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COHERENT LIGHT AMPLIFICATION AND OPTICAL PHASE CONJUGATION WITH PHOTOREFRACTIVE MATERIALS

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Résumé La dépendance entre le champ électrique et l'écartement des franges du transfert d'énergie stationnaire entre deux faisceaux traçants lors de la formation en volume d'un hologramme de phase avec des matériaux photoconducteurs électro-optiques ($\text{KNbO}_3:\text{Fe}^{2+}$, $\text{Bi}_{12}(\text{Si,Ge})\text{O}_{20}$,...) a été étudiée. Les résultats expérimentaux sont comparés avec les expressions théoriques décrivant l'influence de différents champs de charge-espace photoinduits dans des domaines photoréactifs. Un pic d'accroissement exponentiel $\Gamma = 15 \text{ cm}^{-1}$ a été atteint dans le cas de $\text{KNbO}_3:\text{Fe}^{2+}$ avec un choix approprié de paramètres expérimentaux (tels que l'enregistrement de la longueur d'onde, l'espacement des franges, champ électrique appliqué etc...). Les résultats de la dépendance du champ électrique à la réflectivité du front d'onde ρ dans des expériences de mélange de quatre ondes dégénérées sont rapportés et discutés suivant une analyse théorique similaire à celle des expériences de mélange de deux ondes. Il est montré que des pics de réflectivité avec $\rho = 25\%$ ont été atteints pour $\text{KNbO}_3:\text{Fe}^{2+}$, que des fronts d'onde complexes conjugués peuvent être générés et que des fronts d'onde déformés peuvent être effectivement corrigés par des expériences de mélange de quatre ondes dans $\text{KNbO}_3:\text{Fe}^{2+}$.

Abstract The electric field and fringe spacing dependence of the stationary energy transfer between two writing beams in volume phase-hologram formation with photoconductive electro-optic materials ($\text{KNbO}_3:\text{Fe}^{2+}$, $\text{Bi}_{12}(\text{Si,Ge})\text{O}_{20}$,...) has been studied. Experimental results are compared with the theoretical expressions describing the influence of the different photoinduced space-charge fields in photorefractive media. A peak exponential gain of $\Gamma = 15 \text{ cm}^{-1}$ has been reached in $\text{KNbO}_3:\text{Fe}^{2+}$ for the proper choice of experimental parameters such as recording wavelength, fringe spacing, applied electric field etc.). Results of the electric field dependence of the wavefront reflectivity ρ in degenerate four-wave mixing experiments are reported and discussed in terms of a similar theoretical treatment as the two-wave mixing experiments. It is shown, that peak reflectivities of $\rho = 25$ percent have been reached for $\text{KNbO}_3:\text{Fe}^{2+}$, that conjugate complex wavefronts can be generated and that distorted optical wave fronts can be effectively corrected by four-wave mixing experiments in $\text{KNbO}_3:\text{Fe}^{2+}$.

1. INTRODUCTION

Photoinduced refractive index changes in electro-optic materials can be used for recording phase holograms in the volume of the photorefractive crystals¹. The recording of such thick volume holograms permits the interference of an incident light beam with its own diffracted beam inside the recording material. This effect causes the continuous recording of a new grating that may add to or subtract from the initial grating that is not uniform through the thickness of the material, and which can be phase shifted to the initial grating². The energy redistribution between writing beams primarily depends on the phase shift between the fringe pattern and the recorded grating. Stationary energy transfer has been shown to be forbidden if there is a phase shift of zero or π between the interference pattern and the recorded hologram, due to

destructive interference caused by the $\pi/2$ phase shift of the Bragg diffracted beam with respect to the recording beam travelling in the same direction. A maximum energy transfer is therefore obtained, if there is an additional phase shift of $\pi/2$ between the undiffracted signal beam and the diffracted reference beam. This is the case for a stationary grating phase shift of $\pi/2$ with respect to the intensity distribution². Such a grating phase shift can be achieved, if diffusion is the dominant recording mechanism or if the photoconductive transport length L_E is comparable or larger than the fringe spacing Λ (violation of quasineutrality)³. The energy transfer between interacting beams within the photorefractive material can be used for the amplification of coherent light beams, including amplification of beams carrying optical information and for other applications in optical information processing. The first part of this paper deals with the light amplification in materials where quasineutrality can be violated at moderate applied electric field strength. Several interesting applications of nonlinear optical materials for the restoration of phase distorted images (adaptive optics) have been proposed recently⁴. It will be shown in this paper, that a series of these applications can be performed better by degenerate four-wave mixing (FWM) with photorefractive materials as the nonlinear element. Whereas the optical nonlinearity in nonlinear optical materials is due to the "instantaneous" electronic response, in electro-optic materials it is due to both the electronic and lattice polarizability due to photoinduced space-charge fields, the latter being non-instantaneous. The advantage of the electro-optic technique is that it need much less optical power than in materials with electronic response only and thus it can be used efficiently with continuous lasers too.

For the experiments described in this paper we used a reduced $\text{KNbO}_3:\text{Fe}$ crystal. The optimum crystal orientation as well as other relevant materials parameters and procedures for reducing the Fe-doped crystals have been described in a series of earlier papers^{3,9,10}.

Reduced KNbO_3 as a ferroelectric material with photoconductivity comparable with or larger than that in $\text{Bi}_{12}\text{SiO}_{20}$ and $\text{Bi}_{12}\text{GeO}_{20}$ ^{9,10} offers many advantages compared with the other more commonly used photoconductors $\text{Bi}_{12}\text{SiO}_{20}$ and $\text{Bi}_{12}\text{GeO}_{20}$. First, the rather large value of the spontaneous polarization gives large electro-optical effects leading to large steady-state photoinduced refractive-index changes. Second, the photoconductivity is high enough that even at wavelengths near 600 nm these steady-state refractive index changes are reached in less than a second at moderate optical power densities, i.e. 2 orders of magnitude less than in $\text{Bi}_{12}\text{GeO}_{20}$. Whereas beam coupling in $\text{Bi}_{12}\text{GeO}_{20}$ and $\text{Bi}_{12}\text{SiO}_{20}$ is substantially reduced because of the large optical activity of these materials, the optical polarization remains the same along the light path for polarization directions parallel to the crystallographic directions.

Photorefractive recording is much more efficient in $\text{KNbO}_3:\text{Fe}^{2+}$ than in BaTiO_3 , another material with large electro-optic effects, with typical response time T of the order of 10 msec at recording wavelengths of $\lambda=488$ nm and T of the order of 100 msec at $\lambda=592$ nm in $\text{KNbO}_3:\text{Fe}^{2+}$ for $I=1\text{W}/\text{cm}^2$. These values compare favorably with $T=1$ sec measured in BaTiO_3 ¹¹ for $\lambda=514$ nm.

2. THE PHOTOREFRACTIVE EFFECT IN PHOTOCONDUCTIVE MATERIALS

The light induced changes of refractive indices in electro-optic crystals are based on the spatial modulation of photocurrents by non-uniform illumination. The electrons or holes which are excited from the impurity centers by light of suitable wavelength, are upon migration, retrapped at other locations leaving behind positive or negative charges of ionized trap centers. The resulting space-charge field between the ionized donor centres and the trapped charges modulates the refractive indices via the electro-optic effect.

In photoconductive materials the photocurrent along one dimension (z) with electrons as carriers is given by:

$$(1) \quad j(z,t) = \frac{e\phi\alpha}{h\nu} I(z,t) \cdot L_d - \frac{\mu r \phi \alpha k T}{h\nu} \frac{dI(x,t)}{dz},$$

where $L_d = \mu\tau \cdot E_0$ is the drift length, e the electronic charge, ϕ the quantum efficiency for exciting an electron, α the absorption constant, $h\nu$ the photon energy, μ the electron mobility, τ the lifetime of photoexcited electrons, $I(z,t)$ the light intensity distribution and E_0 the applied electric field.

Elementary refractive index gratings recorded by a setup shown in Fig. 1 using the photorefractive effect are in phase with the interference pattern produced by the recording beams if drift is the recording mechanism. For recording by diffusion the photocurrent depends on the first spatial derivative of the intensity distribution and thus there is a $\pi/2$ -phase shift between the recorded refractive index grating and the intensity pattern. It has been shown, that $\pi/2$ phase shifted gratings are obtained also in photoconductive recording, if the drift length L_d becomes comparable to the grating spacing Λ (violation of quasineutrality).

Electro-optic materials with sufficient photoconductivity to allow "violation of quasineutrality" at moderate electric field strengths are listed in Table I together with photoconductivity data and the electro-optic coefficients describing the electro-optic activity of the materials.

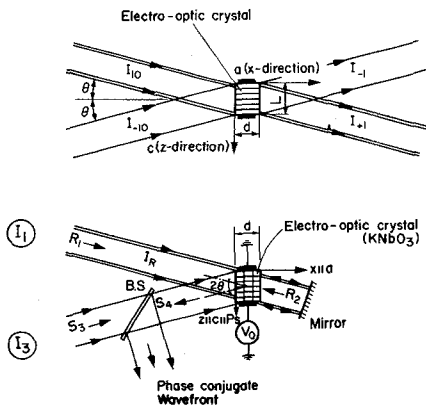


FIGURE 1: Experimental setup for (a) coherent light amplification by two-wave mixing and (b) optical phase conjugation by degenerate four-wave mixing (B.S.: beam splitter)

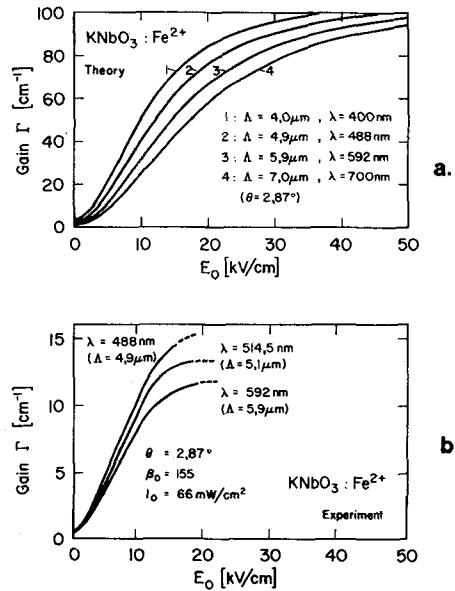


FIGURE 2: Electric field dependence of the gain Γ for different fringe spacings for $\text{KNbO}_3:\text{Fe}^{2+}$. a: theoretical; b: experimental (smoothed curves with 5 % relative errors)

Phase shifted volume holograms recorded in these materials permit an efficient dynamic energy redistribution between two or more recording beams and can thus be used for coherent light amplification of weak beams ² or complex conjugate (time reversed) wave generation. ⁵

3. COHERENT LIGHT AMPLIFICATION

Self-diffraction of recording beams in the volume of a dynamic material with nonlocal response can lead to an efficient optical energy transfer. The intensities of two beams emerging a photorefractive material of thickness d is given by ²:

TABLE 1: Photoconductive electro-optic materials

Material	Photoconductivity $I_d/E \left[\frac{\mu\text{m}}{\text{kV/cm}} \right]$	Electro-optic coefficient $r_{ij} \left[\frac{\text{pm}}{\text{V}} \right]$
$\text{Bi}_{12}\text{SiO}_{20}$	1	5
$\text{Bi}_{12}\text{GeO}_{20}$	0.8	3.4
$\text{KNbO}_3:\text{Fe}^{2+}$	0.3	64
$\text{K}(\text{NbTa})\text{O}_3 (T_c=40^\circ\text{C})$	0.01	~ 500
$\text{Ba}_{.25}\text{Sr}_{.75}\text{Nb}_2\text{O}_6$	0.8	45

$$(2) \quad I_{-1} = \frac{I_o \beta_o}{1 + \beta_o e^{\Gamma d}} \exp(\Gamma - \alpha) d \approx I_{-10} \cdot \exp(\Gamma - \alpha) d \quad (\beta_o \ll 1)$$

$$(3) \quad I_{+1} = \frac{I_o}{1 + \beta_o e^{\Gamma d}} \exp(-\alpha d),$$

where $I_o = I_{-10} + I_{+10}$ is the total intensity incident to the crystal, $\beta_o = \frac{I_{-10}}{I_{+10}}$ and Γ the exponential gain characterizing the energy transfer. The gain Γ is determined by the $\pi/2$ -shifted component of the refractive index grating with amplitude Δn_1 :

$$(4) \quad \Gamma(x) = \frac{4\pi \beta_o(x) \Delta n_1(x)}{\lambda \cos \theta} \sin \phi_g(x)$$

(λ = recording wavelength, 2θ = angle between recording beams, ϕ_g = grating phase shift).

A general expression for the gain Γ which describes the relative influence of the different charge transport processes has been derived by Kutchtarev and Vinetsky ²:

$$(5) \quad \Gamma = C \cdot E_D \frac{1 + (E_D/E_q) + (E_o^2/E_D E_q)}{[1 + (E_D/E_q)]^2 + (E_o^2/E_q)^2},$$

where C is a proportionality constant describing the electro-optic activity of the material, $E_D = \frac{2\pi kT}{\Lambda e} = A/\Lambda$ is the diffusion field and $E_q = \frac{dN_A \Lambda}{2\pi \epsilon \epsilon_o} = B \cdot \Lambda$ the maximum space-charge field limited by the trap concentration N_A and ϵ is the dielectric constant along the direction of the space-charge field.

It can be seen from eq.(2) that effective amplification of a weak beam is obtained if the exponential gain is larger than the absorption constant ($\Gamma > \alpha$). With a detailed knowledge of the photorefractive recording mechanisms it is possible to optimize the grating phase shift with a suitable choice of physical parameters (applied electric field, fringe spacing, recording wavelength etc.) and to control the gain in energy transfer experiments with these parameters (eq.(5)). The experimental results of such measurements are shown in Fig. 2b (electric field dependence) and Fig. 3b (fringe spacing dependence). These curves can be interpreted by (5) using the materials parameters shown in Table 1^{3,9} and $\epsilon = 50$, $N_A = 5.9 \cdot 10^{15} \text{ cm}^{-3}$. Theoretical curves $\Gamma(E_o)$ and $\Gamma(\Lambda)$ obtained by using (5) with these parameters are shown in Figs. 2a and 3a. The fringe spacing dependence enters in eq.(5) through the dependence of E_D and E_q on Λ .

The electric field dependence of the gain Γ shows saturation for $E_o \rightarrow 20 \text{ kV/cm}$, indicating, that indeed the charge transport length ($L_d = 6 \mu\text{m}$ for $E_o = 20 \text{ kV/cm}$) is com-

parable with the fringe spacing ($\Lambda=5 \mu\text{m}$) and that therefore quasineutrality is violated. The saturation of $\Gamma(E_0)$ in the theoretical plots is reached at smaller field strengths for the shorter wavelengths since the fringe spacing is smaller at these wavelengths and a $\pi/2$ -phase shift is reached for smaller drift lengths.

The fringe spacing dependence of the gain Γ shows a peak near fringe spacings satisfying the condition $K_{\text{peak}} \cdot L_d = 1$; i.e. at $\Lambda_{\text{peak}} = 2\pi m E_0$. For $E_0=16 \text{ kV/cm}$ and $\Lambda \approx 7 \mu\text{m}$ a maximum gain of $\Gamma=11.5 \text{ cm}^{-1}$ has been obtained in the experiment. Using the optimized recording parameters a 25-fold amplification of an optical image in quasi

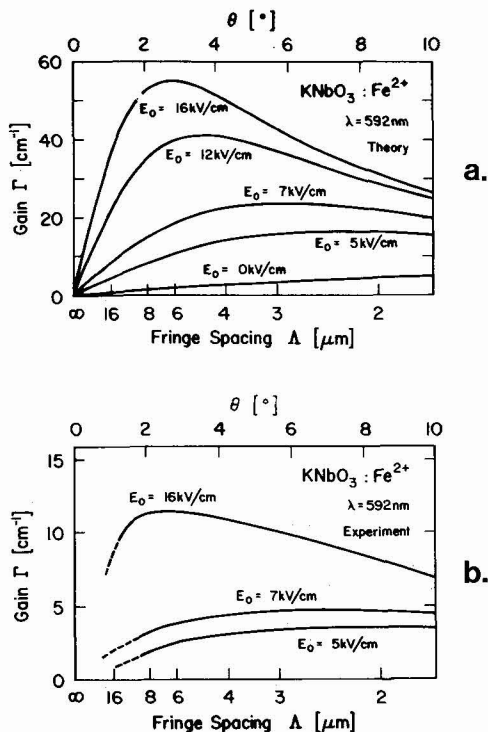


FIGURE 3: Gain Γ as a function of the angle of interacting beams θ for different electric fields for $\text{KNbO}_3:\text{Fe}^{2+}$
 a: theoretical (eq.(5))
 b: experimental (smoothed curves with 5% relative errors).

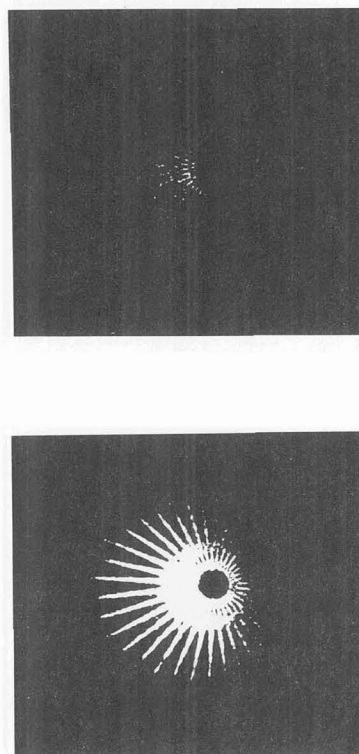


FIGURE 4: a. Original image transmitted through the reduced KNbO_3 crystal
 b. 25x amplified image.

real-time (recording time $\approx 10 \text{ ms}$) has been achieved without significant image quality reduction due to crystal inhomogeneities⁶ (Fig. 4).

The quantitative disagreement between theory and experiment in our opinion is mainly due to the fact that in the experiment Fresnel reflected beams from crystal surfaces also interfere with the recording beams and reduce the fringe contrast, whereas the theoretical treatment is correct only for two-beam interaction. However the good qualitative agreement between theory and experiment indicates that in a first approximation under certain conditions also multiple beam mixing can be described at least qualitatively with an approach similar to the one used above.

4. OPTICAL PHASE CONJUGATION BY DEGENERATE FOUR-WAVE MIXING (FWM)

In FWM two counter-propagating pump waves with complex amplitudes R_1 and R_2 and a weak signal wave S_3 interact in the volume of nonlinear media to produce the fourth wave S_4 , complex conjugate to the signal wave (Fig. 1b). The first observations of phase conjugation by four-wave mixing in photorefractive materials were reported for $\text{Bi}_{12}\text{SiO}_{20}$ in Ref. 5 and for LiNbO_3 and LiTaO_3 in Ref. 7. Recent experiments with BaTiO_3 ⁸ show, that in materials with large electro-optic coefficients CW phase conjugate wave generation with simultaneous amplification can be achieved.

Using the holographic approach to four-wave mixing proposed by Yariv⁴ and developed in⁵, the appearance of the fourth wave can be interpreted as a consequence of the diffraction of one of the pump beams on the refractive index grating recorded by the two other beams.

The wavefront reflectivity $\rho = I_4(x=0)/I_3(x=0)$ also depends on the recording process and can be expressed in terms of the fields characterizing the charge transfer processes. In a first approximation (ρ small,...) one gets⁵:

$$(6) \quad \rho \approx R \frac{I_2}{I_1} \frac{E_O^2 + E_D^2}{(1 + E_D/E_q)^2 + E_O^2/E_q^2},$$

where R is a proportionality constant which depends on the electro-optical properties of the material mainly¹².

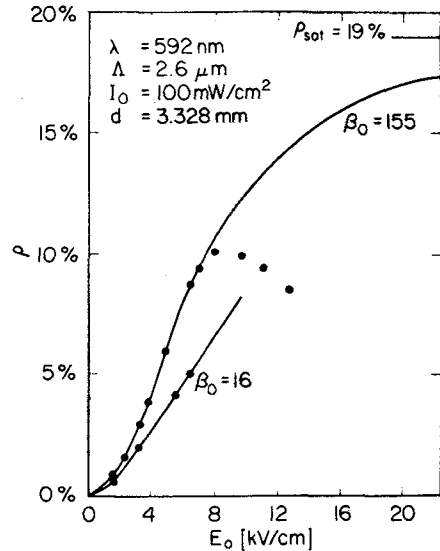


FIGURE 5 : Wave-front reflectivity ρ versus applied electric field for different intensity beam ratios for $\text{KNbO}_3:\text{Fe}^{2+}$.

The electric-field dependence of the wavefront reflectivity for different values of the beam ratio $\beta_0 = I_1/I_3$ is shown in Fig. 5. The other parameters used in these experiments are indicated in Fig. 5 and represent the optimum data determined in two-wave mixing experiments.

In the limit of $E_O < E_q$, a quadratic-field dependence of the wavefront reflectivity has been observed, in agreement with Eq. (6). For very large electric fields $E_O > E_q$, the wave-front reflectivity should saturate since, according to Eq. (6), $\rho \propto E_q^2$ for $E_O \gg E_q$, independently of E_O . The expected field dependence (Eq. (6)) with $E_q = 17$ kV/cm. has been plotted in Fig. 5. A peak reflectivity of 19 % is expected.

For large electric fields, however, the experimental results of Fig. 5 show a saturation of $\rho(E_O)$ for E_O near 8 kV/cm. For larger field strengths ρ shows a decrease that cannot be explained by the theoretical relation (Eq. (6)) since this theory is valid only for weak beam coupling and small photoinduced phase changes, and this assumption seems to be violated at large electric fields. Also, self-interference inside the crystal volume between the phase-conjugate generated wave and retroreflected reference beam R_1 may not be neglectable if wave-front reflectivity becomes larger.

In the experiments described above, I_2 was simply the retroreflected pump wave I_1 , which on being transmitted through the recording crystal was attenuated because of

absorption and reflection losses, yielding $I_2(x=d)/I_1(x=0) \approx 0.6$ only. By increasing the retroreflected pump wave intensity up to $I_2 \approx 200 \text{ mW/cm}^2$ ($I_2(x=d)/I_1(x=0) \approx 2$), the wave-front reflectivity ρ increased up to $\rho = 25\%$ for the parameters given in Fig. 5 and for $E_0 = 8 \text{ kV/cm}$. This value represents one of the largest phase conjugate reflectivities measured up to now. We believe that, with more sophisticated experimental arrangements such as the ones used in BaTiO_3 ¹¹, phase conjugation with simultaneous amplification could be achieved at much shorter response times and for the visible wavelength range $\lambda = 400\text{--}600 \text{ nm}$.

The effect of wave-front restoration of the laser spot distorted by a roughened glass plate by four-wave mixing in reduced KNbO_3 is illustrated in Fig. 6. Besides static phase distortions, dynamic phase inhomogeneities occurring, e. g., in the photorefractive medium itself, can be compensated for since the recording time is of the order of 100 msec at the power levels used above and at $\lambda = 600 \text{ nm}$. This response time can be further decreased by increasing the light intensity or by using light sources with shorter wavelengths.

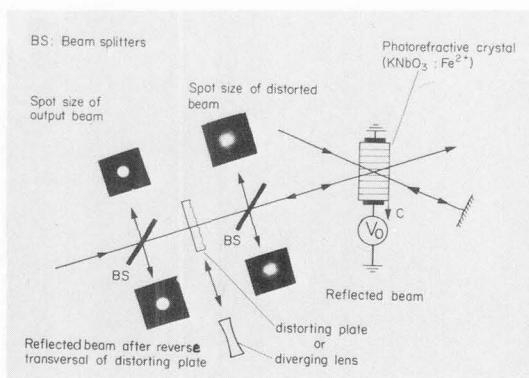


FIGURE 6: Experimental setup for phase-conjugate wave generation and for correction of phase distortions by four-wave mixing in photorefractive materials.

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