NONLINEAR DYNAMIC THEORY FOR PHOTOREFRACTIVE
PHASE HOLOGRAM FORMATION

by

Dae M. Kim, Rajiv R. Shah,
T.A. Rabson, and F.K. Tittel

January 1976

TECHNICAL REPORT #7602

NONLINEAR DYNAMIC THEORY FOR PHOTOREFRACTIVE PHASE HOLOGRAM FORMATION*

Dae M. Kim, Rajiv R. Shah, T.A. Rabson, and F.K. Tittel
Department of Electrical Engineering
Rice University, Houston, Texas 77001

A nonlinear dynamic theory is developed for the formation of photorefractive volume phase holograms. A feedback mechanism existing between the photogenerated field and free electron density, treated explicitly yields the growth and saturation of the space charge field in a time scale characterized by the coupling strength between them. The expression for the field reduces in the short time limit to previous theories and approaches in the long time limit the internal or photovoltaic field. Additionally, the phase of the space charge field is shown to be time dependent.

Photorefractive volume phase holography in electro-optic crystals such as $LiNbO_3$ has recently been investigated for optical data storage and display.[1,2] Amodei[3] proposed a theory for the photorefractive effect in the limit of electron drift or diffusion with the assumption that the electron migration length is short. For the same limits, Young et al.[4] removed this assumption and considered its effect on the phase shift between space charge field ($\mathcal{E}$) and the light modulation pattern. Both these theories are linear in that they do not consider the effect of photogenerated $\mathcal{E}$ back on to the electron drift or diffusion. Thus, they are only valid in the short writing time regime and do not explain the saturation behavior exhibited by these crystals[5].

In this Letter, we present a nonlinear theory of phase hologram formation. By adopting Amodei's general framework of electron drift and diffusion, we derive the coupled equation of the photogenerated electron density (n) and the resultant $\mathcal{E}$, and obtain a closed form solution. The continuity equation of n is given along the c-axis by

$$(\frac{\partial}{\partial t} + \frac{1}{\tau} - D \frac{\partial^2}{\partial x^2} + \mu E \frac{\partial}{\partial x} + \mu \frac{\partial}{\partial x} \mathcal{E})n = g(1 + m\cos Kx) \qquad (1)$$

where τ, D, μ, n, E denote, respectively, the electron trapping time, diffusion coefficient, mobility, and internal as well as bulk photovoltaic field.[6] The magnitude of the generation rate g is given in terms of photon number flux, absorption cross section σ, and the donor concentration N by $g = I_o \sigma N/\hbar\omega$; m is the depth of intensity modulation and $K = 4\pi\sin\theta/\lambda$ the grating wavevector with θ the writing half angle. Note that we have included in

Eq. (1) the modification in the electron drift arising from the generated space charge field. Also we have taken the field, E, to be directed along the negative c-axis[4,5].

We proceed by partitioning the electron density n as

$$n = n^{(o)} + \tilde{n} \tag{2a}$$

$$n^{(o)} = g\tau + n_1^{(o)} e^{iKx} + n_{-1}^{(0)} e^{-iKx} \tag{2b}$$

$$n_{\pm 1}^{(o)} = \tfrac{1}{2} mg\tau / (1 + K^2 L'^2 \pm iKL) \tag{2c}$$

Here $n^{(o)}$ represents the electron density produced <u>directly</u> by illumination and hence is obtained from Eq. (1) by setting $\mathcal{E} = 0$. Note that $n^{(o)}$ also contains transient terms which, however, decay rapidly within a few trapping times, τ. We have defined the electron drift and diffusion length as $L = \mu E\tau$, $L' = (D\tau)^{\frac{1}{2}}$ as in Ref. 4. It is to be noted that the previous theories[2,3] regarded $n^{(o)}$ as the total electron density giving rise to the space charge field, $\mathcal{E}$. However, it is important to realize that $n^{(o)}$ initially gives rise to $\mathcal{E}$, <u>which in turn affects the motion of electrons, i.e., the overall n</u>. This modification of n due to $\mathcal{E}$ is in our theory incorporated in the $\tilde{n}$-term, which then affects $\mathcal{E}$ and so forth. This nonlinear interaction between $\tilde{n}$ and $\mathcal{E}$ significantly affects the evolution in time of phase holograms.

We investigate this feedback mechanism by the coupled mode approach in which we represent $\tilde{n}$ and $\mathcal{E}$ by

$$\tilde{n} = \sum_{p=-\infty}^{\infty} \tilde{n}_p(t) e^{ipKx} \tag{3a}$$

$$\mathcal{E} = \sum_{p=-\infty}^{\infty} \mathcal{E}_p(t) e^{ipKx} \tag{3b}$$

Upon inserting Eq. (3) into Eq. (1) and Coulomb's law , $(\partial/\partial x)E_{total} = \rho/\epsilon$, one can obtain a hierarchy of coupled equations for each mode. We will consider the fundamental modes of $\widetilde{n}$ and $\mathcal{E}$, the equations of which read[7]

$$\left(\frac{\partial}{\partial t} + \frac{1}{\tau} + DK^2 + i\mu EK\right)\widetilde{n}_1 = i\mu K(g\tau\mathcal{E}_1 + \mathcal{S}) \tag{4a}$$

$$\left(\frac{\partial}{\partial t} + \frac{e\mu g\tau}{\epsilon}\right)\mathcal{E}_1 = -\frac{e}{\epsilon}(\mu E + iDK)(n_1^{(o)} + \widetilde{n}_1) - \frac{e\mu}{\epsilon}\mathcal{S} \tag{4b}$$

$$\mathcal{S} = \mathcal{E}_o(n_1^{(o)} + \widetilde{n}_1) + \mathcal{E}_2 n_{-1}^{(o)} + \sum_{p=1}^{\infty}(\mathcal{E}_{p+1}\widetilde{n}_{-p} + \mathcal{E}_{-p}\widetilde{n}_{p+1}) \tag{4c}$$

Equations (4) clearly show that $\widetilde{n}_1, \mathcal{E}_1$ are driven by the dominant terms, $n_o^{(o)} = g\tau$, $n_{\pm 1}^{(o)}$ and are coupled to each other via the electron drift, μE and diffusion, DK. We point out here that a new time scale, $\tau_g = (e\mu g\tau)/\epsilon)^{-1}$ naturally enters Eq. (4b) from first principles. τ_g is analogous to Rabi spin flipping time and provides a measure of time in which $\mathcal{E}_1$ grows and saturates. The $\mathcal{S}$-term represents higher order coupling between the fundamental modes under consideration and other components in $\widetilde{n}$ and $\mathcal{E}$.

One can analyze Eq. (4) by a straightforward perturbation scheme in which the unperturbed term $n^{(o)}$ drives $\mathcal{E}$ which then gives rise to the first order modification to $\widetilde{n}$ and so forth. Alternatively, one can use the Laplace transform technique and deal with a truncated matrix equation whose rank is determined by the number of coupled modes one chooses to consider. An exact solution in this truncated matrix equation is equivalent to summing over dominant contributions in each order of perturbation series in

the long time approximation[8] (which in our case is longer than the trapping time τ). By using the second method, we analyzed in great detail Eq. (4) as well as other harmonic components. In this Letter, we discuss the major term of the space charge field due to $\mathcal{E}_{\pm 1}$ and will report the detailed analysis in a later publication. The space charge field was found to be

$$\mathcal{E}(t) = m\mathcal{F}_c(t)\cos Kx + m\mathcal{F}_s(t)\sin Kx \qquad (5)$$

with the in-phase and quadrature amplitudes,

$$\mathcal{F}_c(t) = E[1 - e^{-ft/\tau_g}\cos(f't/\tau_g)] - \frac{DK}{\mu} e^{-ft/\tau_g}\sin(f't/\tau_g) \qquad (5a)$$

$$\mathcal{F}_s(t) = Ee^{-ft/\tau_g}\sin(f't/\tau_g) + \frac{DK}{\mu}[1 - e^{-ft/\tau_g}\cos(f't/\tau_g)] \qquad (5b)$$

and the time scale

$$\tau_g = (e\mu g\tau/\varepsilon)^{-1} \qquad (5c)$$

$$f = (1 + K^2 L'^2)/[(1 + K^2 L'^2)^2 + K^2 L^2] \qquad (5d)$$

$$f' = KL/[(1 + K^2 L'^2)^2 + K^2 L^2] \qquad (5e)$$

Equations (5) explicitly describe the dynamic formation of the fundamental component of $\mathcal{E}$ in terms of writing time, t; the internal or photovoltaic field, E; the generation rate, g; and the electron drift (L) and diffusion (L') lengths. Here we considered the electron drift and diffusion contribution simultaneously. Note that in the short writing time limit, i.e., linear regime ($t \ll \tau_g$), our space charge field yields an expression identical to those in Ref. 4 for drift and diffusion only cases. (See their equations (3) and (4).) On the other hand, in the long time limit, $\mathcal{E}$ attains asymptotically the steady state value mE in phase with the intensity pattern and the value mDK/μ in quadrature. In this paper, we have not included the small modifica-

tions in these steady state values arising from the subsequent generation of harmonic terms in $\mathcal{E}$ and $\tilde{n}$. In order to relate the modulation of index of refraction in space and time to that of intensity pattern, it is convenient to recast Eq. (5) in the form

$$\mathcal{E}(t) = A(t)\cos[Kx - \varphi_g(t)] \tag{6a}$$

with

$$A(t) = m[\mathcal{F}_c^2(t) + \mathcal{F}_s^2(t)]^{\frac{1}{2}}; \varphi_g(t) = \tan^{-1}[\mathcal{F}_s(t)/\mathcal{F}_c(t)] \tag{6b}$$

The significant information contained in Eqs. (5) and (6) is summarized as follows: (1) The time scale in which $\mathcal{E}$ grows and saturates, determined by τ_g, f, f', is mainly affected by the generation rate, the mobility and trapping time and to a lesser extent by E . Since g is proportional to the light intensity as well as the donor concentration, one can control the time required to form an efficient hologram; (2) the relative phase, φ_g, between $\mathcal{E}$ and intensity pattern is determined by the relative contribution of drift and diffusion to $\mathcal{E}$ and even more importantly, φ_g undergoes a significant change as a function of writing time.

To illustrate the time dependence of the quantities involved, we present two theoretical curves. In Fig. 1a are shown the magnitude (A) and the relative phase (φ_g) of $\mathcal{E}$ as a function of writing time for a fixed field and for three different generation rates, g. As g increases, A(t) rises rapidly. reaching its steady state value in a short time interval. For E = 80KV/cm and g = 5×10^{15}/cm^3 sec, the space charge field attains the mE value within 40 seconds. Additionally, $\varphi_g(t)$ decreases at a much faster rate with increasing g . In Fig. 1b are shown A(t) and $\varphi_g(t)$ for a fixed generation

rate and various field strengths, E. In this case, $\varphi_g(t)$ decreases monotonically at about the same rate and A(t) rises with about the same time scale. The absolute value of A(t) at a given time is proportional to E.

In conclusion, a nonlinear dynamic theory of photorefractive volume phase hologram formation has been developed. Our general expression for the space charge field includes the results of previous theories in appropriate limits. The nonlinear growth in time of the space charge field amplitude (or equivalently the index of refraction modulation) shows the general trend experimentally observed in Ref. 5. Needless to say, further contact of $\mathcal{E}$ with experimentally measured quantities can be made in terms of the diffraction efficiency of a hologram, η. Recently a closed form solution of η was obtained[9] and the linear (in time) results of $\mathcal{E}$ in Ref. 4 were used to fit the data with nonlinear time dependence of $\mathcal{E}$ introduced phenomenologically. The expression for η in Ref. 9 considers the modification of the refractive index due to $\mathcal{E}$. We feel, however, that a closed form expression of η should include a similar modification in the local absorption coefficient due to the time varying space charge field[10,11]. This discussion will be presented elsewhere.

REFERENCES AND FOOTNOTES

1.   D. Chen and J.D. Zook, Proc. IEEE 63, 1207 (1975).

2.   D.L. Staebler and W. Phillips, Applied Optics 13, 788 (1974).

3.   J.J. Amodei, RCA Rev. 32, 185 (1971).

4.   L. Young, W.K.Y. Wong, M.L.W. Thewalt, and W.D. Cornish, Appl. Phys. 24, 264 (1974).

5.   F.S. Chen, J. Appl. Phys. 40, 3389 (1969).

6.   A.M. Glass, D. von der Linde, and T.J. Negran, Appl. Phys. Lett. 25, 233 (1974).

7.   Note that the time independent component in $\widetilde{n}_1$ is zero.

8.   I. Prigogine, Nonequilibrium Statistical Mechanics V.1, Interscience Publishers (1962), Chapter 2.

9.   D.W. Vahey, J. Appl. Phys. 46, 3510 (1975).

10. H. Kogelnik, Bell Syst. Tech. J. 48, 2909 (1969).

11. Y. Ninomiya, J. Appl. Phys. 63, 1124 (1973).

FIGURE CAPTIONS

Fig. 1a   The time dependence of the amplitude A(t) (solid line) and the rela-
tive phase (dashed line) in units of 2π for three different electron
generation rates and a constant field, E .

Fig. 1b   The time dependence of the amplitude A(t) (solid line) and the rela-
tive phase $\varphi_g(t)$ (dashed line) in units of 2π for a constant genera-
tion rate and for different values of electric field.

Axis labels:
- Left y-axis: $A(t)$ (kV/cm), ranging from 1 to 100
- Right y-axis: $\phi_g(t)/2\pi$, ranging from 10^{-4} to 10^{-2}
- x-axis: TIME (sec), 20 to 100

Curve labels:
- $g = 5 \times 10^{15}/cm^3$ sec
- $g = 10^{15}/cm^3$ sec
- $g = 5 \times 10^{14}/cm^3$ sec

$E = 80$ kV/cm

(a)

(b)