Internal transitions of negatively charged magnetoexcitons in quantum dots

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Abstract
We report calculations of oscillator strengths for the far infrared absorption of light by the excitonic complexes $X^{-n}$ (the excess charge, $n$, ranging from one to four) confined in quantum dots. The magnetic field is varied in an interval which corresponds to 'filling factors' between $2$ and $3/5$. Electron–hole interaction effects are seen in the deviations of the peak positions from the Kohn lines, and in the spreading of the oscillator strengths over a few final states. Transition densities are used as an additional tool to characterize the absorption peaks.

The lateral confinement potential of electronic quantum dots is known to be, to a good approximation, parabolic \cite{1}. Thus, the far infrared (FIR) absorption of light by the dots exhibits a single peak, the Kohn mode, whose position is split in a magnetic field:

$$\Delta E_{\pm} = \hbar \Omega_e \pm \frac{\hbar \omega_{ce}}{2}. \quad (1)$$

In equation (1), $\omega_{ce}$ is the electron cyclotronic frequency, $\Omega_e = \sqrt{\omega_{0e}^2 + \omega_{ce}^2 / 4}$, and $\omega_{0e}$ is the frequency of the lateral confinement potential. The $\pm$ signs in $\Delta E$ correspond, respectively, to the absorption of circularly polarized photons $\sigma^\pm$.

If a valence hole is added, but the electron–hole (e–h) interaction is neglected for the moment, a similar mode appears for the hole:

$$\Delta E_{\pm} = \hbar \Omega_h \mp \frac{\hbar \omega_{ch}}{2}. \quad (2)$$

The e–h interactions, however, destroy this simple picture of FIR absorption by spreading out the oscillator strength over a few final states, whose positions are moved out from the Kohn lines (1) and (2). In the present paper, we study these effects by computing the FIR absorption of light by $X^{-n}$ complexes confined in a quantum dot. The excess charge, $n$, is varied from 1 to 4, that is from the so-called trion (2 electrons, one hole) to the 5-electron, one-hole system.

Experimentally, blue shifts of the main $\sigma^+$ peaks of charged excitons were observed in doped quantum wells as the background electron density in the well increases \cite{2}. The
experimental technique used is the detection of FIR resonances by measuring changes in the intensity of a single photoluminescence line when a FIR radiation is superposed. No experimental results for quantum dots, to the best of our knowledge, exist, and the only theoretical computation is that of the trion in relatively high magnetic fields [3].

We employ a simple two-dimensional two-band model to describe the dot. The magnetic field is applied normally to the dot plane. GaAs electron- and heavy-hole masses, and dielectric constant are used: \( m_e / m_0 = 0.067 \), \( m_h / m_0 = 0.115 \), \( \kappa = 12.5 \). The lateral confinement is \( \hbar \omega_0 = 1.8 \text{ meV} \). This means that ‘filling factors’ near one are reached with relatively low magnetic fields, \( B = 3.3 \text{ T} \). The latter was verified in a 5-electron system in which the Coulomb interactions are weakened by a factor 0.6 in order to account for the quasibidimensionality of the dot (we have in mind dots etched from a 20–25 nm wide quantum well, for example). For holes, we choose \( m_e \omega_0 = m_h \omega_0 \), leading to the same confinement length as for electrons. Calculations include 10–15 Landau levels (LLs) in the smallest system, the trion, and 3–4 LLs in the largest one, which correspond, respectively, to bases spanned by approximately 40 000 and 350 000 two-dimensional Slater determinants. The estimated errors are 0.02 meV for the excitation energies, and 0.02 for the normalized oscillator strengths.

We summarize in figure 1 the results of our calculations for the trion. The exact meaning of each panel in this figure is as follows. Let us consider, for example, the absorption of \( \sigma^+ \) photons by the trion from the lowest state with total electronic spin \( S_e = 0 \) (singlet state), figure 1(b). According to the selection rules, transitions to singlet states in which the angular momentum projection is increased by one unit, \( \Delta L_z = 1 \), take place. The normalized oscillator strengths are represented as circles centred at the positions of the resonances, and with radii proportional to the strengths. The strength normalization is based on the energy-weighted sum

\[ \Delta E \text{ [meV]} \]

Figure 1. Oscillator strengths for the trion in spin triplet and singlet states and \( \sigma^\pm \) polarized light.
At low magnetic fields, the oscillator strengths are concentrated on e-bands, whereas for high \( B \) only the e-bands in \( \sigma^+ \) polarization, and the h-bands in \( \sigma^- \) polarization survive.

Most of these features can be understood on simple grounds. Let us consider, for example, \( B = 0 \) and we make the rough approximation in which the e–h interaction is \( V_{e-h} = -(N e^2/\kappa/|\vec{x}_h - \vec{X}|) \), where \( \vec{x}_h \) and \( \vec{X} \) are hole and electron centre of mass coordinates. For simplicity, we take \( \omega_h = \omega_0 = \omega \). Then, the \( N \)-electron, one-hole Hamiltonian, which includes interaction with the FIR radiation, takes the following form:

\[
H = \left\{ \frac{p^2}{2M} + \frac{1}{2} M \omega^2 X^2 + \frac{1}{2} \mu \omega^2 X^2 \right\} + \left\{ \frac{p^2}{2\mu} + \frac{1}{2} \mu \omega^2 \vec{X}^2 \right\} N_e + \frac{N(m_e + m_h)}{Nm_e + m_h} \vec{E}(t) \cdot \vec{x} + H_{int}^{(e)},
\]

where we introduced \( \vec{x} = \vec{x}_h - \vec{X}_e, \vec{X} = (m_h \vec{x}_h + N_m \vec{X}_e)/M, M = m_h + N m_e, \mu = N m_e m_h/M, \vec{E} \) is the light polarization vector, and \( H_{int}^{(e)} \) describes the internal motion of the electronic subsystem, which is decoupled in this approximation. Then, as follows from (4), only the position of hole resonances will be affected by the e–h interactions, whereas the oscillator strength of the electron resonance will be stronger. That is qualitatively the same as happens in figure 1, with the h-bands deviating strongly from the Kohn lines, and the e-bands concentrating the oscillator strengths at low \( B \).

On the other hand, for high magnetic fields one can compute a sum rule like (3) with the inclusion of only single-particle states in the first LL. The right-hand side of this sum rule is given by \( 2\hbar(\Omega_e - h\omega_0)/2)N_e + N m_e/m_h \). This means that transitions within the first LL are suppressed as \( B \) increases. Only e-bands in \( \sigma^+ \) polarization and h-bands in \( \sigma^- \) polarization have nonvanishing strengths in this limit.

The fact about the blueshift of singlet bands has also a simple interpretation. Indeed, in singlet states, the electronic cloud is more compactly localized around the hole than in triplet states. Consequently, e–h interactions are stronger and the collective motion of the electrons, for example, will have higher excitation energies. With respect to the excited, e^*-bands, and the change of slopes, we ascribe these features to mode mixing, i.e. to e- and h-band admixtures. Oscillations of the electron cloud in counterphase with the hole cloud are the dominant resonances in the FIR absorption of neutral systems [5, 6]. In charged excitons, the Kohn modes dominate, but there are states resembling these counterphase oscillations, which can be viewed as mode admixtures.
To conclude the analysis of the trion, we stress that for bands whose energy increases with $B$, the number of levels below the band also increases. This means that the ‘transfer’ of oscillator strength between resonances in a band goes through ‘level collisions’ as the magnetic field varies. A similar situation was reported for the biexciton [6].

Excitonic complexes with excess charge $n > 1$ exhibit the same qualitative features (i)–(iv) mentioned above for the trion. There are, in addition, new aspects which can be summarized as follows.

(I) These complexes undergo ground-state (gs) rearrangements as $B$ is varied between 1 and 5 T. Thus, resonances in a band may correspond to transitions which start at different initial states.

On the other hand, as a function of $n$:

(II) The main $e$-band in $\sigma^+$ polarization first experiences a redshift (for $n = 3$ and spin-polarized states it is already below the Kohn line), and then for $n = 4$ (and presumably for larger values also) is blueshifted.

(III) The main $h$-band in $\sigma^-$ polarization moves down in energy from $n = 1$ up to 4 staying, however, above the Kohn line.

(IV) The $e$- or $e^*$-bands in $\sigma^-$ polarization become redshifted up to $n = 4$.

We show in figure 2 the results for the FIR absorption by the $X^{4-}$ complex in $S_e = 5/2$ (spin-polarized) and $S_e = 3/2$ states, and $\sigma^\pm$ light polarizations. The main $e$- and $h$-bands, and even the $e^*$-bands, the blueshift of $S_e = 3/2$ bands with respect to $S_e = 5/2$ bands, etc are observed.

**Figure 2.** Oscillator strengths for the $X^{4-}$ complex. See the explanation in the main text.
Let us consider the question about gs rearrangements and their consequences. In the trion, no such phenomenon exists. The lowest triplet state is always a state with $L_z = -1$, and the lowest singlet state is a state with $L_z = 0$. Mode mixing proceeds continuously, leading to change of band slopes at low magnetic fields, etc. In the larger complexes, gs rearrangements may lead to discontinuous variations of the properties of a resonance along a band, because a given oscillation mode may not be supported by the new gs. As a consequence, the oscillator strength is transferred to a state with different properties.

This effect is seen in the transition densities [4], $\langle \nu | \hat{\rho}(\vec{r}) | i \rangle$, where $\hat{\rho}(\vec{r})$ is the one-particle density operator. Transition densities enter the expression for the time-dependent density, which is the result of a perturbation (the external FIR radiation field) around the initial state:

$$\rho(\vec{r}, t) = \rho_i(\vec{r}) + \sum_{\nu} [\langle i | \hat{\rho}(\vec{r}) | \nu \rangle C_{\nu} e^{-iE_{\nu}t/\hbar} + \langle \nu | \hat{\rho}(\vec{r}) | i \rangle C^*_{\nu} e^{iE_{\nu}t/\hbar}] + \ldots,$$

(5)

where the $C_{\nu}$ are the coefficients of the spectral decomposition of the perturbed state. In figures 3(a) and (b), we present the two leading e-resonances at $B = 1$ T in the spin-polarized $S_e = 5/2$ system, and $\sigma^+$ polarization. Both resonances are characterized by dominant electronic oscillations and counterphase oscillations of electrons and holes. At $B = 1.6$ T we observe the first gs rearrangement. The gs angular momentum jumps from $L_z = -5$ to $-10$. The transition density of the leading e-resonance is given in figure 3(c), showing an additional nodal line. The qualitative form of the transition density for the leading e-resonance persists up to $B = 4$ T. At $B = 5$ T a new gs rearrangement takes place, in which the momentum jumps...
to $L_z = -14$. The leading e-resonance is shown in figure 3(d). The qualitative behaviour changes again. The conclusion is that, in the larger complexes, resonances along a band may correspond to transitions from different initial states, and represent qualitatively different oscillation modes against these gs.

Finally, let us stress that the shift of resonance bands as a function of the excess charge $n$ is a result of Coulomb interactions in these few-body complexes. Unlike the quantum-well case [2], we obtain a redshift of the main e-band in $\sigma^+$ polarization for $n = 3$. For $n = 4$, the band moves again to higher excitation energies. A qualitative understanding of the behaviour of these bands is still lacking.

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