Appl. Phys. B 68, 907–910 (1999) / DOI 10.1007/s003409900032

Applied Physics B
Lasers
and Optics
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Transmission behavior in a photorefractive polymer film

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Received: 18 November 1998/Revised version: 10 February 1999/Published online: 12 April 1999

Abstract. When a *p*-polarized beam propagates through a high-performance photorefractive polymer composite, poly-(*N*-vinylcarbazole:2,4,7-trinitro-9-fluorenone:1-n-butoxyl-2,5-dimethyl-4-(4′-nitrophenylazo)benzene, its transmission behavior is influenced by three effects: the electroabsorption, the photorefractive coupling with the reflected beam from the rear surface, and the amplified scattering. From the measurements on the incidence angle dependence as well as the applied-electric-field dependence of the three effects, some conclusions are obtained. At a small incidence angle with a low applied electric field, both the amplified scattering and the electroabsorption are small whereas the coupling between the incident beam and the reflected beam plays a principal role. At a large incidence angle or with a high poling electric field, the transmission is influenced mainly by the amplified scattering and the electroabsorption. A poling electric field asymmetric loss to the amplification scattering is also observed.

PACS: 42.65.Hw; 42.70.Jk

In the first two years after the demonstration of the photorefractive (PR) effect in a polymer composite in 1991 [1], almost all the reported organic PR composites exhibited very low performance, for example, the two-beam coupling coefficient was even smaller than that of inorganic crystals [2–4]. This was attributed to the high glass transition temperature (T_g) of the samples, in which the PR effect was contributed only by the Pockels effect. A milestone improvement was the use of a plasticizer [5], which lowered the T_g of the composite to enable the electro-optic chromophore to reorient in situ at room temperature, and thus the orientationally enhanced PR effect dominated [6]. Till now, several PR polymer composites have be reported to exhibit a two-beam coupling coefficient Γ higher than 200 cm^{-1} and the gain index $\Gamma L \geq 2$ [7–9]. For such high-gain PR materials, an amplified scattering (beam fanning) effect often occurred [10], which possessed many important applications such as self-pumped phase conjugator [10, 11]. Recently, Grunnet-Jepsen et al. have observed an amplified scattering in a high-gain polymer composite of PVK:PDCST:BBP:C_{60} when a beam traveled through the sample [12]. And then a self-pumped phase conjugation was demonstrated [13]. In their work, the influence of the scattering amplification on the transmitted beam was investigated at a certain incidence angle. They observed a distinct electric-field asymmetric transmission and gave an explanation on amplification of the scattering [12]. More recently, another phenomenon about the intensity change of the transmitted beam has been reported by Meerholz et al. [14, 15] in a four-component PR composite PVK:DMNPAA:TNF:ECZ. Several PR gratings, including the PR gratings formed between two incident beams, between the incidences and their multiple reflected beams, and between the reflected beams were observed [14]. The applied-electric-field dependence of the competition between these PR gratings was also reported. Similar to the report in [12], when the diameters of the two incident beams became larger, the beam fanning effect was observed [15]. Further, for the applied-electric-field direction in which light could not be coupling to the polymer layer, the originally Gaussian-shaped interacting beams exhibited a shoulder or even split into two due to beam fanning. The field dependence of the beam fanning and the coupling of the two writing beams as well as the first-order non-Bragg self-diffraction showed a field asymmetry [15]. However, the incidence angle dependence of both the scattering amplification and multiple coupling have not been investigated in organic PR materials till now to our best knowledge.

In our previous report [9], a high-PR-performance three-component low-T_g composite, PVK:BDMNPAB:TNF, was reported to exhibit a two-beam coupling coefficient as high as 195 cm^{-1} at an applied electric field of 92.4 V/μm. In this report, the transmission behavior of a *p*-polarized beam traveling through this three-component PR composite is studied. Strong incidence angle dependence of transmission is observed. The different contributions from three effects, including a strong electroabsorption, coupling between the incident

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Dedicated to Prof. Dr. Eckard Krätzig on the occasion of his 60th birthday.

beam with its reflected beam and amplified scattering, at various incidence angles are studied.

1 Experiments and discussions

The PR composite film, consisting of poly(*N*-vinylcarbazole):2,4,7-trinitro-9-fluorenone:1-n-butoxyl-2,5-dimethyl-4-(4′-nitrophenylazo)benzene at a weight ratio of 55:1:44, was fabricated at a thickness of 100 μm by using spacers [9]. The experimental apparatus (involving the sample film configuration) is presented in Fig. 1. A *p*-polarized beam from a He-Ne laser at a wavelength of 633 nm with an intensity of 40 mW/cm^2 was incident upon the sample film at an incidence angle θ_{air} in air. As the diameter of the incident beam ($\approx$ 2 mm in air) is much larger than the thickness of the polymer layer (only 100 μm) and the glass piece (1 mm), all the reflected beams traveling through the sample can interact with the incident beam. As to our sandwiched sample, we could only detect the reflected beams from two air/glass surfaces. The reason for this might be the small differences between the refractive indices of the glass, the ITO, and the polymer. Thus, we consider only the reflected beam on the rear glass surface (beam 2 in Fig. 1).

We define the direction of the applied poling electric field **E** as follows. When the rear ITO electrode is the anode, the electric field inside the sample film is positive, i.e., $\mathbf{E} > 0$, as shown in Fig. 1. The measurement procedure is as follows. First, we detected the transmitted and reflected intensities without the applied electric field and set them as the units. Then the laser beam was blocked and a dc electric field was applied. After the EO chromophore was poled sufficiently the beam was turned on to pass through the sample, and the intensities of both the transmitted and reflected beams were measured.

Figure 2 shows the dynamic behaviors of both transmitted and reflected intensities carried out at $\theta_{air} = 22.5°$ with $|\mathbf{E}| = 72.6$ V/μm. For $\mathbf{E} > 0$, the intensity of the transmission has been increased obviously at the moment of tuning on the beam. This may be considered as the existence of a distinct electroabsorption because there are not any PR gratings at this moment. Then, the intensity decreased as a typical dynamic two-beam coupling behavior, while the reflected intensity increased. Because the transmitted intensity ($\approx$ 600 μW) is

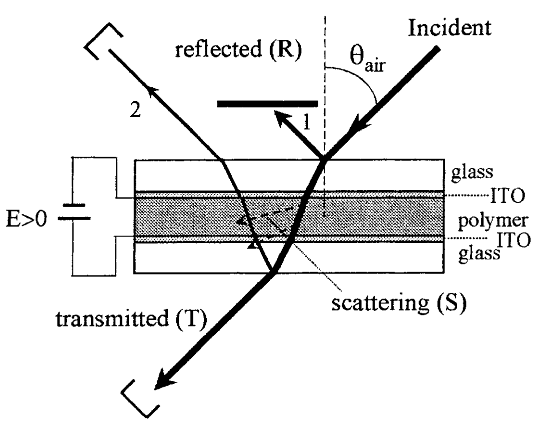

Fig. 1. The experimental setup for the measurements of the transmission behavior

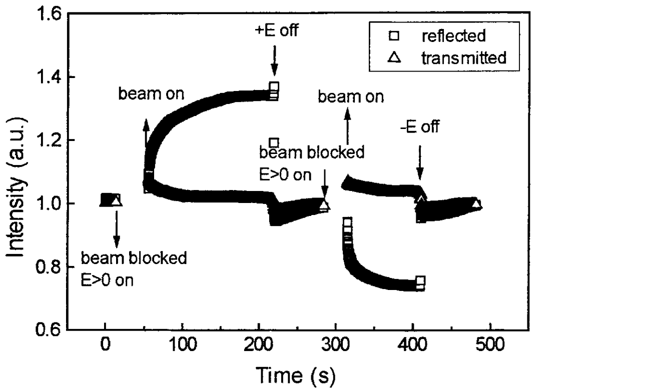

Fig. 2. The measurement procedure at $\theta_{air} = 22.5°$ and $|\mathbf{E}| = 72.6$ V/μm. Details: see the descriptions in the text

much larger than the reflected intensity ($\approx$ 10 μW), it can be considered as an undepleted-pump process in their coupling. From Fig. 2, however, it is obvious that the decrement of the transmitted intensity is much larger than what the reflected beam gained. The explanation for this may be the appearance of the scattering amplification. This is confirmed further from the experimental results for $\mathbf{E} < 0$. If there exists only the coupling between the reflected and the transmitted beams, for example, no amplified scattering, the transmission should be increased because the energy coupling direction is reverse for $\mathbf{E} < 0$. However, from the results presented in Fig. 2, the transmission decreased even when the reflected intensity decreased for $\mathbf{E} < 0$. The different decrements at $\mathbf{E} > 0$ and $\mathbf{E} < 0$ suggest a field-asymmetric amplified scattering, as pointed out by Grunnet-Jepsen et al. [12] and Meeerholz et al. [15]. To lower the amplitude of the scattering PR grating formed by the scattering beam with laser beams inside the polymer, and thus decrease the loss of the transmitted beam to the scattering, we introduced an erasure beam with *s*-polarization from another He-Ne laser in the direction of the normal of the sample surface. The beam diameter is 4 mm and the intensity is 30 mW/cm^2. Increments on both the reflection and transmission were observed as shown in Fig. 3. This implies that both the reflected and the transmitted beams con-

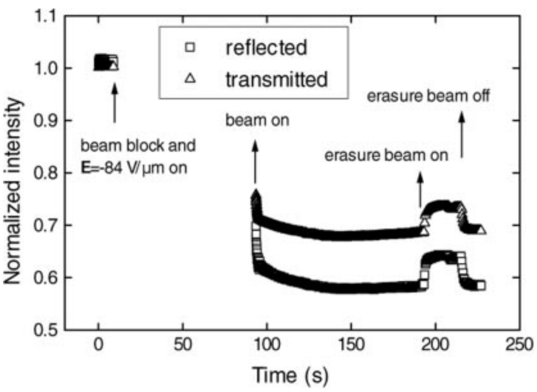

Fig. 3. Experimental results with and without the erasure beam at $\theta_{air} = 67°$ and $\mathbf{E} = -84$ V/μm. When both intensities were stabilized, opening the erasure beam to increase the modulation degree of the noise grating and thus decrease the amplification of the scattering, intensities of both the reflected and the transmitted beams increased. This result confirmed the existence of the strong amplified scattering

tributed their energies to the amplified scattering by forming the scattering PR grating with the scattering beam.

As mentioned above, a distinct electroabsorption exists in our sample [16]. An electroabsorption has been observed in PVK:TNF complex [17], however, this could not be the only cause of such an extremely strong electroabsorption. Meanwhile, a strong incidence angle dependence was observed as shown in Fig. 4a. No obvious difference is observed for $\mathbf{E} > 0$ and $\mathbf{E} < 0$. At $|\mathbf{E}| = 84\,\mathrm{V/\mu m}$ and $\theta_{\mathrm{air}} = 78.7°$, an electroabsorption coefficient as high as $\approx 25\,\mathrm{cm}^{-1}$ was detected. The applied-electric-field dependence of $\Delta\alpha$ at $\theta_{\mathrm{air}} = 22.5°$ was also measured and is shown Fig. 4b. A theoretical fit suggests that there exist a linear- and a quadratic-electroabsorption simultaneously. However, the physical mechanism of such an electroabsorption behavior is unclear yet. The possibility is the reorientation of the chromophore under the applied electric field [15].

Apart from the electroabsorption, the change of the reflected intensity is a result of the couplings with the incident beam and the scattering beam. From the standard photorefractive model, its gain can be calculated by [7, 9]

$$\Gamma^{(R,T)}(E) = \frac{\cos\theta}{d} \times \ln\left(\frac{R(E, t=\tau)R(E)}{R^2(E, t=\tau) + R(E, t=\tau)T(E, t=\tau) - R(E, t=\tau)R(E)}\right), \tag{1}$$

where R and T are reflected and transmitted intensities, $t = \tau$ represents the moment when the laser beam was turned on (no beam interaction but the electroabsorption occurred), θ is the incident-angle inside the polymer, and d the polymer film thickness.

At $|\mathbf{E}| = 84\,\mathrm{V/\mu m}$, we measured the incidence angle dependence of the gain of the reflected beam, which was calculated from (1). The results are represented in Fig. 5a. As θ_{air} increases, a decreasing trend for $|\Gamma|$ is observed for both cases of $\mathbf{E} > 0$ and $\mathbf{E} < 0$. This is reasonable because a larger θ_{air} corresponds to a smaller grating wave-vector and smaller effective interaction volume. Meanwhile, a field asymmetry also appeared as shown in the figure. At $\mathbf{E} > 0$, Γ decreased quickly and then entered the negative region. At $\mathbf{E} < 0$, $|\Gamma|$ decreased slowly and then seemed to increase again. This confirms that the reflected beam loses its energy through amplifying the scattering too. The loss to the amplified scattering increased with the increasing of θ_{air}. Furthermore, the field dependence shown in Fig. 5b is not in agreement with the prediction of $\Gamma \propto |\mathbf{E}|^2$ from the PR theory [9], which should be attributed to higher loss to the amplified scattering at higher applied-electric-field. As pointed out by Meerholz et al. [15], Γ has lost its original physical meaning due to the amplified scattering.

As the transmitted intensity is much higher than that of the reflected beam, as mentioned above, almost any observable loss of the transmitted energy should be attributed to the amplified scattering, which has been confirmed in Fig. 3. To describe the energy loss by amplifying the scattering, we define a gain coefficient G of the transmission by

$$T(E) = E(E, t=\tau)\exp(Gd/\cos\theta)\,. \tag{2}$$

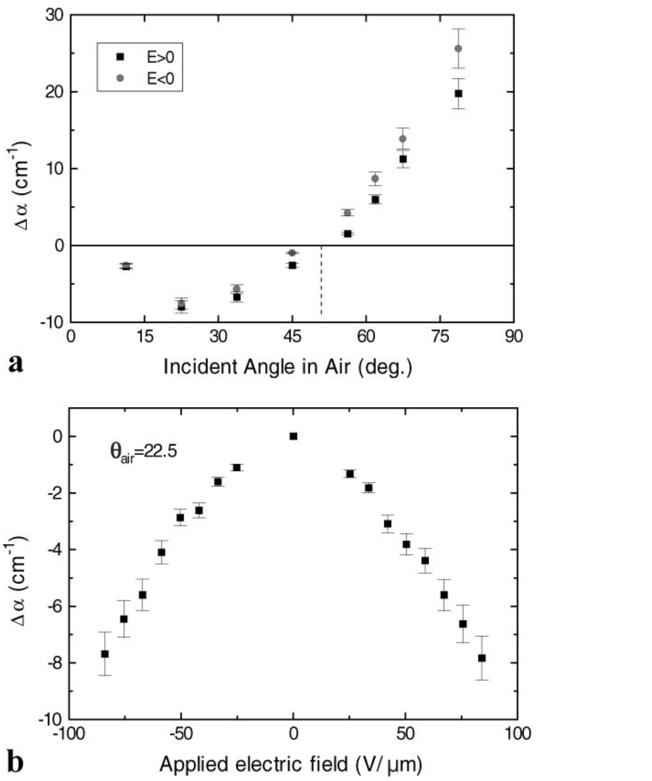

Fig. 4a,b. The electroabsorption coefficient as a function of **a** the incidence angle at $|\mathbf{E}| = 84\,\mathrm{V/\mu m}$; **b** the applied electric field at $\theta_{\mathrm{air}} = 22.5°$

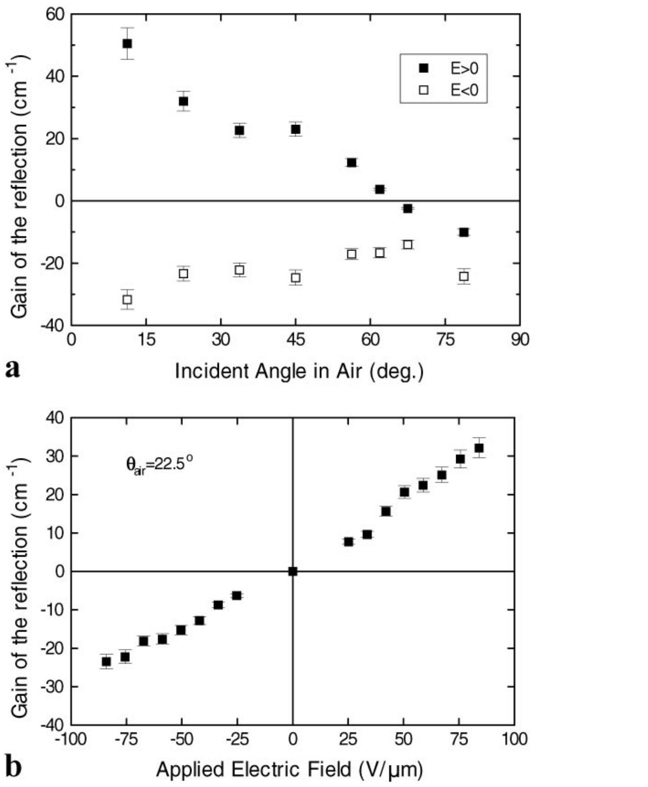

Fig. 5a,b. Gain of the reflected beam as a function of **a** the incidence angle at $|\mathbf{E}| = 84\,\mathrm{V/\mu m}$; **b** the applied electric field at $\theta_{\mathrm{air}} = 22.5°$

Using the parameter of gain G defined, we measured this loss as functions of both θ_{air} and E. Results are shown in Fig. 6a,b, respectively. The amplified scattering appears in only one side where energy transfers from incident beam to scattering beam due to the PR nature of directional coupling. In our configuration shown in Fig. 1, the left-hand scattering is amplified and the right-hand is forbidden at $\mathbf{E} > 0$, whereas only the right-hand is amplified at $\mathbf{E} < 0$. Figure 6a indicates a strong field asymmetry. This suggests the amplified scattering at $\mathbf{E} > 0$ is much stronger than that at $\mathbf{E} < 0$, which means that the coupling coefficient between the incident beam and its left-hand scattering beam is larger than that between the incident- and its right-hand scattering beams. This is reasonably attributed to the differences of the effective electro-optic coefficient, the amplitude of the wave-vector, and the component of the applied electric field in the wave-vector direction of the scattering PR grating for the two cases. This asymmetry is decreased or even reversed by increasing the incidence angle θ_{air}, as shown in Fig. 6b.

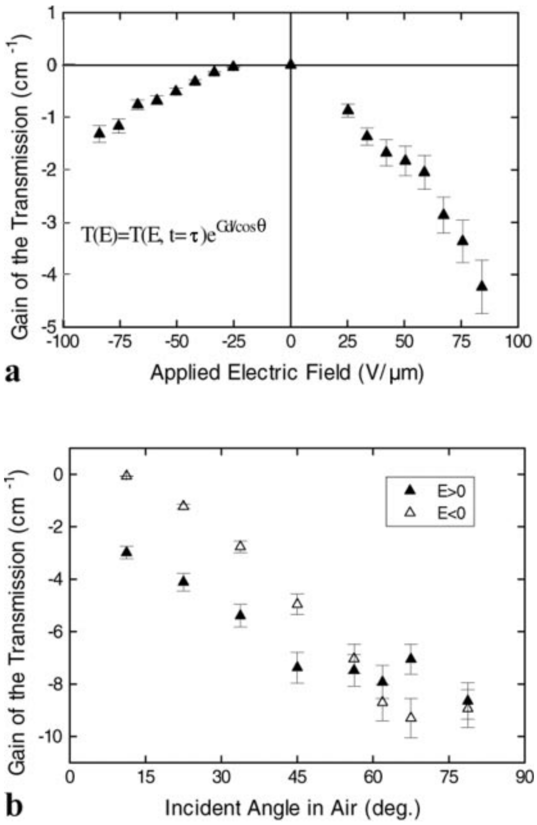

Fig. 6a,b. Gain of the transmitted beam as a function of **a** the applied electric field at $\theta_{air} = 22.5°$; **b** the incidence angle at $|\mathbf{E}| = 84\ \mathrm{V/\mu m}$. The gain was mainly caused by the amplified scattering

2 Conclusions

The influence of three effects, including electroabsorption, coupling with the reflected beam, and loss to the amplified scattering, on the transmission character of a p-polarized beam passing through a sandwiched three-component PR polymer composite film was observed. At a small incidence angle with a low applied electric field, the loss of the transmission is mainly due to the two-beam coupling between the incident beam and its reflected beam from the rear surface. For a large incidence angle or with a high poling electric field, the loss to the amplified scattering and electroabsorption become the dominant. All the electroabsorption, coupling with its reflected beam and with the scattering beams, are strongly incidence angle and applied-electric-field dependent. The asymmetry of the amplified scattering can be reversed by changing the incidence angle.

Acknowledgements. F.W. is grateful to Miss H. Yang for her assistance. This work was supported by the Science Foundation of Post-Doctor of China and NNSFC through grants No. 1950412.

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