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Third-order nonlinear optical susceptibility and photoionization of an exciton in quantum dots

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ABSTRACT

In this study, an investigation of an exciton in a disk-like quantum dot has been carried out by using the matrix diagonalization method and the compact density-matrix approach. The dependences of binding energies of excitons for the light and heavy hole masses on the size of the confining potential are analyzed explicitly. The third-order nonlinear optical susceptibility of third harmonic generation as well as the photoionization cross section has been calculated for the light-hole and heavy-hole excitons. The results are presented as a function of the incident photon energy. The results show that the third-order nonlinear optical susceptibility and the photoionization cross section of an exciton in a quantum dot are strongly affected by the dot size and the hole mass.

1. Introduction

Much attention has been paid recently to optical properties of low-dimensional semiconductors due to the fact that these systems offer a promising class of materials for a variety of optoelectronic device applications [1–5]. Since excitonic effects play an important role in optical phenomena in semiconductor materials, the behavior of excitons in low-dimensional systems has been extensively investigated both theoretically and experimentally. In low-dimensional semiconductors the electron–hole interaction of an exciton is enhanced owing to the confinement effect.

Quantum dots (QDs) confine charge carriers in all three space dimensions. And their size, shape, and other properties can be controlled in experiments. The three-dimensional nanoscale confinement

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of charge carriers gives rise to a full quantum nature of these structures. One elementary excitation of QDs is an exciton which plays an important role in semiconductor optical properties [6]. Meanwhile, the dependence of the optical transition energy on the confinement strength (or quantum size) allows the tunability of the resonance frequency. Hence, the optical properties of excitons in QDs have attracted an enormous interest in recent years [7–11]. These results show that intersubband optical transitions have very large optical nonlinearity in QDs. Both linear and nonlinear optical properties can be used for practical applications in photodetectors and high-speed electron–optical devices [12]. The results show that the cross section depends strongly on the binding energy and wave function of low-dimensional systems.

In low-dimensional semiconductors, the nonlinear optical susceptibilities associated with intersubband transitions are known to be considerably enhanced as compared with those in bulk semiconductors. In 1987, Hanamura studied the third-order optical polarization due to excitons in semiconductor microcrystallinity, and showed that optical nonlinearity is very large when one considers excitonic effects [13]. Recently, Karabulut and coworkers investigated the excitonic effects on the nonlinear optical properties, such as second harmonic generation, third harmonic generation, nonlinear absorption coefficient and refractive index changes in small QDs [14]. They found that the smaller confining potential frequency values will yield larger nonlinear optical susceptibilities in QDs. Among the nonlinear optical processes in semiconductor QDs, the attention has been paid to third-order nonlinear optical properties, such as third-order nonlinear optical absorption and third-harmonic generation (THG). This is because the second-order nonlinear processes does not happen in a lot of cases. The second-order nonlinear process happens only when the quantum system demonstrates significant asymmetry [8].

On the other hand, the photoemission is a useful experimental method for the determination and the study of the electronic states of low-dimensional semiconductors. The knowledge of the cross section of a material can help to detect the electronic and optical properties in these structures. In recent years, a substantial increase in the number of photoionization cross section of a hydrogenic impurity has been reported for low-dimensional electronic structures [15–20]. The cross section depends strongly on the binding energy and wave function of low-dimensional systems. Hence, in present work, we will focus on studying the size and the hole mass effects of the binding energy, the third-order nonlinearity susceptibility of THG, and the photoionization cross section of an exciton in a disk-like QD.

2. Theory

Let us consider an exciton moving in a disk-like QD with the confined parabolic potential. Hence, the Hamiltonian of the system within the effective-mass approach is given by

$$H = -\frac{\hbar^2}{2m_e}\nabla_e^2 + \frac{1}{2}m_e\omega_{e0}^2 r_e^2 - \frac{\hbar^2}{2m_h}\nabla_h^2 + \frac{1}{2}m_h\omega_{h0}^2 r_h^2 - \frac{e^2}{\epsilon|\vec{r}_e - \vec{r}_h|}, \tag{1}$$

where m_e (m_h) is the effective electron (hole) mass, $\vec{r}_e$ ($\vec{r}_h$) is the position vector of the electron (hole), ω_{e0} (ω_{h0}) is the frequency of the parabolic confining potential of the electron (hole), and ϵ is the dielectric constant.

Introducing, as usual, the center-of-mass coordinate

$$\overrightarrow{R} = \frac{m_e\vec{r}_e + m_h\vec{r}_h}{M}, \tag{2}$$

and the relative coordinate of the electron–hole pair

$$\vec{r} = \vec{r}_e - \vec{r}_h, \tag{3}$$

then Eq. (1) can be rewritten as

$$H = -\frac{\hbar^2}{2M}\nabla_R^2 + \frac{1}{2}M\omega_1^2 R^2 - \frac{\hbar^2}{2\mu}\nabla_r^2 + \frac{1}{2}\mu\omega_2^2 r^2 - \frac{e^2}{\epsilon r} + \mu(\omega_{e0}^2 - \omega_{h0}^2)\overrightarrow{R}\cdot\vec{r}, \tag{4}$$