Exciton coherence in a layer of CdSe/CdS/ZnS semiconductor quantum dots under continuous laser excitation and two-quantum exciton transient holograms

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Abstract. We discuss the possibility of exciting a stimulated exciton induction in a thin film of semiconductor quantum dots and analyze the possibility of writing and reading out the two-quantum exciton transient holograms in such samples.

1. Introduction
Semiconductor quantum dots (QDs) are increasingly used in quantum optics [1]. We have been most impressed by the work of the experimental group from the Institute of Spectroscopy of the Russian Academy of Sciences (Moscow, Troitsk) with double-shell semiconductor QDs CdSe/CdS/ZnS (manufactured by QD-Light, Russia) 3–7 nm in size [2–8]. Note that these quantum dots were grown by colloidal synthesis so that the emitting CdSe core was surrounded by two shells of CdS and ZnS, which improve the photostability and quantum yield of these QDs. According to [9], upon excitation of such QDs by laser radiation at a wavelength of 580 nm, corresponding to the maximum of exciton absorption, the irreversible relaxation time at a temperature of 10 K is equal to 0.75 ps.

Quantum dots of the CdSe/CdS/ZnS type are characterized by a discrete set of energy states, like atoms, which is why they are often called "artificial atoms", and their size practically coincides with the size of Wannier–Mott excitons [10] (see also monographs [11, 12]). Therefore, we made an assumption [13, 14] about the excitonic nature of the coherent phenomena observed in [6–9]. The novelty of this theoretical consideration lies in the fact that the working objects in this case are the excitons of QDs, and not impurity centers. We first discuss the possibility of exciting the stimulated excitonic induction (SEI), and then analyze the possibility of recording and reading out the two-quantum excitonic transient holograms.

2. Stimulated excitonic induction
Here we follow [15], in which the Heisenberg equations of motion for the components of the exciton electric polarization vector under continuous laser excitation and their solutions are written. Then, an expression was obtained for the SEI intensity per unit solid angle $\Delta \Omega$ in the direction of the excitation wave vector $\vec{\omega}$:

$$\Delta I_{SEI} \sim \frac{N^2}{8\pi^2 d^2} \sigma d \frac{E_0^2 T_2^2 (1 + \Delta \omega^2 T_2^2)}{A^2} \Delta \Omega,$$

where $N$ is the concentration of QDs, $d$ is the thickness of the film, $E_0$ is the amplitude of the excitation field, $T_2$ is the decay time of the exciton, and $A$ is the area of the sample.
where $N_{x}$ is the number of "working" excitons with the wave vector $\vec{k}$, $V$ is the volume of the quantum dot, $\vec{d}_{f}$ is the electric dipole moment of the resonant transition, $E_{0}$ is the amplitude of the electric field of the exciting light wave, $T_{1}$ is the transverse relaxation time, $\Delta \omega_{x}$ is the frequency detuning, $A = 1 + \Delta \omega_{x}^{2}T_{2}^{2} + d_{f}^{2}E_{0}T_{1}T_{2}$. An extremum study of $\Delta I_{SEI}$ shows that for $\Delta \omega_{1,2} = \pm \frac{1}{T_{2}} \sqrt{|d_{f}^{2}E_{0}T_{1}T_{2} - 1|}$ there are two maxima, equal to $(\frac{2\pi}{\omega_{B}^{2}})d_{f}^{2}(\frac{\omega_{2}}{T_{1}})$, where $T_{1}$ is the longitudinal relaxation time. Let us discuss how the spectrum of exciton absorption (emission) of the semiconductor QDs changes when taking into account the fact that the statistics of excitons differs from the bosonic one in the case of interaction of excitons, for example, with one branch of optical vibrations $\Omega$ (without taking into account a dispersion). Following [16], we obtain

$$
\partial(\vec{k}, t) = 2\pi N^{-1} \sum q(\xi \Omega)^{2} \{ \langle V_{q} \rangle - \langle N_{x} + q \rangle + \frac{2}{N} \sum k \langle N_{k} \rangle \} \cdot \delta(h \omega - E_{x} + q + h \Omega) + 
$$

$$
+ \left[ 1 + \langle V_{q} \rangle + \langle N_{x} + q \rangle - \frac{2}{N} \sum k \langle N_{k} \rangle \right] \cdot \delta(h \omega - E_{x} + q + h \Omega),
$$

where $\langle V_{q} \rangle$ and $\langle N_{k} \rangle$ are the average values of the number of phonons with the wave vector $\vec{k}$ and the number of excitons with the wave vector $\vec{k}$, $\zeta$ is the coupling parameter of the electronic and vibrational excitations, $\vec{k}$ and $\omega$ are the wave vector and frequency of laser excitation, $N$ is the number of semiconductor QDs.

So, as a result of laser action, the temperature of QDs rises and phonons appear, the interaction of excitons with which causes broadening of spectral lines and a change in the formula for $\Delta I_{SEI}$. Of course, this issue still requires its detailed consideration.

3. Excitonic transient holography on CdSe/CdS/ZnS quantum dots

Let us discuss the possibility and conditions of excitation of the two-quantum transient holograms on the semiconductor QDs, following A.K. Rebane with colleagues [17]. Excitons are excited in the QDs in a two-quantum mode by two picosecond laser beams, one of which is an object one and the other is a reference one. As the coherent responses carrying a holographic wave front, we chose correlated free induction decay (FID) signals, as was done in [18]. The formation of such a wave front is considered in [19]. The excitation wavelength in the case of the CdSe/CdS/ZnS QDs should be 1160 nm.

According to [18], the electric field of correlated FID signals excited in the two-quantum mode in the approximation of small pulse areas can be written as:

$$
\vec{E}_{FID}(t) = \vec{E}_{0}^{2}h^{-1} \Delta t Q \Phi_{FID} \hbar \omega \frac{h \omega}{2k_{0}^{2}} f \sum f \left[ e^{-i(\vec{k}_{FID} - \vec{k}_{1} + \vec{k}_{2})\vec{r}_{j}} + e^{-i(\vec{k}_{FID} - \vec{k}_{1} + \vec{k}_{2} + \vec{k}_{1})\vec{r}_{j}} \right],
$$

where $\vec{E}_{0}^{2}$ is the electric field amplitude, $\Delta t$ is the duration of the exciting pulses, $Q$ is the two-quantum excitation parameter, $\Phi_{FID}$ is the FID form factor, $\omega$ is the excitation frequency, $k_{0}$ is the Boltzmann parameter, $T$ is the operating temperature, $f \approx 1$ is a parameter that controls the dimension, $N$ is the number of participating excitons, the angle between the wave vectors $\vec{k}_{1}$ and $\vec{k}_{2}$ is 60°.

When forming the two-quantum correlated transient holograms (CTH), one of the exciting beams is an object one and has a nonplanar wavefront:

$$
\vec{E}_{1}(\vec{r}, t) = \sum_{\eta=1}^{m} \vec{E}_{0\eta}^{(1)} \exp[i(\omega t - \vec{k}_{1\eta} \vec{r} - \varphi_{1}(\vec{r}))],
$$
(where $\tilde{E}_{0}^{(1)}$, $\omega$ and $\varphi_{1}(\vec{r})$ are the electric field amplitude, excitation frequency and phase of the object wave) and, according to the rules of transient holography, its electric field must be decomposed into a Fourier series of plane waves. The second beam is a reference beam with a plane wavefront

$$\tilde{E}_{2}(\vec{r}, t) = \tilde{E}_{0}^{(2)} \exp[i(\omega t - \vec{k}_{2}\vec{r} - \varphi_{2}(\vec{r}))],$$

and does not require decomposition. Here $\tilde{E}_{0}^{(2)}$ is the electric field amplitude, $\omega$ is the excitation frequency, $\varphi_{2}(\vec{r})$ is the phase of the reference wave. The parameters $\varphi_{1}(\vec{r})$ and $\varphi_{2}(\vec{r})$ define the wavefronts of the corresponding beams.

According to [19], the electric field of the two-quantum correlated transient hologram has the form:

$$\tilde{E}_{CTH}(\vec{r}, t) = \tilde{E}_{0}(\vec{r}) \times$$

$$\times \left\{ \exp[-i(\vec{k}_{1} - \vec{k}_{2} - \vec{k}_{CTH})\vec{r} - \omega(t - \Delta t) - (\varphi_{CTH}(\vec{r}) - \varphi_{1}(\vec{r}) + \varphi_{2}(\vec{r}))] + $$

$$+ \exp[-i(\vec{k}_{2} - \vec{k}_{1} - \vec{k}_{CTH})\vec{r} - \omega(t - \Delta t) - (\varphi_{CTH}(\vec{r}) - \varphi_{2}(\vec{r}) + \varphi_{1}(\vec{r}))] \right\},$$

where $\tilde{E}_{0}(\vec{r}) = E_{0}^{(1)}E_{0}^{(2)}\Delta Q\Phi_{CTH}\theta^{2}\frac{\hbar^{2}}{2\hbar\omega^{2}}e^{-\frac{(t-\Delta t)}{2\tau_{2}}}T$, $\tau_{2}$ is the phase relaxation time. One can see that the correlated signals of excitonic free induction decay (EFID1 and EFID2) propagate in mutually opposite directions:

$$\vec{k}_{EFID1} = \vec{k}_{1} - \vec{k}_{2} \quad \text{and} \quad \vec{k}_{EFID2} = \vec{k}_{2} - \vec{k}_{1}$$

and have reversed wave fronts:

$$\varphi_{CTH1}(\vec{r}) = \varphi_{1}(\vec{r}) \quad \text{and} \quad \varphi_{CTH2}(\vec{r}) = -\varphi_{1}(\vec{r}).$$

4. Conclusion

Above, the possibility of recording and reading out the two-quantum excitonic transient holograms was shown. The question arises about the nature of excitons in semiconductor QDs of CdSe/CdS/ZnS type, which are discussed in [7–9]. If we were talking about free Wannier–Mott excitons, then the irreversible relaxation time $T_{2}$ would be 5 fs [20]. In our case, $T_{2}$ of excitons is about 1 picosecond, i.e. we are dealing with localized Wannier–Mott excitons. Therefore, the theory of such transient holograms on an ensemble of excitons is the same as for impurity centers [21] (see also monograph [22]). Note that in the case of thin films containing QDs it is convenient to perform laser excitation and detection of the coherent responses using total internal reflection prisms [23].

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