Dynamic Stark effects in a semiconductor quantum dot exhibiting a two-hole species valence band

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Abstract. We have analytically explored the nature of Stark shift and self energy correction in the presence of two discrete excitonic states i.e., the heavy-hole and light-hole valence bands, below the fundamental absorption edge of semiconductor quantum dots. The time dependent perturbation technique for a three-level system, under the near band gap resonant excitation regime, has been employed; with the incorporation of the relaxation and dephasing mechanisms phenomenologically. The detuning spectra of Stark shift and self energy correction and their dependence on a moderately strong electric field, have been analyzed. The numerical and analytical estimations in a weak noncentrosymmetric CdTe quantum dot show a decrease in Stark shift and an enhancement in self energy correction for both heavy-hole and light-hole excitons.

1. Introduction
During the past three decades, the physics of low-dimensional semiconductors have become a vital part of present-day research. Semiconductor quantum dots (QDs) represent ideal quantum systems because their electrons are three dimensionally confined, resulting in a discrete density of states. The combination of high nonlinear properties and fast response time make applications in all-optical switching devices possible with semiconductor QDs. Electric field has become an interesting probe for studying the physical properties of low-dimensional structures, from both the theoretical and technical points of view. The ac Stark shift also known as dynamical Stark shift arises in an active medium when a rapidly oscillating field of an electromagnetic wave is applied to shift the atomic, molecular or electron energy levels. The measurement of Stark shift is utilized to calculate the transition dipole moments and relaxation parameters of the crystal [1]. The self energy correction indicates that the damping mechanisms are modulated in presence of the electric field amplitude of the pump radiation. The observation of the Stark effect in semiconductors is normally hinded by the fast dephasing rates in the vicinity of the excitonic transitions. With the recent advances in ultrashort pulse spectroscopy, it is now possible to observe such phenomena induced by intense coherent laser beams in bulk as well as in low-dimensional semiconductor structures. In 1985, Frohlich and coworkers first demonstrated experimentally, the optical Stark effect in Cu2O and Miller et al explained theoretically the quantum-confined Stark effect (QCSE) in QWs [2]. In 2004, Unold et al [3] reported the first experimental study of the optical Stark effect in a single semiconductor QD.
As CdTe is a weak noncentrosymmetric crystal. The occurrence of noncentrosymmetry (NCS) has been classically explained by the local fields existing in the crystal and due importance should be given to its contribution while studying the stark effects in a specific semiconducting crystal. To the author’s knowledge, so far no systematic attempt has been made to investigate the dependence of nonlinear transient’s phenomena on applied electric field in NCS QDs.

In the present analytical study of optical nonlinearities of QDs, we have selected the QD sizes to be much smaller than the bulk exciton Bohr radii. Such QD sizes are classified as belonging to the strongly confinement regime (SCR). In a semiconductor QD, the confinement induced exciton binding energy increases abruptly when its geometrical radius becomes smaller than the bulk exciton Bohr radius $R_B$. Under the SCR, apart from exciton, biexcitons also play an important role in determining the optical properties of semiconductor QDs. It has been shown by Hu et al. [4] that the ground state exciton energy $\hbar\omega_{\text{ex}}$ scaling as R-2 dominates distinctly over the electron-hole (e-h) Coulomb interaction and remains practically independent of the e-h effective mass ratio for $R \leq 0.6 \; a_B$.

In most semiconductors, the conduction band can be approximated by a parabolic band model but the valence band represents a complicated structure with heavy-hole (hh) and light-hole (lh) bands and split-off bands. The split-off bands are far away from the band edge. The hh and lh bands are degenerate at $k = 0$ and strongly coupled at $k \neq 0$. This coupling leads to a nonparabolic subband structure. The inclusion of valence-band mixing in the analysis of the optical properties of the active material makes the calculation complicated. So the intrasubband mixing in the valence bands as well as split-off energy band have been neglected. Hence the photo induced band-to-band electronic transitions are considered from both the heavy-hole (hh) and light hole (lh) valence bands to the lowest (1s) exciton state below the fundamental absorption edge. The special feature of the present analysis is the in-depth study of the transient optical phenomena viz. dynamical Stark shift and self energy correction in QDs under a moderately strong electric field.

2. Theoretical Formulations

The band structure of direct gap semiconductor is taken to be isotropic, parabolic and nondegenerate. The three energy levels considered are the crystal ground state $|a\rangle$, the lowest (n = 1, 1s) single electron-hole pair (exciton) state $|b\rangle$ is described in terms of both heavy-hole and light-hole subbands, and the biexcitonic state $|c\rangle$ arising due to the formation of bound exciton pairs. The energy of the lowest (1s) excitons in the presence of finite electron-hole coulomb interaction in a QD of radius R is expressed as

$$\hbar\omega_{\text{ex}} = \hbar\omega_{\text{ex}} + \left(\frac{\hbar^2 \pi^2}{2m_j R^2}\right) - \left(3.527a_B/R\right) + \Delta E_{\text{ex}}$$

where $\hbar\omega_{\text{ex}}$ and $\Delta E_{\text{ex}}$ are the bulk band gap energy and exciton Rydberg energy respectively. $a_B$ is the Bohr radius and $m_j$, is the effective reduced mass of the exciton with $\lambda = \text{hh or lh}$. The light-hole and heavy-hole effective masses can be represented in terms of Luttinger parameters $\gamma_1$ and $\gamma_2$ as $m_{\text{lh}} = m_h / (\gamma_1 + 2\gamma_2)$ and $m_{\text{hh}} = m_h / (\gamma_1 - \gamma_2)$ with $m_h$ being the free electron mass. Also, the exciton-biexciton transition energy with biexciton binding energy $\Delta E_{\text{ex}}$ defined as $\hbar\omega_{\text{ex}} = \hbar\omega_{\text{ex}} - \Delta E_{\text{ex}}$. We have incorporated the relaxation and dephasing mechanisms phenomenologically through the dephasing parameters $\Gamma_h$ and $\Gamma_c$.

Using time dependent perturbation treatment and rotating wave approximation, the equations of motion of the probability amplitudes in these three states of an NCS system are given by

$$\dot{a}(t) = i\Omega_{ab}e^{-i\omega_{ab}t}a(t) + i\sum_k \Omega_{ab}e^{-i\omega_{ab}t}b(k,t)$$

(1)
The third one being numerically at \( \gamma \) is obtained \((89.0 \text{\Omega}^2)\) in the equations of motion. In a NCS system, the field dependent Rabi flopping frequencies corresponding to the level-to-level transitions \( |a\rangle \leftrightarrow |b\rangle \) and \( |b\rangle \leftrightarrow |c\rangle \) respectively, defined by \( \Omega_{ab} = \mu_{ab} E_0/2\hbar \). Assuming the transition dipole moment operators \( \mu_{ab} \) and \( \mu_{bc} \) to act parallel to the applied electric field \( E(t) = (t/2) E_0 \exp(i\omega t) + \text{c.c.} \). Using the standard approach, \( [6] \) a cubic equation in \( \Omega \) is obtained \(( \Omega \) is introduced in the solution for \( a(t) \) given by \( a(t) = A \exp(\Omega_{ab}(\omega t)/\omega - i\omega t) \) as

\[
\Omega^3 - \Omega^2 (\omega_{NB} + \omega_{NC}) + (\omega_{NB} \omega_{NC} + \omega_{\omega N} - \Omega_{ab} - \Omega_{bc}) + \omega_{NC} \Omega_{ab}^2 = 0, \tag{4}
\]

where \( \omega_{NC} = \omega_{bc} + \omega_{ab} - 2\omega + (\Omega_{ab} - \Omega_{bc}) \cos \omega t - i[(\Omega_{ab} - \Omega_{bc}) \sin \omega t - \Gamma_{bc}] \); \( \omega_{NB} = \omega_{ab} - \omega + (\Omega_{ab} - \Omega_{bc}) \cos \omega t + i[(\Omega_{ab} - \Omega_{bc}) \sin \omega t - \Gamma_{bc}] \); \( \omega_{Nab} = (\Omega_{ab} + \Omega_{bc}) \cos \omega t + i[(\Omega_{ab} - \Omega_{bc}) \sin \omega t - \Gamma_{bc}] \).

The solution of the above equation yields three complex roots. Among these three roots, the magnitude of two are comparable to the dephasing frequency \( \Gamma_{bc} \) and third one being numerically at least three orders of magnitude lower than the other two becomes almost trivial. Thus to solve Eq. (4) analytically, we have taken \( \Omega > \omega_{NC} \Omega_{ab}^2 \) such that the cubic equation reduces to the following quadratic equation,

\[
\Omega^2 - \Omega (\omega_{NB} + \omega_{NC}) + (\omega_{NB} \omega_{NC} + \omega_{\omega N} - \Omega_{ab}^2 - \Omega_{bc}^2) = 0. \tag{5}
\]

Equation (8) yields two complex solutions as

\[
\Omega_{1,2} = \frac{\omega_{NB} + \omega_{NC}}{2} \pm \sqrt{\frac{\omega_{NB}^2}{4} + \frac{\omega_{NC}^2}{4} - \omega_{\omega N} + \Omega_{ab}^2 + \Omega_{bc}^2}. \tag{6}
\]

Following the approach of Zimmermann [7], the negative solution i.e., \( \Omega_{2} \) may be utilized to calculate the Stark broadening in a semiconductors. The real and imaginary part of the \( \Omega_{2} \), yield the Stark shift and self energy correction respectively.

### 3. Results and Discussions

In order to explore the occurrence of Stark shift and self energy correction in QDs, we have made numerical estimates for a weak NCS material viz., CdTe. The material parameters of CdTe are: \( h_0 g = 1.607 \text{eV}, \ a_b = 2.9 \text{nm} \) and \( m_e = 0.096 m_0 \) and Luttinger parameters are taken \( \gamma_1 = 5.29 \) and \( \gamma_2 = 1.89 \). We have assumed the average size of the QDs to be \( R = 0.6 a_b = 5 \text{nm} \) with pump frequency \( \omega = (\omega_{ab} - 10 F_s) = 2.73 \times 10^{15} \text{sec}^{-1}, \ T_s = 150 \text{fs} \). The contribution of NCS effect is introduced through \( \Omega_{ab,bb,cc} \) assuming \( \Omega_{ab} = \gamma \Omega_{ab} \) and \( \Omega_{bc} = \Omega_{cc} = 0.8\Omega_{ab} \); the NCS factor \( \gamma = 0.01 \). The system becomes centrosymmetric for \( \gamma = 0 \).
Figure 1. Detuning spectra of Stark shift $\Omega_r$ (A) and self energy correction $\Omega_i$ (B), at $t = 2\text{fs}$ for different strength of electric field.

Figure 2. Electric field dependence of Stark shift $\Omega_r$ (A) and self energy correction $\Omega_i$ (B) for the transitions from hh and lh valence subband.

Figures 1 show that both the nonlinear optical processes are diminished through the increase in electric field intensity. The behavior of Stark shift is symmetric with respect to the central frequency ($\omega = \omega_{ab}$) while the self energy correction deviates from such symmetric responses and its magnitude is large under off-resonant transitions below the band edge ($\omega < \omega_{ab}$) than the corresponding magnitude above the band edge ($\omega > \omega_{ab}$). Figures 2 depict a decrease in Stark shift and an enhancement in self energy correction for both hh and lh excitons. It is clear that hh contribution dominates over lh contribution significantly.

4. Conclusions
To conclude, we have analyzed theoretically the role of excitons and biexcitons on the Stark shift and self energy correction in a weak NCS CdTe QD under strong confinement regime. The two topmost valence subbands have been assumed to participate in the photoinduced band-to-band transitions in the direct gap semiconductor QDs. Increase in electric field strength reduces both Stark shift and self energy correction. The self energy correction show distinct asymmetry. The asymmetric signal has its origin in the strong biexcitonic and excitonic resonances. It is worthy to note that hh contribution dominates over lh contribution significantly. A more realistic theoretical treatment incorporating intrasubband mixing in the valence band will be the subject matter of a future publication.

5. References
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