Hyperfine Splitting and Quantum Beats in CdSe Quantum Dots

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ABSTRACT
The hyperfine splitting and consequent phenomenon of quantum beat in CdSe crystal has been discussed analytically. The origin of hyperfine splitting in CdSe quantum dot is assigned to the electron spins in exciton which may be either parallel or antiparallel to hole spin. Understanding such mechanism is important because of the observation of much smaller hole spin relaxation times as compared to the electron spin relaxation times in semiconductors. The degeneracy of the electron energy levels is lifted due to the hole angular momentum and a three-level transition picture is considered to explain the quantum beat phenomenon. We found that the quantum beats can be observed only at specific analyzer axes. Our results are well in agreement with the recently reported experimental observations.

Keywords: Hyperfine splitting, quantum beats, degeneracy, excitons and quantum dots.

1. INTRODUCTION
The quantum dots (QDs) are envisaged as vital active components in spintronics where informations can be processed by the characteristic polarization states of the input/output radiation. During photoinduced resonant electronic transitions in a QD, the polarization property of the emitted light is governed by the specific selection rules determined by the electron and hole angular momenta and respectively. The resultant gives rise to polarization states of the field output. The information carried by the polarization state of the radiation will not be corrupted as long as the coherence of the quantum states is maintained. Thus knowledge of the dephasing times of the energy states is of fundamental importance in a quantum dot system. Recently, the study of dynamics of coherent states in quantum dots has gained considerable momentum.

Quantum beat spectroscopy is one of the powerful tools to determine precisely the energy levels in atoms, molecular crystals and semiconductor nanostructures. In addition, the time resolved spectroscopy allows the investigation of the dynamics of coherent states. The underlying physical principle of these spectroscopic techniques is essentially based upon the fact that initial interaction of coherent radiation with matter sets up a coherent macroscopic polarization which decays within the dephasing time . If more than one states are excited by the radiation, the radiative outputs interfere to yield beating in the output radiation. The superposition of quantum states has been established in the study of the fine structure of the energy states in the zero-dimensional semiconductor QDs.

The fine structure of excitonic energy states in a QD is a matter of debate. The origin of splitting has so far been assigned to the mechanisms such as (i) mixing of light and heavy hole states, (ii) asymmetries in quantum dot potential and (iii) electron-hole exchange interaction. The distinction between long and short range exchange interactions are influenced by shape and size of the quantum dots. Efros and his coworkers made detailed investigations of the effects of quantum dot shape and size on luminescence polarization who inferred that the above anisotropies have been responsible for selective absorption by the electron-hole pair states. These observations agree well with the results obtained by Gammon et al who found a splitting of 20-50 µeV in a single GaAs quantum dot. The fine structure splitting has also been studied by Puls et al in CdSe quantum dot where they found the energy splitting within the allowed doublet ~200 µeV and that in case of forbidden doublet ~ 20 µeV. According to them, the splitting of the allowed doublet arises due to long range exchange interaction. Another interesting feature that confirms the polarization properties of the output signals was observed by Flissikowski et al in an experiment on quantum beat spectroscopy of CdSe quantum dot. In their experiment, Flissikowski et al found that the beats were observable only at a specific angular positioning of the analyzer showing that the interfering signals must be polarized. The beat picture was explained on the basis of lifting of degeneracy of the exciton energy doublet due to asymmetry in the CdSe quantum dot having spread of 10 nm and height of 1.6 nm. The energy splitting of the doublet was found to be ~13 µeV. The model proposed by Efros et al to calculate exciton fine structure in quantum dots is based upon degenerate valence band structure and incorporating the nonsphericity of the crystal shape and intrinsic hexagonal asymmetry. For
the CdSe quantum dot studied by Flissikowski, Efros model\textsuperscript{7-9} yields an energy splitting \( \sim \) meV. This shows that the observed energy splitting of 13 \( \mu \)eV must be having its origin in some microscopic interactions other than shape and size asymmetry.

To understand the mechanism more critically, in the present letter, we have considered the magnetic field effects arising due to hole spin on electron energy in the conduction band. It may be recalled that the hyperfine splitting of energy levels in a hydrogen atom can be explained on similar ground except the fact that in a \( \text{H} \) atom, one uses the infinite mass assumption for proton. Hyperfine interaction in semiconductors generally refers to the coupling of nuclear magnetic moment to that of the magnetic moment of electrons. In III-V semiconductors like GaAs, the nuclear magnetic moment is large enough to induce hyperfine interaction\textsuperscript{11-12}. On the contrary, in case of II-VI semiconductors like CdSe the nuclear magnetic moment is zero for most of the isotopes of Cd. Thus the cause of hyperfine interaction in CdSe QD can be assigned to interaction of hole magnetic moment with the electron magnetic moment. The splitting of exciton energy levels in a quantum dot leads to complex multicomponent time scenario where the contributions from electron and hole spins can be hardly separated. In fact, spin flip of any one of the carriers in the exciton will switch the exciton from an allowed state to a forbidden state. Due to the complicated valence band structure, the spin relaxation of holes is faster than that of electrons. As a consequence, one may find two possible energy levels for electrons with electron spin parallel as well as antiparallel to that of the hole. Under such situations, one may choose a common ground state in which hole spin is either unknown or indeterminate due to very small hole spin relaxation time. The possible excited states of electrons corresponding to opposite spins will yield oppositely polarized (RCP and LCP) radiative outputs. We have made an attempt to establish the fact that the superposition of these outputs at the analyzer axis yields the quantum beats observed in Flissikowski’s experiment\textsuperscript{1}. Accordingly, the objective of the present paper is two fold: (i) analysis of the hyperfine splitting of heavy hole excitons in CdSe quantum dots and (ii) the study of the radiative outputs and consequent beating phenomena in CdSe QD using the results of the above analysis.

2. HYPERFINE SPLITTING IN CdSe QD

The spin states of the excitons in a quantum dot can be analyzed using Dirac’s relativistic wave equation. In an exciton, the relative motion of electron can be described in a centre of mass reference frame. In Dirac equations, the relativistic correction terms are incorporated in a perturbative manner and the corrected Hamiltonian can be written as\textsuperscript{13}

\[
H = H_0 + W .
\]

In eq. (1), \( H_0 \) is the unperturbed Hamiltonian while \( W \) consists of terms arising due to relativistic corrections and accounts for the modification in the kinetic energy of electrons, spin-orbit coupling and Fermi contact term. Similar to the hydrogen atom, the physical origin of Fermi contact term in a semiconductor lies in the fact that in presence of the magnetic field effect of holes on electrons, the electron energy levels are modified. Thus, in order to incorporate only the magnetic field effects, we describe the perturbative term \( W \) in eq. (1) as

\[
W = \psi \left( \frac{\mu_e \cdot \mu_h}{m_e c^2} \frac{(r_e \cdot r_h)}{m_e m_h c^2} \right) \hat{J} \psi \left( r_e, r_h \right) (\hat{r}) .
\]

Here, \( \hat{S} \) and \( \hat{J} \) represent the spin and the total angular momentum quantum number operators of the electron and hole, respectively, \( c \) is the velocity of light while \( \psi \left( r_e, r_h \right) \) is the exciton wave function and is given by\textsuperscript{7-9}

\[
\psi \left( r_e, r_h \right) = \psi \left( r_e, a \right) \psi_s \left( r_h, a \right) \left| \phi \left( r_e - r_h \right) \right| ^2 ,
\]

with

\[
\psi \left( r_e, a \right) = \left( \frac{2}{a} \right) ^{1/2} \pi \frac{\sin \pi r / a}{r} Y_{00} ,
\]

and

\[
\psi_M \left( r_h, a \right) = 2 \sum_{\lambda = 0, 2} R_\lambda \left( r_h, a \right) \sum_{m + \mu = \lambda} \left( \frac{3/2}{\mu} \frac{\lambda}{m} \frac{3/2}{-M} \right) \lambda \mu U_\mu .
\]
Here, $a$ is the average quantum dot size. The radial function $R_a(r)$ is given by

$$R_a(r,a) = \frac{A}{a^{3/2}} \left\{ j_k(kr/a) - \frac{(-1)^{3/2} j_0(k) j_{3/2}(k,\sqrt{B} r/a)}{j_0(k,\sqrt{B})} \right\}$$

(6)

where $j_k(x)$ is the spherical Bessel function. $M, \mu = \pm 3/2, \pm 1/2$ and $k$ is the first root of

$$j_0(k) j_{3/2}(\sqrt{B} k) + j_2(k) j_0(\sqrt{B} k) = 0.$$  

(7)

Also, $A$ is the normalization constant and $\beta$ is the mass ratio of the light and heavy holes. The hyperfine splitting term takes into account only the spin angular momentum part and is given by

$$W = \frac{e^2 \eta^2}{4 m_e m_h c^2} \int |\psi(r_e,a)|^2 \left[\psi(r_e,a) \right]^2 \left[\psi(r_e-a) \right]^2 d^3 r.$$  

(8)

Using eq. (8) we have calculated the hyperfine splitting term for a CdSe quantum dot of 10 nm size with a height of 1.6 nm. The splitting energy is found to be 14 $\mu$eV and corresponds to the energy difference between the $\pm 1/2$ spin states of electrons.

3. QUANTUM BEAT PHENOMENA IN CdSe QD

We now consider the interaction of linearly polarised light with the quantum dot. The linearly polarised light comprises of right ($\sigma_+$) and left ($\sigma_-$) circularly polarised components, respectively. Each component causes selective excitations of heavy hole states. The radiative deexcitation from each state interferes to produce quantum beats. We denote the $|\pm 1/2\rangle$ states of electrons by $|a\rangle$ and $|b\rangle$, respectively. We assume that the degeneracy of these states is lifted by the magnetic field arising due to hole magnetic moment. The common ground state arising due to small hole spin relaxation time is denoted by $|c\rangle$. Thus the transitions between the states $|a\rangle, |b\rangle$ and $|c\rangle$ can be represented by the V type transitions picture. We consider that the excited states $|a\rangle$ and $|b\rangle$ are populated during the interaction of CdSe QD and near band gap resonant linearly polarized coherent radiation. This sets up a macroscopic polarization in the medium which decays within the carrier dephasing time $T_2$. The superposition of individual responses from the two excited states may give rise to the phenomenon of interference. Since the radiative outputs from these states are polarised therefore if an analyser is tuned with a variable angle $\phi_0$ (say) relative to the QD axes, the components of the emitted radiation propagating along the analyser axis will superpose to produce a beat signal. This signal will carry information about the coherence of the electronic states involved. We address ourselves to the understanding of such quantum beat phenomena in the semiconductor QD in our forthcoming discussions.

The occurrence of quantum beat phenomena can be examined analytically following the standard approach under Wigner-Weiskopf approximation. The time dependence of probability amplitudes in states $|a\rangle, |b\rangle$ and $|c\rangle$ are given by

$$\psi_a(t) = -\frac{i\mu_a E}{\eta} e^{i\delta_a t} e^{-\gamma_a t/2} c,$$

$$\psi_b(t) = -\frac{i\mu_b E}{\eta} e^{i\delta_b t} e^{-\gamma_b t/2} c,$$

(9)

(10)

and

$$\psi_c(t) = \frac{-i\mu_a E}{(\delta_a + \gamma_a / 2) \eta} \left( e^{-(\delta_a + \gamma_a / 2) t} - 1 \right) - \frac{i\mu_b E}{(i\delta_b + \gamma_b / 2) \eta} \left( e^{-(\delta_b + \gamma_b / 2) t} - 1 \right).$$  

(11)
In the above equations, \( \hat{\mu}_{ac} \) and \( \hat{\mu}_{bc} \) are the transition dipole moments related to the transition momentum matrix element \( |p_{ac}| = |p_{bc}| = |p| \) as

\[
|\hat{\mu}_{ac}| = |\hat{\mu}_{bc}| = \frac{|e| |p|}{m_e}.
\]  

(12)

Here, \( m_e \) is the electron rest mass. \( \hat{x} \) and \( \hat{y} \) are the unit vectors along \( x \)- and \( y \)-axes, respectively. Also, \( E_+ \) and \( E_- \) represents the right and left circular polarized components of the electric field. We have assumed the transition dipole moment operators \( \hat{\mu}_{ac} \) and \( \hat{\mu}_{bc} \) to act parallel to the field \( E_\pm \). Further, \( \delta_a \) and \( \delta_b \) are the detuning parameters while \( \gamma_a / 2 \) and \( \gamma_b / 2 \) represent the dephasing frequencies for states \( |a\rangle \) and \( |b\rangle \), respectively. Using rotating wave approximation and following Meystre\(^{14}\), we obtain the ensemble averages of the induced transition dipole moment as

\[
\left\langle \hat{\mu}_{ac} \right\rangle = e^{-\gamma_a t/2} \frac{|a(0)|^2 (\mu_x \hat{x} + \mu_y \hat{y} \cdot \frac{\partial}{\partial t} E_+) \times e^{-i(\delta_a + \gamma_a t)/2} - 1}{\eta(\delta_a - i\gamma_a / 2)}
\]

(13)

and

\[
\left\langle \hat{\mu}_{bc} \right\rangle = e^{-\gamma_b t/2} \frac{|b(0)|^2 (\mu_x \hat{x} + \mu_y \hat{y} \cdot \frac{\partial}{\partial t} E_+) \times e^{-i(\delta_b + \gamma_b t)/2} - 1}{\eta(\delta_b - i\gamma_b / 2)}
\]

(14)

The total induced dipole moment will be given by the vector summation \( \left\langle \hat{\mu}_T \right\rangle = \left\langle \hat{\mu}_{ac} \right\rangle + \left\langle \hat{\mu}_{bc} \right\rangle \). These transition dipoles will emit radiation at frequencies \( \delta_a \) and \( \delta_b \), respectively, and each of them will have opposite sense of polarization. The output signal will be proportional to the square of the effective induced polarization. When the signal is scanned by an analyser making an arbitrary angle \( \phi \) with the \( x \) axis, the \( x \) and \( y \) components of the induced dipole moment will contribute to the total intensity of the signal. Consequently, the field output at the analyser axis at an angle \( \phi \) is obtained as

\[
\mathcal{E}_{\text{output}}(\phi) \propto \left[ \mu_x (\mu_x E_x + \mu_y E_y) |a(0)|^2 e^{-\gamma_a t/2} \left\{ e^{-i(\delta_a + \gamma_a t)/2} - 1 \right\} \right] \\
+ \mu_x (\mu_x E_x + \mu_y E_y) |b(0)|^2 e^{-\gamma_b t/2} \left\{ e^{-i(\delta_b + \gamma_b t)/2} - 1 \right\} \cos \phi \\
+ \mu_y (\mu_x E_x + \mu_y E_y) |a(0)|^2 e^{-\gamma_a t/2} \left\{ e^{-i(\delta_a + \gamma_a t)/2} - 1 \right\} \sin \phi
\]

(15)

where we have taken \( \mu_x = \mu_y = \mu \) and \( \Omega_{ab} = (\delta_{ab} - i\gamma_{ab}/2) \).
In eq. (16), the first two terms correspond to the nutating output for optical transitions from |a⟩ and |b⟩ states, respectively to the ground state |c⟩ while the third term gives rise to the beat signal. The first term maximises at φ = π while it is minimum at φ = π/2. On the other hand, the second term maximizes at φ = π/2 and minimizes at φ = π. Thus the two orthogonal positions of the analyser yields nutating signals due to the two spin states as considered in the present model. The third term maximizes at φ = π/4 and varies sinusoidally as cos(δa − δb)t. Thus at θ = π/4 axis of the analyser, we obtain the beat signal.

To conclude, we have presented a simplistic theoretical model to explain successfully the hyperfine splitting of the excited electronic states as small as 13 µeV observed experimentally in CdSe quantum dot by Flissikowski et al. Moreover, the emergence of a photon beat signal from the same nanostructure has been interpreted in terms of the interaction of circularly polarized coherent radiation with the semiconductor quantum dot.

REFERENCES


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