has its maximum possible value for that family (cf. with translationally invariant states in 2D electron systems in strong B).[14] The normalized daughter
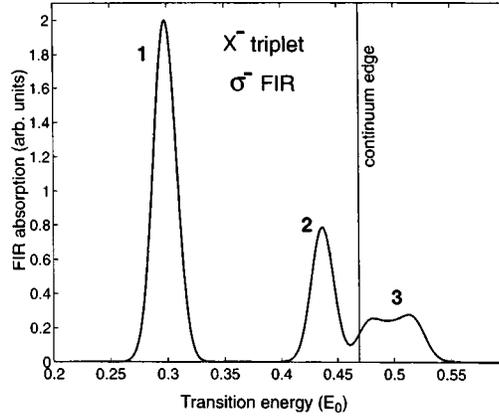
FIG. 2. Energies (counted from $\hbar\omega_{ch}$, in units of $E_0 = \sqrt{\pi/2}\, e^2/\epsilon l_B$) and dipole matrix elements of the inter-LL $\Delta N_h = 1$ transitions (Fig. 1) from the ground triplet X_{t00}^- state in the high-field limit. The spectra have been convoluted with a Gaussian of width $0.02E_0$.

states with $k = 1,2,\ldots$ in the ith family, $|\Psi_{M_z'}^{(D_i)}\rangle$, are constructed from the PS with the help of the ladder operators:

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

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

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

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 it characterizes the two families of states.

This selection rule, due to the rich structure of the continuum, is easily satisfied for bound-to-continuum transitions. However, considering many families of the bound X_{t01}^- states in the next hole LL $(N_e N_h) = (01)$, we see that there are only two PSs that are connected by a FIR transition (Figs. 1 and 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 $X_{t00}^- \to X_{t01}^-$ transitions in the σ^- polarization, both lying above the h–CR (cf. Ref. 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 LLs are genuinely discrete only in a sufficiently strong B [cf. (4)]. With decreasing B, the discrete states merge with the MX continuum of lower LLs and become resonances. Such a situation is also typical for bulk 3D systems, where X^- (and H^-) states in higher LLs always merge with the continuum of the unbound internal z-motion in lower LLs.[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 have 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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