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Optical phase conjugation via degenerate four-wave mixing in copper chloride

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We present a study of optical phase conjugation through degenerate four-wave mixing in CuCl, as a function of light frequency, intensity, polarization and sample temperature.

The phase-conjugate signal exhibits two maxima at the frequencies of the Γ_5 exciton (one-photon resonance) and of the biexciton (two-photon resonance). By appropriate combinations of the input beam polarizations, it is possible to single out the various terms contributing to the signal (small-spaced, large-spaced population grating and two-photon coherent excitation).

The absolute value of $\chi^{(3)}$ responsible for the two-photon resonance has been determined ($\chi^{(3)}(-\omega, \omega, \omega, -\omega) = 3.10^{-7}$ e.s.u.‡). It leads to 'mirror' efficiencies of the order of 10 % for input pump intensities $I_0 \approx 10^5$ W cm^{-2} and sample thickness $d \approx 10^{-4}$ cm. At higher I_0, a saturation of the reflection takes place. The variation of the conjugate beam with temperature has been studied in the range 15 K < T < 70 K. Finally, the phase-conjugate nature of the signal has been verified by inserting an aberrator in the path of the input probe beam, and by checking the reconstruction of the reflected beam.

The mechanism of conjugate wavefront generation has stirred a considerable interest, in view of its possible applications in optical imaging, owing to the fact that the generated wave has the property of time reversal with respect to the incident wave (Fisher 1983). This allows, in particular, the reconstruction of aberrated wavefronts. A practical method of creating a conjugate mirror is via degenerate four-wave mixing in a nonlinear medium. A current thrust of research is to discover materials with high efficiency. This can be achieved by making use of the resonant enhancement of the third-order nonlinear susceptibility $\chi^{(3)}(-\omega, \omega, \omega, -\omega)$ describing the four-wave mixing process. Among the resonant terms, those based on a two-photon coherence (t.p.c.) are particularly attractive, since they have potentially very fast response times.

In a recent letter (Chase *et al.* 1983), we have reported an experimental study of phase conjugation in CuCl. A t.p.c. signal has been observed in samples as thin as 1 μm, with efficiencies exceeding 5 %. In this paper, we present additional results concerning the polarization and temperature variation of the signal. The beams configuration is shown in the inset of figure 1. The two counter-propagating pump beams 2 and 3, of equal intensity I_0 (maximum intensity 10^7 W cm^{-2}), as well as the probe beam 4, of intensity $5 \times I_0/100$, are derived from the same nitrogen-pumped dye laser (pulse duration, 5 ns). Propagation directions of the beams are shown in figure 1, where the external angle α is either 3° or 90° with respect to pump beam 2. The generated beam 1 (the signal beam) is directed, through

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‡ 1 e.s.u. = 1 cm^3 erg^{-1} ≡ 1.4 × 10^{-8} m^2 V^{-2}.

a small aperture, to a photomultiplier tube. To discriminate the signal from scattered pump light, mechanical chopping of the probe beam is used together with phase-sensitive detection.

Figure 1 shows the spectral dependence of the signal intensity, recorded in the vicinity of the exciton region of CuCl, for various configurations of polarization of the beams. In case $V_1V_2V_3V_4$ (where V_i((H_i) stands for vertical (horizontal) polarization direction of beam i) two

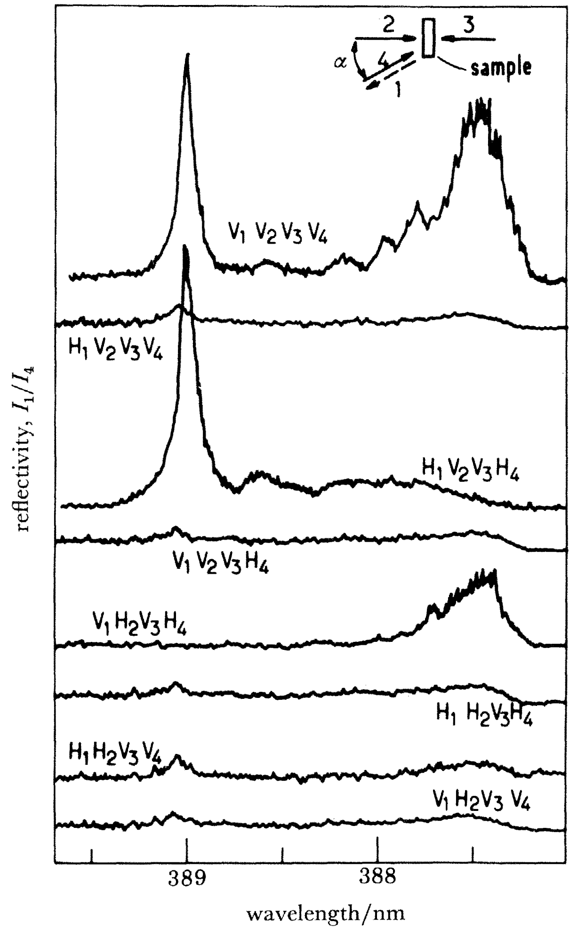

FIGURE 1. Wavelength dependence of the phase-conjugate signal at $T = 2$ K, for various polarization configurations of the optical beams. $V_i(H_i)$ stands for vertical (horizontal) polarization of beam i; $I_p = 1$ MW cm^{-2}, $t = 1.25$ μm, $\alpha = 3°$.

maxima are apparent. From their position and polarization characteristics, they may be readily interpreted. The peak at $\lambda \approx 387.5$ nm falls near the $n = 1$ exciton absorption line, formed with a hole of the upper Γ_7 valence band and an electron of the lowest Γ_6 conduction band. This peak disappears if the probe polarization is orthogonal to both pumps. Such behaviour is expected from a one-photon resonance term, resulting in the formation of a real exciton population volume grating. Of more direct concern to us is the peak located at 389 nm, the position of which corresponds exactly to half the biexciton energy $\hbar\omega_B$. As is well known (Hanamura 1973; Gale *et al.* 1974), the biexciton state leads to a giant two-photon transition cross-section, and therefore should also be effective in t.p.c. phase conjugation. Given the biexciton symmetry Γ_1, one can show that the signal from the biexciton two-photon resonance should be proportional to the scalar product of the fields $\boldsymbol{E}_2 \cdot \boldsymbol{E}_3$, independent of the orientation of $\boldsymbol{E}_4$. Furthermore, $\boldsymbol{E}_1$ should be parallel to $\boldsymbol{E}_4$. These predictions are well obeyed and give support to the assignment of the peak.

In figure 2, we have plotted the intensity dependence of the signal as a function of the lattice temperature. The data were recorded on a 1 μm thin sample, with a pump intensity

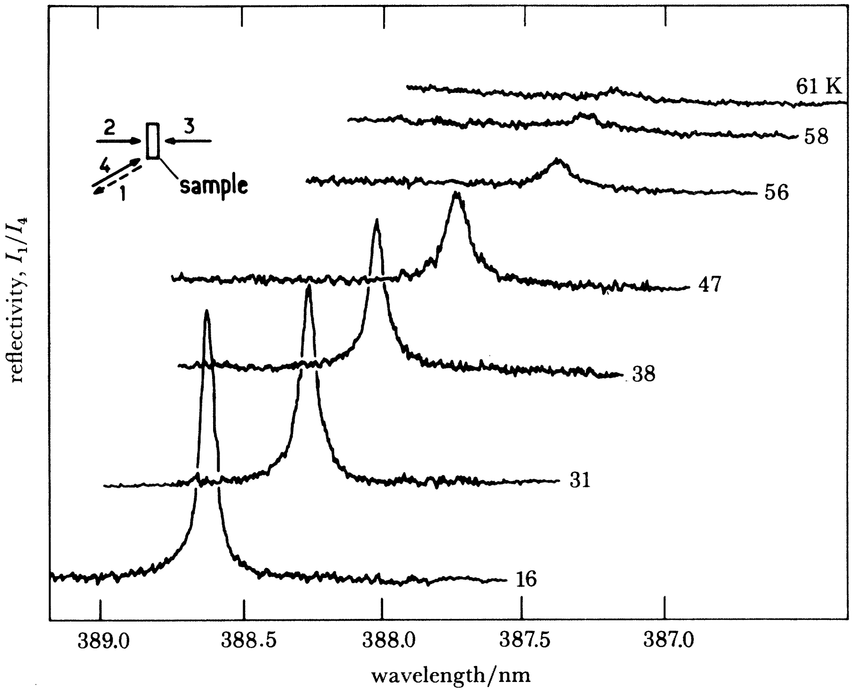

FIGURE 2. Temperature dependence of the t.p.c. conjugate signal in a 1 μm thick CuCl sample for $H_1 V_2 V_3 H_4$.

$I_0 \leqslant 10^6$ W cm^{-2}. As can be seen, the signal is clearly observed up to temperatures of the order of 60 K, with a noticeable broadening above $T = 40$ K. A similar behaviour in function of temperature has been reported and discussed by Itoh *et al.* (1980) for the biexciton two-photon absorption line of CuCl.

Finally, as mentioned by Chase *et al.* (1983), we have verified the phase-conjugate nature of the reflected wave by inserting an aberrator in the path of the probe. Also, we have determined the absolute value of $\chi^{(3)}(-\omega, \omega = \frac{1}{2}\omega_B, \omega, -\omega) = 3 \times 10^{-7}$ e.s.u. This very large nonlinearity suggests the possibility of achieving mirror efficiencies exceeding 100% with samples of thickness 10^{-3} cm and pump beam intensities $I_0 \approx 10^6$ W cm^{-2}. So far, we have obtained efficiencies of the order of 20% without taking any special precaution to ensure good quality and spatial overlap of the optical beams.

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