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# Nonlinearity of biexciton waves in CuCl

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**Abstract**
We propose an experimental scheme to evaluate the intrinsic anharmonicity of the biexciton ensemble by extending the conventional four-wave mixing technique to coherent biexciton waves, which can be prepared by two-photon absorption of ultrashort optical pulses. The anharmonicity originates from the two-body interaction of the bosonic quasi-particles at low densities and leads to Kerr-type nonlinearity for the coherent biexcitonic waves generated in the crystal. We discuss the feasibility of determining the two-body biexciton interaction energy in CuCl by measuring the anharmonicity of the biexcitonic ensemble.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

Recent advances in Bose–Einstein condensation (BEC) of neutral atoms have made it possible to develop a new technique for manipulating materials with wave optical methods such as interference and diffraction. The two-body interaction of particles, which accounts for the density-dependent correction to the energy of an atom in the condensate, is the crucial parameter which determines the stability of the macroscopic coherence and leads to collective quantum-mechanical phenomena such as superfluidity. The density proportional contribution to the BEC atom energy also permits the nonlinear regime in matter wave optics that gives rise to such phenomena as four wave mixing and solitons [1–3]. The quantum information is another important aspect of the two-body interaction, which can produce the entanglement and squeezing of BEC atoms [4] leading to quantum optics with matter waves.

An ultracold atomic cloud is not a unique object for the study of nonlinear wave phenomena in an ensemble of degenerate bosons. Excitons and biexcitons, which can be created by

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irradiating semiconductor crystals with light, offer interesting opportunities to probe and exploit the macroscopic quantum effects in the ensemble of massive bosonic particles. Excitons are composed of an electron and a hole, and under favourable conditions may behave like massive Bose particles resembling positronium or hydrogen atoms. If the separation between excitons is large enough in comparison with the exciton radius, they are nearly ideal bosons and show linear optical response. With an increase in the exciton density the fermionic character of the exciton constituents manifests itself as a weak exciton–exciton interaction. The spin-dependent interaction between excitons gives rise to optical nonlinearity of the electron–hole system. Correspondingly, information on the interaction between excitons can be obtained by using methods of nonlinear optical spectroscopy (e.g. the four wave mixing technique) [5].

Since the exciton effective mass is of the order of the free electron mass, macroscopic quantum effects can be observed in the macroscopic excitonic ensemble at higher temperatures than in atomic gases. For instance, Bose–Einstein condensation is expected to appear at around 10 K for an exciton density of $10^{17}$ cm$^{-3}$, compared to $10^{-3}$ K for an atomic gas of similar density.

Recent observations of macroscopic quantum coherence in quantum wells [6, 7] and bulk semiconductors [8] have revealed, however, that BEC experiments are severely complicated by the composite nature of quasi-particles in semiconductors. In particular, the electron–hole pairs are created by an interband excitation, which inevitably results in partial transfer of the photon energy to the crystal lattice, leading to the heating of the sample at a finite exciton concentration. Moreover, the finite lifetime of the quasi-particles, their usually strong dipole coupling with photons and short diffusion length makes it difficult to create the excitonic ensemble in a high density and low temperature state.

In bulk semiconductors, the macroscopic quantum effects can be studied for long-living quasi-particles that are not resonantly coupled with light. These are paraexcitons in $Cu_2O$ [9, 10] and biexcitons [8, 11]. In CuCl, the biexciton with a ground energy of 6.372 eV at 2 K has a lifetime of 50 ps and can be created by a two-photon process with a giant cross section [12, 13]. Its wavefunction is totally symmetric with s-like symmetry and its effective mass has been found to be $m = 5.29m_0$ [14]. Luminescence studies have shown that a biexciton ensemble behaves like an ideal Bose gas. The ensemble is spread in momentum space according to the Maxwell distribution at low densities, evolving towards a Bose distribution at higher densities and lower temperatures [15].

We have recently shown [8] that a pulsed coherent light source tuned to half the biexciton energy, 3.186 eV, can create a large number of biexcitons in a very small number of momentum states, thereby effectively producing a supercooled bosonic gas. In the conventional scheme of laser cooling, light from a laser (the ensemble of coherent photons with low entropy) gradually reduces the temperature of atoms through a series of absorption–emission processes [16]. In contrast with such a laser cooling scheme, in CuCl, an ensemble of massive cold particles are effectively created quasi instantaneously by an ultrashort laser pulse tuned to half the biexciton energy, $\hbar\omega_0 = \hbar\omega_b/2$. Since the biexciton binding energy in CuCl is as large as 32 meV, the optical polarization of the medium at the frequency $\omega_0$ is off-resonant from the exciton and no significant one-photon absorption takes place. For a two-photon process, the energy and momentum conservation conditions can be fulfilled not only for two degenerate photons with energy $\hbar\omega_0$, but also for non-degenerate photon pairs in the pulse spectrum. Specifically, the high frequency component of the incident pulse couples with the low frequency component satisfying energy and momentum conservation. As can be seen in figure 1, the momentum spread of the created biexcitonic wave is very narrow, $\delta k \simeq 10^3$ cm$^{-1}$ around $2k_0 \simeq 8.9 \times 10^5$ cm$^{-1}$, where $k_0$ is the wavevector of the photon with energy $\hbar\omega_0$. Since the creation process is very short, corresponding to the pulse duration, it is possible to follow

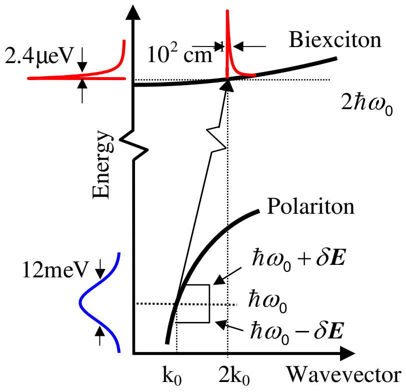

**Figure 1.** Creation of a supercooled population of biexcitons by two-photon excitation in CuCl. A photon with excess energy $\delta E$ can find a partner photon of equivalent energy defect $\delta E$ to complete a two-photon transition and convert into a biexciton while still conserving energy and momentum. Also shown is the calculated distribution of biexcitons in momentum space created by a femtosecond laser pulse. The corresponding initial biexcitonic effective temperature is estimated to be $T \approx 3 \times 10^{-2}$ K.

the subsequent evolution of the ultracold biexciton population. In CuCl, biexciton–acoustic phonon coupling is weak at low temperature [17] and thus biexcitons, which preserve their initial small momentum spreading for several tens of picoseconds, are effectively thermally isolated from the lattice. Therefore this is a unique system for investigating the wave nature of an ensemble of biexcitons in a highly degenerate state. In our previous paper, we demonstrated the linear interference of coherent biexciton waves [8] and the non-linear interaction with photon field [18]. With an increase of the amplitude of biexciton waves, interaction between the biexcitons becomes pronounced resulting in the anharmonicity of the biexcitonic ensemble. In this regime the nonlinear behaviour of the biexciton wave takes place. Correspondingly, the energy of the two-body biexciton interaction can be obtained from the cross-section of the nonlinear scattering of coherent biexciton waves.

In this paper, we discuss the Kerr-like nonlinearity of biexciton waves caused by the two-body interaction of biexcitons and the four-wave mixing of the biexcitonic waves. We show that the biexcitonic wave exhibits a nonlinear phase shift (which is close to $\pi$ at a biexciton density of $10^{16}$ cm$^{-3}$) similar to that for the intense light wave in a Kerr-like medium.

## 2. Model

Up to Mott densities, a biexciton ensemble in CuCl can be described in terms of the weakly interacting Bose gas with positive scattering length (the Bogolubov approximation). Taking into account the two-body biexciton–biexciton interaction the system Hamiltonian can be presented in the following form [19, 20]:

$$H_{\mathrm{b}} = \hbar\omega_{\mathrm{b}} b^{\dagger} b + \hbar W (a^3/V) b^{\dagger} b^{\dagger} b b, \qquad (1)$$

where the $\omega_{\mathrm{b}}$ is the biexciton frequency, $a$ is the exciton Bohr radius, $V$ is the crystal volume and $W$ is the energy of the two-body biexciton interaction. The calculation taking into account the non-momentum-transfer interaction between biexcitons returns $W \simeq 17.4R$ [19, 20], where $R = 213$ meV is the exciton Rydberg for the $Z_3$ exciton in CuCl. Such a cubic anharmonicity of the coherent biexcitonic ensemble leads to various possible nonlinear processes, which are well known in optics. In order to illustrate this, let us consider the self-phase-modulation of the coherent biexcitonic wave. In nonlinear optics, it is well known that self-phase-modulation is the most general manifestation of the third-order optical nonlinearity of the medium. The temporal evolution of the biexciton annihilation operator can be obtained from equation (1) in the following form:

$$\partial b/\partial t + \mathrm{i}\omega_{\mathrm{b}} b = -2\mathrm{i}W(a^3/V)b^{\dagger}bb. \tag{2}$$

At the given biexciton population, which is determined by the intensity of the incident light pulse and two-photon absorption coefficient, a self-phase-modulation of the biexcitonic wave takes place,

$$b = b_0 \exp(-\mathrm{i}\omega_{\mathrm{b}} t - 2\mathrm{i}na^3 Wt), \tag{3}$$

where $n = \langle b^{\dagger}b\rangle/V$ is the biexciton density, which does not change in time, according to equation (2). $\langle\cdots\rangle$ stands for statistical averaging.

To examine the feasibility of detecting the Kerr-type nonlinearity, let us estimate the magnitude of the nonlinear phase shift which the biexciton wave acquires during the dephasing time $\tau_{\mathrm{b}}$,

$$\Phi = 2na^3 W\tau_{\mathrm{b}}. \tag{4}$$

By substituting $R = 200$ meV, $a = 7$ Å and $\tau_{\mathrm{b}} = 50$ ps we can conclude that in CuCl, the $\pi$ phase may be accumulated in the crystal at a biexciton density of about $10^{16}$ cm$^{-3}$, which is considerably lower than the Mott density. We would like to note that our recent experiments [8, 18] have not revealed the effects of the biexcitonic nonlinearity. This is because these experiments have been carried out at a biexciton density below $10^{14}$ cm$^{-3}$, i.e. the nonlinear biexciton phase shift was of the order of $10^{-2}$ rad.

The two-order difference between the biexciton density necessary to achieve the nonlinear wave regime (i.e. $10^{16}$ cm$^{-3}$) and the Mott density (i.e. $10^{18}$ cm$^{-3}$) provides a range large enough to study various third-order nonlinear phenomena. In particular, one may perform the four wave mixing (FWM) of coherent biexciton waves in the conventional counter-propagation geometry (compare with the recent experiment in BEC [1]). In nonlinear optics, the degenerate FWM process involves mutual interaction of four light waves of the same frequency. In the biexcitonic counterpart of such a process, four biexcitonic waves with different wavevectors but the same frequency $\omega_{\mathrm{b}}$ are involved.

## 3. Discussion

The proposed scheme is based on the long dephasing time of biexcitons in CuCl that are created in CuCl by two-photon absorption of ultrashort pulses, with the central frequency tuned to the two-photon resonance. A large biexciton binding energy in CuCl ($E_{\mathrm{b}} = 32$ meV) ensures large detuning of the central frequency of the incident pulse from the exciton resonance. Therefore, the incident pulse does not create a finite exciton population, i.e. excitonic polaritons exist in the crystal only during the pulse duration. This allows us to detect the interaction of the biexcitonic waves when the light has already left the crystal, avoiding the direct interference of the incident light pulses, which creates ‘pump’ and ‘test’ biexcitonic waves. In contrast, coherent biexcitons, which are created by the resonant two-photon absorption process and have a dephasing time of several tens of picoseconds, survive even when the incident light pulses have left the crystal.

In the biexciton FWM process, two counter-propagated ‘pump’ biexciton waves are prepared by a pair of intense light pulses with frequency $\omega_0 = \omega_{\mathrm{b}}/2$ and wavevector $k_0$

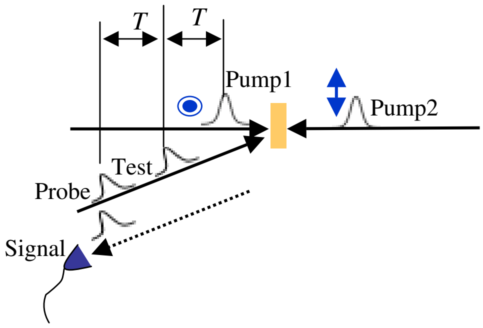

**Figure 2.** A sketch of the proposed experiment. Two orthogonally polarized pump pulses create counter-propagated biexciton waves. Another biexciton wave created by the test pulse, which is delayed on time $T$ with respect to the pump pulses in order to avoid the optical interference between the pump and test pulses. The scattering of the 'pump' and 'test' biexciton waves on the Kerr-type biexciton Hamiltonian results in the creation of the FWM biexciton wave, which propagates in the opposite direction with respect to the test wave. The probe light pulse, which is co-propagated with the test one, is scattered on created biexciton wave creating the signal pulse.

propagating along the $z$ and $-z$ directions. In order to avoid the direct interference of the light waves in the crystal, the incident light pulses are cross-linear polarized (see figure 2). The third light pulse of the same frequency propagating at some angle with respect to the first two pulses (e.g. along the $x$ axes) acts as a 'test' biexcitonic wave. This pulse should be delayed with respect to the second pulse to avoid the light interfering in the crystal. The time delay $T$ should be longer than the pulse duration $\tau_p$ and propagating time $\tau_L = L/c$. On the other hand, $T$ should be much shorter than the biexciton dephasing time $\tau_b$.

The 'test' biexciton wave, which is created by the third pulse, interacts with the counter-propagated 'pump' biexcitonic waves and produces the 'signal' biexcitons with a wavevector along the $-x$ direction. When the time delay between the pump and test pulses is short enough to neglect the biexciton dephasing, the amplitude of the 'FWM' biexciton wave emerging in the crystal with length $L$ is

$$\langle b_{\mathrm{FWM}} \rangle = -2\mathrm{i}\pi\sqrt{n_1 n_2}a^3 W \tau_L \langle b_{\mathrm{test}} \rangle \mathrm{e}^{-\mathrm{i}(\phi_1+\phi_2)}, \tag{5}$$

where $n_{1,2}$ and $\phi_{1,2}$ are the biexciton densities and phases of the counter-propagated 'pump' biexciton waves, respectively. In the simplest model of no-momentum-transfer interaction between biexcitons, the phase conjugate reflectivity $r = |\langle b_{\mathrm{FWM}} \rangle / \langle b_{\mathrm{test}}^{\dagger} \rangle|$ at $n_1 = n_2 = 10^{16}$ cm$^{-3}$ can be estimated by the ratio $L/c\tau_b$, which is of the order of $10^{-3}$ in the CuCl crystal with a length of several tens of micrometres.

The generated biexciton wave with momentum $-\mathbf{k}_{\mathrm{test}}$ can be detected by measuring the intensity of the diffracted probe light pulse (see figure 2). The intensity of the diffracted signal pulse is given by

$$I_{\mathrm{diff}}/I_{\mathrm{probe}} = \left(\frac{8\pi^2 \mu_{\mathrm{ex}} \mu_{\mathrm{bi}} L}{E_b \lambda n}\right)^2 \frac{n_{\mathrm{FWM}}}{v_{\mathrm{cell}}} \sim W^2 n_1 n_2 n_{\mathrm{test}}, \tag{6}$$

where $n_{\mathrm{test,FWM}} = \langle b_{\mathrm{test,FWM}} \rangle^2 / V$ is the biexciton density in the 'test' and 'FWM' waves, $v_{\mathrm{cell}}$ is the unit cell volume, $\mu_{\mathrm{ex}}$ and $\mu_{\mathrm{bi}}$ are the dipole moments of the exciton and biexciton, respectively, and $\lambda$ is the signal pulse wavelength. One can observe from equation (6) that the diffraction efficiency is governed by the biexciton anharmonicity parameter $W$, which can be measured directly in the proposed experiment. Assuming $n_{\mathrm{FWM}} = 10^{10}$ cm$^{-3}$, which corresponds to the phase conjugate biexciton reflectivity $r = 10^{-3}$, the diffraction efficiency for the 30 $\mu$m single CuCl crystal can be estimated from (6) as $I_{\mathrm{diff}}/I_{\mathrm{probe}} \approx 10^{-4}$ for the following parameters $\mu_{\mathrm{ex}} = 1\ e$Å, $\mu_{\mathrm{bi}} = 10\ e$Å and $\upsilon_{\mathrm{cell}} = (5.41\ \text{Å})^3/4$ [19]. The obtained diffraction efficiency indicates that the biexciton anharmonicity in CuCl can be found experimentally at relatively low excitation densities.

## 4. Conclusions

We show that coherent biexcitons, which can be created in CuCl by ultrashort laser pulses and survive for several tens of femtoseconds, have strong potential for storing information carried by photons. The intrinsic anharmonicity of biexcitons in CuCl offers an interesting opportunity to demonstrate nonlinearity in coherent bosonic waves. We propose an experimental scheme, which is based on the measurement of the phase conjugate reflectivity of biexciton waves, to detect this nonlinearity and to perform a quantitative study of the biexciton–biexciton interaction.

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