Appl. Phys. B 57, 199–201 (1993)

Applied Physics B Photophysics and Laser Chemistry

Thermally Induced Optical Bistability in a New Polymeric Blend at Room Temperature

U. Bernini [1], L. De Stefano [2], P. Mormile [2], G. Pierattini [2], P. Russo [1]

[1] Dipartimento di Scienze Fisiche, Universita' di Napoli Federico II, Pad. 20 Mostra d'Oltremare, I-80125 Napoli, Italy
[2] Istituto di Cibernetica del C.N.R., Via Toiano 6, I-80072 Arco Felice (Napoli), Italy (Fax: +39-81/5267654)

Received 4 May 1993/Accepted 2 June 1993

Abstract. The transition from the transmission to the reflection regime for an Ar^+-laser beam propagating in the new polymeric blend PMMA–EVA at a nonlinear interface has been observed. A comparison between the experimental data and a calculation of the input optical intensity at which this transition should occur (1.45×10^7 W m^{-2}) is presented using Kaplan's theory. The results suggest the presence of thermally induced optical bistability in PMMA–EVA.

PACS: 68.35, 61.40, 42.70

The reflection of light at the interface between two dielectrics, one of which is linear and the other one has an intensity-dependent index of refraction (so-called nonlinear interface), is still the subject of a great deal of research activity for its potential applications (optical switch, bistable operation, or logic element) in the field of nonlinear optics [1]. It is well known that the optical bistability phenomenon is produced by the combination of a feedback process and a nonlinearity of the sample. In the case of light propagation at a nonlinear interface such a feedback mechanism is due to the change of the intensity-dependent refractive index when increasing the optical input power, which determines the switch of the incident wave from the transmission to the reflection regime. In this case, a bistable behaviour of the transmitted wave occurs at a glancing angle θ_g. All possible cases of transmission and reflection regimes for media with either negative or positive nonlinearities have been investigated [2, 3]. In this paper we present experimental results on the propagation of a laser beam through a nonlinear interface and report the results of a numerical analysis that confirms the experimental data. The interface considered is that between PMMA and PMMA–EVA. The first one is regarded as a linear dielectric whereas PMMA–EVA has nonlinear thermal properties.

In a related paper [4], we proposed a poly(methylmethacrylate) (PMMA) with 7 wt.% of poly(ethylene-co-vinylacetate) (EVA) blend (PMMA–EVA) as novel material for application in nonlinear optics of glassy polymers [5–8]. This polymeric blend consists of a finely dispersed rubbery phase in a glass PMMA matrix. It is characterized by an interpenetrated network morphology, with very small rubbery particles, and at the same time it retains a large part of the optical transparency of PMMA [9]. The microstructure which develops at the end of the fabrication process is a multicore-shell structure. Recently [10], we measured the thermal coefficient of the refractive index dn/dt of PMMA–EVA. Its value was so large ($dn/dt = -1.41 \times 10^{-4}\,^\circ$C^{-1}, at 30° C) and temperature dependent so as to suggest a nonlinear thermal behaviour of PMMA–EVA.

1 Theoretical Description

Let us consider a TE polarized plane wave incident at the interface between a linear medium and a nonlinear medium with refractive indices n_1 and n_2, respectively. The incident wave can be written as:

$$E_\mathrm{I}(x,z) = E_0\,\mathrm{e}^{\mathrm{i}k_0\beta x}\,\mathrm{e}^{\mathrm{i}k_0\gamma_1 z}\,,$$

where $\gamma_1 = (\varepsilon_1 - \beta^2)^{1/2}$ and $\beta = n_1 \sin\Theta_1$,

whereas the amplitude of the transmitted and reflected waves are, respectively,

$$E_\mathrm{T}(x,z) = t_{12}E_0\,\mathrm{e}^{\mathrm{i}k_0\beta x}\,\mathrm{e}^{\mathrm{i}\gamma_2 k_0 z}\,,$$
$$E_\mathrm{R}(x,z) = r_{12}E_0\,\mathrm{e}^{\mathrm{i}k_0\beta x}\,\mathrm{e}^{-\mathrm{i}\gamma_1 k_0 z}\,,$$

where $\gamma_2 = (\varepsilon_2 - \beta^2)^{1/2}$ and t_{12}, r_{12} are the Fresnel coefficients for the nonlinear interface. For a TE wave $t_{12} = 2\gamma_1/(\gamma_1+\gamma_2) = |E_\mathrm{T}|^2/|E_\mathrm{I}|^2$, and $r_{12} = (\gamma_1-\gamma_2)/(\gamma_1+\gamma_2)$.

In our case n_2 is not a constant but is a function of $|E_\mathrm{T}|^2$, which gives rise to an increase of the blend temperature. The refractive index may be written as:

$$n_2 = n_0 + n_\mathrm{NL}|E_\mathrm{T}|^2 = n_0 + (dn/dt)_p\langle T\rangle\,,$$

where $\langle T\rangle$, averaged along the beam path, is calculated from the heat equation near the beam axis, assuming that the beam intensity varies for successive little steps so that thermal equilibrium holds in the sample. This temperature is a function of the thermal parameters of the PMMA–EVA blend, i.e. its diffusivity $D = 1.14 \times 10^{-7}\,\mathrm{m^2\,s^{-1}}$ and $k = 1.93 \times 10^{-1}\,\mathrm{W\,m^{-1}\,K^{-1}}$ is its thermal conductivity.

It can be demonstrated that the equation for $\langle T\rangle$ predicts initially a linear rise of the temperature near the beam axis, whereas for times $t > t_D = a^2/4D$ ($a = 1$ cm is the boundary of the medium), thermal equilibrium is attained. In this case the equilibrium of the temperature T_e is proportional to the incident pump power [11]:

$$T_e = \frac{P_0}{4\pi k d}\,[1 - \exp(-d\alpha)]\ln\left(\frac{2\gamma a^2}{w^2}\right),$$

where $P_0 = \pi w^2 I_0/2$, the beam waist $w = 100\,\mu$m, I_0 is the incident intensity of the laser beam, $\alpha = 0.9\,\mathrm{cm^{-1}}$ is the measured total attenuation coefficient of the blend at 514.5 nm, $d = 0.98$ mm is the sample thickness and $\gamma = 1.781$. We can easily obtain equations for E_R and E_I by writing $E_R = E_T r_{12}/t_{12}$ and $E_I = E_T/t_{12}$. The transmitted field E_T is totally reflected at the second boundary and then refracted at the first one. Taking into account the optical path of the laser beam, with the incident and refractive angles and the Fresnel coefficients, $\langle T\rangle, n_2, r_{12}$ and t_{12} are determined, for a given value of $|E_T|^2$. Consequently, $T = |t_{12}|^2, R = |r_{12}|^2$ and $|E_I|^2 = |E_T|^2/|t_{12}|^2$ are calculated. The transmittivity T versus the input intensity is obtained from the last equation (see below).

2 Experimental Results

In Fig. 1 a schematic diagram of the experimental setup is shown. A Pockels cell is used to modulate the power of a cw Ar^+-laser beam ($\lambda = 514.5$ nm) between 0 mW and 300 mW. A beam-splitter directs a fraction of the input power onto a photodiode. The transmitted power is measured by another photodiode. The signals from both detectors are plotted on an X-Y recorder to obtain the input–output characteristic. The light beam propagates through a prism of PMMA (the linear medium, with refractive index $n_1 = 1.497$ at $\lambda = 514.5$ nm) and is focused by a lens (spot $= 10^4\,\mu\mathrm{m}^2$) at a fixed angle on the boundary of a thin (0.98 mm) slab of PMMA–EVA (the nonlinear medium).

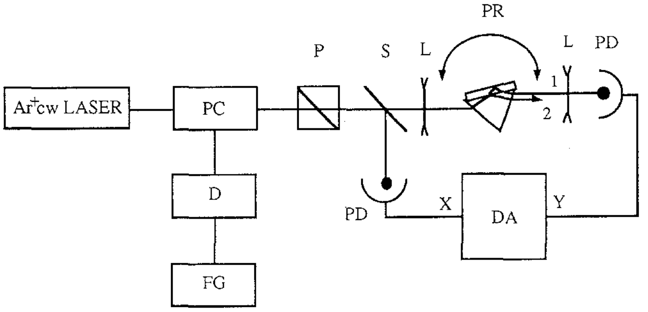

Fig. 1. Experimental arrangement for measuring output power vs input power; PC = Pockels cell, D = driver, FG = function generator, P = polarizer, S = beam splitter, L = lens, PD = photodiode, PR = rotating prism, DA = data acquisition

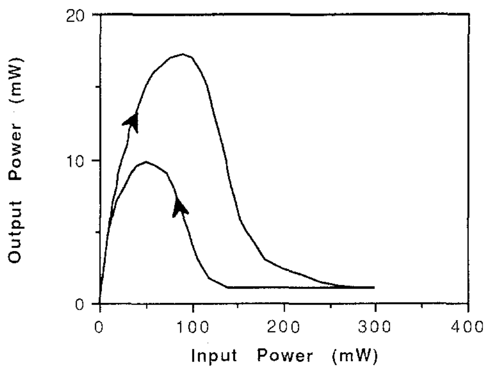

Fig. 2. Measured output vs input power for PMMA/PMMA–EVA interface

The refractive index of PMMA–EVA at constant pressure p is

$$n_2 = n_0(T = 20°\,\mathrm{C}, \lambda = 514.5\,\mathrm{nm}) + (dn/dt)_p\langle T\rangle$$
$$= 1.499 - 1.41 \times 10^{-4}\,°\mathrm{C}^{-1}\langle T\rangle\,.$$

Switching the beam on, initially $n_2 > n_1$: the beam is partially transmitted into the nonlinear medium at the interface, reflected back and then collected by the photodetector (Fig. 1, beam 1). Beam 2 is stopped. As the incident power increases, $\langle T\rangle$ increases and the effective refractive index n_2 decreases, giving rise to a decrease of the collected beam 1 intensity. The threshold for the switching-off of the output power occurs at about 100 mW. Then, by decreasing the input power the switching-on of the output power occurs at a lower power value, giving rise to an hysteresis cycle (optical bistability) (Fig. 2). In all measurements the rate of variation of the incident power was about $1\,\mathrm{mW\,s^{-1}}$. Such a low rate allows for the settling of thermodynamical equilibrium in the sample at all levels of input power.

3 Conclusion

The theoretical curves of the transmittivity and reflectivity as a function of the incident intensity, obtained for a 57° incidence angle, show optical bistability as predicted by Kaplan's theory, in the case of light propagation at a nonlinear interface. Here, we report the results (Fig. 3) of our numerical analysis starting from such a theory, considering only the case of a negative nonlinearity. For such numerical elaboration we assumed the thermal coefficient of the refractive index of the blend to be dependent on the incident light intensity as reported before, utilizing the thermo-optical parameters of the polymeric sample previously measured. In Fig. 4 we report a plot of experimental results refered to the transmittivity versus incident intensity for an incidence angle of 57°. The data well agree with theory. The threshold value of the incident light intensity (I_{crit}) at which there is the jump from the transmission regime to the reflection is about $1.15 \times 10^7\,\mathrm{W\,m^{-2}}$. The agreement with the value theoretically calculated ($1.45 \times 10^7\,\mathrm{W\,m^{-2}}$) is good. In conclusion our measurements suggest the existence of optical bistability at the nonlinear interface between PMMA and PMMA–EVA when sweeping the input intensity through the

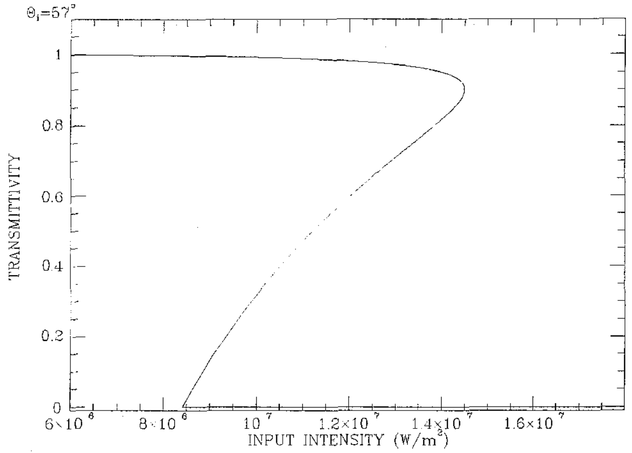

Fig. 3. Theoretical curve of the transmittivity as a function of input intensity for an incidence angle $\theta = 57°$

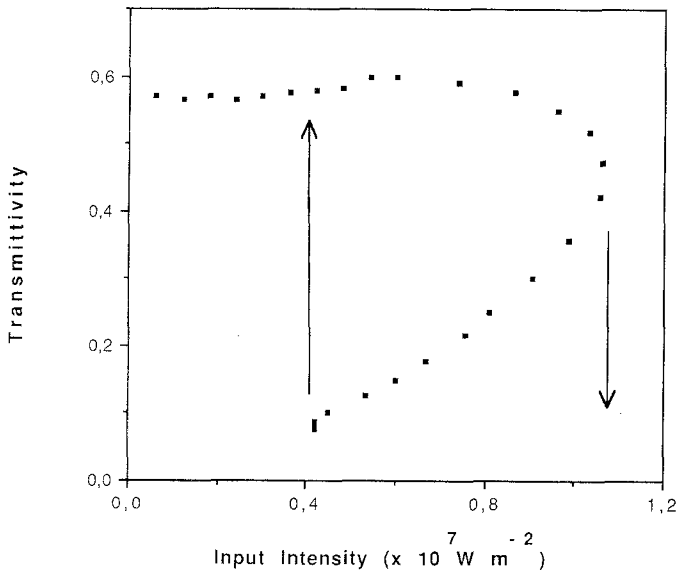

Fig. 4. Experimental input–output characteristics of the transmittivity at $\theta = 57°$

threshold value which determines the change of the refractive index of PMMA–EVA. However, it is to remark that the low thermal conductivity of PMMA–EVA blend leads to a substantial heating of the sample even at low input intensities; that is, the mechanism of the nonlinearity in our case is thermal. Therefore the employment of some polymeric materials, such as PMMA–EVA blend is limited to those optical devices where the temperature change plays a dominant role in the thermally induced optical bistability effect.

Acknowledgements. The authors wish to thank Mr. S. Piscitelli and E. Casale for their technical assistance. This work was partially supported by a grant from C.N.R., "Progetto Finalizzato Tecnologie Elettroottiche".

References

1. I.C. Khoo, Ping Zhou: Nonlinear interface tunneling phase shift. Opt. Lett. **17**, (1992)
2. V.S. Butylkin, A.E. Kaplan, Yu.G. Khronopulo, E.I. Yakubovich: *Resonant Nonlinear Interactions of Light with Matter* (Springer, Berlin, Heidelberg 1989) pp. 301
3. A.E. Kaplan: Theory of plane wave reflection and refraction by the nonlinear interface, in *Optical Bistability*, ed. by C.M. Bowden, M. Ciftan, H.R. Robl (Plenum, New York 1991)
4. U. Bernini, G. Carbonara, M. Malinconico, P. Mormile, P. Russo, M.G. Volpe: Investigation on the optothermal properties of a new polymeric blend: polymethylmethacrylate-polyethylene-co-vinylacetate (PMMA–EVA). Appl. Opt. **31**, (1992)
5. C. Noel, C. Friedrich: In *Proc. Europ. Symp. on Polymeric Materials*, Lyon, France, (1987)
6. D.J. Williams (ed.): *Nonlinear Optical Properties of Organic and Polymeric Materials*, Symp. Ser., Vol. 233 (ACS, Washington, DC 1983)
7. M. Sorm, L. Dvoranek, L. Machova, J. Svantner, V. Kubanek: In *Proc. Europ. Symp. on Polymeric Materials*, Lyon, France (1987)
8. J.E. Sohn, K.D. Singer, M.G. Kuzyk, H.E. Katz, C.W. Dirk: In *Proc. Europ. Symp. on Polymeric Materials*, Lyon, France, (1987)
9. P. Laurienzo, M. Malinconico, G. Ragosta, M.G. Volpe: Angew. Makromol. Chem. **170**, 137 (1989)
10. G. Carbonara, P. Mormile, M. Malinconico, M.G. Volpe, G. Abbate, U. Bernini, P. Maddalena: The measurement of the thermal coefficient of the refractive index of a new high performance polymeric blend, in IUPAC, Int'l Symp. Speciality Polymers, Singapore (1990)
11. G. Abbate, U. Bernini, P. Maddalena, S. De Nicola, P. Mormile, G. Pierattini: Interferometric technique for the thermal nonlinearities in semiconductor glasses. Opt. Commun. **70**, 15 (1989)