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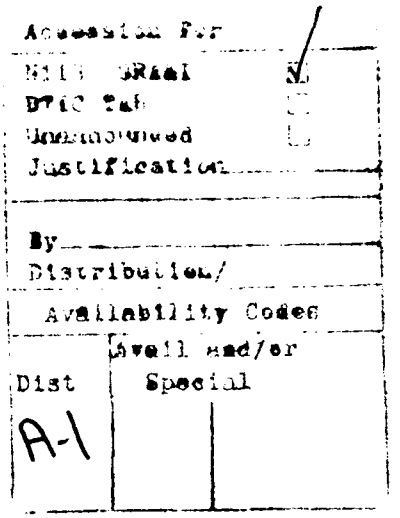

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Two-Beam Coupling Measurements of Grating Phase in a Photorefractive Polymer

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ABSTRACT: We investigate in detail the grating properties of a recently discovered organic photorefractive polymer as a function of electric field using two-beam coupling. We present measurements of index and absorption grating phase relative to the intensity interference pattern as well as the amplitude of both the absorption and index gratings. We find that in low electric fields, a weak in-phase grating (possibly photochromic) and the low electro-optic coefficient prevent observation of a phase-shifted photorefractive grating. However, at moderate to high electric fields, a much stronger photorefractive index grating with phase shift approaching 90° dominates. The field dependence of the index grating phase is discussed in terms of a standard photorefractive model. The presence of an index grating with a 90° phase shift at high fields provides strong evidence that these polymers are indeed photorefractive.

I. INTRODUCTION

Recent observations of the photorefractive effect (PRE) in a doped organic nonlinear crystal[1] and in doped nonlinear polymers [2-4] have opened up a new class of materials which offers the promise of reasonable nonlinearity with small dc dielectric constant, that is, improved values of the figure-of-merit n^3r/ε, with n the optical index of refraction, r the electro-optic coefficient, and ε the dc dielectric constant. In these materials, a host matrix that is optically nonlinear by virtue of either poling or crystal growth is made photoconducting by the addition of dopant molecules which allow for charge generation, transport, and/or trapping. In comparison to organic crystals, the poled polymer materials in general offer additional flexibility in that they are easily doped, readily formed into thin films for waveguide experiments, and made acentric by poling. The detailed properties of the polymer materials are therefore of current interest so that the ultimate limits of performance of this new materials class can be determined.

Many of the intriguing possible applications for PRE [5,6], such as beam fanning[7], novelty filtering [8,9], and self-phase conjugation [10, 11] rely upon the presence of a nonzero phase shift between the index of refraction grating and the optical intensity (interference) pattern, or equivalently, asymmetric two-beam coupling. In addition, as long as moving gratings and/or ac electric fields are not used [12-14], a nonzero phase shift is in fact firm proof that the grating formed is indeed due to charge generation, transport, and trapping in an electro-optic material, i.e., the true photorefractive effect and not a purely local photochromic, heating, or other type of dynamic grating[15].

This paper reports grating phase measurements as a function of applied field for the photorefractive polymer composed of the nonlinear epoxy polymer bisA-NPDA (bisphenol-A-diglycidylether 4-nitro-1,2-phenylenediamine[16]) doped with 30 weight % of the hole transporting molecule, DEH (diethylamino-benzaldehdye diphenylhydrazone). The photorefractive effect in these polymers was first demonstrated using field- and polarization-dependent volume holography as reported earlier [3,17]. Here we add the additional evidence provided by two-beam coupling measurements of grating phase, which shows that the phase

of the index grating is close to 90 ° at moderate to high fields and the absorption grating amplitude under these conditions is small relative to the index grating. At zero field, only a weak, in-phase grating was observed, presumably due to photochromic effects. The convincing observation of nonzero phase shift in this polymeric photorefractive should lead to future experiments which directly require the presence of asymmetric two-beam coupling. In addition, this work illustrates a direct experimental test which should be applied to future candidate organic photorefractive materials to rule out sources for grating formation other than photorefractivity.

II. EXPERIMENTAL

In previous studies in photorefractive and nonphotorefractive materials, several techniques for the determination of grating phase have been demonstrated, including external phase shifting of one beam by mirror motion[18], phase-modulation approaches[19], and electro-optic shifting of the interference pattern [20] to name a few. A recent technique, fast displacement of the sample along the grating wavevector after grating formation[21] is convenient for the detection of asymmetric two-beam coupling even when the overall diffraction efficiency is small, as it naturally is in the relatively thin (350 μm) samples used here. This is essentially a result of the homodyne enhancement of the diffracted beam by the transmitted beam in the same direction.

Samples for this study were fabricated by mixing the partially crosslinked epoxy polymer bisA-NPDA and the charge transport agent DEH in a solvent, drying, and placing the doped polymer between two glass slides with transparent electrodes as reported elsewhere[3]. The geometry of the grating formation is shown in Figure 1. A tilted grating with oblique incidence angles is required in order to yield a reasonable projection of the grating wavevector along the poling (E_{dc}) axis of the sample. Two equal-intensity p-polarized Gaussian beams (beam 1 and beam 2) with vacuum wavelength λ_v = 647.1 nm from a Kr^+ ion laser were crossed in the sample at external incidence angles of 30 ° and 60 °, respectively, writing a grating with spacing Λ_g = 1.6 μm oriented 25 ° from the plane of the film. The various angles inside the sample are defined as follows: θ_g is the angle between the grating vector

K_g (which is parallel to the space-charge field E_{sc}) and the film plane; θ_1 is the angle of incidence for beam 1; θ_2 is the angle of incidence for beam 2; and $2\theta_o = \theta_2 - \theta_1$ is the crossing angle between beams 1 and 2. The growth of the grating gives rise to two diffracted beams which propagate in the same direction as the other writing beam. Grating phase measurements were performed with externally applied dc fields E_{dc} ranging from 0 - 114 kV/cm. For each field, a thoroughly erased or new spot on the sample was used to prevent interference from pre-existing gratings.

The two-beam coupling and grating phase measurements were performed using a slight modification of the method of Sutter et al.[21]. A grating was formed using two equal intensity writing beams with the tilted grating geometry as described above. The intensity of each writing beam was 2.5W/cm^2. The total transmitted intensity of each beam was recorded using identical photodetectors. The transmitted intensities were normalized for laser power fluctuations using a pair of electronic ratiometers, low-pass filtered, and fed into a transient waveform recorder. After writing the grating for 3-5 minutes, the grating was moved along the plane of the sample using a precision translator driven by a stepper motor with 0.1 μm resolution (Klinger UT 50.20). Both beams 1 and 2 were diffracted by the grating and the diffracted beams interfered with the transmitted beams propagating in the same direction. The translation of the sample thus resulted in a modulation of the transmitted intensities at the detectors. Although the diffraction efficiencies of the gratings in bisA-NPDA/DEH were typically 10^{-4} to 10^{-5}, the relative amplitudes of the intensity modulations were 10^{-2} to 3×10^{-3} due to the homodyne detection.

In this work, the sample was translated 6.7 μm in 2.1 s, minimizing grating erasure during translation. The normalized transmissions of the two beams as a function of grating displacement were recorded simultaneously, as is shown in Figure 2 for an applied field of 86 kV/cm. The irregularities in the traces near the beginning of motion were due to slight imperfections in the initial motion of the translator. Measurements were also performed with (i) a different translation stage driven by a 20,000 step-per-revolution stepper motor and (ii) a Polytec P280.2 piezoelectric block translator, but these devices suffered from (i) more uncertainty in the initial motion of the stage due to backlash and (ii) nonlinear displacement,

respectively. Nevertheless, grating phase determinations from both of these alternate translators provided results similar to those reported here for the 0.1 μm translation stage. Overall, the scans with the latter stage were found to be the most reproducible. In addition, it is evident in Fig. 2 especially before the translation begins and after it stops that there is a slow drift in the dc levels of the ratiometers over the course of the scan. This and the translational inaccuracies contributed some scatter to the determinations of the grating phase when the grating amplitude is small (see Figure 3 below).

III. THEORY and DATA ANALYSIS

This section presents a generalization of the analysis of Sutter et al.[21] for the case of p-polarized diffraction from a slanted grating, using the general coupled wave theory of Kogelnik[22]. After the grating is formed, both beams 1 and 2 in Fig. 1 act as reading beams whose electric field amplitudes are labeled with the letters R_1 and R_2, respectively, and the diffraction from the grating produces a pair of scattered beams labeled S_1 and S_2. We assume that the grating formed during the writing interval by the intensity pattern of modulation depth $m \simeq 1$ given by $I = 2I_o[1 + \cos(\mathbf{K_g}\cdot\mathbf{r})]$ produces (separate) index and absorption gratings so that

$$n(\mathbf{r}) = n_o + \Delta n \cos(\mathbf{K_g}\cdot\mathbf{r} - \phi_n)$$

$$\alpha(\mathbf{r}) = \alpha_o + \Delta\alpha \cos(\mathbf{K_g}\cdot\mathbf{r} - \phi_\Lambda). \qquad (1)$$

with α_o the field amplitude absorption coefficient, n_o the background refractive index (= 1.63) $\mathbf{r}$ a vector in the material, and I_o the intensity in each of the two writing beams. (In fact, as will be shown below, equal intensities for the two writing beams outside the sample yields unequal intensities inside the sample, and the modulation index will only be near 1 rather than equal to 1. This only affects the ultimate sizes of $\Delta\alpha$ and Δn and may be ignored without loss of generality.) The index and absorption grating amplitudes will only be related by a Kramers-Kronig relation if they arise from the same physical process; otherwise, the two

amplitudes can be independent. The phases ϕ_P and ϕ_Λ are the desired phase shifts of the index and absorption gratings, respectively.

First considering readout by beam 1, the fields just before exiting the polymer are then given by

$$E^{(1R)} = E_{o1} R_1 \exp[i(\omega t - \mathbf{k}_1 \cdot \mathbf{r})],$$

$$E^{(1S)} = E_{o1} S_1 \exp[i(\omega t - \mathbf{k}_2 \cdot \mathbf{r})], \tag{2}$$

where $\mathbf{k}_1$, $\mathbf{k}_2$ and E_{o1}, E_{o2} are the propagation vectors and initial field amplitudes for the reading and scattered beams just inside the polymer, respectively. We consider the limit of small diffraction efficiencies, which means that the amplitude of the reading (R) beam is affected only by optical losses in the material, and that the contributions of the index and absorption gratings to the diffracted beam amplitude simply add. Further, since beams 1 and 2 formed the grating, readout is automatically Bragg-matched. In this case,

$$R_1 \equiv D = \exp\left[\frac{-\alpha_o d}{2}\left(\frac{1}{c_r} + \frac{1}{c_s}\right)\right]. \tag{3}$$

with d the sample thickness, and $c_r = \cos\theta_1$, $c_s = \cos\theta_2$ are the obliquity factors when beam 1 is the reading beam. The scattered beam coefficient contains terms from both the absorption and index gratings:

$$S_1 = -D[iP \exp(-i\phi_P) + \Lambda \exp(-i\phi_\Lambda)]. \tag{4}$$

with the diffraction amplitudes P and Λ given by

$$P = \frac{(\hat{\mathbf{r}}\cdot\hat{\mathbf{s}})\pi d\Delta n}{\lambda_v(\cos\theta_2 \cos\theta_1)^{1/2}},$$

$$\Lambda = \frac{(\hat{\mathbf{r}}\cdot\hat{\mathbf{s}})d\Delta\alpha}{2(\cos\theta_2 \cos\theta_1)^{1/2}} \tag{5}$$

with λ_v the free-space wavelength. In these relations, the factor resulting from p-polarized readout $\hat{r}\cdot\hat{s} = \cos(\theta_2 - \theta_1)$ is given by the dot product of the two field polarization vectors $\hat{r}$ and $\hat{s}$ (see Eq. (90) of Ref. 22). Furthermore, the scattered beam factor S_1 has been multiplied by the factor $(c_s/c_r)^{1/2}$, in order to satisfy the requirement for power flow conservation in the direction perpendicular to the film (see Eqs. (24) and (40) of Ref. 22), i. e., the scattered beam and the readout beam to which it is referenced are propagating in different directions.

Similarly, when beam 2 is the reading beam,

$$E^{(2R)} = E_{o2}D \exp[i(\omega t - \mathbf{k}_2\cdot\mathbf{r})],$$

$$E^{(2S)} = E_{o2}S_2 \exp[i(\omega t - \mathbf{k}_1\cdot\mathbf{r})], \tag{6}$$

and

$$S_2 = -D[iP \exp(+i\phi_P) + \Lambda \exp(+i\phi_\Lambda)], \tag{7}$$

with P and A given by Eqs. (5) and where the positive signs in the exponential arguments of S_2 reflect diffraction in the opposite direction relative to the grating phase shift.

Linear superposition allows simple addition of the field amplitudes for each of the two beams just before exiting the polymer to give

$$E^{(1T)} = D[E_{o1} - E_{o2}(iPe^{-i\phi_P} + \Lambda e^{-i\phi_\Lambda})],$$

$$E^{(2T)} = D[E_{o2} - E_{o1}(iPe^{+i\phi_P} + \Lambda e^{+i\phi_\Lambda})]. \tag{8}$$

where the superscript T means "total". Since beams 1 and 2 are incident at different angles, the amplitudes E_{o1} and E_{o2} will not be equal in general. The intensity in each of the two beams at the exit edge of the polymer may then be computed to be (dropping terms of order P^2, Λ^2)

$$I^{(1)} = (1/2)D^2 n_o \varepsilon_o [E_{o1}^2 - E_{o1}E_{o2}(2\Lambda \cos\phi_\Lambda + 2P \sin\phi_P)],$$

$$I^{(2)} = (1/2)D^2 n_o \varepsilon_o [E_{o2}^2 - E_{o1}E_{o2}(2\Lambda \cos\phi_\Lambda - 2P \sin\phi_P)], \tag{9}$$

where ε_o is the permittivity of free space (SI units). These useful relations show that the contributions to the two beams from index gratings are 180° out of phase, while the contributions from absorption gratings are in phase.

It is clear that the oblique incidence and unequal reading field amplitudes complicate the analysis; however, an important simplification can be made by factoring out the geometric mean of the intensity in the two reading beams $I_g = (1/2)D^2 n_o \varepsilon_o E_{o1} E_{o2}$ and forming the sum and the difference of the intensities:

$$I^+ = I_g\left[\left(\frac{E_{o1}^2 + E_{o2}^2}{E_{o1}E_{o2}}\right) - 4\Lambda \cos\phi_\Lambda\right], \tag{10}$$

$$I^- = I_g\left[\left(\frac{E_{o1}^2 - E_{o2}^2}{E_{o1}E_{o2}}\right) - 4P \sin\phi_P\right]. \tag{11}$$

Using the Fresnel factors for beams 1 and 2 entering the sample to compute E_{o1} and E_{o2} from the value of the incident electric field amplitude E_o outside the sample, it can easily be verified for our geometry that the first term in the brackets of Eqn. (10) is equal to 2 within a few percent. In addition, computation of the intensity transmission coefficients for propagation of the two beams out of the exit side of the sample through the cover plate and into the air yields a transmission coefficient of 0.98 for beam 1 and 0.99 for beam 2. Thus, without loss of generality, we may take Eqns. (10) and (11) to apply to the actual detected intensities. The final complicating factor, the first term in the brackets in Eq. (11) is not zero as it is in the non-oblique case [21] and can thus be left as a fitting parameter.

As the sample is translated in a time much less than the grating growth or decay times, the effect is to replace the static phase shifts $\phi_{\Lambda,P}$ by $(\phi_{\Lambda,P} + 2\pi\xi \cos(\theta_g)/\Lambda_g)$ where ξ is the displacement increasing linearly with time t from t = 0. Thus, the sum and difference of the two transmitted intensities can then be written

$$I^{+}(\xi) = I_g[2 - 4\Lambda \cos(\phi_\Lambda + 2\pi\xi \cos(\theta_g)/\Lambda_g)].$$

$$I^{-}(\xi) = I_g[F - 4P \sin(\phi_P + 2\pi\xi \cos(\theta_g)/\Lambda_g)], \quad (12)$$

with F a fitting parameter. To summarize, using data like that in Figure 2, the values of A, ϕ_A, and I_g were determined by fitting the sum of the two traces to Eq. (11) with a nonlinear least-squares algorithm, and the values of P, ϕ_P, and F were determined by fitting the difference to Eq. (12). This procedure was repeated at each value of applied dc field. Direct inspection of the high-field data in Fig. 2 reveals that the intensities are 180° out of phase indicating the dominance of an index grating. Equation (12) for I^- shows that the trace should start out at $\xi = 0$ as a sine function; since the data start out as a cosine shape, ϕ_P is 90 degrees.

IV. RESULTS AND DISCUSSION

The index grating phase ϕ_A for data from several samples as a function of applied field along the grating wavevector is shown in Figure 3 (symbols). We see that at zero field, the phase of the index grating formed is near zero degrees (an in-phase grating). Since standard models of PRE give a 90° phase shift due to diffusion in zero field, this grating is not photorefractive in nature, but may be due to local processes such as photochromism.

We recall that in bisA-NPDA, the polymer is not fully crosslinked; thus the low glass transition temperature allows alignment of the nonlinear chromophores at room temperature by the applied field E_{dc}[3]. (For this reason, all the measurements reported in this paper were taken several minutes after any external electric field changes in order that the nonlinear chromophores might reach near-equilibrium orientation before writing a grating.) Since the electro-optic coefficient of the poled polymer approaches zero as the applied field goes to zero, any charge separation by diffusion (i.e., the PRE) that might be present would be extremely difficult to detect.

However, as the field is increased (and hence, r), the phase of the index grating increases until moderate to high fields (25 - 50 kV/cm), where it plateaus near 90°. This index grating shifted by 90° at high fields is certainly photorefractive in nature, because no moving gratings or frequency shifts were present during grating formation.

Figure 4 shows the absorption grating phase, ϕ_Λ, also as a function of the applied field along the grating wavevector. At zero field, we see that the in-phase index grating of Fig. 3 is accompanied by an in-phase absorption grating. At intermediate fields, competition between the two underlying grating formation mechanisms prevents detailed interpretation, but at high fields the phase of the absorption grating approaches 90°. This absorption grating at high fields may be the Kramers-Kronig companion of the strong index grating (see below).

In order to further understand the various contributions to the measured signals, we have plotted in Figure 5 the amplitudes of both the index grating P and the absorption grating A in the dimensionless units of Eq. (5). The amplitude of the absorption grating does not change with applied field, but the amplitude of the index grating is strongly field-dependent. We emphasize that the analysis technique of Section III assumed only one index grating and only one absorption grating; therefore, interpretations of the data will only be valid in that limit. At the highest fields, the index grating is dominant and is more than a factor of ten larger than the absorption grating, while at zero field the amplitudes are small and comparable. This with the behavior of the index grating phase in Fig. 3 provides evidence for the presence in our polymer of two distinct physical sources for gratings, one which produces a weak and in-phase grating at zero field, and one which leads to a strong phase-shifted index grating at high fields, i.e., the photorefractive mechanism. We note that in inorganic PRE materials, absorption gratings can also arise due to free-carrier effects [23] or to the presence of multiple absorbing levels[24]. Further work is necessary to conclusively identify the origin of the weak in-phase gratings in bisA-NPDA/DEH; however, a photochromic source is a reasonable possibility.

According to the standard PRE model [25, 26] the space charge field generated leads directly to an index grating. Further, the phase of the space charge field (and thus the phase of the index modulation) may be calculated using the complex form for E_{sc}:

$$\text{phase}(E_{sc}) = \text{phase}\left[\frac{m(E_{g,dc} + iE_d)}{1 + E_d/E_q - iE_{g,dc}/E_q} \right] \tag{13}$$

where $E_{g,dc} = E_{dc} \sin \theta_g$ is the component of the applied field along the grating vector, as indicated in Figure 1. Values for the diffusion field E_d = 1.0 kV/cm and the trap-limited space charge field E_q = 29.4kV/cm have been previously estimated[3]. Using these parameter values, Fig. 3 includes a plot of Eq. (13) (solid curve) along with the experimental data points. This curve shows that in the case of a purely photorefractive grating, the phase shift with no applied field should be 90°, then the phase shift departs from 90° as diffusion and drift compete, and finally the phase shift approaches 90° again in the high field limit. However, in a conventional photorefractive crystal, the electro-optic coefficient is not varying strongly with the applied field, as is the case with our poled polymer. Moreover, rather than being a fixed constant, the charge generation rate may increase strongly at higher fields due to Onsager geminate recombination[27]. Finally, Eq. (13) only strictly applies to the small m limit; corrections for m approaching 1 may be found in Ref. 28. Thus it is not surprising that the experimental points in Fig. 4 only roughly approach the PRE curve. The interpretation of our data at low fields where the photorefractive grating is weak is further complicated by the presence of the in-phase grating. It is clear that future models for the PRE in organic materials need modification to account for not only field-dependent generation and weak in-phase gratings, but also field-dependent mobility, even if the r-coefficient is locked in by a large glass transition temperature.

We can estimate values for Δn and $\Delta\alpha$ in our materials using Eq. 5. At high fields, P $\approx$ 0.01, which gives $\Delta n = 4.5 \times 10^{-6}$. At all fields, $\Lambda \approx 5 \times 10^{-5}$ which gives $\Delta\alpha = 2 \times 10^{-3} cm^{-1}$. While this Δn is not extremely large, one can easily expect improvements in Δn simply by using polymers with larger nonlinearity. Furthermore, the two-beam coupling experiment reported here also enables estimation of the gain coefficient Γ. We may

write in the usual fashion $\Gamma \approx (1/d) \ln(I_1/I_2)$. The data of Fig. 2 show that as the grating is formed, one transmitted beam increases in intensity and one decreases by approximately the same amount, $\simeq$ 1 %, so that $\Gamma \simeq 0.57\ cm^{-1}$. While this gain is not nearly as large as the values observed for inorganic crystals[6], it nevertheless would be expected to increase by a large factor when nonlinear polymers with higher r coefficients than bisA-NPDA are used, or when larger fields are used for poling. Nonlinear polymers with ten times larger r coefficients are becoming numerous; if some of these materials can be made photorefractive it is potentially possible that a photorefractive polymer with reasonable gain will be forthcoming.

These two-beam coupling studies of grating phase in the bisA-NPDA/DEH system show that at high fields, an index grating is dominant with phase shift near 90 °, thus providing final proof of the presence of the PRE in these materials. At low fields, the presence of a weak, in-phase grating suggests a local (perhaps photochromic) process is also operative. We suggest that grating phase measurements are useful in organic photorefractive materials not only for determining grating phase, but also for further elucidation of the grating formation mechanisms in these materials.

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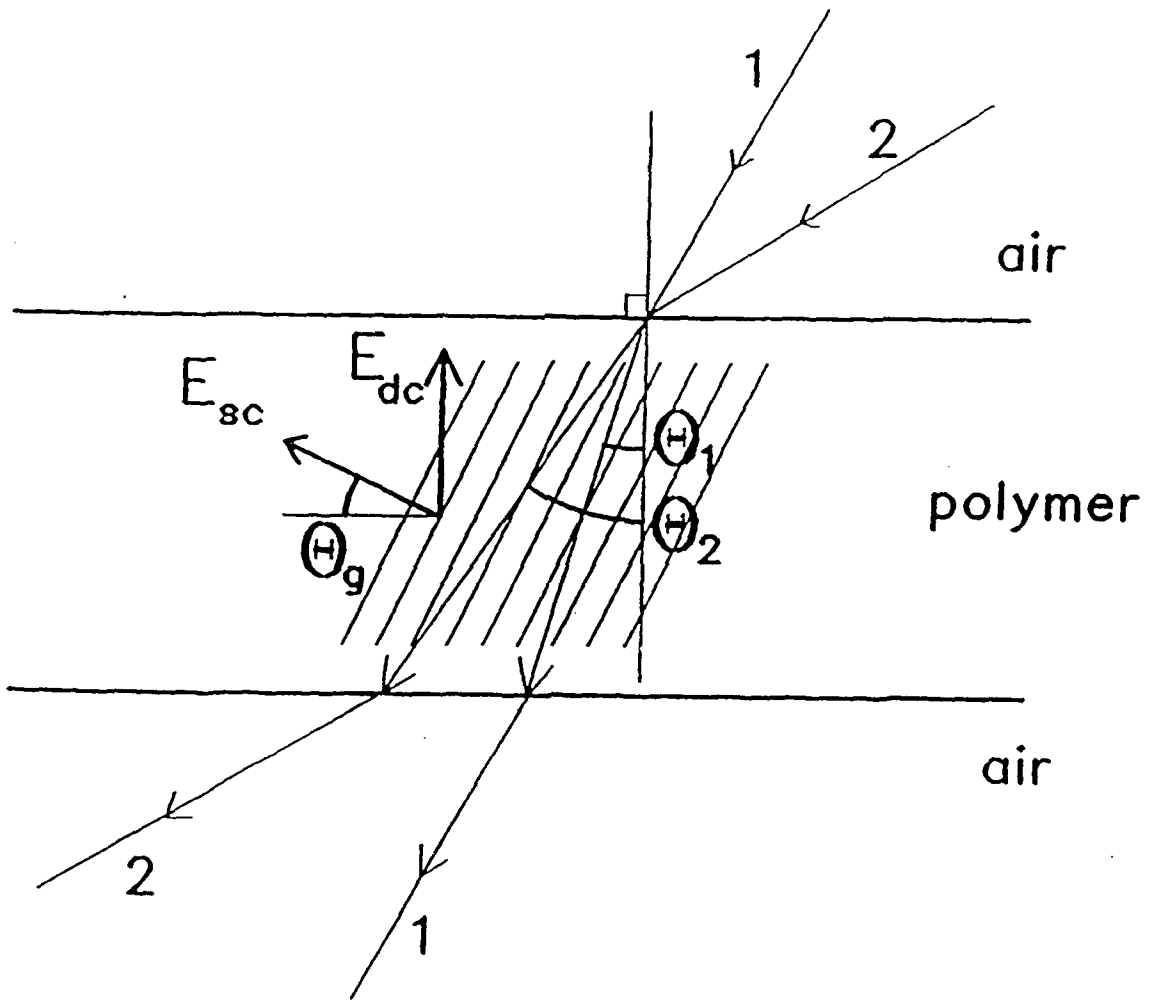

Figure 1. Geometry used for writing the grating and determining the grating phase using two-beam coupling. E_{dc} is the externally applied dc poling field, and the space charge E_{sc} is in the direction of the grating wavevector, K_g. The angles are defined in the text.

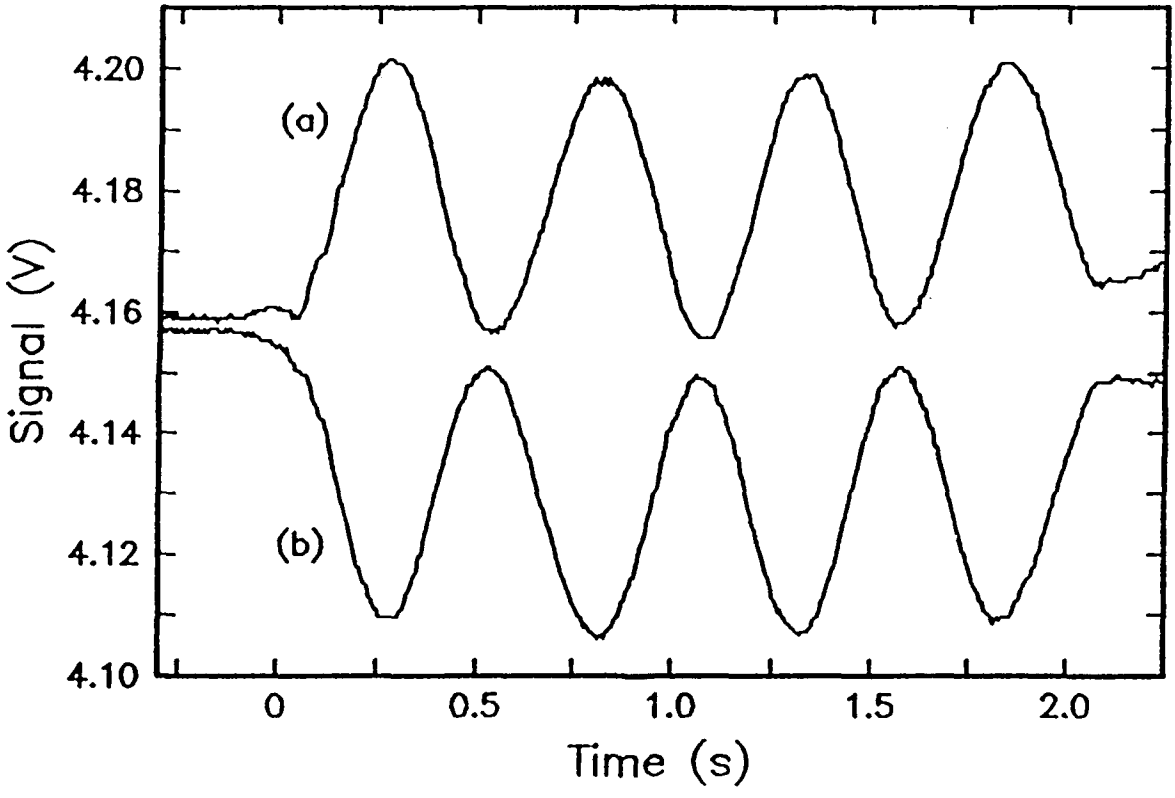

Figure 2. Transmitted intensities of beam 1 (a) and beam 2 (b) as the grating is translated. The rate of translation is 3.2 μm/s. Motion begins at time = 0 and stops at $\simeq$ 2.1 s. These data have been taken in the high field regime ($E_{g,dc}$ = 37 kV/cm) and the shape of the modulations indicate an index grating with 90° phase shift. Trace (b) has been offset downward by 0.02 V for clarity.