Theoretical Investigations on the Effect of Laser and Magnetic field on the Donor and Exciton states in Quantum Core/Shell Nanostructures

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Abstract. Donor and exciton characteristics of different core/shell nanostructures (CSNs) in presence of external perturbing laser and magnetic field is presented. The zero-dimensional spherical CSNs are considered and their properties are explored using variational technique in effective mass approximation. The confinement potential modified by the laser irradiance greatly alters the onset of tunneling. The impact of laser on the donor in type I GaAs/Ga$_{0.7}$Al$_{0.3}$As and reverse type I Ga$_{0.7}$Al$_{0.3}$As/GaAs CSNs are examined for different donor sites. The indirect to direct exciton transition using THz laser radiance is realized in type II CdSe/CdTe CSN. The interaction of magnetic ions with the carrier of semi-magnetic CdMnTe/CdTe CSN forms Bound/Exciton Magnetic Polaron and shows enhanced shift in Zeeman energy. The screening effect of laser reduces magnetization and polarization of associated magnetic polaron. The induced electric and magnetic dipoles due to the combined laser and magnetic fields, is shown to reduce the spin exchange interaction between the carrier and Mn ions. These studies will give an insight on using an appropriate CSN for opto-electronic device applications.

INTRODUCTION

Research on semiconductor CSNs is of great attention owing to its multi-functional nature in tuning their opto-electric properties [1]. The CSN is a class of hybrid nanoparticles consisting of two different materials at nanoscale namely the inner core and the outer shell layer. The properties of such CSNs are highly tailored by controlling the size and composition of both the core and shell materials. Compared to single nanoparticles, CSNs are realized to show enhanced quantum yield [1,2]. They have been widely used in many applications such as solar cells, LED, biosensors, drug delivery systems etc [1]. In this line, the objective of the thesis is to explore some essential electric and optical properties by investigating the donor and exciton states of zero dimensional spherical CSNs. The functionality of CSNs through perturbing fields like THz laser radiance and magnetic field are examined using variational technique in effective mass approximation. To study some interesting magnetic properties of CSNs, the semi-magnetic system (SMS) with Mn doped CdTe barrier is used in presence of external magnetic field. Further a comparative study on the donor of non-magnetic and semi-magnetic CSNs is conferred. Moreover, the diamagnetic nature of donor and exciton has been used to figure out their dynamics for the first time in CSN.

THEORY

In effective mass approximation, the Hamiltonian of a carrier of CSNs in presence of external laser and magnetic fields along the polar axis \( (z = r \cos \theta) \) is defined as [3],

\[
\hat{H}(\vec{r}, t) = \frac{1}{2m_e} \left( \vec{p} + e \vec{A} \right)^2 + V_c(\vec{r}) - \frac{e^2}{\epsilon} \frac{\vec{p} \cdot \vec{r}}{\epsilon \ell} \tag{1}
\]
The radial distance between the carriers is termed as \( \mathbf{r}_{\text{ei}} \) with \( l \) as the position of hole for exciton and it is the location of dopant atom for donor. The subscript \( i = e, h \) specifies the electron and hole, \( j = c, s \) denotes the core and shell materials. The effective mass of carriers is represented as \( m_i^* \), \( \varepsilon_i \) is the high frequency dielectric constant. The confinement potential for carriers is labeled as \( V_i \). The time dependent Hamiltonian can be translated to a unitary transformation in the Kramers-Henneberger (KH) accelerated frame [3-6]. The laser dressed parameter \( \alpha \) is used to define the intensity of applied laser field. It has the dimension of length defined as [3-6],

\[
\alpha = \left( \frac{I}{\omega e^2} \right) \left( \frac{e}{m_i^*} \right) \left( \frac{8\pi}{c} \right)
\]

(2)

I is the intensity, \( \omega \) is the angular frequency of the applied laser field, \( e \) is the charge of an electron and \( c \) is the velocity of light. For exciton, \( \alpha \) will rely on the reduced mass \( \mu \) of the carrier [6]. While the external magnetic field is defined by the dimensionless parameter \( \gamma \) written as [5-8],

\[
\gamma = \frac{\hbar \omega_c}{2 R_B^*}
\]

(3)

Where, \( \omega_c \) is the cyclotron frequency of carrier with respect to the applied magnetic field and \( R_B^* \) is the effective Rydberg energy written as \( R_B^* = \frac{e^2}{2 \varepsilon a_B^*} \) with \( a_B^* = \frac{\hbar^2 \varepsilon}{m_i^* e^2} \) is the effective Bohr radius of the carrier.

The space translated effective Hamiltonian of a carrier in the unit of \( R_B^* \) and \( a_B^* \) is written as [3-6],

\[
H_{\text{dot}} = -\nabla_i^2 + \gamma L_z + \frac{1}{4} \gamma^2 \rho_i \sin^2 \theta_i + V_i(r_i, \alpha) - \frac{2}{r_{ej}^2} + \hat{H}_{\text{ex}}
\]

(4)

\[
H_{\text{barriere}} = \sigma_i \times \left[ -\nabla_i^2 + \gamma L_z + \frac{1}{4} \gamma^2 \rho_i \sin^2 \theta_i \right] + V_i(r_i, \alpha) - \rho_i \times \frac{2}{r_{ej}^2} + \hat{H}_{\text{ex}}
\]

(5)

Where, \( L_z = m\hbar \) is the z component of angular momentum, \( \sigma = m_d^*/m_b^* \) and \( \rho = \varepsilon_d/\varepsilon_b \) are the dimensionless parameters with \( m_d^* \), \( m_b^* \) are the effective mass of electron/hole in the dot and barrier; \( \varepsilon_d, \varepsilon_b \) are the optical high frequency dielectric constants. The spin exchange interaction between the carriers and Mn ions is represented as \( \hat{H}_{\text{ex}} \).

It is an expression with sum over the sites of Mn ions, \( R_k \) with \( S_e = 1/2, J_e = 3/2 \) and \( S_M = 5/2 \) as the spins of electron, hole and the Mn ions respectively; \( \beta_e, \beta_h \) are the exchange constant of electron and hole. In the KH frame, the carrier is largely subjected to the time averaged potential for the high frequency laser which is expressed as [6,7]

\[
V(r, \alpha) = \frac{\alpha^2}{\pi} \int_0^\infty V(r, \alpha(t)) \, dt = \frac{V_i}{\pi} \arccos \left( \frac{R - r}{\alpha} \right)
\]

(7)

Further by expanding and neglecting the higher order terms, the laser dressed potential is approximated as,

\[
V(r, \alpha) = \frac{V_i}{2} \left[ \Theta(r - (R + \alpha)) + \Theta(r - (R - \alpha)) \right]
\]

(8)

Where, R is the radius of dot and \( \Theta \) is the Heaviside (step) function, \( V_i \) is the conduction/valence band offset i.e., the magnitude of barrier potential for electron/hole. These laser dressed confinement potentials under frequency limit are validated only for \( \alpha \leq R \). By variational method, the trail wave function of a carrier in CSN is chosen as,

\[
\psi_{el} = N_c \psi_i(r_i) \exp \left( -\lambda r_{ej}^2(\alpha) \right)
\]

(9)

Where, \( N_c \) is the normalization constant and \( \lambda \) is the variational parameter and \( \psi_i(r_i) \) are the respective ground state wave functions of electron/hole. It is the linear combinations of spherical and modified spherical Bessel functions can be obtained by solving the respective Schrödinger equation. The binding energy of a donor/exciton is defined as,

\[
BE = E_i - \left\langle \psi_{el} \middle| \hat{H}(\lambda) \middle| \psi_{el} \right\rangle_{\text{min}}
\]

(10)

Here, \( E_i \) is the subband energy level of electron/hole which is calculated using Ben-Daniel Duke boundary conditions at the interface of core and shell material. The minimum value of \( E_i \) obtained on varying \( \lambda \) gives the exact
ground state energy of carriers. Based on the Langevin theory of diamagnetism, the diamagnetic susceptibility ($\chi_{\text{dia}}$) of exciton is obtained from the relation involving the inter particle distance ($r_{ij}$) is given by,

$$
\chi_{\text{dia}} = -\frac{e^2}{6 m_i \epsilon_i, j c^2} \frac{|\psi_{el}|^2}{|\langle \psi_{el} | \psi_{el} \rangle|}
$$

In a semi-magnetic CSNs, the interaction between the spins of confined carrier and Mn ions is defined using mean field theory with Brillouin function and the respective giant Zeeman energy shift is given by the relations [1],

$$
B_{Mn} (y) = \frac{2 S_{Mn}}{2 S_{Mn}} + 1 \text{Coth} \left( \frac{2 S_{Mn}}{2 S_{Mn}} + 1 \right) y - \frac{1}{2 S_{Mn}} \text{Coth} \left( \frac{y}{2 S_{Mn}} \right)
$$

$$
\Delta E_{\text{ZE}} = \frac{N_{Cd}}{2} \left( \frac{\beta_e - \beta_h}{2} \right) x S_0 (x) \left\{ B_{Mn} \left( \frac{S_{Mn} (\beta_e - \beta_h) |\psi_{el}|^2}{2 k_B [T + T_0 (x)]} + \frac{g \mu_B S_{Mn} B}{k_B [T + T_0 (x)]} \right) \right\}
$$

Where $N_{Cd}$ is the concentration of Cd ions, $g$ is the Lande factor, $\mu_B$ is Bohr Magneton, $k_B$ is Boltzmann constant, $x$ is the concentration of Mn ions, $S_0 (x)$ and $T_0 (x)$ are the effective spin and temperature known as phenomenological parameters in response to the paramagnetic behaviour of Mn ions. The magnetization is the result of fluctuating magnetic moments of carriers in the mean field approximation using $B_{Mn} (\eta_i)$ is given by[35],

$$
M = -N_{Cd} x g \mu_B \left\{ S_z \right\} \approx -N_Cd x g \mu_B \left\{ -S_{Mn} B_{Mn} \left( \frac{g \mu_B B}{k_B [T + T_0 (x)]} \right) \right\}
$$

RESULTS AND DISCUSSION

Incitement of laser brings coherent control over the quasi-energy levels of CSNs and alters the confinement potential of carriers. The laser dressed confinement potential mutates the size of core and shell by intruding the barrier into dot and vice versa to the extent of $\alpha$ as in equation (8). On the other hand, the external magnetic field ($B$) will bring two major effects in CSNs namely Landau quantization and magnetic confinement. The change in density of energy states under $B$ provides an additional confinement to the carriers. Also, the band gap which varies with $B$ reduces the height of barrier potential. Further in semi-magnetic CSNs, the interaction of carrier with the induced magnetic moment of magnetic ions plays a significant role in controlling the carrier dynamics.

Comparative study on the approximated laser dressed confinement potentials

FIGURE 1. (a) Laser dressed confinement potentials for $\alpha = 25$ Å (b) Binding energy of donor in type I CSN as a function of $\alpha$

The time averaged laser dressed confinement potential which is an arccos function as in equation (7) and the approximated step like potential using Heaviside function as in equation (8) are compared and illustrated in figure
The effects of these two confinement potentials on the BE of donor in type I CSN is presented in figure 1(b) for different $\alpha$ and $R_c$. It is observed that the results are merely equal for both the confinement potentials. The maximum variation in BE is nearly 1 meV for $\alpha = 5$ Å and $R_c = 70$ Å. These variations in BE is negligible for $\alpha < 15$ Å. Thus the approximated step like confinement potential is used in the present thesis to ease the numerical calculations.

**Effect of laser on the Carriers of Non-Magnetic Core/Shell Nanostructures**

FIGURE 2. Binding energy at different impurity locations for various laser field when $R_s = 100$ Å and $R_c = 70$ Å. Figure 2(a) for type I and figure 2(b) for reverse type I, (c) Diamagnetic susceptibility of exciton as a function of $\alpha$ for different $R_s$

The BE of donor in both type I and reverse type I decreases as $\alpha$ increase. The dressed energy states by the applied laser highly screens the carriers and weakens the Coulomb interaction. This laser dressing increases with increase of laser intensity and in turn reduces the BE as shown in figures 2(a) and 2(b). While the BE energy of exciton shows diverse behavior with increase in $\alpha$. This can be witnessed through the diamagnetic susceptibility ($\chi_{dia}$) of exciton as in figure 2(c). Alike BE, the $\chi_{dia}$ is directly related to the inter-particle distance between the carriers as defined in equation (11). It is observed that on enhancing $\alpha$, the probability of finding exciton inside the core decreases and it is more like to reside in the shell. Hence for smaller $\alpha < 10$ Å, the value of $\chi_{dia}$ is the one that corresponds to the core material. While for $\alpha > 10$ Å, the wave function of exciton starts conquering the shell and it affords the results of shell material. On further increasing $\alpha$, the screening effect of laser around the carriers increases and thereby decreases $\chi_{dia}$. Thereby the laser radiance excites the exciton transition from indirect to direct in a type II CSN.

**Magnetic response of Non-Magnetic and Semi-magnetic Core/Shell Nanostructures**

FIGURE 3. Magnetization ($M$) of reverse type I (a) Non-magnetic and (b) Semi-magnetic CSNs against core radius and (c) Binding energy of donor in non-magnetic and semi-magnetic CSNs beside core to shell radii under different magnetic field

The diamagnetic nature of non-magnetic system shows negative $M$ with the applied $B$ as presented in figure 3(a). In a non-magnetic CSN, for $R_c$ under very small $B < 15$ T, the impact of induced magnetic moment dominates the confinement effect of CSN. Thus, the $M$ of this domain increases with increases in $R_c$ and $B$. For $R_c > 100$ Å under high $B \geq 15$ T, the confinement effect of CSN plays an active role in controlling the $M$. As $R_c$ grows above 100 Å,
the declining shell region along with the strong $B$ enhances the kinetic energy of electron. Hence, the spatial extent of the carriers increases that in turn reduces $M$. While in SMS, $M$ increases with increase in $B$ as shown in figure 3(b). The high density of permanent and induced magnetic moments brings out larger $M$. As $R_c$ increases, the localized donor wave function of narrow shell region starts penetrating into the core. Actually, the core is the potential barrier of system with very high concentration of Mn$^{2+}$ of about 0.3. Thus, the extending core region becomes much sufficient to avail its Mn$^{2+}$ for $M$. Due to this, the concentration of Mn ions that lie with in the vicinity of extended donor orbit increases and enhances the induced magnetic moments resulting in larger $M$.

It is perceived from figure 3(c) that the BE of donor in a non-magnetic system increases with $B$ while in magnetic system the BE decreases with $B$. On enhancing $B$, the electron is likely to reside near the donor which brings strong confinement. Whereas in a SMS, the barrier height get reduced and enhances free energy of electron which results in smaller BE. It is interesting to note that the maximum value of BE in a non-magnetic system falls in the same region of core. But in SMS maximum value shifts to larger core which is the characteristic of SMS. In both CSNs, irrespective of $B$, drastic change in BE is observed for different $R_c/R_s$ ratio. When $R_c/R_s$ equals zero, it is just entirely a shell material with no core i.e., $R_c = 0$ and it gives the value of BE that belongs to the shell material. On increasing $R_c/R_s$ from 0 to 0.8, the core starts growing and reduces the shell width ($R_s - R_c$). For $R_c/R_s > 0.8$, the shell becomes much finer and it becomes almost insignificant due to the presence of very large core. At $R_c/R_s = 1$, the system contains purely the core material and hence the value of BE corresponds to the core material.

**Combined effect of laser and magnetic field on Semi-magnetic Core/Shell Nanostructures**

**FIGURE 4.** Variation of (a) Zeeman energy shift and (b) Magnetization against $\alpha$ under different magnetic field and $R_s$

The effect of laser is more prominent under smaller $R_s$ while the applied $B$ shows wide variation in BE for larger $R_s$ as shown in figure 4(a). As discussed earlier, carriers have more space to move in larger shell so the exciton wave function is well extended on increasing $R_s$. The barrier potential is reduced due to increase in magnetic field. The confinement region gets reduced via barrier intrusion due to applied laser and this supports a strong tunneling possibility of carriers for smaller $R_s$. These combined effects are responsible for the enormous change in BE for smaller $R_s$. From figure 8.8, it is found that the effect of laser is more pronounced on $\Delta E_{ZE}$ and $M$ than the applied magnetic field. Only a small change in $\Delta E_{ZE}$ and $M$ is observed on enhancing $B$. This is because of the dynamic behavior of exciton in the laser induced confining potential so that the associated ferromagnetic interaction with Mn ions is highly varied resulting in abrupt change in $\Delta E_{ZE}$ and $M$.

**REFERENCES**