Stark effect on the exciton complexes of individual quantum dots

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(March 23, 2022)

Abstract

The emission spectrum of exciton complexes formed in individual self-assembled quantum dots (QDs) embedded into a p-n junction is theoretically studied using an effective mass model. We calculate the particle Coulomb interactions, electron-hole overlaps and transition energies of exciton complexes for the different strength and direction of electric field. Both redshifts and blueshifts are observed in exciton, trions and biexciton. The Stark effect may be applied to manipulate the spontaneous emission rate of individual QDs embedded in microcavities.
I. INTRODUCTION

Recently, many efforts have devoted into the studies of single-photon sources (SPS) composed of quantum dots (QDs), which provide the potential applications of quantum cryptography and quantum computing\textsuperscript{1–3}. For small size QDs, the strong three dimensional confinement effect of QDs causes the large energy level separation. Consequently, we can manipulate electrons and holes to occupy the lowest energy level of QDs and operate emitted photons from exciton, trion or biexciton formed in QDs. The antibunching feature of SPS was demonstrated in ref\textsuperscript{1,2} where electrons and holes of the QD are excited using optical pumping, only a handful of studies employed the electrical pumping to demonstrate the antibunching behavior of SPS.

From the practical point of view, the electrically driven SPS has been suggested by Imamoglu and Yamamoto considering individual QDs embedded into a semiconductor p-n junction\textsuperscript{4}. To fabricate a single QD of nanometer size at specific location is one of challenged techniques in the realization of electrically driven SPS. Self-assembled quantum dots (SAQDs) combined with selective formation method may solve such difficulty\textsuperscript{5}. Apart from the electrically driven, it is very important to manipulate the the spontaneous emission rate of SPS in the applications of quantum cryptography. According to the Purcell effect\textsuperscript{6–9}, the enhancement of the spontaneous emission rate due to cavity (1/$\gamma_{cav}$) with respect to free-space is

$$
\frac{1}{\gamma_{cav}} = \frac{1}{\gamma_{free}} \frac{3Q(\lambda_c/n)^3}{4\pi^2 V_{eff}} \frac{\omega_c^2}{4(\omega - \omega_c)^2 + \Delta \omega_c^2} \frac{|E(r_c)|^2}{|E_{max}|^2 \eta^2},
$$

where $\eta = \frac{d.E(r)/|d||E(r)|}{d.}$ describes the orientation matching of dipole $d$ and electric field of emission light $E$, and where the factor 3 from a 1/3 averaging factor accounting for the random polarization of free-space modes with respect to the dipole $d$. The quality factor of cavity is given by $Q = \omega_c/\Delta \omega_c$, where $\omega_c$ and $\Delta \omega_c$ denote, respectively, the angular frequency and linewidth of a cavity supporting a single-mode. Other notations $V_{eff}$, and $n$ are the effective volume of cavity and the refractive index at the field maximum. Although a number of experiments have already demonstrated the potential of QD-based solid-state cavity quantum electrodynamics (QED) in applications such as single-photon sources\textsuperscript{1–2,10–11}. Nevertheless, because it is very difficult to pre-determine the exact
resonance energy and location of an optically or electrically active QD, all of the prior QD-based cavity QED experiments relied on a random spectral and spatial overlap between QDs and cavity modes. Most recently, in spite of Badolado et al have proposed a method to solve above difficulty\textsuperscript{12}, The studies of optimization of Purcell effect for electrically driven SPS are still lack.

Compared with optical pumping, the electrically driven SPS provide a tunable emission spectrum of exciton complexes arising from the Stark effect. Such extra degree of freedom (tunable wavelengths) provides a possibility to reach $\omega = \omega_{c}$ of eq. (1)condition. Although the Stark effect on exciton peaks formed in QDs was well investigated experimentally and theoretically\textsuperscript{13–19}, the Stark effect on the trion and biexciton peaks is still not very clear. Thompson et al. demonstrated that the single photon generated by the biexciton state is significantly less in emission time than the single exciton state\textsuperscript{20}. From this point of view, the biexciton state is favored for the application of single photon generation. Therefore, it is worth studying the physical parameters of biextion state.

The modest purpose of this article is to theoretically study the Stark effect on the transition energies of exciton complexes. Besides, we will also investigate the electric field effect on the particle Coulomb interactions and oscillator strengths of QDs. The formal determines the charging energies for electrons and holes, which are crucial parameters for the electrically driven SPS. The latter influences the spontaneous emission rate of QDs in free space.

II. FORMALISM

Because a number of experiments used InAs/GaAs SAQD system to fabricate SPS, we consider such SAQDs embedded into a p-n junction shown in Fig. 1. To understand the emission spectrum of an InAs/GaAs SAQDs, we need to calculate the electronic structures for various electron-hole complexes, including exciton ($X$), biexciton ($X^2$), negative trion ($X^{-}$), and positive trion ($X^{+}$), which can occur in this system. Due to the large strain-induced splitting between heavy-hole and light-hole band for InAs on GaAs, we only have to consider the heavy hole band (with $J_{z} = \pm 3/2$) and ignore its coupling with light-hole band caused by the QD potential. Thus, we can treat the heavy hole as a spin-1/2 particle with $\sigma = \uparrow, \downarrow$ representing $J_{z} = \pm 3/2$. Meanwhile, we adopt the Hartree-
Fock (HF) approximation to describe the many-particle systems for trions and biexciton. This is a reasonable approximation for many particles confined in a small QD, because the particle correlation effect is greatly suppressed by the lack of available low-energy excitations, which are coupled to the HF ground state via the configuration interaction. In two-electron atomic systems such as H−, He and Li+, it is well known that the correlation energy for all these systems is around 0.11 Ry. A major part of it can be accounted for within the unrestricted Hartee-Fock (UHF) approximation by the so-called “in-and-out correlation”, which arises because the two electrons can occupy two orbitals with very different radii. The remainder of the correlation is due to the “angular correlation”, which may be described by the coupling of the HF ground state to the two-particle excited states with non-zero single-particle orbital angular momentum (while keeping the total orbital angular momentum to be zero). Both the “in-and-out correlation” and the “angular correlation” are significantly suppressed in the QD system with strong particle confinement.

The strengths of $U_e$, $U_h$, and $U_{eh}$ in excitons, trions, or biexcitons are determined via the self-consistent HF calculation within a simple but realistic effective-mass model. We consider an InAs/GaAs self-assembled QD (SAQD) with conical shape. The QD resides on a monolayer of InAs wetting layer and the whole system is embedded in a slab of GaAs with finite width. The slab is then placed in contact with heavily doped p-type and n-type GaAs to form a p-i-n structure for single photon generation. Within the effective-mass model, the electron and hole in the exciton ($X$), trions ($X^-$ or $X^+$), or biexciton ($X^2$) in the QD are described by the coupled equations

$$\left[-\nabla \frac{\hbar^2}{2m_e^*(\rho, z)} \nabla + V_{QD}^e(\rho, z) - eFz + V_{sc}(\rho, z)\right]$$

$$\psi_e(\rho, \phi, z) = (E_e - E_e)\psi_e(\rho, \phi, z), \quad (2)$$

$$\left[-\nabla \frac{\hbar^2}{2m_h^*(\rho, z)} \nabla + V_{QD}^h(\rho, z) + eFz - V_{sc}(\rho, z)\right]$$

$$\psi_h(\rho, \phi, z) = E_h\psi_h(\rho, \phi, z), \quad (3)$$

and

$$V_{sc}(\mathbf{r}) = \int d\mathbf{r}' \frac{e^2[n_e(\mathbf{r}') - n_h(\mathbf{r}')]}{\epsilon_0 |\mathbf{r}' - \mathbf{r}|}, \quad (4)$$
where $E_c$ denotes the energy of the GaAs conduction band minimum, which is $1.518\,\text{eV}$ above the GaAs valence band maximum (defined as the energy zero), $m^*_e(\rho,z)$ (a scalar) denotes the position-dependent electron effective mass, which takes on values of $m^*_{eG} = 0.067m_e$ for GaAs and $m^*_{eI} = 0.04m_e$ for InAs. $m^*_h(\rho,z)$ denotes the position-dependent effective mass tensor for the hole. Due to the strong spin-orbit coupling and the large strain-induced splitting between heavy-hole and light-hole band, we can neglect the coupling of the heavy-hole band with the split-off band and light-hole band. Consequently, it is a fairly good approximation to describe $m^*_h(\rho,z)$ in InAs/GaAs QD as a diagonal tensor with the $x$ and $y$ components given by $m^*_h^{-1} = (\gamma_1 + \gamma_2)/m_e$ and the $z$ component given by $m^*_h^{-1} = (\gamma_1 - 2\gamma_2)/m_e$. $\gamma_1$ and $\gamma_2$ are the Luttinger parameters. Their values for InAs and GaAs are taken from Ref. [24]. $V_{eQD}(\rho,z)$ ($V_{hQD}(\rho,z)$) is approximated by a constant attractive potential in the InAs region with value determined by the conduction-band (valence-band) offset and the deformation potential shift caused by the biaxial strain in the QD. These values have been determined by comparison with results obtained from a microscopic model calculation\textsuperscript{25} and we have $V_{eQD} \sim -0.42\,\text{eV}$ and $V_{hQD} \sim -0.3\,\text{eV}$. The $eFz$ term in Eqs. (2) and (3) arises from the applied voltage, where $F$ denotes the strength of the electric field. $V_{sc}(r)$ denotes the self-consistent potential caused by the electrostatic interaction with the charge densities $[n_e(r)$ and/or $n_h(r)]$ associated with the other particles in the system. $\epsilon_0$ is the static dielectric constant of InAs. The image force is ignored here due to the small difference in dielectric constant between InAs and GaAs.

Eqs. (2) and (3) are solved self-consistently via the Ritz variational method by expanding the ground-state wave functions in terms of a set of nearly complete basis functions, which are chosen to be products of Bessel functions (with axial symmetry) and sine waves

$$\psi_{nlm}(\rho,\phi,z) = J_0(\alpha_n \rho) \sin(k_m(z + \frac{L}{2})), \quad (5)$$

where $k_m = m\pi/L$, $m=1,2,3...$ $L$ is taken to be $300\,\text{Å}$. $J_0$ is the Bessel function of zero-th order and $\alpha_n R$ is the $n$-th zero of $J_0(x)$ with $R$ taken to be $400\,\text{Å}$, which is large enough to give convergent numerical results. Forty sine functions multiplied by fifteen Bessel functions are used to diagonalize the Hartree-Fock Hamiltonian.

The ground state energy of system $i$ ($i = X, X^2, X^-, X^+$) is given by
\[ E_i = N^i_e E^i_e + N^i_h E^i_h + N^i_e N^i_h U^i_{eh} \]
\[ -N^i_e N^i_h U^i_{eh} - N^i_e U^i_e - N^i_h U^i_h, \]

where \( E^i_e \) and \( E^i_h \) denote the ground state HF single-particle energy for electron and hole obtained from solving Eqs. (2) and (3), respectively. \( N^i_e \) and \( N^i_h \) denote the electron and hole particle number in system \( i \). \( U^i_e, U^i_{eh} \) and \( U^i_h \) are the electron-electron interaction, hole-hole interaction and electron-hole interaction, respectively. The transition energies for the electron-hole recombination in \( X, X^2, X^-, \) and \( X^+ \) are given by \( E_X, E_{X^2} - E_X, E_{X^-} - E^0_e, \) and \( E_{X^+} - E^0_h \), respectively. Here \( E^0_e \) and \( E^0_h \) are the free-particle ground state eigen-values for Eqs. (2) and (3) with \( V_{sc} \) set to zero.

**III. RESULTS**

For electrically driven SPS, electrons and holes are injected from the electrodes. To create biexciton state, the applied voltage needs to overcome the charging energies of electrons and holes, which are determined by \( U_e \) and \( U_h \). The calculation of \( U_e, U_{eh}, \) and \( U_h \) is shown in Fig. 2, where the solid curves denote \( X^2 \), the dotted curves denote \( X^- \), and the dashed curves denote \( X^+ \) in conical SAQDs with base radius \( R_0 \) varying between 60 Å and 110 Å. Here, the QD height \( h \) is varied between 15 Å and 65 Å, while keeping the ratio \( (h - 15\text{Å})/(R_0 - 60\text{Å}) = 1 \). The electron-hole Coulomb energy \( U_{eh} \) in the exciton \( X \) is almost the same as that in \( X^2 \) and is omitted in this plot. The strengths of Coulomb interactions are, in general, inversely proportional to the QD size, since the charge densities in smaller QDs are more localized. However, as the QD size decreases below a threshold value (around \( R_0 = 70 \text{ Å} \)), the Coulomb interactions become reduced as a result of the leak out of charge density for small QDs, which tend to be more serious for the electron than for the hole due to the smaller effective mass for the electron. This manifests the effects of finite potential barrier of the QD. This effect will not appear if one adopts an infinite barrier approximation as in some previous studies of excitons in QDs. In the large dot limit \( U_e, U_{eh} \) and \( U_h \) all approach the same value, indicating similar degree of localization for the electron and hole in the large dot. Note that the magnitude of charging energies for electrons or holes in the same order of thermal energy of room temperature \( k_B T \).
where $k_B$ is a Boltzmann constant. This indicates that the operation temperature of system should be much lower than room temperature.

Owing to the applied bias crossing the QD, the electric field effect is not negligible in the variation of particle interactions. We adopt the size of QD with radius 7.5 nm and height 3 nm to study the electric field effect on the particle Coulomb interactions. We assume that the z axis is directed from the base to the apex of the dot. Fig. 3 shows the Coulomb interaction strengths as functions of electric field for different exciton complexes. We see that $U_h$, $U_{eh}$ and $U_e$ display asymmetric behavior of electric field as a result of geometry of the dots. Increasing electric field, the deduction of $U_{eh}$ indicates that the electron-hole separation increases. However, we note that $U_h$ increases in the positive direction of electric field since the wave functions of holes become more localize. Even though the enhancement of $U_e$ is observed in the negative direction of electric field, it only exists at very small electric field region. When the electric field is larger than a threshold value, the wave functions of electrons become delocalize and leak out the quantum dot. Consequently, electron-electron Coulomb interactions becomes weak. As mentioned, $U_e$ and $U_h$ denote the charging energies of QD for electrons and holes, respectively. Therefore, the constant interaction model used extensively in the Anderson model is valid only for small electric field case, otherwise we should take into account bias-dependent Coulomb interactions.

Based on eq. (1), the larger $1/\gamma_{free}$, the better for $1/\gamma_{cav}$. Hence we also attempt to investigate the oscillator strength of QDs, which is proportional to the electron-hole overlap squared. Fig. 4 shows the squared electron-hole overlap function $A(F = 0)$ for $X$, $X^2$, $X^-$, and $X^+$ as functions of the QD radius, $R_0$ (with the height $h$ varying in the same way as in Fig. 2). For all four complexes, the electron-hole overlap increases with the QD size, indicating that the electron and hole wave functions approach each other as they become fully confined in the QD. The electron-hole overlaps in exciton and biexciton are very similar since they are both charge neutral. Meanwhile, the $A(F = 0)$ of biexciton is slightly above that of the exciton. For small size QDs, the electron-hole overlap is the smallest in negative trion ($X^-$), and the largest in positive trion ($X^+$). This is due to the fact that the Coulomb repulsion between the two electrons in $X^-$ causes the electron wave function to be more delocalized.
than in the charge-neutral complexes (X and $X^2$), thus reducing the electron-hole overlap. However, the same effect in $X^+$ causes the hole wave function to be more delocalized, which becomes closer to the electron wave function, and therefore enhances the electron-hole overlap. The behavior is reversed for large QDs with a cross-over occurring at $R_0 \sim 95 \text{ Å}$.

In fig. 4 we did not include the electric field effect for electron-hole overlap function $A(F)$. Using the size of QD considered in Fig. 3, we show $A(F)$ as functions of electric field for four complexes in Fig. 5. $A(F)$ of each complex is enhanced at small field region for $F > 0$, whereas the decline of $A(F)$ appears when the electric field is larger than a threshold field $F_s$, which depends on exciton complexes. For example, $F_s$ is about 45 $kV/cm$ for $X^+$, but 30 $kV/cm$ for X. On the other hand, $A(F)$ declines as the electric field is increased from 0 to 120$kV/cm$ for negative applied field direction. We observe that the variation of $A(F)$ is the largest in negative trion $X^-$ since the electron wave function is more delocalized for $X^-$ than other complexes. Even though the results of Fig. 5 shows that $A(F)$ of positive trion and biexciton states is larger than that of exciton at a given electric field. However, the creation of biexciton (trion) state requires the applied field higher than that of exciton for the overcome of charging energies, so that it is possible to observe that the spontaneous emission rate of biexciton or trion is smaller that of exciton in this electrically driven SPS.

Finally, we attempt to study the transition energies for exciton complexes without and with electric field. Fig. 6 shows the transition energy for the electron-hole recombination in a biexciton, positive trion and negative trion relative to the exciton transition energy ($E_X$) as functions of the QD radius, $R_0$ (with the height $h$ varying in the same way as in Fig. 2). The exciton transition energy ($E_X$) as a function of $R_0$ is also displayed (dashed curve) with the scale indicated on the right side of Fig. 6. The biexciton transition energy is consistently above the exciton transition energy ($E_X$) for the range of $R_0$ considered here, which is still considerably smaller than the free exciton Bohr radius in InAs ($\sim 300\text{Å}$). For very large QDs (with $R_0 > 300\text{Å}$), the correlation effect will become important, and the biexciton transition energy can become lower than $E_X$. The Positive (negative) trion appears at significantly higher (lower) energy than the exciton for small QDs, but approaches the biexciton quickly as the QD size increases. This behavior can be understood by examining the difference in
$U_h$ ($U_e$) and $U_{eh}$ as shown in Fig. 2. The biexciton peak displaying a blue shift with respect to the exciton peak (showing an antibinding biexciton) is also consistent with the observation reported in ref.[3,20]. Recently, studies of the binding and antibinding of biexcitons were reported in Ref. 31. Our calculation given by Eqs. (2) and (3) provides only the antibinding feature of biexciton. In Ref. 31 it is pointed out that the biexciton complex changes from antibinding to binding as the QD size increases. For QDs with dimension larger than the exciton Bohr radius, the correlation energy becomes significant. In this study, we have not taken into account the correlation energy. This is justified as long as we restrict ourselves to small QDs.

According to eq. (1), it is one of challenged techniques to obtain $\omega = \omega_c$. If the frequencies of emitted photons contributed from exciton complexes can be highly tuned, the possibility of optimizing spontaneous emission rate will be enhanced. Fig. 7 (a) and (b) shows Stark shifts of exciton complexes with the strength and direction of the electric field for different quantum dot size. In diagram (a) the size of QD corresponds to that of Fig. 3. In diagram (b) we adopt the size of QD with the radius $R_0 = 8.5nm$ and height $h = 3nm$. The exciton complexes display asymmetric shift around zero field and red shift in the negative field direction. We see that in the positive field direction the exciton complexes display the blue shifts at small field region. The blue shift of positive trion is relatively large if compare with that of other exciton complexes. For charge neutral exciton $X$ and $X^2$, their responses to Stark effect are almost the same. The comparison of diagram (a) and (b) shows that the tunable range of emitted photon frequencies depends on the size of quantum dot. In our case, Stark shifts of exciton complexes are near 2.8 $meV$ and 3.5 $meV$ for the QD of the radius $R_0 = 7.5nm$ and height $h = 3 nm$ and the QD of the radius $R_0 = 8.5nm$ and height $h = 4 nm$, respectively. This remarkable frequency shifts arising from the Stark effect lead the SPS with electric pumping to readily reach the resonant condition of $\omega = \omega_c$.

IV. CONCLUSION

In this article we have calculated the following three physical parameters of exciton complexes formed in the ground state of QD embedded into a p-n junction: particle Coulomb interactions,
electron-hole overlaps and transition energies. In the typical volume of grown SAQDs, we found that the magnitude of electron-electron Coulomb interactions as well as hole-hole Coulomb interactions is just near the thermal energy of room temperature. Therefore, it is difficult for the InAs/GaAs system to provide single-photon sources at room temperature. We note that the suppression of electron-hole overlaps always exist for electrically driven SPS. Although the decaying of biexciton to exciton can generate single photons, we need to apply relatively higher voltage to overcome the charging energies for electrons and holes to create the biexciton state. Such higher voltage is possible to suppress the spontaneous emission rate of the biexciton state and make it smaller than that of exciton at lower voltage. Consequently, the exciton state is preferable for single-photon generation.

Owing to the asymmetric shape of QDs, the direction-dependent Stark effects are observed in the measurable transition energies of exciton complexes. We note that the Stark shifts of complexes are smaller in the positive direction field than negative direction field. Besides, the Stark shifts also depend on the size of QDs. In particular, the largest blueshifts are occurred in the positive trion state, on the other hand, the largest redshifts are observed in the exciton state. The Stark shifts of exciton complexes may be applied to manipulate the resonant condition of eq. (1) and to control the spontaneous emission rate of individual QDs embedded in a cavity.

Acknowledgments

This work was supported by National Science Council of Republic of China Contract No. NSC 92-2112-M-008-053
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Figure Captions

Fig. 1: Energy-band structure of n-i-p junction for a cross section through the quantum dot along z axis. The quantum dot is a small conical structure located in the i layer between n and p semiconductors. Electrons (black) and holes (white) to tunnel cross the barriers with rate $\Gamma_e$ and $\Gamma_h$, respectively. $R_{eh}$ is the electron-hole recombination rate.

Fig. 2: $U_e$, $U_{eh}$, and $U_h$ as a function of the QD size for biexciton (solid), negative trion $X^-$ (dotted) and positive trion $X^+$ (dashed). Note that the ratio $(h - 15\text{Å})/(R_0 - 60\text{Å}) = 1$ is used.

Fig. 3: Particle Coulomb interactions as a function of the strength and direction of electric field for the QD with radius 7.5 nm and height 3 nm. Solid line denotes the biexciton configuration, dashed line and dotted line denote, respectively, positive trion and negative trion.

Fig. 4: Electron-hole overlap squared as a function of the QD size for exciton $X$, biexciton $X^2$, negative trion $X^-$ and positive trion $X^+$.

Fig. 5: Electron-hole overlap squared as a function of the strength and direction of electric field for the QD with radius 7.5 nm and height 3 nm.

Fig. 6: Transition energy as a function of the QD size for exciton $X$, biexciton $X^2$, negative trion $X^-$ and positive trion $X^+$.

Fig. 7: Transition energy as a function of the strength and direction of electric field for different QD size. Diagram (a) and (b) denote, respectively, the QD of radius 7.5 nm and height 3 nm and the QD of radius 8.5 nm and height 4 nm.
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