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Nonlinear optical properties in double-sided nonlinear media with Z-scan technique based on the Huygens–Fresnel principle*

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We present a theoretical model to analyse the propagation of a Gaussian laser beam through double-sided nonlinear media. This model is based on the Huygens–Fresnel diffraction integral method. This theoretical model is not only consistent with the cascade structure model for a small nonlinear phase-shift but also can be used for a large nonlinear phase-shift. It has been verified that it is suitable to characterize the double-sided nonlinear media compared with the cascade structure model. A good agreement between the experimental data and the results from the theoretical model is obtained. It will be useful for the design of multi-sided nonlinear materials.

Keywords: optical nonlinearity, double-sided nonlinear media, Z-scan
PACC: 4265, 4270, 4285F

1. Introduction

The Z-scan technique introduced by Sheik-Bahae *et al* can be used to measure both the nonlinear refractive coefficient and the nonlinear absorption coefficient of a sample.[1] Its simplicity makes this technique particularly useful in researching third-order optical materials. The simplest analysis of the Z-scan technique is under the assumption of a Gaussian TEM_{00} beam and a sample that is thin compared with the Rayleigh length of the beam.[1,2] Since the Z-scan technique was introduced, many theoretical models have been proposed, including the Huygens–Fresnel (HF) formalism,[3] the Fresnel–Kirchhoff diffraction theory,[4] the Gaussian decomposition method (GDM),[5] the fast Fourier transform (FFT)[6] and complex ray-tracing[7] methods. The analytical formulas for the on-axis normalized Z-scan transmittance have also been derived in the first-order approximation for the thin and thick samples in a weak nonlinear regime.[2,8,9] The above Z-scan theoretical models are mostly applied to the single nonlinear sample structures. As both the thin and thick samples are of single sample structure, the analytical expressions are simple.

However, when samples are complex in configurations, for example, multiple-layer samples and samples with the nonlinear optical parameters, such as nonlinear refractive index and absorption coefficient, varying along the propagation direction of the laser beam, it seems difficult to obtain analytical solutions except for some particular cases.[10]

Recently, it is found that the electrostatic self-assembly films demonstrate the enhancement of third-order optical nonlinearities.[11,12] Most electrostatic self-assembly films are double-sided, which can be considered as two identical nonlinear films which a substrate is sandwiched in between. Can the nonlinear optical properties of the double-sided films be characterized with the previous Z-scan theories? Therefore, to answer the question, we present a theoretical model based on the HF diffraction integral method (DIM) to characterize the Z-scan curves of the double-sided nonlinear media. This theoretical model is compared with the cascaded structure model provided by Zang *et al.*[13,14]

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2. Z-scan theory for double-sided samples

We present a model to describe the nonlinear propagation of beam through the double-sided nonlinear media as shown in Fig.1. The model is based on the propagation of a wave front and described by using the Fresnel diffraction integral and the HF principle.[15] For simplicity, we consider a double-sided medium with a purely refractive nonlinearity, and the thickness of the each-sided nonlinear medium is equal.

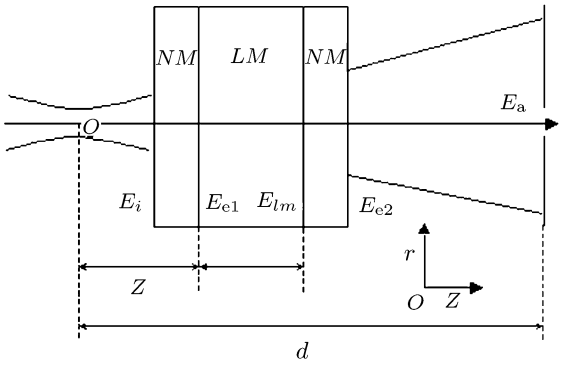


Fig.1. Scheme of the coordinates in the calculation of the electric field. NM: nonlinear medium, LM: linear medium (substrate).

Assuming a TEM_{00} Gaussian beam of waist radius ω_0 to travel in the $+z$ direction, the incident electric field E_i can be written as

$$E_i(z,r,t) = E_0(t)\frac{\omega_0}{\omega(z)} \times \exp\Big(-\frac{r^2}{\omega^2(z)} - \frac{\mathrm{i}kr^2}{2R(z)}\Big)\cdot \mathrm{e}^{-\mathrm{i}\phi(z,t)}, \quad (1)$$

where $\omega(z) = \omega_0(1 + z^2/z_0^2)^{1/2}$ is the beam radius, $R(z) = z(1 + z_0^2/z^2)$ is the radius of curvature of the wave front at z, $z_0 = \pi\omega_0^2/\lambda$ is the diffraction length of the beam, and $k = 2\pi/\lambda$ is the wave vector. $E_0(t)$ denotes the electric field at the focus and contains the temporal envelope of the laser pulse. The $\mathrm{e}^{-\mathrm{i}\phi(z,t)}$ term includes all the radially uniform phase variations.

For the double-sided nonlinear media with purely refractive nonlinearity, after the beam has passed through the first nonlinear medium, the nonlinear phase-shift $\Delta\phi_1$ is obtained. Here, the nonlinear media can be assumed to be thin samples. Then the complex electric field E_{e1} at the exit plane of the first nonlinear medium is given by

$$E_{e1}(z,r,t) = E_i(z,r,t)\mathrm{e}^{-\alpha_0 L/2}\mathrm{e}^{-\mathrm{i}\Delta\phi_1(z,r,t)}. \quad (2)$$

The nonlinear phase-shift $\Delta\phi_1$ at the exit plane of the first nonlinear medium can be expressed as[1]

$$\Delta\phi_1(z,r,t) = \frac{\Delta\Phi_0}{1 + z^2/z_0^2} \times \exp\Big(-\frac{2r^2}{\omega^2(z)}\Big)\exp\Big(-\frac{t^2}{\tau^2}\Big). \quad (3)$$

The on-axis phase-shift at focus, $\Delta\Phi_0$, is defined as

$$\Delta\Phi_0 = k\gamma I_0 L_{\mathrm{eff}}, \quad (4)$$

where τ is the duration of laser pulse, γ the nonlinear refraction coefficient, $L_{\mathrm{eff}} = [1 - \exp(-\alpha_0 L)]/\alpha_0$ the effective thickness of the each-sided nonlinear medium, L the thickness of the each-sided nonlinear medium, α_0 the linear absorption coefficient, and I_0 the laser intensity at the focal point.

The light beam will propagate through the middle linear medium between the nonlinear media after passing through the first nonlinear medium. The electric field at the exit plane of the linear medium is obtained by applying the HF propagation integral[16]

$$E_{lm}(z+l,r,t) = \frac{2\pi}{\mathrm{i}\lambda l}\exp\Big(\frac{\mathrm{i}\pi r^2}{\lambda l}\Big) \times \int_0^{+\infty} r_1 \mathrm{d}r_1 E_{e1}(z,r_1,t) \times \exp\Big(\frac{\mathrm{i}\pi r_1^2}{\lambda l}\Big) J_0\Big(\frac{2\pi r r_1}{\lambda l}\Big), \quad (5)$$

where l is the thickness of the linear medium, and J_0 is the zeroth-order Bessel function.

The complex electric field E_{e2} at the exit plane of the second nonlinear medium is given by

$$E_{e2}(z+l,r,t) = E_{lm}(z+l,r,t)\mathrm{e}^{-\alpha_0 L/2}\mathrm{e}^{-\mathrm{i}\Delta\phi_2(z+l,r,t)}, \quad (6)$$

and the nonlinear phase-shift $\Delta\phi_2$ at the exit plane of the second nonlinear medium is expressed as

$$\Delta\phi_2(z+l,r,t) = k\gamma L_{\mathrm{eff}} I_{lm}(z+l,r,t), \quad (7)$$

where I_{lm} is the light intensity at the exit plane of the linear medium.

Now, the complex electric field E_a at the aperture plane can be obtained through the second HF integral,

$$E_a(z,r,t) = \frac{2\pi}{\mathrm{i}\lambda(d-z-l)}\exp\Big[\frac{\mathrm{i}\pi r^2}{\lambda(d-z-l)}\Big] \times \int_0^{+\infty} r_2 \mathrm{d}r_2 E_{e2}(z+l,r_2,t) \times \exp\Big[\frac{\mathrm{i}\pi r_2^2}{\lambda(d-z-l)}\Big] J_0\Big[\frac{2\pi r r_2}{\lambda(d-z-l)}\Big], \quad (8)$$

where d is the separation between the aperture and the focal plane. Then the normalized transmittance $T(z,S)$ can be obtained by

$$T(z,S)=\frac{c\varepsilon_0 n_0\pi\int_{-\infty}^{+\infty}\mathrm{d}t\int_0^{r_\mathrm{a}}|E_\mathrm{a}(z,r,t)|^2 r\mathrm{d}r}{S\int_{-\infty}^{+\infty}P_i(t)\mathrm{d}t}, \qquad (9)$$

where$P_i(t)=\pi\omega_0^2 I_0(t)/2$ is the instantaneous input power, and $S=1-\exp(-2r_\mathrm{a}^2/\omega_\mathrm{a}^2)$ is the aperture linear transmittance.

3. Comparison with current theory

To confirm that the theoretical equations above are valid, we simulate the closed-aperture Z-scan curves with the above theoretical model and the cascaded structure model,[12] which is based on the Gaussian decomposition method (GDM).[4] To conveniently compare the results obtained from two theoretical models, the Z-scan theory for the double-sided samples and the cascaded structure model are simply referred to as the DS HF DIM and the DS GDM, respectively. According to Ref.[13] for small nonlinear phase-shift, the on-axis normalized transmittance $T(z,\Delta\Phi_0)$ is expressed as

$$T(z,\Delta\Phi_0)=1+\frac{4\Delta\Phi_0 x}{(x^2+1)(x^2+9)}+\frac{4\Delta\Phi_0(x+L_{lm})}{[(x+L_{lm})^2+1][(x+L_{lm})^2+9]}, \qquad (10)$$

where $x=z/z_0$, $L_{lm}=l/z_0$, and $\Delta\Phi_0$ is the on-axis nonlinear phase-shift of the each-sided medium.

Figure 2 shows the calculated curves resulting from the DS HF DIM (shown by the solid line) and the DS GDM (shown by the dashed line). For a small nonlinear phase-shift ($\Delta\Phi_0$ = -π/10) and small linear transmittance of the aperture ($S=0.02$), the curves obtained from the DS HF DIM and the DS GDM overlap almost completely in the whole interval as shown in Fig.2(a), proving that the DS HF DIM is valid. When the thickness of the substrate l is more than two times longer than the diffraction length z_0, a two-peak-two-valley pattern is obtained, which is the same as the result reported by Zang *et al.*[13]

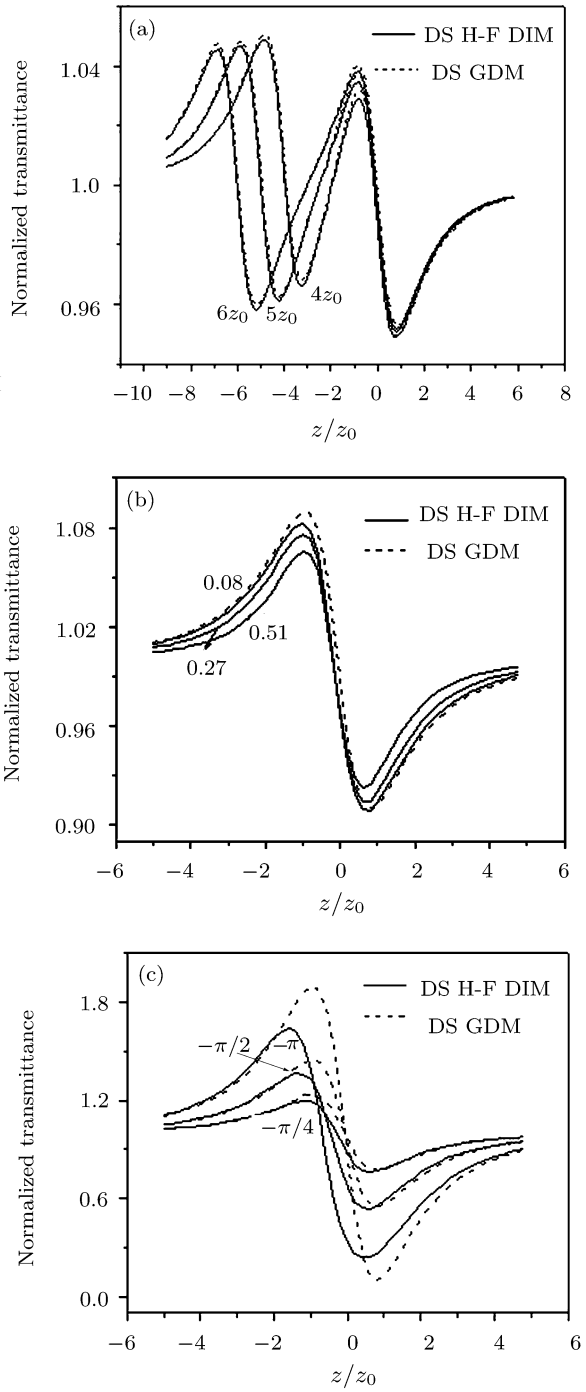


Fig.2. The comparison between the DS HF DIM (the solid line) and the DS GDM (the dashed line). (a) the graph depicts the results for $S=0.02$, $\Delta\Phi_0=-\pi/10$, $l=4z_0$, $5z_0$ and $6z_0$, (b) the graph depicts the results for $l=0.14z_0$, $\Delta\Phi_0=-\pi/10$, $S=0.08$, 0.27 and 0.51, (c)the graphs depict the results for $S=0.02$, $l=0.14z_0$, $\Delta\Phi_0=-\pi/4$, $-\pi/2$ and $-\pi$.

Figures 2(b) and (c) represent the effects of the linear transmittance of the aperture S and the nonlinear phase-shift $\Delta\Phi_0$ on the Z-scan cures for the same

thickness of the substrate ($l = 0.14z_0$). In the case of small l, the Z-scan curve still has only one peak and valley. For a fixed $\Delta\Phi_0$ ($-\pi/10$), with S increasing, both the peak and valley of the curves obtained from the DS HF DIM are asymmetrically suppressed compared with the curves based on the DS GDM as shown in Fig.2(b). Large aperture size reduces the sensitivity in the Z-scan technique since the peak–valley separation becomes smaller. Figure 2(c) shows the simulation results for the nonlinear phase-shifts $\Delta\Phi_0$ equal to -$\pi/4$, -$\pi/2$, -π, but the aperture size is fixed ($S = 0.02$).

It can be clearly seen from Fig.2(c) that with the increase of $|\Delta\Phi_0|$, the curves obtained from the DS HF DIM become asymmetric, and when $\Delta\Phi_0 = -\pi$, the valley is obviously broadened. However, the curves obtained from the DS GDM are symmetric. Apart from the valley, the transmittance is enhanced and the peak is suppressed, both the peak and the valley shift toward the left.

The important parameter to determine the nonlinearity is the difference between the normalized peak transmittance and normalized valley transmittance, $\Delta T_{\rm pv}$, which is a function of not only the nonlinear phase-shift $|\Delta\Phi_0|$ but also the linear transmittance of the aperture S. Figure 3 shows the percentage difference in normalized transmittance between the DS HF DIM and the DS GDM with $l = 0.14z_0$. From this figure we can see that the percentage difference in normalized transmittance between the two theoretical curves increases with $|\Delta\Phi_0|$ and S. When $S = 0.02$ and $|\Delta\Phi_0| \le \pi/2$, the percentage difference is smaller than 10%. When $S = 0.2$ and $|\Delta\Phi_0| = \pi$, it is close to 40%.

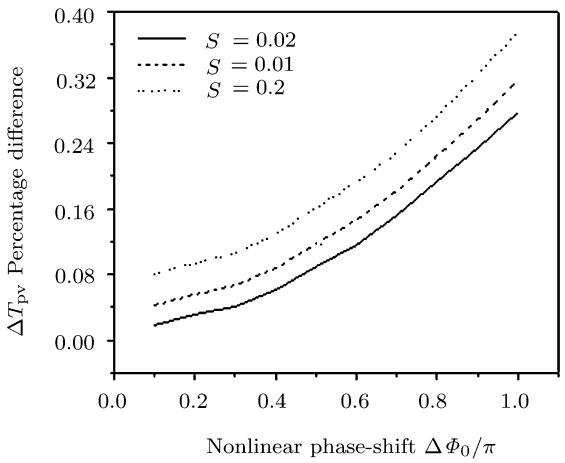


Fig.3. Percentage difference $(\Delta T_{\rm pv(DSGDM)} - \Delta T_{\rm pv(DSDIM)})/\Delta T_{\rm pv(DSDIM)}$ as a function of the nonlinear phase shift with $l = 0.14z_0$.

4. Application

The nonlinear refraction in a 20-bilayer double-sided TTAF/PSS electrostatic self-assembly film is measured with an 8 ns Nd:YAG laser system at 532 nm with a repetition rate of 1 Hz by using the closed-aperture Z-scan technique. The input energy and the linear transmittance of the aperture are 48 μJ and 0.2, respectively. The fabrication of the samples is described elsewhere.[17]

The 20-bilayer TTAF/PSS film structure is shown in Fig.4. Since the 20-bilayer TTAF/PSS film is ultrathin, it can be considered as an integral nonlinear medium.

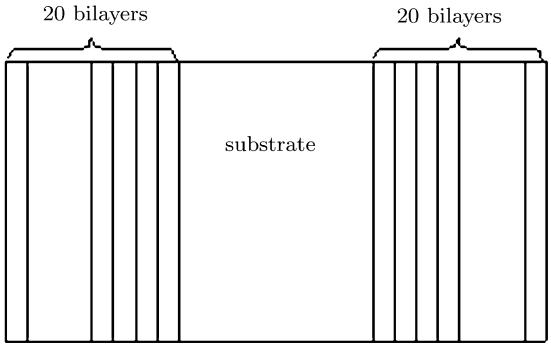


Fig.4. The 20-bilayer film structure.

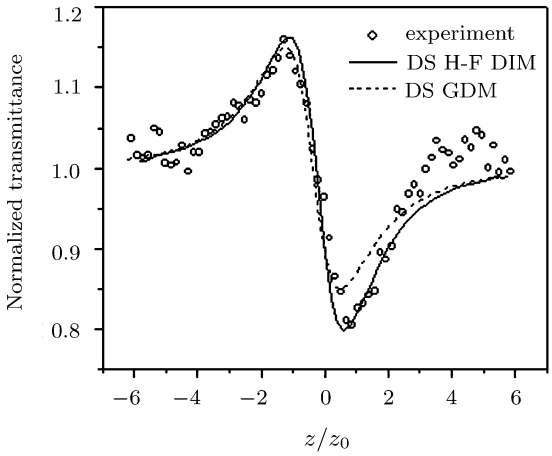


Fig.5. The normalized closed-aperture Z-scan transmittance in a 20-bilayer TTAF/PSS film.

Figure 5 shows the experimental Z-scan transmittance (data represented by open circles) in the 20-bilayer TTAF/PSS film. In the experiment, the separation between the aperture and the focal plane is taken as $d = 40$ cm. The thickness of the quartz substrate used is 1 mm, and the average thickness

of each bilayer for the TTAF/PSS films is measured to be 2 nm by using a step instrument. Both the DS HF DIM and the DS GDM are used to characterize the experimental data. It is clearly shown in Fig.5 that the DS HF DIM can fit both the peak and the valley well, while the DS GDM can fit only the peak of the experimental curve. Furthermore, $\Delta\Phi_0 = -0.70(\gamma = -2.0 \times 10^{-12}$ m^2/W) and $\Delta\Phi_0 = -0.53(\gamma = -1.5 \times 10^{-12}$ m^2/W) are obtained from the DS HF DIM and the DS GDM, respectively. The deviation of the latter from the former is about 25%. Thus the DS HF DIM is more suitable to fit the experimental data from the double-sided samples.

Under the condition of $S = 0.2$, the effect of the linear transmittance of the aperture on the closed-aperture Z-scan curve must be considered. It may induce a valley of a Z-scan curve whose amplitude is larger than that of its peak.[4] In the DS GDM, several approximations, such as the slowly varying envelope approximation, a thin sample, a small nonlinear phase shift and an on-axis approximation must be satisfied. The approximations of the first three terms are basically satisfied, but the approximation of the last term cannot be satisfied, so the DS GDM cannot fit the experimental data well.

5. Conclusions

Based on the Huygens–Fresnel diffraction integral method, we have presented a model to analyse the propagation of a Gaussian laser beam through double-sided nonlinear media. By comparing with the cascaded structure model, we find that our model is not limited to the case of small nonlinear phase-shifts and paraxial approximation, but more suitable to the case of large nonlinear phase-shifts and far-axis. Two theoretical models are used to fit the experimental data, and the results show that our theoretical model is more suitable to fit the experimental data from the double-sided electrostatic self-assembly films. It will be useful for the design of multi-sided nonlinear materials.

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