A NONLINEAR THEORY OF LIGHT AMPLIFICATION IN PHOTOREFRACTIVE MATERIALS*

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Summary

The basic physical principles responsible for two-wave interaction via an impressed grating in photorefractive materials is discussed. The main elements of the theory and the various types of solutions are outlined. Experimental results showing the amplification as a function of input beam ratio for a range of grating spacings are given and compared with the predictions of the theory.

Introduction

Photorefractive crystals have been the subject of extensive investigations ever since 1968 when the first experiments were performed [1]. The term photorefractive is more recent, perhaps ten years old. The name implies that the crystal is both photoconductive and electro-optic: the first property makes possible the generation of charge carriers and the second property is responsible for the change in refractive index in response to incident light.

What can photorefractive materials be used for? There is a large number of potential applications but none of them has so far reached the ultimate glory of commercial exploitation. The most attractive application is for erasable storage media with a potential capacity of about 10^{12} bits/cm³ [2]. Unfortunately, the realisation is hampered by its high sensitivity to tolerances. Another set of applications can be found in the field of real time signal processing as for example logical operations [3, 4] edge enhancement [5, 6], convolution and correlation [7, 8], time average and double-exposure interferometry [9, 10], components in integrated optics circuits [11], phase conjugation [12] and coherent light amplification [13].

The first material to exhibit these properties was LiNbO [1] but a number of others followed including BaTiO [14] KNbO [15], $Bi_{12}SiO_{20}$. $Bi_{12}GeO_{20}$ [16], CdS [17], GaAs [18, 19]. InP [18].

^{*} Dedicated to Professor Károly Simonyi on the occasion of his Seventieth Birthday

The present paper is concerned with coherent light amplification in the transmission configuration when two beams are symmetrically incident on a crystal (Fig. 1) and the frequency of one of them is shifted by a piezo-electric mirror. The amplification involved is of the parametric type when power transfer takes place between two oscillatory systems.



Fig. 1. Schematic representation of the illumination of a photorefractive crystal by two light beams. The frequency of the reference beam may be changed by applying a voltage to the piezoelectric mirror

A qualitative treatment

For simplicity we shall assume that the crystal has no dark conductivity, i.e. there are no mobile carriers present in the dark. There are however negatively charged acceptor atoms (density, N_A^-) and positively charged donor atoms (density, N_D^-) to which we shall refer as ionised acceptors and donors. For charge neutrality

$$N_A^- = N_D^+ . \tag{1}$$

It is further assumed that only a small fraction of the donors have lost electrons whereas all the acceptor atoms are ionised $(N_A = N_A^- \ll N_D)$ and that their density may be regarded constant.

Let us now take the simplest case when the crystal is illuminated by uniform light. The only effect is then the liberation of electrons from the donor atoms resulting in the new balance

$$N_A^- + n_0 = N_{D0}^+ . (2)$$

For most practical situations (except when high power lasers are used) $n_0 \ll N_A^-$.

Let us next consider the two beam case as shown in Fig. 1. The input light then forms an interference pattern in the crystal. The intensity variation may be

described as

$$I = I_0 (1 + m \cos Ky) \tag{3}$$

where

$$I_0 = I_R + I_S, \qquad m = 2(I_R I_S)^{1/2} / I_0$$
(4)

and I_R and I_S are the intensities of the reference and signal beams respectively.

The excited electron and ionised donor distribution will initially follow the light intensity distribution, i.e. we may expect

$$N_D^+ = N_{D0}^+ + N_{D1}^+ \cos Ky$$
 and $n = n_0 + n_1 \cos Ky$ (5)

with

$$n_1 = N_{D1}^+$$
 (6)

However there is now a density gradient and the electrons being mobile will move so as to reduce that gradient. The resulting current is of course the diffusion current. But as soon as the electrons move the balance of charge is disturbed and an electric field arises leading to the appearence of a conduction current. If, in addition, a voltage is applied across the crystal as shown in Fig. 1 then there is a further component of conduction current. To complete the story on the materials side we should also take into account that the resulting electron and donor distributions must be such that under stationary conditions the rate of electron generation balances the rate of recombination. The final situation is that all three, namely the electron density, the donor density and the electric field are shifted by various phase angles relative to the interference pattern.

As mentioned in the Introduction the crystal is electro-optic, i.e. an electric field causes a change in the refractive index. Thus we end up with a refractive index variation of the same period as the interference pattern but with a different phase.

The essential interaction is that the magnitude of the refractive index variation depends on the optical field amplitudes of the individual beams and, conversely, the individual field amplitudes depend on the refractive index variation. The relationships are nonlinear leading to a parametric type of interaction in which one beam is amplified at the expense of the other. The resulting amplification is strongly dependent on the phase shift. There is no amplification when the refractive index variation is in phase or anti-phase with the interference pattern, and maximum amplification occurs when they are 90 degrees out of phase.

It may be shown that the interaction is further enhanced if the frequency of one of the input light beams is changed slightly (of the order of a few tens of Hertz) resulting in a moving interference pattern, i.e. the intensity variation changes to

$$I = I_0 [1 + m \cos K(x - vt)]$$
(7)

where v is the speed of fringe movement.

The resulting interaction, it must be emphasised, is not the travelling wave type used in microwave amplifiers but there is no doubt that it needs both moving electrons and a moving interference pattern. The effect enhances the magnitude of the static electric field and also ensures the right phase relationship.

Theory

The first theory that predicted transfer of power between the beams was formulated by Staebler and Amodei [20] in terms of coupled wave differential equations. It was followed by several theoretical papers which generalised the problem in one or more respects. The currently accepted theory, which has been able to account for all the experimental observations so far, is that of Kukhtarev et al. [21]. Experimental and theoretical results for moving fringes have been given by Huignard and Marrakchi [22] and by Refregier et al. [23, 13].

Kukhtarev's treatment consists of two parts: firstly, the materials equations which describe the dependence of the refractive index distribution upon the light interference pattern, and secondly, the wave equation in which the effect of the refractive index distribution upon the optical qualities is taken into account.

The materials equations

The mathematical relationship between our variables for the onedimensional case may be written in the following form

$$e\frac{\partial N_D^+}{\partial t} = e\frac{\partial n}{\partial t} + \frac{\partial J}{\partial y}$$
(8)

$$\frac{\partial N_D^+}{\partial t} = sI(N_D - N_D^+) + \gamma_R n N_D^+$$
(9)

$$J = e\mu nE_{S} - k_{B}T\mu \frac{\partial n}{\partial x}$$
(10)

$$\frac{\partial E_S}{\partial y} = \frac{e}{\varepsilon_0 \varepsilon_S} \left(n + N_A^- - N_D^+ \right) \tag{11}$$

where J is the current density, E_S is the static electric field comprising of the applied field and of the space charge field, s is the cross section of photoionisation, γ_R is the recombination constant, μ is the mobility, k_B is Boltzmann's constant, T is temperature, ε_0 is the free space permittivity and ε_S is the relative static dielectric constant.

Eqns (8)–(11) are nearly self-explanatory. Eqn (8) is the continuity equation, Eqn (9) contains two terms on the right-hand-side, the first one is the rate of generation (proportional both to input light intensity and to the number of unionised donor atoms) and the second one is the rate of recombination (proportional both to electron density and ionised donor density). Thus the rate of increase of donor density is equal to the difference between the rate of generation and the rate of recombination. Eqn (10) says simply that the current density is equal to the sum of conduction current density and diffusion current density. Finally, Eqn (11) is Poisson's equation (or may be regarded as one of Maxwell's equations).

The next non-trivial question is how to solve the above differential equations. A full numerical solution in conjunction with the optical field equations to be given later, is out of the question even with today's fast digital computers. Therefore one must resort to some kind of analytical approximation. The following possibilities have been considered:

(i) Linearised stationary solution

It is reasonable to assume physically, and it has been shown experimentally, that after a certain time (which may be as long as several seconds) the transients die away and all the material quantities vary at a rate determined by the motion of the fringes. Secondly, one may assume that all the material quantities depend linearly upon the driving term represented by the travelling fringe pattern as expressed by Eqn (7). This means that the $\cos K(y-vt)$ term may be replaced by an $\exp jK(y-vt)$ term and a solution can be written in the form

$$G = G_0 + G_1 \exp jK(y - vt)$$
 (12)

where G may stand for any of the material quantities. Note that G_0 and G_1 are constants and that $G_1 \ll G_0$ is assumed. Whenever the product of two materials quantities comes up (as for example in Eqn (10) where the charge density is multiplied by the electric field) the cross product of the quantities with subscript 1 is neglected.

(ii) Higher perturbation stationary solution

The trial solution is attempted in the form

$$G = \sum_{p=0}^{r} \{G_p \exp[jpK(x-vt)] + \text{c.c.}\}$$
(13)

where r is the order of perturbation, c.c. stands for complex conjugate and G represents again any of the variables. Analytic solutions up to the second order have been found [13]; beyond that the equations appear to be intractable.

(iii) Phenomenological solution

The experimental results (given later in Section 4) clearly indicate that the available gain is reduced as the input power of the signal beam (to be amplified) increases. This suggests that the modulation of the dielectric constant increases less than linearly with the increase of fringe modulation. Guessing a relationship between these two quantities the experimental results may be matched.

(iv) Linèarised transient solution

The trial solution for this case is

$$G = G_0(t) + G_1(t) \exp[jK(y - vt)]$$
(14)

where it is assumed again that the spatially varying term is small in comparison with the spatially constant term. Both approximate analytical (when the reference beam is nondepleting) and numerical solutions have been found.

The wave equation for the optical intensities

In the general case the differential equation to be solved for the optical field vector, \overline{E} , inside the photorefractive crystal is

$$\nabla x \nabla x \overline{E} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \left[(\varepsilon_r + \Delta \varepsilon_r) \overline{E} \right] = 0$$
(15)

where c is the velocity of light, ε_r is the relative dielectric constant and $\Delta \varepsilon_r$ represents the dielectric constant modulation to be given by the materials equations. For most cases of interest the trial solution may be chosen in the

scalar form

$$E = \frac{1}{2} \left[E_R(x) \exp j(\omega_R t - \bar{k}_R \cdot \bar{r}) + E_S(x) \exp j(\omega_S t - \bar{k}_S \cdot \bar{r}) + \text{c.c.} \right]$$
(16)

where E_R and E_S are the normalised field amplitudes $(|E_R|^2 = I_R)$ and $|E_S|^2 = I_S$, ω_R and ω_S are the angular frequencies of the reference and signal beams, k_R and k_S are the respective wave vectors, and \bar{r} is a radius vector.

There is a standard technique to reduce the wave equation (Eqn (15)) to first order coupled wave differential equations in E_R and E_S similar to those obtained for ordinary, non-varying gratings [28]. The essential approximations are the neglect of second derivatives and of higher diffraction orders. The differential equations obtained may then be solved by analytic or numerical methods.

Experimental results and comparison with theory

A rather extensive set of experimental results on $Bi_{12}SiO_{20}$ crystals were obtained recently at the Thomson-CSF Laboratories, Orsay, France [13]. They are shown in Figs 2 and 3 where Γ and γ_0 are plotted against log β . For each measurement (represented by circles, triangles and rectangles) the frequency shift of the reference beam was chosen so as to maximise amplification. The input power in the reference and signal beams is $I_R(0)$ and $I_S(0)$ respectively

$$\beta = I_R(0)/I_S(0)$$
 and $\gamma_0 = I_S(l)/I_S(0)$, (17)

l is the length of the crystal, I(l) is the output power in the signal beam, and Γ is the apparent gain coefficient which for historical reasons is defined as

$$\Gamma = \frac{1}{l} \ln \frac{\gamma_0 \beta}{\beta + 1 - \gamma_0}.$$
(18)

The results in Figs 2a and 2b refer to two different crystal lengths, namely to 0.127 cm and 1 cm respectively. The parameter is the grating spacing Λ . The theoretical curves are the dotted and solid lines which are calculated from the second perturbation and phenomenological theories. The agreement between theory and experiment may be seen to be good.

It is worth noting that the amplification per unit length is larger for the shorter crystal, that the total amplification is over 1000 for the longer crystal and that the highest amplification is obtained for $\Lambda = 23 \,\mu\text{m}$, which is a relatively large grating spacing.



Fig. 2. The apparent gain coefficient Γ and the amplification γ₀ as a function of the input beam ratio for Λ = 23 µm, 10 µm and 3 µm, I₀ = 140 mW/cm², wavelength = 568 nm, applied field = 10 kV/cm. ○, △, □, experimental, -···· theoretical, second perturbation, ______ theoretical, phenomenological.
(a) l=0.127 cm, (b) l=1 cm

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Conclusions

The mechanism of parametric amplification in photorefractive crystals has been discussed both qualitatively and quantitatively. Experimental results for two separate $Bi_{12} SiO_{20}$ crystals and for a wide range of input beam ratios have been presented and shown to be in good agreement with a theory based on the formulation of Kukhtarev et al. It is concluded that high amplification may be achieved when the frequency of the reference beam is shifted resulting in a moving grating.

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