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Correlated signals of cluster superfluorescence under two- and three-photon excitation of CdSe/CdS nanostructures

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Abstract. The possibility and conditions for generation of the correlated signals of cluster superfluorescence (CSF) under two and three-quantum excitation of nanostructured samples CdSe/CdS by two crossed at the angle of 60° femtosecond beams of the Ti:Sapphire laser radiation are investigated. It is shown that intensity of the CSF signals is proportional to the cube of the number of clusters and that these collective signals are generated in mutually opposite directions $\mathbf{k}_1 - \mathbf{k}_2$ and $\mathbf{k}_2 - \mathbf{k}_1$, where $\mathbf{k}_1$, $\mathbf{k}_2$ are the wavevectors of the exciting pulses.

R. Dicke in his basic work [1] devoted to the optical superradiance (SR) also discussed the case of an incoherent pump when at the initial moment of time the radiators (atoms, molecules or clusters) are in the excited state and have no macroscopic polarization. It was shown that under conditions when the linear dimensions of the system are much less than the wavelength λ of the radiated light such a system of the inverted atoms can spontaneously undergo transition to the ground-state during the time which is inversely proportional to the number of atoms N. This case of the spontaneous collective radiation was named in [2] as a superfluorescence (SF). The SF, as well as the SR, is a perfect example of a self-organization [3]. The collective character of photon emission in the SF is due to the indistinguishability of atoms in a system (with linear dimensions $L < \lambda$) interacting with a field. The SF signals appear to be delayed by the time $t_0 = \tau_c \ln N$, where $\tau_c = T_1 / N$ is the time of correlation self-formation, T_1 is the lifetime of the excited state, N is the number of radiators.

The samples studied in the present work are composites consisting of the CdSe/CdS nanoparticles (of core/shell type) dispersed into a PMMA polymer matrix. The average size of grown particles with the spherical or elliptical shape is equal to $5 \cdot 10^{-7}$ cm. The wavelength of the Ti:Sapphire laser is equal to $8 \cdot 10^{-5}$ cm with pulse duration of 40 fs. So, the area of excitation with the linear dimensions of $8 \cdot 10^{-5}$ cm can involve about 160 areas (clusters) emitting the SF signal. For the first time the term “cluster superfluorescence” appeared in paper [4] while studying the quadrupole superradiance [5, 6]. The authors of this paper constructed the generalized Dicke model and showed that the intensity I of the quadrupole cluster superradiation is proportional to N^3. In our case, the situation is more complicated because of the two- and three-photon modes of the excitation by crossed femtosecond laser

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beams. The presence of the angle of 60° between the wavevectors of the first $\mathbf{k}_1$ and the second $\mathbf{k}_2$ exciting pulses leads, according to [7], to the emitting of the coherent CSF signals in the mutually opposite directions $\mathbf{k} = \mathbf{k}_1 - \mathbf{k}_2$ and $\mathbf{k}' = \mathbf{k}_2 - \mathbf{k}_1$.

The peculiarities of the excitation of the coherent responses in the semiconductor CdSe/CdS nanostructures by the Ti:Sapphire were discussed in details in [8]. Earlier, in [9], it was experimentally shown that one needs the three-photon mode in order to excite the electrons of such structures by the radiation of this laser. It was established that both conduction bands of the sample are excited coherently. But then electrons from the upper conduction band decay nonradiatively to the lower one. This is the reason why the nonequilibrium population grating at the energy transition between the valence band and the upper conduction band (257 nm) disappears. But this grating which is important for a calculation of the correlated signals of CSF still exists at the energy transition between the valence band and the lower conduction band (534 nm). Although the excitation has a three-photon character, the presence of the nonradiative transition allows us to simplify the situation and consider the two-photon mode of an excitation. Calculations performed by the method presented in [4] (see also [3]) give us the following formula for the intensity of CSF signals:

$$I(\mathbf{k}_{\mathrm{CSF}}) \sim I_0(\mathbf{k}_{\mathrm{CSF}}) N^3 M A \lambda^2 \,\mathrm{th}^2\left(\frac{\hbar\omega}{2k_{\mathrm{B}}T}\right) e^{-t'/T_2},$$

where $I_0(\mathbf{k}_{\mathrm{CSF}})$ is the intensity of the spontaneous radiation of the isolated cluster in the direction $\mathbf{k}_{\mathrm{CSF}}$, N is the number of active clusters, M is the parameter of quadrupole radiation of a cluster, A is a parameter of two-photon electron absorption, ω is the carrier frequency of the Ti:Sapphire laser, k_{B} is the Boltzmann constant, T is the temperature of the sample, t' is the decay time of CSF signal, T_2 is the time of transverse nonreversible relaxation.

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