Optical phase conjugation based on the giant two-photon resonance of the excitonic molecule in CuCl

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Optical phase conjugation resulting from the two-photon coherence due to the excitonic molecule resonance in CuCl has been observed and studied as a function of frequency, polarization, and pump intensity. Reconstruction of the polarization of an aberrated probe beam is demonstrated.

Degenerate four-wave mixing (DFWM) has been employed for several years as a simple and versatile method for generating phase conjugated, reflected optical beams.[1] The properties of these time-reversed waves, which compensate for wave-front distortions along the path of the input beam, suggest many possible applications.[2] In most of the media that have been utilized for phase conjugation, the nonlinear susceptibility required for DFWM results from either nonresonant, "Kerr-type," electronic and molecular-orientational nonlinearities or one-photon resonant absorption.

The purpose of this Rapid Communication is to present measurements of phase conjugation from DFWM based on the large nonlinear susceptibility resulting from the two-photon absorption (TPA) resonance of the excitonic molecule state in CuCl. The magnitude of this nonlinearity is such that conjugate reflectivities of 10–20% can be obtained at moderate power densities for very small interaction lengths, on the order of one micron. The experimental approach offers the possibility of studying the behavior of the two-photon coherence in analogy with recent experiments in gases.[3,4]

The excitonic molecule in CuCl is a bound state of two excitons which is responsible for a two-photon absorption resonance at a photon energy of 3.186 eV at $T \sim 4.2$ K. The large TPA cross section of this resonance results from the proximity of the Z_3 exciton at 3.202 eV, an intermediate state with a large electric dipole oscillator strength, and from other factors that enhance the electric dipole matrix element coupling this intermediate state to the excitonic molecule state.[5] It is well known[6] that two-photon absorption processes contribute to the third-order optical susceptibility $\tilde{\chi}^{(3)}$, which is the term responsible for DFWM in most media. Our measurements show that the value of $\tilde{\chi}^{(3)}$ at the center of the excitonic molecule resonance in CuCl is a factor of $\sim 10^6$ larger than that of other materials without linear absorption, such as the Kerr-type media typified by CS_2.

A schematic diagram of the experimental setup for optical phase conjugation using DFWM is shown in Fig. 1. Two counterpropagating pump beams with wave vectors $\vec{k}_2$ and $\vec{k}_3 = -\vec{k}_2$ are superposed at the sample S with a probe beam $\vec{k}_4$ which propagates at an angle α to the pump $\vec{k}_3$. The phase conjugated wave has a wave vector $\vec{k}_1 = -\vec{k}_4$ and is a time-reversed replica of the probe.[1] In this process one photon is scattered from each of the pump beams, and a conjugate wave photon and an additional probe photon are emitted. Energy and wave vector are conserved for all values of α.

The distinctive features of two-photon DFWM may be described for a cubic material such as CuCl by writing the relevant terms for the nonlinear polarization $p^{(3)}$ in the form[4]

$$\vec{p}^{(3)} = A(\vec{E}_2 \cdot \vec{E}_4^*)\vec{E}_3 + B(\vec{E}_3 \cdot \vec{E}_4^*)\vec{E}_2 + C(\vec{E}_2 \cdot \vec{E}_3)\vec{E}_4^* , \quad (1)$$

where the electric field subscripts follow the notation of Fig. 1. The first two terms involve the formation of spatial gratings in the material due to the interference between the probe field $\vec{E}_4$ and either of the two pumps. On the other hand, the third term does not involve a spatial grating; it results from the coherent two-photon excitation by the pump waves and vanishes when they are orthogonally polarized. In this case the electric field of the conjugate wave is always parallel to the electric field of the probe wave.

The parameter C in Eq. (1) can be related to the contribution to the third-order nonlinear susceptibility[6] $\tilde{\chi}_3(-\omega, \omega, \omega, -\omega)$ from the biexciton resonance. Since the biexciton is a totally symmetric state, there are two equal

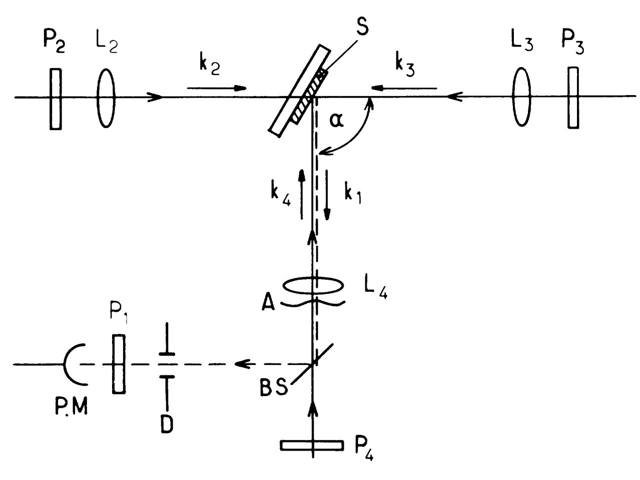

FIG. 1. Schematic diagram of DFWM setup for optical phase conjugation. S = sample, L = lens, P = polarizer, A = aberrator, D = variable aperture, BS = beamsplitter. For the data shown in this paper, $\alpha = 3°$ and the pump beams are normal to the sample surface.

components $\chi^{(3)}_{xxxx}=\chi^{(3)}_{xyyx}$ of the form[6]

$$\chi^{(3)}_{xxxx}=D/(\omega_B-2\omega-i\Gamma_B)(\omega_{ex}-\omega-i\Gamma_{ex})^2 \ , \qquad (2)$$

where A involves some electric dipole matrix elements and nonresonant energy denominators, $\hbar\omega_B$ and $\hbar\omega_{ex}$ are the biexciton and exciton energies, and Γ_B and Γ_{ex} are their damping rates. A comparison of the defining relation $\vec{P}^{(3)}=\tilde{\chi}^{(3)}:\vec{E}\vec{E}\vec{E}$ with Eq. (1) gives $C=6\chi^{(3)}_{xxxx}$.

A measurable quantity in our experiments is the phase conjugate reflectivity which is defined as $R_c=I_1/I_4$. For the simplest geometries, where $\vec{E}_2$ and $\vec{E}_3$ are collinear and either parallel or perpendicular to the probe field, this is given by[7]

$$R_c=\tan^2(2\pi\omega CE_2E_3t/cn) \ , \qquad (3)$$

where t is the interaction length and n is the refractive index.

Measurements were made at 1.8 K on evaporated and annealed thin-film samples with $t\sim 1$ μm and on single-crystal platelets with $t\sim 30$ μm. The exterior angle α in Fig. 1 was either 3° or 90°. A dye laser pumped by a N_2 gas laser supplied 5-nsec-long pulses with an energy of 20 μJ and a linewidth of about 0.1 Å. This was split into the two pump beams with equal intensities and a probe beam with about 5% of the intensity of the pump beams. Absolute measurements of the conjugate reflectivity were made by comparing the signal with that obtained when an ordinary mirror at the position of the sample was used to retroreflect the probe beam. Corrections were made for losses in cryostat windows and the Fresnel coefficients of the film.

Only the results obtained at 1.8 K on a sample of thickness 1.25 μm at pump intensities at and below 1 MW/cm^2 will be presented here. The results obtained with much thicker single crystals or at higher power densities display pronounced effects due to saturation, asymmetric broadening of the biexciton resonance, and distortions caused by linear and two-photon absorption.

The conjugate reflectivity as a function of laser wavelength is shown in Fig. 2 for various combinations of the polarizations. In the nearly collinear geometry the angle $\alpha/n\sim 1°$ between the pump and probe fields when they are polarized in the plane of Fig. 1 is ignored, and they are considered to be polarized along a common y direction. The transmission of one of the pump beams is also shown for comparison. The oscillations in the transmission and in the broad conjugate reflectivity components are caused by Fabry-Perot interferences in the sample, from which a thickness of 1.25 μm has been deduced.

There are two components of different origins in the data of Fig. 2, a sharp line at the TPA wavelength and a broad signal at photon energies just below the exciton absorption at 3870 Å. The polarization behavior of the latter feature shows that it results from contributions to the first two terms of Eq. (1), which are caused by one-photon absorption in the low-energy tail of the exciton absorption, rather than from the pole at ω_{ex} in the TPA contribution of Eq. (2). This grating-induced signal has a magnitude which depends on the grating period, a behavior which may be caused by exciton diffusion.[8]

The sharp line at the position of the biexciton TPA is the signal of interest here. It appears only in the *xxxx* or *xyyx* geometries. The peak values of the reflectivity are equal in

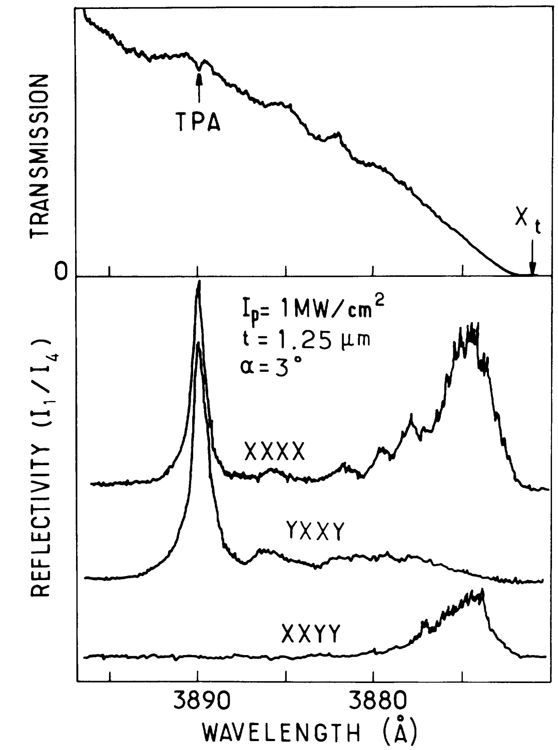

FIG. 2. Laser wavelength dependence of optical-phase-conjugation signal for a 1.25-μm-thick sample at $T=1.8$ K and $I_p=1$ MW/cm^2. The four polarization components are in the order phase-conjugate wave (1), pump (2), pump (3), probe (4). The x direction is orthogonal to the scattering plane and y is in this plane. The upper trace shows the transmission of one pump beam. X_T corresponds to the wavelength of the transverse exciton.

these two cases to within the limits of experimental reproducibility. This result demonstrates that the two-photon coherence, represented by the third term in Eq. (1), is responsible for the phase conjugation. It should be noted that two-photon induced population gratings could also contribute to the phase conjugation in the *xxxx* geometry, where the TPA is allowed for simultaneous absorption of one pump and one probe photon. The equal values of R_c for *xxxx* and *xyyx* show that these population effects are negligible at the pump intensities employed.

The conjugate reflectivity near the TPA wavelength is shown at several pump intensities in the inset of Fig. 3. Below $\sim 10^5$ W/cm^2 the TPA has a constant width $\Gamma\sim 0.2$ meV, presumably due to inhomogeneous broadening. At these lower intensities the reflectivity increases as I_p^2 (the product of the pump beam intensities), as expected from Eq. (3) in the limit $R_c \ll 1$. It has been shown that, for intensities between 10^5 W/cm^2 and 10^6 W/cm^2, the biexciton TPA broadens symmetrically due to collisions, with a width proportional to $I_p^{1/2}$.[9] We find that the linewidth of the phase-conjugate reflectivity signal increases in a similar manner. This broadening is accompanied by a change in the dependence of R_c on I_p.

At pump intensities below 10^5 W/cm^2, where $(R_c)_{\max}\propto I_p^2$, the value of $\chi^{(3)}_{xxxx}$ is constant and has its maximum value. An estimate of $\chi^{(3)}_{xxxx}$ can be made using Eq. (3). Including corrections for the Gaussian beam profiles

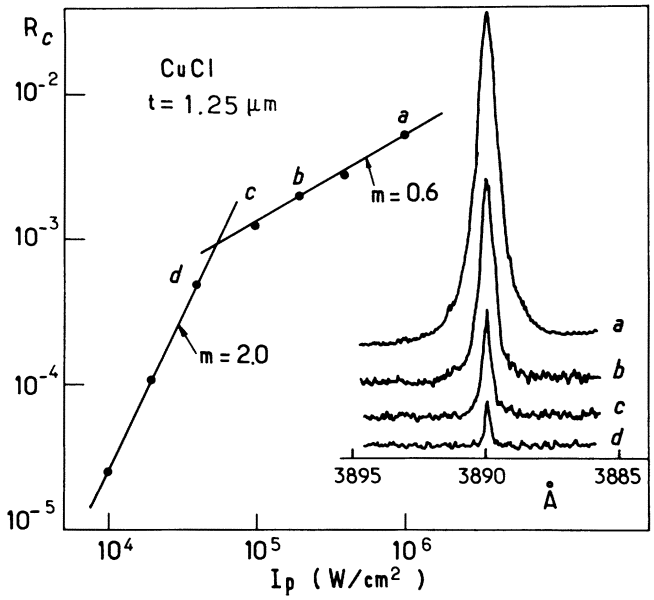

FIG. 3. Peak value of the phase-conjugate reflectivity $(R_c)_{\max}$ as a function of pump beam intensity. The lines correspond to $(R_c)_{\max} \propto I_p^m$. The inset shows the spectral dependence of R_c near the TPA wavelength. The labeled spectra correspond to the identically labeled points in the plot of $(R_c)_{\max}$.

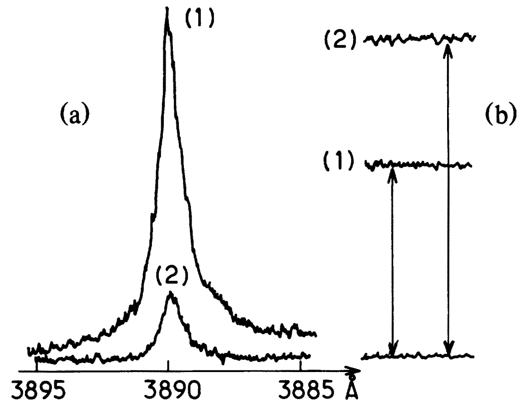

FIG. 4. (a) Spectra of the phase-conjugate reflected beam when an aberrator is inserted at A in Fig. 1; (b) magnitude of the signal observed when the probe beam is retroreflected by a plane mirror interchanged with the sample. In the traces labeled (1) or (2) the measured polarization is parallel or perpendicular, respectively, to that of the original probe beam before its passage through the aberrator. $\alpha=3°$.

the result is

$$\chi^{(3)}_{xxxx}(\omega_B/2)=3\times10^{-7}\text{ esu} .$$

This can be compared with an estimate based on the measured two-photon absorption coefficient, which is proportional to $\text{Im}\chi^{(3)}_{xxxx}$.[10] At a pump intensity I_p-10^5 W/cm^2, 2% of the light is absorbed at the peak of the TPA in a 1-μm-thick sample. This yields an estimated $\chi^{(3)}_{xxxx}\sim6\times10^{-7}$ esu, in reasonable agreement with the value deduced from $(R_c)_{\max}$. These values of $\chi^{(3)}_{xxxx}$ are more than a factor of 10^2 larger than previous estimates.[11]

The phase-conjugate nature of the reflected wave has been verified by inserting a phase and polarization aberrator (marked A in Fig. 1) into the probe beam. The conjugate beam was well collimated after retraversing the aberrator.[12] The polarization of this reconstructed wave is illustrated in Fig. 4 where the conjugate wave signals obtained with the polarized P_1 set parallel and perpendicular to the probe beam polarizer P_4 are compared with the polarization of the beam retroreflected from an ordinary mirror. The original probe polarization is nearly reconstructed in the conjugate beam, whereas the reflection from the mirror actually has a predominant polarization perpendicular to that of the test beam. This property of polarization reconstruction holds for arbitrary pump and probe polarizations provided that the Fresnel coefficients of the sample surface do not alter the polarizations of the probe or conjugate waves. In other media, circularly polarized pump waves and very small values of α are required in order to ensure reconstruction of the polarization.[13].

Additional experiments have been performed in thicker, single-crystal samples, at temperatures up to 60 K and at higher pump intensities up to 20 MW/cm^2. Phase conjugate relfectivities of about 20% have been obtained in a ~30 μm sample. A more complete discussion of these results will be published elsewhere.[8]

Note added in proof. After submission of this paper, we became aware of an independent report of optical phase conjugation in CuCl [G. Mizutani and N. Nagasawa, J. Phys. Soc. Jpn. 52, 2251 (1983)]. Their results are obtained on a 26-μm-thick sample and exhibit the distortions referred to in our text.

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[12]Note that the sample thickness $t=1.25$ μm is still one order of magnitude larger than the light wavelength inside the medium (refractive index $n>3$). Also, the focal diameter of the beams is even much larger, thus allowing for a good reconstruction of the beam.

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