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Comportamiento no lineal óptico de una película fotográfica revelada y blanqueada al caracterizarla bajo la técnica de barrido en z (z-scan)

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RESUMEN
Se aplicó la técnica de barrido para caracterizar una película fotográfica revelada y blanqueada, utilizando laseres de onda continua como el HeNe y Ar. Se observa de las curvas obtenidas que los cambios en el índice de refracción pueden invertirse o permanecer dependiendo de la intensidad, longitud de onda y tiempo de exposición del láser sobre la película, dando una nueva aplicación a este material. Se desarrollo un modelo basándose en la propagación de haces gaussianos para ajustar las curvas experimentales obteniéndose que, no importando las condiciones experimentales, la no linealidad óptica exhibida por la película fotográfica no es del tipo térmico.

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Palabras clave: óptica no lineal, técnica de barrido en z, película fotográfica.

1. INTRODUCTION

Photographic media are photosensitive emulsions based on silver halides. An unexposed photographic film or plate generally consist of a multitude of tiny silver halide (often AgBr) grains suspended in a gelatin support, which is in turn attached to a base consisting typically of acetate for films and glass for plates. When the photosensitive material is exposed to light, the silver halide grains absorb optical energy and undergo a complex physical change. Those grains that have absorbed a sufficient amount of energy are found to contain tiny patches of metallic silver. The exposed film is then subjected to a chemical process called development, during which the exposed zones change to metallic silver. The grains that did not exposed do not undergo such a change. After this the film is fixed by subjecting it to chemical processing that removes the remaining silver halide grains while living the metallic silver. The silver grains are opaque at optical frequencies. Thus, conventional photographic emulsions are used to modulate light primarily through absorption. As a consequence, significant amounts of light are lost. In many applications it is desired to have the photographic film as a phase modulator. Such structures can be realized by chemical bleaching. The bleaching process is one that removes metallic silver from the emulsion and leaves in its place either an emulsion thickness variation or a refractive index variation within the emulsion. A thickness variation results when tanning bleach is used, while a refractive index modulation occurs when nontanning bleach is used. In our case tanning bleach was used. Considering this type of bleach the chemical agents used release certain chemical byproducts as they remove the metallic silver, and these byproducts cause a cross-linking of the gelatin molecules within the emulsion in regions where the silver concentration was high. As the transparency is dried, the hardened areas shrink less than do the unhardened areas, with the result that a relief image is formed, with the thickest regions of the emulsion being where the density was highest, and the thinnest regions where the density was lowest.

Characterization of the nonlinear optical response of a media can be done by many different techniques: degenerate four wave mixing[1] (DFWM), nearly degenerated four and three wave mixing[2], Z-scan[3], nonlinear interferometry[4], ellipse rotation[5], optical third harmonic generation[6], beam distortion measurements[7] and photo-acoustic[8]. Each technique present advantages and disadvantages both in the experimental implementation and in the sensitivity.

The Z-scan technique provides a sensitive and straightforward method for determination of the sign and both real and imaginary part of the nonlinear refractive index of optical materials. The simplicity of both the experimental setup and the data analysis has allowed the Z-scan technique to become widely used. It is based on self-focusing and self-defocusing of an optical beam by a nonlinear sample. In this technique the sample is scanned along the optical axis (*Z*-direction) in the focal region of a single focused laser beam; the transmitted intensity is recorded in the far field by a photodetector with a small aperture. This curve exhibits a prefocus minima (valley) followed by a postfocus maxima (peak), the opposite behavior is expected for a negative nonlinearity. Extensions of the basic technique to allow the use non-Gaussian-beam profiles, thick samples saturable Kerr media or thermal media have been proposed, and certain improvements in sensitivity have been achieved by measuring the total beam profile distortion or through the eclipsing Z-scan method.

The Z-scan technique can be considered an extension of similar experimental technique based on thermal lens effect which was proposed and used for high sensitivity measurements of low optical absorption earlier[9,10]. The model obtained for this technique uses the Gaussian decomposition (GD) method, given by Weaire et al. [11], in which the complex electric field at the exit plane of the sample is decomposed into a sum of Gaussian beams through a Taylor series expansion of the phase term induced by the nonlinear sample with a third or fifth order nonlinearity. The theory allows to obtain an analytical equation for the normalized transmittance for the case of a sample with a cubic nonlinearity and small phase change.

Usually the photographic film after developing and bleaching can not be considered as photosensitive. In this paper we demonstrate that the bleached photographic film present nonlinear response to cw illumination obtained from low power lasers as Argon and He-Ne. The response is characterized using the Z-scan technique. The type of nonlinearity was considered thermal and a model based on Gaussian beam propagation was used to fit the experimental results.

2. EXPERIMENTAL RESULTS

The material used was a Kodak high resolution photoplotter glass plate model 180 RX. This type of film is used to create phototools for precision microelectronics. The film uses fine grain and has a film thickness of 6 μm before processing and 4 μm after developing. The absorption spectrum of the sample used is shown in figure 1.

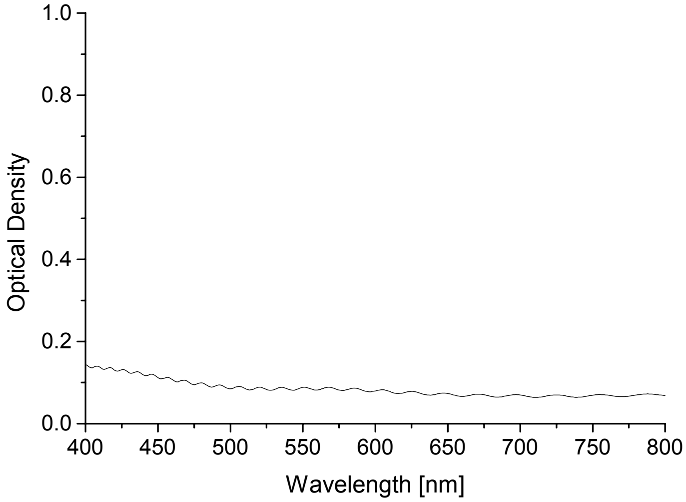


Figure 1. Absorption spectrum of the photographic plate after bleaching.

First experiments were made using a multiline 6 W Argon laser and a 6 cm focal length lens to focus the beam. The photodetector was set to 85 cm from the lens. Under these conditions we can say that we were in the thin sample limit with the photodetector in the far field. The beam was focused to a minimum spot of 15 μm and the total scan was set to 28 mm. The wavelength of the laser was tuned using a prism and the laser power was changed using neutral density filters. We choosen the most intense lines of the laser to characterize the photographic film: 457, 476, 496 and 514 nm. The minimum power used to obtain the Z-scan was 1 mW, after the curve was obtained the power was increased to 5 mW and the maximum incident power used was 10 mW. To observe if some changes were produced in the sample, new Z-scan measurements were made reducing the power to 5 mW and then to 1 mW without changing the sample position. The behavior of the z-scan experiments for the different powers and wavelengths is shown from figures 2 to 5.

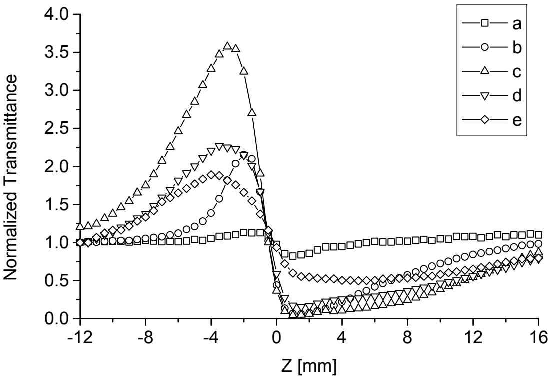


Figure 2. Z-scan at 457 nm for incident power of: a) 1, b) 5, c) 10, d) 5 and e) 1 mW.

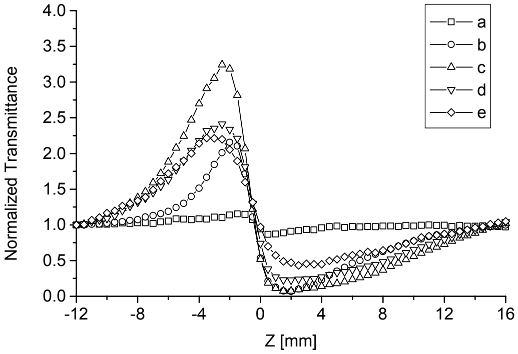
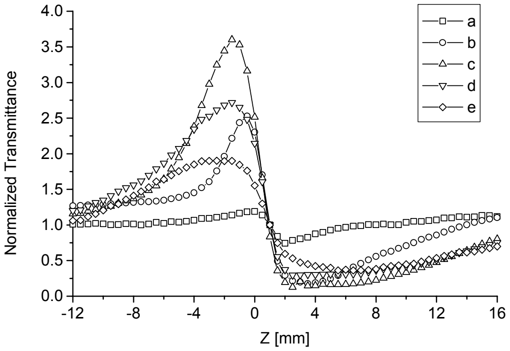


Figure 3. Z-scan at 476 nm for incident power of : a) 1, b) 5, c) 10, d) 5 and e) 1 mW.



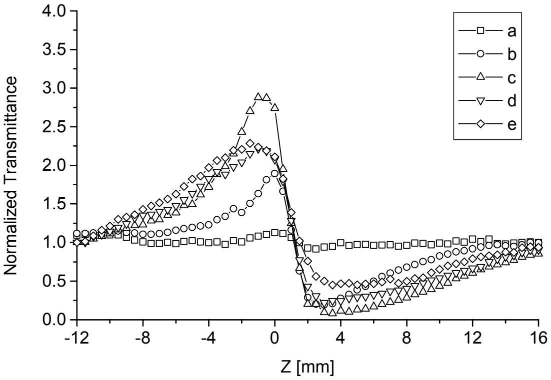


Figure 4. Z-scan at 496 nm for incident power of:
a) 1, b) 5, c) 10, d) 5 and e) 1 mW.
Figure 5. Z-scan at 514 nm for incident power of:
a) 1, b) 5, c) 10, d) 5 and e) 1 mW.

Finally to check the response in the red region of the visible spectrum we use a He-He laser beam of 10 mW with a beam diameter of 0.8 mm. This beam was focused by the same lens used with the Argon laser. The minimum spot size was around to 30 µm. The power was changed in the same way than in the previous experiments obtaining the result shown in figure 6.

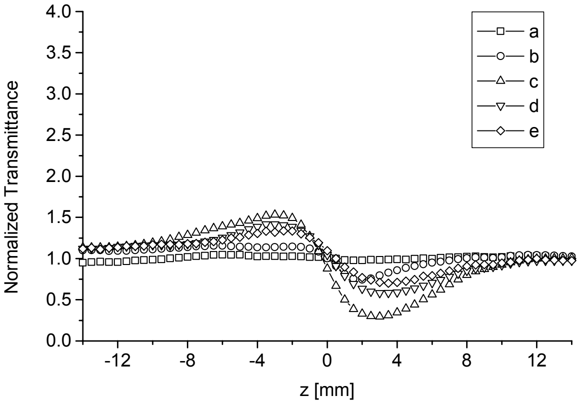


Figure 6. Z-scan at 633 nm for incident power of: a) 1, b) 5, c) 10, d) 5 and e) 1 mW.

From the all shapes of the curves, a prefocus peak followed by a postfocus valley, we can say that the sample behaves as a negative nonlinear media. Note that not necessarily the normalized intensity far from the focus reached the value of one. Probably this effect is due to the storage of some refractive index distribution by the Z-scan itself. When the incident beam power was increased the difference in transmittance was increased too. However, this difference tended to saturate to some maximum value. Reaching the maximum power, 10 mW, and then obtaining the Z-scan curves for lower powers gave rise to changes in the maximum and minimum transmittance and in the overall shape of the z-scan curve. This effect needs more analysis because did not follow the form predicted by Chapple et. al. [12], when a lens, of some focal length, is formed in the sample. However, from the shape of the Z-scan curve as a function of the incident power we can say that the results follow, in a very close way, which reported for materials that present a thermal nonlinearity under illumination of cw sources [13]. For low power, 1 mW, the Z-scan curve is very symmetric with a separation between the peak and valley of $2z_0$. As the incident power is increased the Z-scan curve presents a very well defined peak and a very broad valley.

3. NUMERICAL SIMULATIONS

Trying to obtain the nature of the nonlinear behavior of the photographic film and considering the experimental conditions we assume that the response of the material was of thermal type. Under these assumptions we developed a model based on propagation of Gaussian beams to obtain numerical simulation of the normalized transmittance. The experimental configuration used to obtain the model is shown in figure 7.

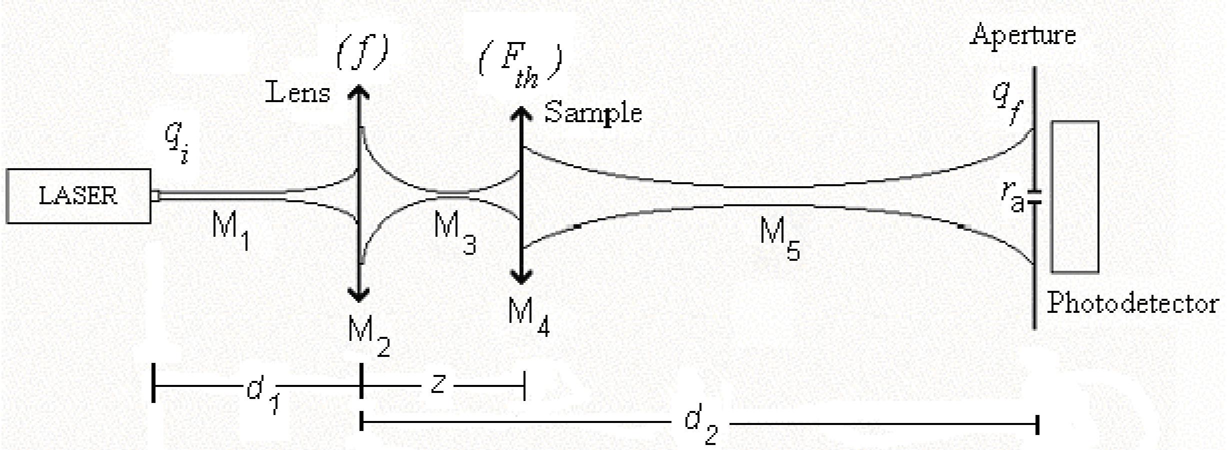


Figure 7. Experimental configuration used to obtain the equivalent *ABCD* matrix and the numerical simulations.

Where the nonlinear material was considered as a thin lens whose focal length was given by [10]:

$$F_{th} = \frac{\pi \kappa \omega^2}{P_{abs}\left(\frac{\partial n}{\partial T}\right)}, \tag{1}$$

where is κ the thermal conductivity, ω is the beam radius in the sample at the position z, P_{abs} is the total light power absorbed by the sample $P_{abs} = P_{in}\left(1 - e^{-\alpha L}\right)$ where P_{in} is the incident laser power, α the absorption coefficient and L the width of the sample), and $\partial n / \partial T$ is the change of the refractive index with the temperature.

Considering Gaussian beam propagation, where we only know the beam waist at the output of the laser, and the equivalent *ABCD* matrix of the optical system, where the elements of this matrix are:

$$A = 1 + \frac{z}{F_{th}} - \frac{d_2}{F_{th}} - \frac{d_2}{f} - \frac{z^2}{fF_{th}} + \frac{zd_2}{fF_{th}}, \tag{2a}$$

$$B = d_1 + \frac{d_1 z}{F_{th}} - \frac{d_1 d_2}{F_{th}} - \frac{d_1 d_2}{f} - \frac{d_1 z^2}{fF_{th}} + \frac{d_1 d_2 z}{fF_{th}} + \frac{z^2}{F_{th}} - \frac{zd_2}{F_{th}} + d_2, \tag{2b}$$

$$C = \frac{z}{fF_{th}} - \frac{1}{f} - \frac{1}{F_{th}}, \tag{2c}$$

and

$$D = \frac{d_1 z}{fF_{th}} - \frac{d_1}{f} - \frac{d_1}{F_{th}} - \frac{z}{F_{th}} + 1, \tag{2d}$$

from it is possible to calculate the characteristics of the Gaussian beam at the position of the photodetector using the *ABCD* law, i.e.:

$$q_f = \frac{Aq_i + B}{Cq_i + D}, \tag{3}$$

where $q_i = iz_0$ and q_f is the *q*-parameter at the position of the photodectector. As it is well known this parameter can be written as

$$\frac{1}{q_f} = \frac{1}{R(z)} - i\frac{\lambda}{\pi w_{NL}^2(z)}, \tag{4}$$

where $R(z)$ is the radius of curvature of the wave front and $w_{NL}(z)$ is the beam radius. Then, the beam radius is related to the imaginary part of ${}^{1}\!/_{q_f}$ and this quantity is proportional to the power captured by the photodetector.

In the same way it is possible to calculate the beam radius when there is not nonlinear sample $w_L(z)$. The normalized transmittance *T* obtained from the Z-scan technique at the position of the detector with an aperture of radius r_a can be calculated from:

$$T = \frac{1 - exp(-r_a / w_{NL}^2)}{1 - exp(-r_a^2 / w_L^2)}. \tag{5}$$

Using a program we calculate the normalized transmittance of a z-scan curve considering the experimental conditions and obtaining the better fit for the experimental results. From the formula for the focal length we know the power absorbed and the beam radius, the value of the ratio $\kappa/(dn/dT)$ was the parameter to fit he experimental data. In figure 8 we present representative results about the correspondence between the simulations and the experiments at *λ* = 457 nm. In this figure we only show the first three incident powers where the general experimental behavior was reproduced by the simulation; the maximum transmittance and shape of the peak had a better correspondence that the same features for the valley. However, it exist remarkable differences between the numerical and the experimental curve.

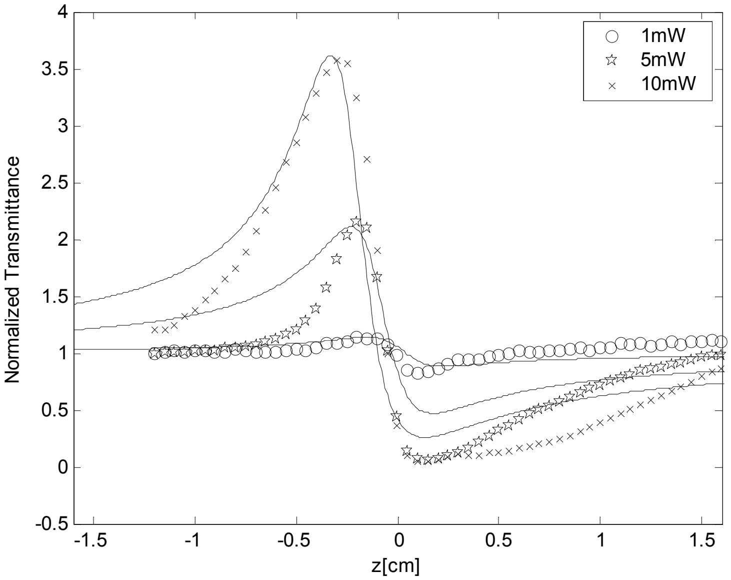


Figure 8. Numerical and experimental comparison. The numerical curves (solid lines) were obtained with the same experimental incident power and the following parameters: λ = 457nm, w_o =15μm, d_1 =60cm, f = 6cm, d_2 = 85cm, αL = 2.8 x10^{-3} , $\kappa/(dn/dT)$= -6.688 x10^{-6} W/cm, and the sample was scanned 28mm.

4. CONCLUSIONS

In this work we have presented a characterization of the nonlinear optical response of developed and bleached photographic film. We use visible cw laser beams at different wavelengths in the range of some tents of mW of incident power, reaching intensities of around 400 W/cm^2, in a z-scan experiment. The results show that the photographic film behaves as a negative nonlinear material where the exhibited nonlinearity was not of thermal type. The last assumption was obtained after comparison of the experimental results with numerical simulations, obtained using Gaussian beam propagation and assuming that the nonlinear media act as a thin thermal lens. Intensities higher than around 200 W/cm^2 created permanent refractive index changes in the photographic film.

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