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TWO-PHOTON OPTICAL NONLINEARITIES IN SEMICONDUCTOR QUANTUM DOTS

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Résumé - Les nonlinéarités optiques dans les points quantiques semiconducteurs sont calculées pour deux régimes de confinement quantique. Nous prédisons une résonance induite à deux photons située énergétiquement au-dessus de la résonance excitonique.

ABSTRACT - Optical nonlinearities of semiconductor quantum dots are computed for two regimes of quantum confinement. An induced two-photon resonance is predicted energetically above the exciton resonance.

I - INTRODUCTION

In recent years it has been shown that the geometric confinement of electrons and holes in semiconductor-quantum-well structures strongly modifies the optical properties in comparison to bulk semiconductors and leads to somewhat enhanced optical nonlinearities in multiple-quantum-wells. These observations stimulated the interest in so-called "quantum dots", i.e. semiconductor microstructures of a radius which is comparable to, or smaller than the exciton Bohr radius in the corresponding bulk material. In contrast to the quantum wells, where the electron-hole motion is restricted only in one spatial dimension, the quantum dots provide confinement in all spatial degrees of freedom. These features are expected to enhance the optical nonlinearities in semiconductor microcrystallites making them interesting not only from the physics point of view but also for possible device applications.

To characterize\(^1,2\) the different size regimes of semiconductor microstructures, one uses the Bohr radii

\[
a_e = \frac{\hbar^2 \epsilon_0}{\mu \epsilon^2}, \quad a_e/h = \frac{\hbar^2 \epsilon_0}{m_e/h \epsilon^2}
\]

of the excitons \((a_o)\), electrons \((a_e)\) and holes \((a_h)\), respectively, in comparison to the radius \(R\) of the quantum dot. Here \(\epsilon_0\) denotes the background dielectric constant of the semiconductor material and \(\mu\) is the reduced mass of the electron-hole-pair. Confinement takes place when one of these radii is comparable to or larger than \(R\). In the present work, we are especially interested in the so-called moderate confinement regime, \(a_h < R < a_e\), where only the electrons are quantum-confined and the much heavier holes move in the Coulomb field of the average electron charge. Since the lowest energy states for an ideal quantum-confined system are discrete,\(^1,3\) we compute the lowest-order optical nonlinearities, \(\chi_3\), including only zero-, one-, and two-pair states. For comparison, we also present the third-order susceptibility for the regime of strong quantum confinement, where the Coulomb interaction between the electrons and holes can be ignored in comparison to the confinement energy.\(^1\)
2 - THIRD-ORDER OPTICAL SUSCEPTIBILITY

In rotating-wave-approximation for the dipole interaction between light field and matter, third-order perturbation theory yields

$$\chi_3 \propto \frac{4A^2}{(i\omega_1 + \gamma)(i\omega_2 + \gamma)} - B^2 \left[ \frac{2}{(i\omega_2 - i\omega_1 + \gamma)(i\omega_1 + \gamma)} \right] + \frac{1}{(i\omega_1 + \gamma)(i\omega_2 + \gamma)} \left[ \frac{1}{i(\omega_2 - \omega_1 + \gamma)} - \frac{1}{i\omega_1 + \gamma} \right]$$

Here, $\gamma$ is the linewidth, $\hbar\omega_1 = E_1 - \hbar\omega$ and $\hbar\omega_2 = E_2 - 2\hbar\omega$, with $E_1$ and $E_2$ being the energies of the lowest one-pair and two-pair states, respectively. The coefficients $A$ and $B$ are given in terms of the matrix elements of the dipole operator between the zero-, one- and two-pair states.

If one treats the electron-hole pair as a true boson, $E_2 = 2E_1$, and $B^2 = 2A^2$, causing $\chi_3$ to be identically zero. However, in the moderate confinement regime this approximation is not valid. The Fermi nature of the constituent particles (electrons and holes) becomes important and the energy structure is modified by the confinement.

To solve the eigenvalue problems for one and two electron-hole pairs we make a series of approximations. In the moderate confinement regime the electron motion is assumed to be determined only by the crystallite boundaries (strongly confined electrons). In this case the electron state is the groundstate of a non-interacting particle in a sphere. The one- and two-pair problems reduce to solving the Schrödinger equations for the motion of a single hole in the average potential of a single electron, and for the motion of two mutually repulsive holes in the average potential of two electrons, respectively. However, due to the presence of dielectric surface charges, the Coulomb interaction potential between electrons and holes inside the microcrystallite is modified. Taking these effects into account, we obtain approximate solutions generalizing the treatment of a single electron-hole pair in Ref. 1. The details of our calculations are presented in Ref. 4.

3 - RESULTS AND DISCUSSION

Our most striking result for the regime of moderate quantum confinement is that the energy $E_2$ of the state with two electron-hole pairs turns out to be larger than twice the energy $E_1$ of a single pair. Hence, in contrast to the case of bulk semiconductors, in microcrystallites the "biexciton" state is higher in energy than twice the "exciton" state. This feature is a direct consequence of the combined Coulomb interaction and quantum confinement which is not simply additive for more particles in the same volume. As a consequence of the confinement, the two-electron-two-hole complex cannot rearrange itself to reduce the repulsive forces between the two holes leading to a net increase of the total energy. In Fig. 1 we present the computed third-order optical susceptibility for the regime of moderate quantum confinement as function of the normalized detuning $(\hbar\omega - E_1)/E_R$ for different linewidths $\gamma$. In addition to the saturation of the one-photon transition at $E_1$, we see in Fig. 1a the increasing absorption around the energy $E_1/2$. This feature is reminiscent of the induced biexciton absorption in bulk semiconductors, but $E_2/2 > E_1$ as a consequence of the quantum confinement. Since the induced absorption occurs on the high-energy side of the saturating one-photon resonance, the dispersive changes corresponding to the absorption saturation and to the induced absorption are both positive in the region $E_1 < \hbar\omega < E_2/2$ and cause the enhancement shown in Fig. 1b.
Fig. 1: Normalized optical susceptibility $\tilde{\chi}_3$ versus detuning of the photon energy $\hbar\omega$ from the lowest electron-hole resonance energy $E_1$. $E_R$ is the exciton Rydberg in the bulk semiconductor material. Fig. 1a shows the imaginary part of $\chi_3$ and 1b shows the real part, respectively. Curves (1) - (3) are for $\gamma/E_R = 0.6, 0.5, 0.4$, respectively. $\epsilon_o/\epsilon_1 = 10$ and $a_h/a_e = 0.2$.

In the regime of strong quantum confinement one may ignore the Coulomb interaction between electrons and holes. Consequently one can easily calculate the full nonlinear optical susceptibility, since the energetically lowest optical transition of the semiconductor in this case is essentially that of a two-level system. For comparison we present in Fig. 2 the numerical evaluation of the third-order susceptibility

$$\chi_3(\omega) \propto \left[ \frac{1}{\omega - E_1 + i\gamma} \left( \frac{1}{(\omega - E_1)^3 + i\gamma} \right) \right]$$

for the same parameters used in Fig. 1. Fig. 2a shows that the imaginary part of $\chi_3$ is strictly negative and resonates at the one-photon transition energy $E_1$ indicating the saturation of the one-pair resonance. The real part of $\chi_3$ in Fig. 2b shows the simple dispersive structure corresponding to the saturating absorption in Fig. 2a. The additional structures present in Fig. 1 are absent in Fig. 2 since the Coulomb effects are negligible in the regime of strong quantum confinement.

To obtain an order-of-magnitude estimate of the expected changes in absorption and refractive index, we consider the case of GaAs ($a_h/a_e \approx 0.13$) quantum dots in vacuum ($\epsilon_o/\epsilon_1 \approx 10.0$). When the surface charge effects are taken into account the absorption change is

$$\frac{\Delta \alpha}{\alpha_{\text{vac}}} \approx \left( \frac{\alpha_{\text{vac}}}{R} \right)^3 \approx 181.0 \, \text{Im} \tilde{\chi}_3$$

and the refractive index change is
\[ \Delta n/I \approx \left( \frac{a_c}{R} \right)^3 1.18 \times 10^{-3} \Re \chi_2. \]

where \( \Delta \alpha \) has the units cm\(^{-1}\), \( I \) is the light intensity in MW/cm\(^2\), and the susceptibilities are normalized as in the figures. The calculated values show that the predicted optical nonlinearities are quite large. Reducing the homogeneous linewidth of the electron-hole transition further increases the magnitude of the nonlinearities. The additional inhomogeneous broadening of the resonances due to the size distribution of the microcrystallites can be estimated in analogy to the Doppler broadening of gases.

In conclusion, our results show that semiconductor microcrystallites with strong or moderate quantum confinement should exhibit large optical nonlinearities for sufficiently small line broadening.

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Fig. 2: Same as Fig. 1 but for the regime of strong quantum confinement

References
3. see, e.g., N. Bloembergen, Nonlinear Optics, W.A. Benjamin Inc., Reading/MA, 1965