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# Two beam coupling gain enhancements in porphyrin:Zn-doped nematic liquid crystals by using grating translation technique with an applied dc field

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#### ABSTRACT

The two beam coupling (TBC) gains in porphyrin:Zn-doped nematic liquid crystals (NLCs) are greatly enhanced by adopting the grating translation technique with applying dc electric field. The maximum gain coefficient of  $\Gamma=624~{\rm cm^{-1}}$ , whose value is at least three times larger than that of no grating translation was obtained under the applied dc field of  $E_0=1.2~{\rm V/\mu m}$  and the grating translation speed of  $v_q=33.4~{\rm \mu m/s}$ . Based on the material and torque balance equations for NLCs taking into account the grating translation, we present the theoretical expressions for TBC gain enhancements, showing excellent agreement with the experimental results.

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#### 1. Introduction

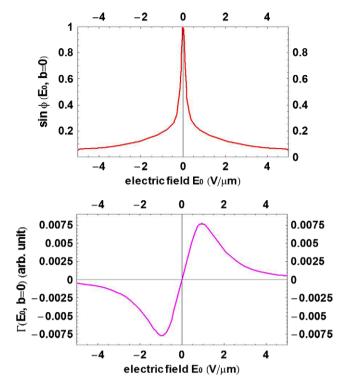
Nematic liquid crystals (NLCs)-based photorefractive materials have been extensively investigated because of their large dielectric and optical anisotropies. Since an orientational photorefractive (OPR) effect in doped NLCs has been predicted and experimentally demonstrated by Rudenko and Sukhov [1] and later by Khoo [2], intensive investigations have been performed on this subject [3-5]. Strong two beam coupling (TBC) energy transfer between two coherent writing beams in dye-doped NLCs was reported [5,6] and Khoo presented a detailed discussion of the optical and electric field induced photorefractivity in NLCs and observed the large optical nonlinearity in fullerene  $C_{60}$  and dye-doped NLCs [2,3]. Recently, we reported on the observation of an OPR effect in porphyrin: Zn doped NLCs by measuring the TBC gain and diffraction efficiency under the influence of an applied dc field [7]. The TBC gain coefficient of over 170 cm<sup>-1</sup> was obtained with the applied dc fields of  $\textit{E}_0 = 0.57 \text{ V}/\mu\text{m}$  and the refractive index change coefficient was  $n_2 \approx 10^{-2}$  cm<sup>2</sup>/W. This value of  $n_2$  is one order of magnitude larger than that of  $C_{60}$ -doped 5CB sample.

In this work, we performed the TBC experiments in porphyrin: Zn doped NLCs by adopting the grating translation technique with dc electric field and presented that the TBC coefficient is

\* Corresponding author. E-mail address: chkwak@yu.ac.kr (C.H. Kwak). greatly enhanced. Under the applied dc field of  $E_0=1.2~{\rm V/\mu m}$  and the grating translation speed of  $\nu_q=33.4~{\rm \mu m/s}$ , the maximum gain coefficient we achieved was  $\Gamma=624~{\rm cm^{-1}}$ , whose value is at least three times larger than that of 170 cm<sup>-1</sup> with no grating translation.

Some techniques for the enhancement of TBC gain in inorganic photorefractive crystals such as BSO, InP:Fe, GaAs and others have been investigated and developed in the past years; applied dc field technique, dc field and moving grating technique, intensity- and temperature-dependent technique, and applied ac field technique [8,9]. The most widely used gain enhancement technique is the moving grating method [8], which is to apply a dc electric field in a sample accompanied by frequency detuning one of the writing beams. The moving grating technique with an applied dc electric field [8] is developed by the interference fringe shift with an external mirror detuned by a PZT transducer for gain enhancement, while the grating translation technique [10] is by the direct movement of the sample by a linear motor for determining the phase and the magnitude of the phase grating and the absorption grating [11], not for enhancing the TBC gain and/or diffraction efficiency.

The aim of this work is to develop the theory of TBC gain enhancement for the grating translation method and to explain the experimental results. From the material equations and the torque balance equation of NLCs taking into account of the grating translation, we derived the expressions for the moving spacecharge field, the refractive index change of the OPR grating and the TBC gain coefficient.



**Fig. 1.** The sine of the grating phase shift,  $\sin\phi(E_0, b=0)$ , and the normalized gain coefficient  $\Gamma(E_0, b=0)$  as a function of applied dc electric field  $E_0$  with no grating translation (i.e., b=0).

#### 2. Theory

To analytically explain the OPR gain enhancement phenomena due to the grating translation, we start with the material equations for NLCs [1]:

$$\frac{\partial n^{\pm}}{\partial t} + \gamma n^{+} n^{-} \pm \frac{1}{e} \nabla \cdot \mathbf{J}^{\pm} = \alpha I, \tag{1a}$$

$$\mathbf{J}^{\pm} = e\mu^{\pm}n^{\pm}\mathbf{E} \mp k_{B}T\mu^{\pm}\nabla n^{\pm} + \sigma_{d}\mathbf{E}, \tag{1b}$$

$$\nabla \cdot \mathbf{E} = \frac{e}{\varepsilon \varepsilon_0} (n^+ - n^-), \tag{1c}$$

where  $n^{\pm}$  are the positive and negative charge carrier densities,  $J^{\pm}$ are the current densities,  $\mu^{\pm}$  are the mobilities,  $\gamma$  is the recombination rate,  $\alpha$  is the charge generation rate, e is the elementary charge,  $k_B$  is the Boltzmann's constant, T is the absolute temperature,  $\varepsilon$  is the relative dielectric constant,  $\varepsilon_0$  is the dielectric constant in the vacuum,  $\sigma_d$  is the dark conductivity, which is assumed to be neglected for small value as compared to the photocurrent, E is the total electric field, consisting of the applied dc electric field  $E_0$  and the induced space-charge field  $E_1$ . In TBC experiments accompanied by the grating translation, the sample is illuminated by the sinusoidally modulated light intensity of  $I(\mathbf{r}, t) = I_0 + I_1 \exp[i(\mathbf{q} \cdot \mathbf{r} - \Omega t)]/$ 2 + c.c., where  $I_0(=I_a+I_b)$  is the total input beam intensity,  $I_a$  and  $I_b$ are the incident intensities of the two coherent writing beams,  $I_1 = mI_0$ , m is the modulation depth, defined  $m = 2\sqrt{I_aI_b}/(I_a + I_b)$ , **q** is the grating vector,  $q = |\mathbf{q}| = 2\pi/\Lambda_g$  is the grating wave number,  $\Lambda_g$  is the grating period,  $\Omega = qv_q$  is the frequency detuning,  $v_a$  is the moving velocity along the grating vector and c.c. denotes the complex conjugate. The physical variables,  $n^{\pm}$ ,  $J^{\pm}$ and E in Eq. (1), are assumed to have the same periodic function with the intensity distribution  $I(\mathbf{r},t)$ . Substituting the physical variables including the intensity grating into Eq. (1) and separating the

variables with subscripts 0 and 1, and following the same procedures of Ref. [12], after some calculations, we finally obtain the time-dependent differential equation for the space-charge field:

$$\frac{\partial E_1}{\partial t} + gE_1 = mh,\tag{2}$$

where g=B/A, h=C/A,  $A=(1+2\tau_d/\tau+E_D/E_q+iE_0v\sin\beta/E_q+i2b)/\tau_d$ ,  $B=(2/\tau_d\tau)(1+E_D/E_M+E_D/2E_q+E_0^2\sin^2\beta/2E_qE_M+E_D^2/2E_qE_M+iE_0-v\sin\beta/2E_q+ib)$ ,  $C=(iE_Dv-E_0\sin\beta)/\tau_d\tau$ ,  $\beta$  is the tilt angle of the grating as shown in Fig. 4,  $n_0^+=n_0^-=n_0=\sqrt{\alpha I_0/\gamma}$ ,  $\tau_d=\varepsilon\varepsilon_0/en_0(\mu^++\mu^-)$  is the Maxwell relaxation time,  $E_q=en_0/q\varepsilon\varepsilon_0$  is the limiting space-charge field,  $E_M=\gamma n_0/\mu q$  is the drift field,  $E_D=qk_BT/e$  is the diffusion field,  $\tau=1/\gamma n_0$  is the photo-ion life time,  $b=\Omega\tau_d=qv_q\tau_d$ ,  $v=(\mu^+-\mu^-)/(\mu^++\mu^-)$  and  $\mu=\mu^+\mu^-/(\mu^++\mu^-)$ . It is noted that in deriving Eq. (2) we neglect the cross products of two quantities with subscript 1, which are the nonlinear driving sources of some interesting phenomena such as higher harmonic gratings and subharmonic gratings [12] and use the slowly-varying amplitude approximation of the space-charge field with time. For steady state, the magnitude of the space-charge field  $|E_1|$  and the grating phase shift  $\phi$  relative to the intensity grating are given by, respectively.

$$|E_1| = \frac{m}{2} \left[ \frac{E_D^2 v^2 + E_0^2 \sin^2 \beta}{X^2 + Y^2} \right]^{1/2}, \tag{3a}$$

$$\phi = \tan^{-1} \left[ \frac{E_D \nu X + E_0 Y \sin \beta}{E_D \nu Y - E_0 X \sin \beta} \right], \tag{3b}$$

where  $X=1+E_D/E_M+E_D/2E_q+E_0^2\sin^2\beta/2E_qE_M+E_D^2/2E_qE_M$ ,  $Y=E_0v\sin\beta/2E_q+b$ . The applied dc electric field  $E_0$  and the dimensionless moving velocity  $b=\Omega\tau_d=qv_q\tau_d$  alter not only the magnitude of the space-charge field, but also the grating phase shift. The translating optical interference pattern creates a moving space-charge field, followed by the optical torque change of the director axis of the NLC molecules, which is caused by the combined effect of the applied dc field and the moving space-charge field. As a result, the director axis reorientation of NLC and the photoinduced refractive index changes occur. The torque balance equation under the one elastic constant K is given by [2,7,12],

$$\gamma_{\mathit{vis}} \frac{\partial \theta}{\partial t} = K \left[ \frac{\partial^2 \theta}{\partial x^2} + \frac{\partial^2 \theta}{\partial z^2} \right] + |\Gamma_{\mathbf{opt}}| \tag{4}$$

where  $\theta$  is the reorientation angle of the director axis,  $\gamma_{vis}$  is the Leslie viscosity coefficient,  $|\Gamma_{\rm opt}| = \Delta \varepsilon \varepsilon_0 | {\bf n} \cdot {\bf E}({\bf n} \times {\bf E})|$  is the magnitude of the director axis torque, which is induced by the optical and electric field, and  ${\bf n}=(\sin\theta,0,\cos\theta)$  is the reoriented director axis vector of NLCs and  $\Delta\varepsilon=\varepsilon_{\parallel}-\varepsilon_{\perp}$  is the dielectric anisotropy. Taking the trial solution of the form  $\theta({\bf r},t)=\theta_1(t)\cos({\bf q}\cdot{\bf r}-\Omega t+\phi)$  with the small reorientation angle approximation (i.e.,  $\theta_1<1$ ) and following the same procedures of the literatures [2,7,12], we have the steady state value of the OPR grating (i.e., the refractive index change of the induced extraordinary wave phase grating)  $\Delta n$  as

$$\Delta n = \delta n_1 \cos(\mathbf{q} \cdot \mathbf{r} - \Omega t + \phi), \tag{5}$$

where  $\delta n_1=(n_\parallel-n_\perp)(n_\parallel/n_\perp)\sin(2\beta)\theta_1$  is the amplitude of the OPR grating,  $\theta_1=E_0|E_1|\cos\beta/(E_C^2+E_0^2)$  is the reorientation angle of director axis of NLCs,  $E_C=\sqrt{Kq^2/\Delta\varepsilon\varepsilon_0}$  is the Freedericksz transition field,  $n_\perp$  is the ordinary index of refraction,  $n_\parallel$  is the extraordinary index of refraction, and  $\Delta\varepsilon=\varepsilon_\parallel-\varepsilon_\perp$ . The theoretical and experimental TBC gain coefficient  $\Gamma(E_0,b)$  are then given by, respectively [13],

$$\Gamma(E_0, b) = \frac{4\pi}{\lambda_w \cos \theta_{inc}} \delta n_1 \sin \phi, \tag{6a}$$

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