

# Femtosecond correlated transient holography in CdS crystal under two-quantum excitation

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**Abstract.** This paper is devoted to the discussion of a possibility of the femtosecond correlated transient holograms recording in CdS crystal under two-quantum excitation.

## 1. Introduction

Let us analyze the situation when the femtosecond correlated transient holograms (FCTH) are recorded into the wave fronts of coherent responses such as the free induction decay (FID) signals formed in CdS crystal under two-quantum excitation. These signals may be used for quantum calculations and are characterized by property of superposition for which there always exists a complete set of eigenstates named as a basis [1]. The property of superposition means that there is no possibility to say which of the two resonant states is realized in practice. For coupled particles there is a correlation (for example, due to the Coulomb interaction of electrons in semiconductor crystal CdS) between them. This correlation leads to the appearance of the correlated states. Electrons in such a state can form an EPR-pair [2].

In our paper [3] we have considered the signals of the correlated free-induction decay (CFID) emitted in CdS crystal under two-quantum excitation by femtosecond 793 nm radiation of a Ti:sapphire laser. The exciting beams propagating in the directions  $\mathbf{k}_1$  and  $\mathbf{k}_2$  at an angle of  $60^\circ$  to each other induce the appearance of two EPR-gratings of nonequilibrium electric polarization  $\mathbf{k}_1 - \mathbf{k}_2$  and  $\mathbf{k}_2 - \mathbf{k}_1$  in the crystal. The reason for formation of two gratings is following. The electrons in the CdS crystal need to absorb the energy of two photons of laser radiation in order to pass from valence zone to conduction one. But we should take into consideration only photons belonging to different laser beams. It should be also noted that two quanta of laser energy ( $2\hbar\omega_L$ ) are equal to the sum of quanta ( $\hbar\omega_L$ ) of CFID coherent responses.

## 2. CFID signals in CdS crystal under two-quantum excitation

Since there are two EPR-gratings the process of simultaneous emission of two correlated responses is similar (but only with respect to  $\sigma_\pm$  polarizations) to the process of the FID formation in a ruby [4]. Omitting the details of calculation let us write the following formula for the CFID electric field in the case of two-quantum excitation:



$$\mathbf{E}(t, \mathbf{r}) = 2 \int_0^\infty d(\Delta\omega) g(\Delta\omega) \sum_j^N W(\Delta\omega, \mathbf{r}, t) e^{-i\mathbf{k}_{\text{CFID}} \mathbf{r}}, \quad (1)$$

where  $\Delta\omega = \omega_e - 2\omega_L$  is the detuning,  $\omega_e$  is the frequency of the two-quantum energy transition of an electron,  $\omega_L$  is the laser frequency,  $g(\Delta\omega)$  is the distribution function of the detuning  $\Delta\omega$ ,  $W(\Delta\omega, \mathbf{r}, t) = -\frac{i}{2} \sin \theta e^{-i\Delta\omega(t-\Delta t)}$ ,  $\Delta t$  is the pulse duration,  $\theta$  is the pulse area which under the two-quantum excitation is described as

$$\begin{aligned} \theta &= \hbar^{-1} \Delta t \left[ \frac{\langle e | \mathbf{p} \mathbf{E}_0 e^{i(\omega_L t - \mathbf{k}_1 \mathbf{r})} | \gamma \rangle \langle \gamma | \mathbf{p} \mathbf{E}_0 e^{i(\omega_L t + \mathbf{k}_2 \mathbf{r})} | g \rangle}{\hbar(\omega_\gamma - \omega_L + i\Delta_\gamma)} + \frac{\langle e | \mathbf{p} \mathbf{E}_0 e^{i(\omega_L t - \mathbf{k}_2 \mathbf{r})} | \gamma \rangle \langle \gamma | \mathbf{p} \mathbf{E}_0 e^{i(\omega_L t + \mathbf{k}_1 \mathbf{r})} | g \rangle}{\hbar(\omega_\gamma - \omega_L + i\Delta_\gamma)} \right] \\ &= \hbar^{-1} \Delta t Q \mathbf{E}_0^2 \left[ e^{-i(\mathbf{k}_1 - \mathbf{k}_2) \mathbf{r}} + e^{-i(\mathbf{k}_2 - \mathbf{k}_1) \mathbf{r}} \right]. \end{aligned} \quad (2)$$

Here  $Q = \frac{\langle e | \mathbf{p} | \gamma \rangle \langle \gamma | \mathbf{p} | g \rangle}{\hbar(\omega_\gamma - \omega_L + i\Delta_\gamma)}$  is the parameter of two-quantum excitation,  $\langle e | \mathbf{p} | \gamma \rangle$  is the electric dipole moment of the transition,  $|g\rangle$ ,  $|\gamma\rangle$  and  $|e\rangle$  are the ground, intermediate and excited states of an electron,  $\omega_\gamma$  and  $\Delta_\gamma$  are the frequency and the width of the intermediate state  $|\gamma\rangle$ ,  $\mathbf{E}_0$  is the amplitude of the electric field of the exciting pulses,  $\mathbf{k}_1$  and  $\mathbf{k}_2$  are the wave vectors of the exciting pulses which are at the angle  $\varphi=60^\circ$  to each other. Then the expression (1) for the electric field of CFID for small  $\theta$  takes the form

$$\mathbf{E}(\mathbf{k}_{\text{CFID}}, t) = E_0^2 \hbar^{-1} \Delta t Q \Phi_{\text{CFID}} \text{th}^2 \frac{\hbar \omega_e}{2k_B T} f \sum_j^N \left[ e^{-i[\mathbf{k}_{\text{CFID}} - \mathbf{k}_1 + \mathbf{k}_2] r_j} + e^{-i[\mathbf{k}_{\text{CFID}} - \mathbf{k}_2 + \mathbf{k}_1] r_j} \right], \quad (3)$$

where  $\Phi_{\text{CFID}}$  is the form-factor of CFID, which depends on the excitation conditions. The corresponding phase matching conditions follow from formula (3):

$$\begin{aligned} \mathbf{k}_{\text{CFID}} &= \mathbf{k}_1 - \mathbf{k}_2, \\ \mathbf{k}'_{\text{CFID}} &= \mathbf{k}_2 - \mathbf{k}_1. \end{aligned} \quad (4)$$

### 3. Correlated transient holograms

In the experimental work [5] A.K. Rebane with colleagues has shown the possibility of recording of the femtosecond two-quantum holograms in the organic samples. The method was demonstrated on the simplest holographic example – the recording of the frequency-domain gratings, when the first optical beam with the wave vector  $\mathbf{k}_1$  and phase  $\varphi_1$  is a referent wave and the second beam with the wave vector  $\mathbf{k}_2$  and phase  $\varphi_2$  is an object one. Since the pulses in both beams are femtosecond we assume that they reach the sample simultaneously. It should be noted that in the case of ordinary transient holography on one-quantum transitions (see review [6]) the exciting beams are divided by time intervals smaller than the  $T_1$  and  $T_2$  relaxation times. Rebane with colleagues has demonstrated that the time interval between the referent and the object beams can be zero under two-quantum excitation. Let us now examine the possibility of the femtosecond recording and reading of the correlated transient two-quantum holograms in the semiconductor crystal CdS at low temperature. In

the area of exciting beams' intersection on the surface of the sample there are the following two optical waves :

*referent wave*

$$\mathbf{E}_1(\mathbf{r}_j, t) = \mathbf{E}_0^{(1)}(\mathbf{r}_j) \exp\left\{i\left[\omega_L t - \mathbf{k}_1 \mathbf{r}_j - \varphi_1(\mathbf{r})\right]\right\}, \quad (5)$$

*object wave*

$$\mathbf{E}_2(\mathbf{r}_j, t) = \sum_{\eta=1}^m \mathbf{E}_{0\eta}^{(2)} \exp\left\{i\left[\omega_L t - \mathbf{k}_{2\eta} \mathbf{r}_j - \varphi_2(\mathbf{r})\right]\right\}. \quad (6)$$

Here  $\mathbf{E}_0^{(1)}(\mathbf{r}_j)$ ,  $\omega_L$ ,  $\mathbf{k}_1$  and  $\varphi_1(\mathbf{r})$  are the amplitude, laser frequency, wave vector and phase of the referent wave.  $\mathbf{E}_{0\eta}^{(2)}$ ,  $\omega_L$ ,  $\mathbf{k}_{2\eta}$  are the amplitude, laser frequency, wave vector of  $\eta$ -th component in a Fourier transform of the object wave,  $\varphi_2(\mathbf{r})$  is the phase of object wave. The electric field of the referent wave together with each plane Fourier-component of the object wave forms the plane Fourier-response of FID. The sum of such Fourier-responses gives us the wave front of the total FID response named as a transient hologram. Now one principal moment should be noted. Since there are two EPR-gratings in CdS crystal then two scattered FID responses will be scattered by these gratings into two opposite directions. So, the formula for CFID electric field can be used for calculation of electric field of the transient holograms. But we will use another representation of the electric field under calculation by formula (2). According [7] the electric field of pulses is conveniently expressed over the wave fronts:

$$\begin{aligned} \mathbf{E}_1(\mathbf{r}_j, t) &= \mathbf{E}_0^{(1)}(\mathbf{r}_j) \exp\left\{i\left[\omega_L t - \mathbf{k}_1 \mathbf{r}_j - \varphi_1(\mathbf{r})\right]\right\}, \\ \mathbf{E}_2(\mathbf{r}_j, t) &= \mathbf{E}_0^{(2)}(\mathbf{r}_j) \exp\left\{i\left[\omega_L t - \mathbf{k}_2 \mathbf{r}_j - \varphi_2(\mathbf{r})\right]\right\}, \end{aligned} \quad (7)$$

where  $\varphi_1(\mathbf{r})$  and  $\varphi_2(\mathbf{r})$  are the wave fronts of the first and the second pulses. Here a wave front is a surface of a same phase. Then, the electric field of the correlated transient two-quantum hologram is written as

$$\mathbf{E}_{\text{CTH}}(\mathbf{r}, t) = \mathbf{E}_0(\mathbf{r}) \left[ e^{-i(\mathbf{k}_1 - \mathbf{k}_2 - \mathbf{k}_{\text{CTH}})\mathbf{r} - \omega_L(t - \Delta t) - [\varphi_{\text{CTH}}(\mathbf{r}) - \varphi_1(\mathbf{r}) + \varphi_2(\mathbf{r})]} + e^{-i(\mathbf{k}_2 - \mathbf{k}_1 - \mathbf{k}_{\text{CTH}})\mathbf{r} - \omega_L(t - \Delta t) - [\varphi_{\text{CTH}}(\mathbf{r}) - \varphi_2(\mathbf{r}) - \varphi_1(\mathbf{r})]} \right]. \quad (8)$$

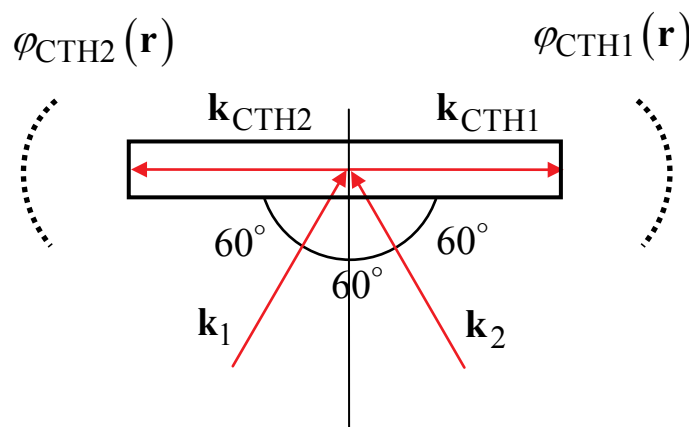
where “CTH” stands for “correlated transient hologram”,  $E_0(\mathbf{r}) = E_0^{(1)} E_0^{(2)} \hbar^{-1} \Delta t Q \Phi_{\text{CTH}} \text{th}(\hbar \omega_L / 2k_B T) f e^{-(t - \Delta t)/T_2}$ ,  $T_2$  is the total dephasing time (for CdS  $T_2 = 680$  fs), which is due to the exciton-exciton interaction [7]. The corresponding phase matching conditions follow from formula (8):

$$\begin{aligned} \mathbf{k}_{\text{CTH1}} &= \mathbf{k}_1 - \mathbf{k}_2, \\ \mathbf{k}_{\text{CTH2}} &= \mathbf{k}_2 - \mathbf{k}_1. \end{aligned} \quad (9)$$

The wave fronts  $\varphi_{\text{CTH}}(\mathbf{r})$  of the correlated transient holograms are equal to

$$\begin{aligned}\varphi_{\text{CTH1}}(\mathbf{r}) &= \varphi_1(\mathbf{r}) - \varphi_2(\mathbf{r}), \\ \varphi_{\text{CTH2}}(\mathbf{r}) &= \varphi_2(\mathbf{r}) - \varphi_1(\mathbf{r}).\end{aligned}\tag{10}$$

Since the wave front of the referent wave can be zero, the wave fronts of the holograms CTH1 and CTH2 are reversed. The situation is explained in figure1.



**Figure 1.** Formation of the correlated transient holograms “CTH1” and “CTH2” in CdS crystal.

#### 4. Conclusion

Above, the possibility of the correlated transient two-quantum holograms in the CdS crystal is shown. These holograms are reversed. For the spherical wave fronts we obtain [8]

$$\begin{aligned}\frac{1}{R_{\text{CTH1}}} &= \frac{1}{R_1} - \frac{1}{R_2}, \\ \frac{1}{R_{\text{CTH2}}} &= \frac{1}{R_2} - \frac{1}{R_1},\end{aligned}\tag{11}$$

where  $R_1$  and  $R_2$  are the radius of curvature of the corresponding exciting waves. The holograms “CTH1” and “CTH2” are placed in the wave fronts of signals CFID1 and CFID2. The results of this paper can be used in quantum optics [9].

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