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Charged excitons in quantum dots: novel magnetic behavior and Auger processes

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Abstract

We describe theoretically multiply-charged excitons interacting with a continuum of delocalized states. Such excitons exist in relatively shallow quantum dots and have been observed in recent optical experiments on InAs self-assembled dots. The interaction of an exciton and delocalized states occurs via Auger-like processes. To describe the optical spectra, we employ the Anderson-like Hamiltonian by including the interaction between the localized exciton and delocalized states of the wetting layer. In the absence of a magnetic field, the photoluminescence line shapes exhibit interference effects. When a magnetic field is applied, the photoluminescence spectrum demonstrates anticrossings with the Landau levels of the extended states. We show that the magnetic-field behavior of charged excitons is very different to that of diamagnetic excitons in three and two-dimensional systems.

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Keywords: Quantum dot; Exciton; Magnetic field

In voltage-tunable structures, self-assembled quantum dots (QDs) are embedded between two contacts [1,2]. This makes it possible to control the number of electrons in a QD by application of voltage U_{gate} (Fig. 1a). In addition, the self-assembly process means that the QDs are formed on a thin wetting layer (WL) (Fig. 1a). The WL acts as a two-dimensional (2D) quantum well. The photoluminescence (PL) spectrum of such structures demonstrates narrow peaks due to localized charged excitons X^{n-}. Such an exciton contains $n+1$ electrons and one hole. In the exciton, the hole is optically generated; the electrons are supplied by tunneling from the back contact and by the optical excitation. With increasing gate voltage, the exciton charge changes abruptly at particular values of U_{gate}. Thus, the PL spectrum as a function of the gate voltage has regions where the excitonic charge is constant. In the conventional model [3], a QD is described with an infinitely deep quantum well. However, real self-assembled QDs have a finite depth and therefore the QD excitons can communicate with the delocalized states of the WL under some conditions. Such a coupling of localized excitons and delocalized states has been studied in several experiments [4,5].

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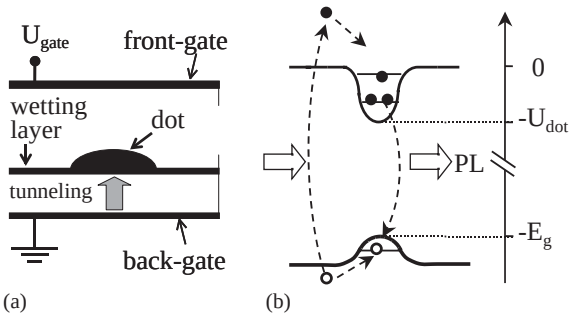

Fig. 1. (a) Schematic of the heterostructure with a QD embedded between front and back gates; (b) The band diagram of a QD.

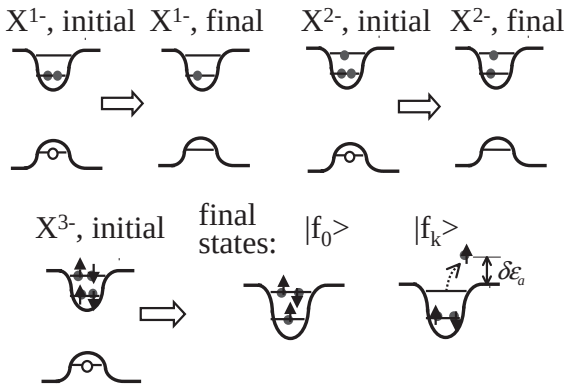

Fig. 2. Electron configurations for the initial and final exciton states. Spins are shown only for the exciton X^{3-}.

In Ref. [4], the coupling was observed as an interaction between a localized exciton state and Landau levels in the WL. Another type of coupling exists in the presence of a Fermi sea in the WL [6] and manifests itself as voltage-tunable exciton lines [1,4]. The interaction with the extended states of the WL can also be observed in the form of up-conversion [5]. One mechanism of coupling originates from Auger processes which can be either incoherent [7] or coherent [4,6]. Here we will focus on intra-band Auger processes in the PL spectra of charged excitons. We find that under some conditions, such Auger processes can be coherent.

Auger processes are a peculiar property of excited quantum states. In the initial state of the PL process, the exciton is in its ground state because of fast energy relaxation. At the same time, the final states of the photon-emission process can be excited because the optical recombination process involves the s-states in the conduction and valence bands (Figs. 1b, 2). After such a process, the final state of few electrons turns out to be excited. Therefore, *intra-band* Auger processes can play an important role for the final states. It is important to emphasize that we consider here the *intra-band* Auger processes. Let us now consider only the two lowest bound states in a QD, the s-state and lower p-orbital. Typically, self-assembled QDs are anisotropic and therefore the p-states are not degenerate. We start from charged excitons with a small number of extra electrons. In the case of X^0, the final state does not contain any particles; obviously, Auger processes do not exist for the exciton X^0. Fig. 2 shows electron configurations for the initial and final states of charged excitons. For the excitons X^{1-} and X^{2-}, the Auger-processes induced by the intra-dot Coulomb interaction are not possible because the number of excited electrons in the final state is not sufficient. For the Auger process, the system has to have at least two excited p-electrons in the final state; this case is realized in the multiply-charged excitons X^{3-}, X^{4-}. Here we will focus on the exciton X^{3-}, which was studied in detail in a recent experiment [4]. In the final state of X^{3-}, one of the p-electrons can make a transition to the s-state, while another p-electron becomes excited into the continuum (Fig. 2). Such a process can occur with conservation of energy. Qualitative results obtained below can also be applied to the excitons with $n > 3$.

It is interesting to note that the Auger processes in the final state of X^{3-} can be spin-sensitive [4]. In our model, the QD is anisotropic and the p-states are not degenerate. Hence, the electronic spin in the ground initial state of X^{3-} is zero, $S^{e}_{initial} = 0$ (the two electron levels are completely occupied).[1] Then, one of the p-electrons recombines and the spin of the final state

[1] The ground state of X^{3-} has zero spin if the *asymmetry* of a QD is strong enough. Namely, it happens when the splitting between single-particle energies of the p-states, $E_{p2} - E_{p1}$, is larger than the exchange–interaction energy for the p-states. The exchange energy found in experiments [1,4] is about 2–3 meV. In this paper, the single-particle splitting $E_{p2} - E_{p1}$ is taken as 5 meV. Thus, the ground initial state of X^{3-} for our range of parameters has the zero electronic spin. Note that, if a QD is almost symmetric, the electron states obey the Hund's rule and $S^{e}_{initial} = 1$ [1,4].

$S^{e}_{final} = \frac{1}{2}$. Since the Auger process conserves the spin, the final state $|f_k\rangle$ should contain the completely filled s-orbital and a delocalized electron with the spin $\frac{1}{2}$. Thus, we see that the Pauli principle allows the Auger processes for X^{3-} in an *asymmetric* QD. However, it was demonstrated in Ref. [4] experimentally and theoretically that, in *symmetric* QDs, the Auger process for X^{3-} is forbidden at zero magnetic field due to the spin selection rules.

The final states of X^{3-} are linear combinations of the three-electron states $|f_0\rangle$ and $|f_k\rangle$, where $|f_0\rangle$ is the localized state and $|f_k\rangle$ is a state with one delocalized electron (Fig. 2). To find the final states, we employ the Anderson-like Hamiltonian [8],

$$\hat{H} = E_{fo}|f_0\rangle\langle f_0| + \sum_k E_{fk}|f_k\rangle\langle f_k| + \sum_k W_{0k}|f_k\rangle\langle f_0| + W^*_{0k}|f_0\rangle\langle f_k|,$$

where k is the index (wave vector) of an electron delocalized state in the WL, and E_{fo} and E_{fk} are the energies of corresponding states; W_{0k} is the Auger matrix element: $W_{0k} = \langle f_0|U_{Coul}|f_k\rangle$, where $U_{Coul} = \sum_{i<j} e^2/\varepsilon|r_i - r_j|$ is the Coulomb interaction, and ε is the dielectric constant of the host semiconductor. In our model, we include only two bound states, the s-state and lower p-orbital. To calculate numerically the parameters, we represent a QD as an anisotropic harmonic oscillator taking the electron (hole) oscillator frequencies as 25 and 20 (12.5 and 10) meV, typical values for self-assembled QDs. In the spirit of Anderson model, the single-electron delocalized state is represented by a plane wave function, $\exp(ikr)$. The energies of the above states can be written as $E_{fo} = \varepsilon_0^{dot}$ and $E_{fk} = \varepsilon_1^{dot} + \varepsilon_k$, where $\varepsilon_0^{dot}(\varepsilon_1^{dot})$ is the intra-dot energy of the state $|f_0\rangle(|f_k\rangle)$ and ε_k is the single-particle energy of a delocalized state in the WL. In our approach, the quantization in a QD is assumed to be strong so that the Coulomb interaction can be included with perturbation theory.

The optical emission spectrum at zero temperature is given by the correlation function:

$$I(\omega) = \mathrm{Re}\int_0^\infty \mathrm{d}t e^{-i\omega t}\langle X^{3-}|\hat{V}^+_{opt}(t)\hat{V}_{opt}(0)|X^{3-}\rangle,$$

where $|X^{3-}\rangle$ is the initial state, the operator $\hat{V}_{opt} = V_{opt}(\hat{b}_{s,-3/2}\hat{a}_{s,\uparrow} + \hat{b}_{s,+3/2}\hat{a}_{s,\downarrow})$ describes the photon emission process that involves the s-states of electrons and heavy holes, the operators $\hat{a}_{s,\uparrow(\downarrow)}$ and $\hat{b}_{s,\pm 3/2}$ are related to electrons, and to holes with angular momenta $J_z = \pm\frac{3}{2}$, respectively. The operator $\hat{V}_{opt}$ couples the initial state X^{3-} only with the state $|f_0\rangle$. An exact expression for the optical spectrum takes the form [6]

$$I(\omega) = V^2_{opt}\mathrm{Re}\,\frac{-i}{\tilde{\omega} - \Sigma_1 - i\gamma_{ph}},$$

$$\Sigma_1 = \int_0^\infty \frac{|W_{0\varepsilon}|^2\rho(\varepsilon)\,\mathrm{d}\varepsilon}{\tilde{\omega} - \delta\varepsilon_a + \varepsilon - i\gamma_{ph}}, \quad (1)$$

where $\tilde{\omega} = \omega - \Delta E_{X^{3-}}$ and $\Delta E_{X^{3-}} = E^{initial}_{X^{3-}} - E_{fo}$ is the PL energy of the X^{3-} exciton in the absence of Auger coupling; $\delta\varepsilon_a = \varepsilon_0^{dot} - \varepsilon_1^{dot}$ is the excess kinetic energy in the Auger process, and $\rho(\varepsilon)$ is the 2D density of states (DOS); γ_{ph} is a phonon-induced broadening. The self-energy Σ_1 in Eq. (1) was calculated according to the diagram shown in Fig. 3. For the matrix element $W_{0\varepsilon}$ we assume $W_{0\varepsilon} = W_a$ in the interval $0 < \varepsilon < D$ and $W_{0\varepsilon} = 0$ elsewhere.[2] In our model, the excess kinetic energy $\delta\varepsilon_a$ depends on the QD depth

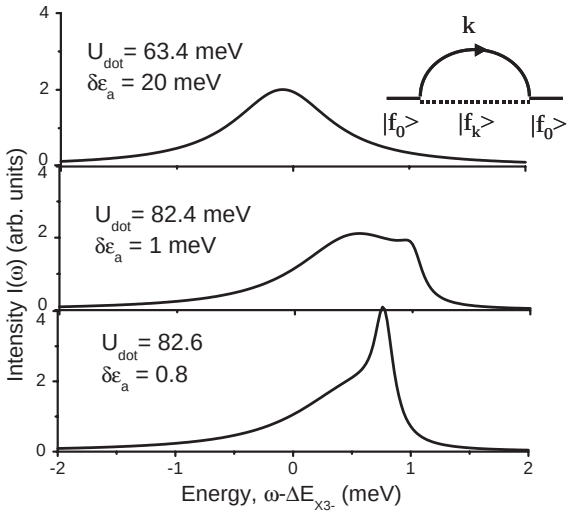

Fig. 3. Emission lines for several QD depths. Here we assume that $\gamma_{ph} = 0.1$ meV; this is the typical value in the experiments [1,4].

[2] The Auger matrix element $W_{0\varepsilon}$ decreases with increasing kinetic energy ε_k since the wave functions of delocalized states oscillate. The cut-off parameter D can be estimated as $\hbar^2/m^*L^2$, where L and m^* are the QD lateral size and effective mass, respectively.

$U_{\rm dot}$. Fig. 3 shows the calculated emission spectrum for $\Delta_{\rm a} = \pi W_{\rm a}^2 \rho_0 = 0.4$ meV, $D \approx 30$ meV and several values of the parameter $U_{\rm dot}$; here ρ_0 is the 2D DOS at zero magnetic field. The energy $\Delta_{\rm a}$ is the broadening of the emission peak due to Auger processes. For the parameters used in Eq. (1), we take the experimental values: $\Delta_{\rm a} \approx 0.4$ meV and $\delta\varepsilon_{\rm a} \approx 20$ meV [4]. If $\delta\varepsilon_{\rm a} \gg \Delta_{\rm a}$, the spectrum is close to a Lorentzian. Interesting shapes of the emission line appear when a QD is sufficiently deep and therefore $\delta\varepsilon_{\rm a} \approx \Delta_{\rm a}$. In the latter case, the emission line shows Fano-like quantum interference features. Thus, in contract to the limit $\delta\varepsilon_{\rm a} \gg \Delta_{\rm a}$, the Auger-coupling becomes partially coherent in the case $\delta\varepsilon_{\rm a} \approx \Delta_{\rm a}$. In addition, we note that the Auger matrix element can be estimated as $W_{0\varepsilon} = (e^2/\sqrt{S}\varepsilon)\int {\rm d}\vec{r}_1\, {\rm d}\vec{r}_2 \psi_{\rm s}(\vec{r}_1)\exp(i\vec{k}\vec{r}_2)\psi_{\rm p}(\vec{r}_1)\psi_{\rm p}(\vec{r}_2)/|\vec{r}_{12}|$, where $|\vec{r}_{12}| = \sqrt{(\vec{r}_1 - \vec{r}_2)^2\, l_z^2}$, $\psi_{\rm s(p)}$ is the lateral wave function related to the s(p)-state, $r_{1(2)}$ is the in-plane position vector, S is the surface area, and l_z represents an effective width of the QD in the vertical direction. With the above expression, we obtain a slight overestimate of the broadening;[3] $\Delta_{\rm a} \approx 0.7$ meV for $\varepsilon_{\rm a} \approx 20$ meV and $l_z = 2$ nm.

In the presence of a magnetic field B, the spectrum $I(\omega)$ is dramatically changed.[4] In this case, the DOS becomes strongly modulated due to the formation of Landau levels (LLs). Here we model the DOS of LLs with Gaussian functions: $\rho(\varepsilon) = \rho_0 \hbar\omega_{\rm c}/\sqrt{\pi}\gamma_{\rm LL} \sum_{n=0,1,2,\ldots} \exp(-(\varepsilon-\varepsilon_n)^2/\gamma_{\rm LL}^2)$, where $\varepsilon_n = \hbar\omega_{\rm c}(n+1/2)$ is the LL energy and $\gamma_{\rm LL}$ is the LL broadening. In the spirit of Anderson model, the extended states involved in the Hamiltonian should correspond to the uniform system. We take into account the inhomogeneity of the system by introducing the broadening of LLs. The emission spectrum $I(\omega)$ demonstrates anticrossing between the energies $\hbar\omega = \Delta E_{X^{3-}}(B)$ and $\hbar\omega = \Delta E_{X^{3-}}(B) + \delta\varepsilon_{\rm a}(B) - \varepsilon_n(B)$ [4,6] (Fig. 4). The last equality corresponds to the condition when the excess kinetic energy of the p-electron in the final state becomes equal to the LL energy (Fig. 4, inset). Such an interaction between the localized exciton and LLs has a coherent character

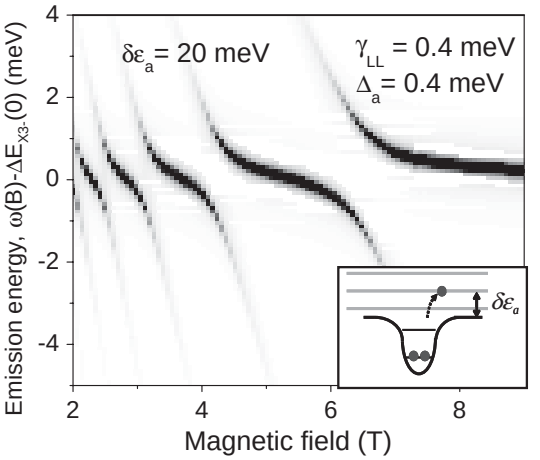

Fig. 4. Gray-scale plot of the emission spectrum as a function of magnetic field; $\Delta E_{X^{3-}}(0)$ is the exciton peak energy at $B = 0$.

since it induces anticrossings and does not lead to an essential broadening. It was recently observed experimentally in single *asymmetric* QDs [4].

To conclude, the exciton X^{3-} in self-organized QDs can strongly couple with the continuum of delocalized states via an Auger process. It is worthwhile to note that the magnetic behavior of the X^{3-} exciton qualitatively differs from the excitons X^{0-}, X^{1-}, and X^{2-}. The excitons with $n = 0, 1$, and 2 demonstrate the usual diamagnetic behavior [9], just like excitons in 3D systems [10].

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[3] We overestimate $\Delta_{\rm a}$ because the plane-wave function used above should be modified inside the QD.

[4] This is true also for symmetric dots because the magnetic field induces a transition from $S^{\rm e}_{\rm initial} = 1$ to $S^{\rm e}_{\rm initial} = 0$, allowing the Auger process in the final state to become active [1,4].

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