THEORY OF GENERATED FOUR-WAVE MIXING FOR TWO-PHOTON CREATION OF EXCITON CONDENSATE IN C\textsubscript{2}O

I. V. Beloussov*, J. B. Ketterson**, L. V. Arapan*, and Y. Sun**

* Institute of Applied Physics, Academy of Sciences of Republic of Moldova, 5 Academiei str., Chisinau, MD-2028, Republic of Moldova
** Department of Physics and Astronomy, Northwestern University, Evanston, Illinois 60208, USA

Abstract

The experimental situation in which excitons in Cu\textsubscript{2}O are pumped directly into a state with wave vector \( \mathbf{k} = 0 \) by two counter-propagating laser pulses, and subsequently probed by a third, time-delayed, pulse has been theoretically investigated. It has been shown that additional electromagnetic waves are excited in the system. A specific configuration of two pump pulses and one probe pulse has been found to generate only one additional electromagnetic wave. The dependence of the time-integrated photon number in this wave on the delay time between pump and probe pulses for some values of the detuning has been investigated.

The possibility of two-photon excitation of a non-equilibrium Bose condensate has been theoretically investigated. We consider the situation, where the incoming laser beam is split into two beams, which are then directed at the sample from opposite directions. The polarization vectors of these beams are chosen in such a way that exciton creation results from the absorption of one photon from each beam; this process is allowed by the selection rules for two-photon absorption by orthoexcitons having the symmetry applicable to Cu\textsubscript{2}O [1]. Spin orbit coupling splits the 1s yellow excitons in Cu\textsubscript{2}O into a threefold degenerate \( ^3\Gamma_{25}^+ \) (orthoexciton) state, with wave functions transforming as xy, yz, zx, and a single \( ^1\Gamma_2^+ \) (paraexciton) state. The two-photon transitions from the ground state of the system into orthoexciton states are allowed, but are forbidden for the paraexciton state.

The state of the system then can be probed by an optically delayed beam extracted from the same laser pulse. If we regard the counter propagating beams 1 and 3, with wave vectors \( \mathbf{k}_1 \) and \( \mathbf{k}_3 = -\mathbf{k}_1 \), respectively, as pump beams which create a uniform \( \mathbf{k} = \mathbf{k}_1 + \mathbf{k}_3 = 0 \) polarization through condensate formation, then the probe beam 2 with wave vector \( \mathbf{k}_2 \) will produce a phase-conjugated beam 4 with wave vector \( \mathbf{k}_4 = -\mathbf{k}_2 \). This resulting beam 4 signal would then serve as a measure of the decay of the condensate amplitude following its formation. Consider the configuration shown schematically in Fig. 1.

![Fig. 1](image-url)
Our aim here is to investigate the propagation through the crystal of two counter-propagating pump pulses and a delayed probe pulse. This process is accompanied by the generation of a phase-conjugated signal. Strictly speaking, to treat this problem one needs to consider solutions of the dynamical equations which are non-uniform, not only in time, but also in space, and which satisfy specific conditions at the crystal boundaries. However we will restrict ourselves to the simpler problem involving only the time evolution of the system. It should be good approximation to regard the total electromagnetic field in the crystal as a superposition of two spatially uniform fields. The first of them, which we will assume has a specific envelope in time, is generated by external sources. The second field arises from a polarization of the medium resulting from exciton creation in the crystal by the first field. The set of equations therefore contains two phenomenological parameters: the source \( j_x(k,t) \), which creates the specific electromagnetic field in the crystal, and the photon damping \( \Gamma_{ph}(k) \), that takes into account the photon escape from the crystal. This approach is valid only for laser pulses in which the spatial space extent exceeds the crystal dimensions. The additional phenomenological constant \( \Gamma_ex(k) \) in the equations takes into account the exciton dephasing.

We obtain classical equations for the macroscopic exciton and photon amplitudes \( a_\alpha(k,t) \) and \( c_\lambda(k,t) \) (\( \alpha = xy, yz, zx; \lambda = 1,2 \)). We choose the orientation of the wave vector \( k \) and polarization vectors \( e^{(i)}(k) \) of the two pump and one probe pulses in such a way, that the set of equations for the amplitudes has the simplest form: \( k_x = -k_i = |k_i| \hat{y}, k_2 = |k_i| \sin \phi \cos \theta, e^{(1)}(k_i) = e^{(1)}(k_2) = -e^{(2)}(k_3) = \hat{x}, e^{(2)}(k_i) = -e^{(1)}(k_3) = -\hat{x}, e^{(2)}(k_2) = \hat{x} \sin \phi - \hat{y} \sin \theta \). Here \( \hat{x}, \hat{y}, \) and \( \hat{z} \) are the unit vectors defining the Cartesian coordinate system. Furthermore, we consider only the case of very small angles \( \phi, \theta \), so \( \cos \phi \approx 1 \). In this approximation we obtain that the dynamics of the system is described only by the amplitudes \( a_{xy}(0,t), a_{yz}(k_2 + k_3, t), a_{zx}(k_1 + k_4, t), c_i(k_4, t) \) (\( i = 1,2,3,4 \)).

From the set of the equations for these amplitudes it follows that: a) the waves with the wave vectors \( k_3, k_1, \) and \( k_2 \) are, in turn, influenced by the excitons that they generate; b) the additional electromagnetic wave with the amplitude \( c_i(k_4, t) \) is excited. For this wave \( k_4 = -k_2, e^{(1)}(k_4) = \hat{x} \cos \phi - \hat{y} \sin \phi \), \( e^{(2)}(k_4) = \hat{z} \).

We define the total number of photons generated during the whole time interval, normalized on the total photon number in each pulse. Note it as \( N_{ph}(k_x,t_d) \). We investigate how \( N_{ph}(k_x,t_d) \) depends on the delay time \( t_d \geq 0 \) for some values of the detuning \( \hbar \Delta = \hbar \omega_{ex}(0) - 2 \hbar \omega_{ph}(k_1) \geq 0 \), where \( \hbar \omega_{ex}(k) \) and \( \hbar \omega_{ph}(k) \) are the energies of the excitons and photons in the states with wave vector \( k \), respectively.

The right-hand sides of the obtained equations are proportional to the dimensionless interaction constant \( \xi = g |C(0)|/ \hbar \omega_{ph}(k_1) \), where \( C(t) \) is the amplitude of the envelope of each of the three pulses 1, 2, and 3. To solve these equations numerically we assume that the pulse envelope has the Gaussian shape and its duration \( \tau_p = 30 \text{ps} \). Furthermore, we choose the value of the \( C(0) \) such as \( \xi = 0.01 \). For the photon life time in the crystal we take the time-of-flight through the sample: \( \hbar / \Gamma_{ph} = \sqrt{\varepsilon_{ph}}(L/c) \). Here \( \varepsilon_{ph} \) is the background permittivity of the medium, \( L \) is the sample thickness, and \( c \) is the speed of light in vacuum.
For $L = 3mm$, $\varepsilon_b = 6.5$ we have $\Gamma_{ph} = 25.8\mu eV$. For the exciton dephasing energy we use $\Gamma_{ex} = 1\mu eV$.

The numerical results are presented in Figs.2-4.

**Fig. 2.** $N_{ph}(k_x, t_d)$ vs. $t_d$ for $\hbar\Delta = 10\Gamma_{ex}$ (thin line) and $\hbar\Delta = 10^2\Gamma_{ex}$ (thick line). Inset: the same, but on different scale.

**Fig. 3.** $N_{ph}(k_x, t_d)$ vs. $t_d$ for $\hbar\Delta = 10\Gamma_{ex}$. Inset: the same, but on different scale.
Fig. 4. $N_{ph}(k,t_d)$ vs. $t_d$ for $\hbar\Delta = 0$. Inset: the same, but on different scale.

Fig. 2 shows that the curves for $\hbar\Delta = 10^3\Gamma_{ex}$ and $\hbar\Delta = 10^2\Gamma_{ex}$ are of the oscillating character and display a peak for $t_d > 0$. As the delay time $t_d$ increases, the period of oscillations initially decreases, but then increases. For the case when $\hbar\Delta = 10^3\Gamma_{ex}$ the $N_{ph}$ magnitude can be three times greater than for the case $\hbar\Delta = 10^2\Gamma_{ex}$.

As can be seen in Fig. 3, $N_{ph}(k,t_d)$ vs. $t_d$ has an oscillatory dependence, as for the cases $\hbar\Delta = 10^3\Gamma_{ex}$ and $\hbar\Delta = 10^2\Gamma_{ex}$, except within the two intervals $24.8 \text{ ps} < t_d < 27.4 \text{ ps}$ and $33.5 \text{ ps} < t_d < 38.7 \text{ ps}$, where multiple, chaotically-located, narrow peaks of various heights are observed. In the first of these intervals the magnitude of $N_{ph}$ reaches its maximal value ($\sim 0.32$), but small changes in $t_d$ can lead to large decrease in $N_{ph}$.

According to Fig. 4 for $\hbar\Delta = 0$ and small delay times ($t_d < 395 \text{ ps}$), $N_{ph}$ depends chaotically on $t_d$. As $t_d$ increases, this dependence becomes smoother, and for $t_d > 816 \text{ ps}$ the pronounced dips appear. The distances between the dips increase as $t_d$ increases. The chaotic behavior of the function $N_{ph}$ presented in Figs. 2 and 3 is due to chaotic dependence of the number of generated photons on the time $t$ at small values of the parameters $\hbar\Delta = 0$ and $t_d$.

References