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Excitation of exciton states on a curved surface

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ABSTRACT

Excitonic transitions on the surface of a sphere have been studied in he presence of external static electric and laser fields. The spectrum and the various coupling matrix elements, $\langle \psi_{l,m} | cos^n \vartheta | \psi_{l',m'} \rangle$ (for $n = 1, 2, 3$), between few states of exciton have been evaluated in the absence and presence of excitonic Coulombic interaction with different values of dielectric constant. Variation of various physical quantities: energy eigenvalues, transition probability, orientational and alignment parameter, has been shown to have strong dependence on the laser field and static electric field.

1. Introduction

Excitonic transitions have been of great importance, as the excitons play important role in physics related to interdisciplinary areas, such as quantum information and quantum computations [1,2]. Excitons are quite stable and have large lifetimes [3] making them important from technological point of view. Their use in photovoltaic and solar cell devices [4] is one of the important applications. An exciting aspect of excitons is that their size and their coupling matrix elements are not only dictated by the electron–hole Coulomb potential but also by the structure on which these excitons are formed. Excitons attract chemists as well as theorists as reported by Scholes and Rumbles [5]. The transitions of excitons to various excited states have been studied for many quantum heterostructures [6–9]. The excitons in quantum wells and dots, of different shapes have been studied in detail [10–15]. Excitons confined in quantum heterostructures have some peculiar properties such as large oscillator strengths and in these structures, excitonic spectrum persists even at room temperature [16–18]. In addition, for almost all quantum heterostructures, it is shown that the binding energies, oscillator strengths etc. of excitonic transitions, increase with increasing confinements [19].

Nowadays, in communication system, transfer of information at high speed is possible with optical fibers by using light, and information processing is done using electronic transistors. The bottleneck in the communication networks is the conversion between optical and electronic signals. One can overcome this problem with the help of optical transistors. New ways are being tried to create optical transistors by excitons and polaritons [20–22]. The size dependent properties of excitons and multiexcitons [23] have given a way to potential applications in the field of emissive displays, solar energy conversion, and biological and biomedical fluorescence imaging [24]. In addition, excitons may dephase into uncorrelated electron–hole pairs that facilitate charge transport [25].

The excitons on the surface of a sphere, in particular for the evaluation of energy levels, have been studied previously by many authors [26–29]. For example, Y. Kayanuma studied the energy levels of excitons confined in a spherical microcrystal [30] by using Ritz variational technique. Y. Kanayuma and N. Saito [26] studied quantum size effect on the Wannier excitons on the surface of a sphere. Kanayuma [31] studied ground state properties of electron–hole system confined in a spherical well, using variational method. Takagahara [32,33] has shown that excitonic optical nonlinearity gets modified in the presence of external fields. Recently the excitonic transitions have attracted renewed interest as in the case of semiconductor quantum heterostructures, the transition of valence band electrons from valance to conduction band can be realized experimentally quite easily, thus creating an exciton. These excitons have some interesting features, that need to be explored. In this paper, we study excitonic states on the surface of a sphere. We solve the resulting Schrödinger equation of such states numerically. As we are considering only excitons on the surface of a sphere, where only angular confinement is possible, hence wavefunctions and matrix elements between different states depend only on angular parameters (i.e., ϑ and ϕ), whereas radius of the sphere comes as a parameter.

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2. Theoretical methods

Consider an electron and a hole confined on the surface of a sphere of radius R. The sphere is considered as an isotropic microcrystal having a reliable dielectric constant ϵ'. The Hamiltonian of such a system is given by:

$$H = \frac{p_e^2}{2m_e^*} + \frac{p_h^2}{2m_h^*} - \frac{e^2}{\epsilon'|\vec{r}_e - \vec{r}_h|} \tag{1}$$

The excitons on the surface of a sphere are analogous to two particle system confined on layered spherical nano-structures [34] with large radii i.e., in such systems the angular motion is quite slow compared to radial electrons motion, hence one can study the angular motion in analogy with the rotational motion of a diatomic molecule. In such a case, the Coulomb interaction between particles depends only on the angular separation between them [35].

It is well known that if the optical isotropy is present, the Schrödinger equation for an exciton confined on the surface of a sphere of radius R is given by:

$$-\left[\frac{1}{\mu_m R^2}\left(\frac{\partial^2}{\partial\vartheta^2} + 2\cot\vartheta\frac{\partial}{\partial\vartheta}\right) + \frac{1}{\epsilon' R\sqrt{(2-2cos\vartheta)}}\right]\psi = E'\psi \tag{2}$$

where ϑ is the relative angular coordinate, μ_m is the reduced mass of the electron–hole system, $\mu_m = \frac{m_e^* m_h^*}{m_e^* + m_h^*}$, where m_e^* and m_h^* are the effective masses of the electron in the conduction band and hole in the valence band respectively and ϵ' is the dielectric constant of the medium.

As shown by Loos [36], for angular momentum $m = 0$ states, only equation to be solved is for relative motion, i.e., equation (2) can be reduced to the following equation for relative motion by a transformation $E \to \mu_m R^2 E'$ and $B_e = \frac{1}{\mu_m R^2}$. (For detailed calculations, please see [34,36].)

$$-\left(\frac{\partial^2}{\partial\vartheta^2} + 2\cot\vartheta\frac{\partial}{\partial\vartheta} + \frac{1}{\epsilon\sqrt{(2-2cos\vartheta)}}\right)\psi = E\psi \tag{3}$$

Now the energy eigenvalues are in the units of B_e and $\epsilon = \frac{\epsilon'}{\mu_m R}$ is the effective dielectric constant.

Having obtained the energy eigenvalues of the excitonic states, we use the eigenfunctions, thus obtained to evaluate the coupling matrix elements $\langle\psi_{l,m}|cos^n\vartheta|\psi_{l',m'}\rangle$ $(n = 1, 2, 3)$. It is well known that the excitonic transitions on the surface of a sphere in external fields depend on these coupling elements. Next, we study the transitions to various excited excitonic states from the ground state in the presence of external static field and a laser field. The static electric field interacts via $cos\vartheta$ term, while the laser field is supposed to interact via $cos^2\vartheta$ term.

The polarizability of an exciton is the extent to which an applied electric field can pull the electron and hole apart, resulting in decrease in the energy of the exciton [37]. The polarizability of electrons and holes are widely studied in quantum heterostructures [38]. It has been shown in numerous studies [39–41] that the polarizability increases with decrease in confinement. Hence it is an important parameter for laser-exciton interaction. We also consider the material of which the semiconducting material is composed of, has some polarizability parameter through which laser field may interact and this interaction is averaged to $(1/2)E_L^2\alpha cos^2\vartheta$ where E_L is the laser field strength, α is the anisotropy parameter and ϑ is the angle between the laser field and the excitonic axis. The resulting Schrödinger equation takes the form:

$$H_T\phi = E_T\phi \tag{4}$$

Table 1
Energies of few lowest states (in units of B_e) of exciton on surface of a sphere for different values of dielectric constant ϵ.

$E_{l,m}$	CT = 0	$\epsilon = 1.5$	$\epsilon = 5$	$\epsilon = 12.5$
$E_{0,0}$	0	−0.589925	−0.171742	−0.068215
$E_{1,0}$	3	2.276154	2.785433	2.914448
$E_{2,0}$	8	7.195036	7.760128	7.904226
$E_{3,0}$	15	14.136452	14.742067	14.896950
$E_{4,0}$	24	23.090542	23.728010	23.891296
$E_{5,0}$	35	34.052786	34.716499	34.886672
$E_{6,0}$	48	47.020723	47.706754	47.882759
$E_{7,0}$	63	61.992862	62.698304	62.879370
$E_{8,0}$	80	78.968231	79.690846	79.876379
$E_{9,0}$	99	97.946161	98.684171	98.873703
$E_{10,0}$	120	118.926171	119.678132	119.871283
$E_{11,0}$	143	141.907904	142.672618	142.869074
$E_{12,0}$	168	166.891088	167.667546	167.867042
$E_{13,0}$	195	193.875512	194.662849	194.865161
$E_{14,0}$	224	222.861005	223.658478	223.863410
$E_{15,0}$	255	253.847432	254.654389	254.861773

where $H_T = H_0 + H'$, H_0 being the Hamiltonian defined by equation (2) and H' is the external perturbation.

It may be noted here, that excitons confined on the surface of a sphere of radius ≥ 200 a.u., the parameter $B_e(= 1/R^2) \simeq 10^{-5}$ a.u. Hence, the system resembles to that of rigid rotor. The period of exciton in different states corresponds to few picoseconds. So the interaction of the laser field may be averaged to $(1/2)E_L^2 cos^2\vartheta$.

$$H' = H_{int}^s + H_{int}^L \tag{5}$$

As mentioned earlier, all terms are represented in terms of B_e. H_{int}^s is the interaction of static electric field with the system, while H_{int}^L is the interaction of the laser field.

$$H_{int}^s = \mu E_s' cos\vartheta, \tag{6}$$
$$E_s = \mu E_s'$$

where μ is the dipole moment operator, and

$$H_{int}^L = -\left(\frac{1}{2}\right)\alpha E_L'^2 cos^2\vartheta, \tag{7}$$
$$E_L = \left(\frac{1}{2}\right)\alpha E_L'^2$$

3. Results and discussion

As mentioned, we study excitonic transitions on the surface of a sphere in external static and laser fields. Also, the interaction terms of the static field and the laser field are taken into account in terms of B_e. It is assumed that the material of which the sphere is composed of, contains some polarizability parameter through which the laser field interaction comes into picture via $cos^2\vartheta$ dependence. The first part of the study contains the evaluation of the spectrum of excitonic states. The Schrödinger equation (2) is solved by 9-point finite difference method. We calculate energy eigenvalues and also the coupling matrix elements $\langle\psi_{l,m}|cos\vartheta|\psi_{l',m'}|\rangle$, $\langle\psi_{l,m}|cos^2\vartheta|\psi_{l',m'}\rangle$ and $\langle\psi_{l,m}|cos^3\vartheta|\psi_{l',m'}'\rangle$ for different values of effective dielectric constant ϵ (1.5, 5.0 and 12.5). It helps to understand how energy levels and coupling elements change with ϵ, and thus helps in explaining the excitation and 'order' parameters $\langle cos\vartheta\rangle$ and $\langle cos^2\vartheta\rangle$ for different excitonic states.

Table 1 shows the values of ground state and first fifteen excited states (in units of B_e) of exciton on surface of a sphere, in the absence of Coulombic term (CT = 0) and presence of the electron–hole Coulombic attraction term, for different values of dielectric constant of the material of which the sphere is made of. In the absence of Coulombic interaction, the ground state energy, $E_{0,0}$, is