Efficient Two-Photon Light Amplification by a Coherent Biexciton Wave

R. Shimano,[1] Yu. P Svirko,[2,*] A. Mysyrowicz,[2,3] and M. Kuwata-Gonokami[1,2,†]

[1]*Department of Applied Physics, University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan*
[2]*Cooperative Excitation Project, ERATO, Japan Science and Technology Corporation, 4-1-8 Hon-cho, Kawaguchi, Saitama 332-0012, Japan*
[3]*LOA, ENSTA, Ecole Polytechnique, Palaiseau, France*

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A reversible coupling between photon pair states and a long-lived, highly coherent biexciton wave in CuCl allows efficient phase-sensitive two-photon amplification or attenuation of ultrashort light pulses. We demonstrate a gain of 350 cm^{-1} for a pump intensity of 1 MW/cm^2, nearly 2 orders of magnitude higher than that achievable with conventional parametric crystal amplifiers. We develop a theoretical model that describes this new type of parametric converter where the light pump is replaced by a coherent biexciton wave and show that it is well suited for the generation of entangled photons and the squeezing of an optical beam with ultrafast time gating.

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Exploiting the quantum nature of light in optical measurements, cryptography and information processing has attracted growing interest in recent years. For instance, squeezed light beams, in which the quantum noise of one quadrature component of the radiation field is reduced below the vacuum limit by projecting it onto the other component, is of paramount importance for ultrasensitive optical detection [1]. Also, entangled photon pairs which show nonlocal quantum mechanical correlation are key components of quantum cryptography and information processing [2]. In quantum cryptography, the well defined total symmetry of the entangled state implies that a polarization measurement performed on one photon of the pair determines the polarization of its partner. This enables one to uncover unwanted signal recording and, therefore, ensures secure transmission of information. In quantum information processing, one can simultaneously accomplish an operation on the entire set of nonseparable states, thereby gaining a large factor in processing time.

Up to now, the most practical method to generate entangled photon pairs is parametric down-conversion using noncentrosymmetrical nonlinear crystals [3,4]. In this process, the incident photons of an incoming pump beam are converted into correlated pairs of photons. Parametric down-conversion can also result in the squeezing of light. Typically, conventional nonlinear crystals used in the parametric process operate in a transparent region for the pump beam, in order to have a large conversion bandwidth [5]. The relatively low quantum efficiency of the down-conversion process characteristic of a nonresonant case is compensated by phase-matching techniques. The achievable degree of squeezing is a few dB in a single path configuration, limited by nonlinear propagation effects [6]. A higher degree of squeezing can be achieved in a confocal cavity geometry [7,8], but at the expense of operational simplicity and bandwidth $\Delta\nu$.

In this Letter, we demonstrate a different mechanism for the efficient generation of correlated photon pairs and the squeezing of photon states in a CuCl crystal. Although it benefits from a high efficiency associated with a resonance effect, its operation bandwidth makes it possible to process laser pulses with duration as short as 60 fs. It relies on the following scheme. The energy of a coherent pump laser pulse is stored in the crystal in the form of a long-lived, highly coherent biexciton wave via a resonant two-photon absorption process. The stored excitation is later exchanged with a second probe pulse, again in a two-photon process. Depending on the phase between the coherent biexciton and the probe wave, either amplification or attenuation of the probe beam occurs, resulting in either two-photon amplification or light squeezing. Since the lowest biexciton state is a totally symmetric state (Γ_1), polarization selection rules of the two-photon transition ensure a generation of nonseparable two-photon states. It is therefore well suited for quantum processing of entangled photon states.

There are several features which make this scheme particularly attractive for light squeezing or production of entangled photon pairs. First, a bandwidth of more than 1 THz is available for the biexciton-mediated amplification process. The bandwidth is determined by the polariton dispersion in the crystal rather than the phase-matching constraint that is crucial in a conventional parametric down-converter. This can be understood from Fig. 1. We create the coherent biexcitons by resonant two-photon excitation with a linearly polarized short pulse of central frequency tuned to half of the biexciton energy. Energy and momentum conservation conditions to fulfill the transition can be satisfied not only with photons of energy $\hbar\omega_0 = \hbar\omega_b/2$, but also with nondegenerate photon pairs in the broad spectrum of the ultrashort pulse [9]. As can be seen in Fig. 1, the resulting momentum spread in the created biexcitonic wave is very narrow, $\Delta k = 10^3$ cm^{-1} around $2k_0 = 8.9 \times 10^5$ cm^{-1}, where k_0 is the wave vector of the photon with energy $\hbar\omega_0$. Conversely, this phase-space compression allows

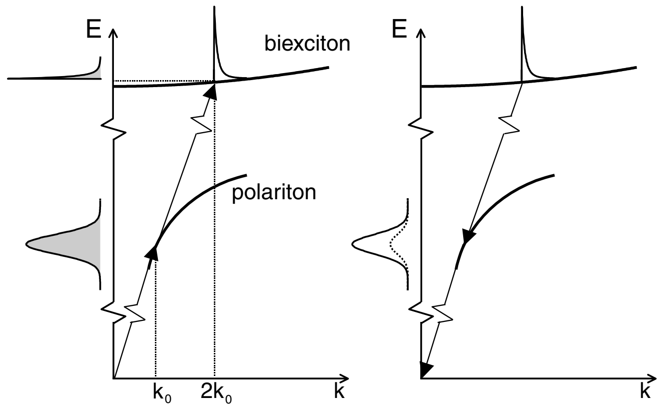

FIG. 1. Schematic illustration of the parametric amplification and deamplification of the optical pulses by a coherent biexciton wave. A coherent biexciton wave prepared by two-photon absorption of a laser pulse, shown in the inset, stores the second order coherence at frequency $2\hbar\omega_0 = 6.372$ eV within the radiative dephasing time of 50 psec. With pulses of 12 meV spectral width, the momentum distribution of created biexcitons is $\Delta k = 10^3$ cm^{-1}. A subsequent pulse, tuned to half the biexciton energy, interacts with the biexciton wave and is parametrically amplified (in phase condition) or deamplified (out of phase condition).

one to amplify ultrashort laser pulses with a highly coherent biexcitonic wave. It should be noted here that a biexciton can be created also by absorption of two photons with nonequal frequencies the wave vectors of which are determined by polariton dispersion. Our analysis shows that the bandwidth of the parametric process is mainly determined by the group velocity dispersion, i.e., by the second derivation of $E(k)$ in Fig. 1. In the frequency degenerate case, nonzero group velocity dispersion limits the bandwidth to 10 THz in CuCl. In the nondegenerate case, the group velocity is different for signal and idler pulses. Such a group velocity mismatch limits the bandwidth and makes it narrower than that of the degenerate case. Second, the equivalent figure of merit in the biexciton-mediated parametric process is very high. The biexciton wave keeps its coherence during a long time, comparable to its lifetime, 50 ps [10], because of the weak exciton-phonon coupling in CuCl at low temperatures. This implies that the coherence carried by incident photon pairs can be stored in the crystal for up to 50 ps, which is equivalent to a cavity with a Q factor of 10^5. Finally, one benefits from the large two-photon cross section associated with the particular transition, in which two photons are simultaneously absorbed by two electrons to create the biexciton. A large gain can be obtained in a very compact system, with a crystal a few microns in thickness.

In the experiment, the frequency-doubled output of a *cw* mode-locked Ti:sapphire laser, with photon energy of $\hbar\omega_0 = 3.186$ eV, pulse duration of 2 ps, repetition rate of 76 MHz, and a spot diameter of 22 μm, is injected in the sample. Since the biexciton binding energy in CuCl is large, $\hbar\Omega_B = 32$ meV, no significant one-photon absorption of the pump takes place. High quality platelet single crystals grown in the vapor phase are used in the experiments. The samples are held strain-free at 7.8 K in a helium gas-flow cryostat. Measuring the four-wave mixing signal of a noncollinear signal pulse monitors the time evolution of the biexciton wave amplitude. A more detailed description of experimental conditions can be found in [9].

For a pump pulse intensity of 0.7 MW/cm^2, 40% of the light energy is transferred to the crystal in the form of a coherent biexciton wave with an initial biexciton density of 10^{14} cm^{-3}. A collinear probe pulse, with an intensity of 46 kW/cm^2, arrives on the sample after a delay of 5.7 ps. The polarization vectors of pump and probe pulses are set orthogonal to each other. A Glan-Thompson prism placed after the sample transmits only the probe pulse to a detector. The temporal separation between the two pulses can be finely adjusted with a piezoelastic element. Figure 2 shows the transmittance of the probe pulse and the squared biexciton amplitude as a function of delay

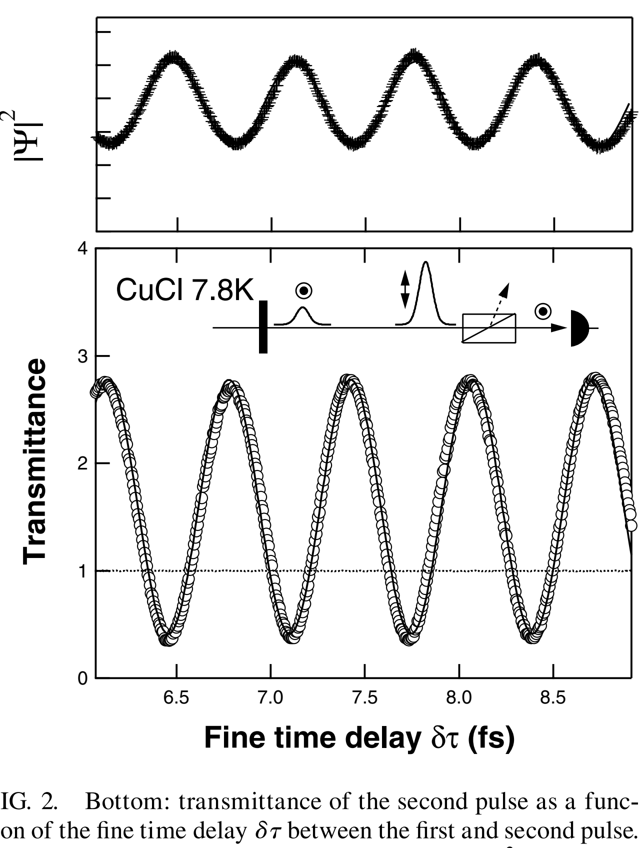

FIG. 2. Bottom: transmittance of the second pulse as a function of the fine time delay $\delta\tau$ between the first and second pulse. The respective intensities are 700 and 46 kW/cm^2. The transmittance shows parametric amplification and deamplification with an oscillation period corresponding to the biexciton frequency $2\omega_0$. A sketch of the experimental configuration is also shown. Top: simultaneously measured four wave mixing intensity, showing the squared biexciton wave amplitude $|\psi|^2$. Sinusoidal wave fits are shown by solid lines.

time. The reversible coherent coupling of the biexciton and light waves is clearly observed as a phase dependent modulation of the transmitted intensity. The energy of the probe laser pulse increases up to 3 times due to phase-coherent amplification. Figure 3 shows the maximum and minimum transmission of the probe pulse as a function of pump pulse intensity.

To model the effect, we first note that the process can be viewed as a parametric process, except that the signal and idler photons (of nearly same energy in this case) are produced by the fission of a biexciton rather than the splitting of a pump photon. The coherent energy exchange between biexciton and light waves can then be described by the following equations:

$$\tau_p\left(\frac{\partial\psi}{\partial t} + \gamma\psi\right) = -iA_+A_-; \qquad \frac{\partial A_\pm}{\partial z} + \frac{1}{c}\frac{\partial A_\pm}{\partial t} = -i\beta\psi A^*_\mp. \tag{1}$$

Here ψ is the normalized amplitude of the biexcitonic wave, $A_\pm$ are amplitudes of the left- and right-circular polarized components of the light wave, γ is the biexciton dephasing rate, and τ_p is the light pulse duration. $\beta = 2\pi^2\omega_0\mu_{ex}^2\mu_{bi}^2\tau_p/c^2n\hbar^3\Omega_B^2 v_{cell}$ determines the resonant two-photon absorption coefficient, where μ_{ex} and μ_{bi} are dipole moments for exciton and biexciton states, respectively; v_{cell} and n are, respectively, the cell volume and refractive index of the crystal. In Eq. (1) we neglect the small momentum spread of the biexciton wave, which is created in the crystal by resonant two-photon absorption. Since τ_p and the time delay between pulses τ are much shorter than the biexciton dephasing time ($\gamma\tau$, $\gamma\tau_p \ll 1$), we can obtain from (1) the following equations for the intensity of the transmitted first pulse and intensity of the second pulse at maximum and minimum transmission:

$$I_1^t = I_1^{in}/(1 + bI_1^{in}); \qquad I_2^{max,min} = I_2^{in}(1 + bI_1^{in})^{\pm 2}, \tag{2}$$

where $b = \beta L/2$, L is the with crystal thickness, $I_{1,2}^{in} = I_{1,2}\exp\{-r^2/a^2\}$ are the intensities of the linearly polarized pump and probe pulses, respectively, a is the beam radius, and r is the transverse coordinate. By integrating the transmitted intensity (2) over the beam area, we arrive at the following equations for the transmittance of the first and second pulses:

$$T_1 = \frac{\ln(1 + bI_1)}{bI_1}; \qquad T_2^{min} = 1/(1 + bI_1); \qquad T_2^{max} = 1 + bI_1 + (bI_1)^2/3. \tag{3}$$

The calculated transmitted intensity of the first pulse and the transmittance of the second pulse are presented in Fig. 3 by solid lines, with $b = 1.75\ \mathrm{cm^2/MW}$ which corresponds to a two-photon absorption coefficient $\beta \approx 10^6$ cm/GW. This allows us to estimate the biexciton dipole moment as $\mu_{bi} = 5.9e$ Å, which is close to the previously obtained values of $4.2e$ Å (biexciton Rabi splitting [11]) and $4.8e$ Å (radiative lifetime measurements [10]).

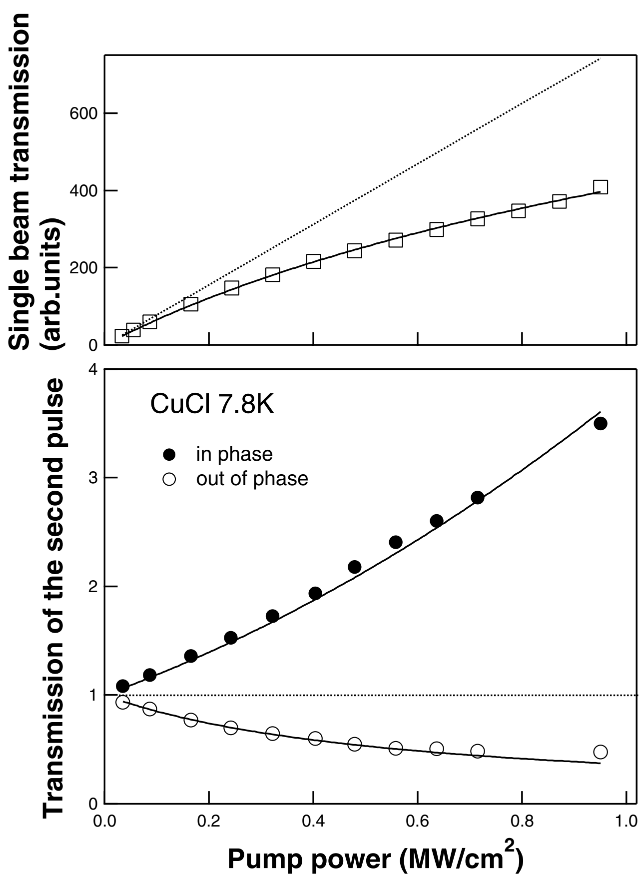

FIG. 3. Bottom: dependence of the maximum (filled circles) and minimum (empty circles) of the second pulse transmittance on the first pulse intensity. The second pulse intensity is kept constant, at 46 kW/cm^2. Top: single beam transmission versus incident pulse intensity, showing two photon absorption due to the biexciton. Theoretical fits are shown by solid lines.

The phase-sensitive interaction of the probe pulse with the coherent biexciton wave may result not only in a change of its amplitude but also in squeezing of light. The situation here resembles the conventional quadrature squeezing in parametric amplifiers, with the biexcitonic wave playing the role of the pump beam. Specifically, let us assume that the ultrashort pump pulse has created the biexcitonic wave with frequency 2ω and wave vector $2k$, which freely evolves in the crystal with negligible dephasing. If the intensity of the pump pulse is high enough to treat the biexciton wave classically, the annihilation operator for the transmitted photons of the second pulse with energy $\hbar\omega$ and momentum $\hbar k$ reduces to the conventional form of parametric down-conversion [1]:

$$a_2 = a_{20}\cosh\beta|\psi|ct - a_{20}^\dagger e^{-2i\omega\delta\tau}\sinh\beta|\psi|ct, \tag{4}$$

where a_{20} ($a_{20}^\dagger$) is the annihilation (creation) operator at

$t = 0$. Therefore, the interaction of the radiation with the classical biexciton wave results in the squeezing of the coherent light beam, as in a conventional parametric down-conversion. In the experiment, we observe a parametric deamplification of 0.4, which corresponds to 2 dB noise reduction from vacuum level at a biexciton density of 10^{14} cm^{-3} with a pump beam of 0.7 MW/cm^2 intensity and 2 ps duration. From Eqs. (3) one can calculate that for a pulse of the same duration a squeezing of 10 dB between the quadrature amplitudes would be obtained with a pump intensity of 57 MW/cm^2. If we choose a longer pump pulse duration the same degree of squeezing can be obtained with lower pulse intensity. For instance, 10 dB squeezing would require only 11 MW/cm^2 for a 10 ps pump pulse. This is because amplification (deamplification) is a function of the product $\tau_p I_1$ in the limit $\gamma\tau_p < 1$ [see Eqs. (1) and (4)].

Based on the observation of the strong parametric gain, we can now address the feasibility of a parametric oscillator based on a CuCl crystal, with a thickness much smaller than that of conventional nonlinear $\chi^{(2)}$ crystals. The oscillation threshold can be obtained from Eq. (2), which describes the amplification of the light wave with frequency ω and wave vector k coupled with the biexciton wave of the same phase velocity. If the reflectance of the ring mirror system is R, the oscillation threshold can be obtained from the following equation:

$$R(1 + bI_{1\mathrm{th}})^2 = 1. \tag{5}$$

That is, the threshold intensity is given by $bI_{1\mathrm{th}} = |R|^{-1/2} - 1$. For reflectivity of the vacuum/crystal interface of $R = 0.1$ this gives a threshold biexciton density of 3×10^{14} cm^{-3}, which can be realized with a rather moderate excitation condition, as already discussed previously. Spherical-shaped micron-size CuCl crystals also offer a remarkable opportunity to achieve a very low threshold parametric oscillation by exploiting the ultrahigh Q value of whispering gallery modes in a very small mode volume [12].

Another intriguing aspect of the parametric processes involving biexciton waves is the opportunity to switch off the parametric gain rapidly. The long-lived coherence of the biexciton wave provides us with a scheme of ultrafast encoding by using the coherent control technique with femtosecond pulses [9]. A phase jump of $\pi/2$ is sufficient to switch from an amplifying to an absorbing state. By using a phase-controlled succession of optical pulses and by inducing a $\pi/2$ shift, one can extinguish the biexciton gain and achieve time gated operation of a squeezed light source with femtosecond accuracy.

In conclusion, we have presented experimental results and a theoretical model describing the coherent coupling between coherent light and a coherent biexciton wave in a thin CuCl single crystal. We have shown that a strong amplification of ultrashort light pulses can take place due to a particular parametric conversion process, where the usual pump beam is replaced by a classical biexciton wave. Finally, we have shown the feasibility to obtain 10 dB squeezing in a single path with a sample a few tenths of microns in thickness and examined the conditions required for a biexciton-mediated parametric oscillator.

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*Also at Physics Department, University of Joensuu, P.O. Box 111, Joensuu, 80101 Finland.
†To whom correspondence should be addressed.
Electronic address: gonokami@ap.t.u-tokyo.ac.jp

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