

# Analyses and computations of asymmetric Z-scan for large phase shift from diffraction theory

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Based on Fresnel-Kirchhoff diffraction theory, we set up a diffraction model of nonlinear optical media to Gaussian beam, which can interpret the Z-scan phenomenon from a new way. This theory is not only well consistent with the conventional Z-scan theory in the case of small nonlinear phase shift, but also can fit for the larger nonlinear phase shift. Numeric computations indicate the shape of the Z-scan curve is greatly affected by the value of the nonlinear phase shift. The symmetric dispersion-like Z-scan curve is only valid for small nonlinear phase shift ( $|\Delta\phi_0| < \pi$ ), but with increasing the nonlinear phase shift, the valley of the transmittance is severely suppressed and the peak is greatly enhanced. Further calculations show some new interesting results.

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Nonlinear refractive index  $n_2$  or Kerr constant  $\gamma$  is a key parameter to the third order nonlinearity of nonlinear optical materials. Among numerous techniques for measurement of nonlinear refraction, Z-scan method, proposed by Sheik-Bahae *et al.* in 1989<sup>[1]</sup>, is a simple, sensitive, single-beam method that uses the principle of spatial beam distortion to measure both the real and imaginary parts of the complex nonlinear refractive indices and their signs. A profocal transmittance maximum (peak) followed by a postfocal transmittance minimum (valley) is the Z-scan signature of a negative refractive nonlinearity. Positive nonlinear refraction, following the same analogy, gives rise to an opposite valley-peak configuration. Recently, the Z-scan theory and technique have also been extended to the measurements of nondegenerate two-photon absorption coefficient and nondegenerate nonlinear refractive index<sup>[2]</sup>, the studies of thermal lensing effect<sup>[3]</sup>, Gaussian beam propagation property and optical limiting of nonlinear media<sup>[4-6]</sup>.

As we know, the conventional Z-scan theory is based on the "Gaussian decomposition" method<sup>[7]</sup>, where several approximations, i.e., slowly varying envelope approximation (SVEA), thin sample, small nonlinear phase change ( $\Delta\phi_0$ ), must be satisfied, then the normalized transmittance  $T(z, \Delta\phi_0) \approx 1 + \frac{4\Delta\phi_0 x}{(x^2+9)(x^2+1)}$  is obtained, here  $x = z/z_0$ . Obviously, for large phase shift ( $|\Delta\phi_0| > \pi$ ), the symmetric equation does not fit the experimental data well. Considering this deficiency, Kwak *et al.* recently developed a new theory by employing an aberration-free approximation of the Gaussian beam<sup>[8]</sup>, which can be applicable to a large nonlinear phase shift.

In this paper, we try to study the Z-scan from a new approach. Here, the thin nonlinear medium is regarded as a phase modulated screen caused by the incident Gaussian beam for purely nonlinear refraction, which conversely diffracts the Gaussian beam<sup>[9]</sup>. Based on the Fresnel-Kirchhoff diffraction integral equation, the complex electric field distribution on the aperture plane can be calculated, then the power through the aperture and the

aperture transmittance can be obtained. Comparing our theory with Sheik-Bahae's theory and Kwak's theory, we find that in the small phase shifts case ( $< \pi$ ), our results are consistent with the formers' very well, in large phase shifts case ( $> \pi$ ), our results agree with the later's. By making numerous calculations, we also find some new interesting results.

Suppose a TEM<sub>00</sub> Gaussian beam traveling in the +Z direction, the coordinate zero is at the beam waist, the incident plane of a nonlinear medium is at  $Z_1$ . If the nonlinear absorption is negligible and the SVEA approximation is met, the complex electric field in the sample can be written as

$$E(r, z) = E(0, 0) e^{-\alpha(z-Z_1)/2} \frac{\omega_0}{\omega(z)} \cdot \exp \left[ -\frac{r^2}{\omega^2(z)} \right] \exp \left[ -\frac{ikr^2}{2R(z)} - i\phi(z) \right], \quad (1)$$

where  $E(0, 0)$  denotes the on-axis amplitude of the electric field at the focus,  $\alpha$  is the linear absorption coefficient,  $\omega_0$  is the beam waist radius,  $\omega^2(z) = \omega_0^2(1 + z^2/z_0^2)$  is the beam radius at  $z$ ,  $R(z) = z(1 + z_0^2/z^2)$  is the radius of curvature of the wavefront at  $z$ ,  $z_0 = \pi\omega_0^2/\lambda$  is the diffraction length of the beam,  $k = 2\pi/\lambda$  is the wave number,  $\lambda$  is the laser wavelength, and  $\phi(z)$  contains all the radially uniform phase variation. According to optical Kerr effect, the refraction index change can be written as  $\Delta n = \gamma I$ , here the laser intensity is  $I = \varepsilon_0 c n_0 |E|^2/2$  and the on-axis intensity at the focus is  $I(0, 0) = 2P/\pi\omega_0^2$ ,  $n_0$  is the linear refractive index,  $P$  is the laser power,  $c$  and  $\varepsilon_0$  are the speed of light and the permittivity in vacuum. The laser-induced refractive index distribution thus can be expressed as

$$\Delta n(r, z) = \frac{2\gamma P}{\pi} \cdot \frac{e^{-\alpha(z-Z_1)}}{\omega^2(z)} \cdot \exp \left[ -\frac{2r^2}{\omega^2(z)} \right]. \quad (2)$$

The transverse phase variation on the exit surface of the sample is the integration of Eq. (2) over the length

of the sample (*d*):

$$\Delta\phi(r, Z_1) = \frac{2\pi}{\lambda} \int_{Z_1}^{Z_1+d} \Delta n(r, z) dz \approx \Delta\phi_0(Z_1) \exp\left[-\frac{2r^2}{\omega^2(Z_1)}\right]. \quad (3)$$

In this equation, in order to analytically integrate, we have supposed  $\omega(z) \approx \omega(Z_1)$ , and it is true for "thin sample" (i.e.  $d < Z_0$ )<sup>[7]</sup>, where  $\Delta\phi_0(Z_1) = \frac{4\gamma}{\lambda} \cdot \frac{1-e^{-\alpha d}}{\alpha}$ .  $\frac{P}{\omega^2(Z_1)}$  is the peak nonlinear phase shift (somewhere simply denoted as  $\Delta\phi_0$ ). Equation (3) indicates that the transverse phase variation on the exit surface of the sample is a Gaussian-like distribution. We can regard this phase distribution as a phase hole<sup>[9]</sup>. The diffraction screen function is  $\tilde{U}(r, Z_1) = \exp[-i\Delta\phi(r, Z_1)]$ . And the radial electric field distribution of the Gaussian beam on the exit surface of the sample can be written as

$$\tilde{V}(r, Z_1) = E(0,0)e^{-\alpha d/2} \frac{\omega_0}{\omega(Z_1)} \exp\left[-\frac{r^2}{\omega^2(Z_1)}\right] \cdot \exp\left[-\frac{ikr^2}{2R(Z_1)}\right]. \quad (4)$$

So the complex electric field distribution in the far field aperture plane can be obtained from the Fresnel-Kirchhoff diffraction integral equation<sup>[10]</sup>

$$\tilde{E}(\rho) = \frac{-i}{\lambda} \int \int F(\theta_0, \theta) \tilde{V}(r) \tilde{U}(r) \frac{e^{ikD}}{D} r dr d\varphi, \quad (5)$$

where  $F(\theta_0, \theta) = (\cos\theta_0 + \cos\theta)/2$  is the inclination factor,  $\theta_0$  is the direction angle of the incident light to the sub-wave source (for vertical incident light,  $\theta_0 = 0$ ),  $\theta$  is the direction angle of the receipt point on the aperture plane to the subwave source.  $D = [Z_2^2 + (r \cos\phi - \rho)^2 + (r \sin\phi)^2]^{1/2}$  is distance from the diffraction point to the receipt point.  $r$  and  $\rho$  are respectively the radial coordinates on the diffraction screen and the aperture plane.  $Z_2$  is the distance between the diffraction screen and the aperture plane.

According to the characteristic of Gaussian function, when  $r = \sqrt{5}\omega(Z_1)$ ,  $\exp[-r^2/\omega^2(Z_1)]$  and  $\Delta\phi(r)$  tend to be 0, the integral limits can be selected as  $r = 0 \rightarrow \sqrt{5}\omega(Z_1)$  and  $\varphi = 0 \rightarrow 2\pi$ . Substituting all the above

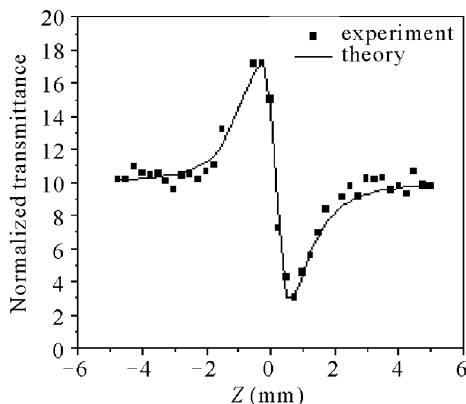


Fig. 1. Experimental and theoretical curves of Z-scan of a  $\pi$ -conjugated polymer (PPNB).

related equations into Eq. (5), we obtain the intensity distribution on the aperture plane as

$$I(\rho, Z_1, Z_2) = \frac{e^{-\alpha d}}{2\pi\lambda^2} \cdot \frac{P}{\omega^2(Z_1)} \cdot \left| \int_0^{2\pi} \int_0^{\sqrt{5}\omega(Z_1)} \left(\frac{1}{D} + \frac{Z_2}{D^2}\right) \exp\left(-\frac{r^2}{\omega^2(Z_1)}\right) \cdot \exp\left\{i\left[\frac{2\pi}{\lambda}D - \frac{kr^2}{2R(Z_1)} - \Delta\phi_0(Z_1) \exp\left(-\frac{2r^2}{\omega^2(Z_1)}\right)\right]\right\} r dr d\varphi \right|^2, \quad (6)$$

and the laser power in the aperture (with radius  $a$ )  $P_A(Z_1, Z_2, a, \Delta\phi_0) = 2\pi \int_0^a I(\rho, Z_1, Z_2) \cdot \rho d\rho$ .

Scanning  $Z_1$ , we can compute the Z-scan curve from Eq. (6). As an example, Fig. 1 shows the experimental and theoretical Z-scan curves of a new type of  $\pi$ -conjugated polymer<sup>[11,12]</sup>. We can see that the theory have a good consistency with the experimental data.

To confirm the validity of the above theoretical equations and the computing programs, we calculate the Z-scan curve for the small nonlinear phase shift  $\Delta\phi_0 = -\pi/2$ , as shown in Fig. 2. Here we should point out that the shape of the conventional Z-scan curve is symmetrical. So the tiny difference in Fig. 2 indicates that the new theory can give a more accurate result. Others validity checks are given in Ref. [13].

Based on this theory, we have studied the effect of the nonlinear phase shift on the shape of the Z-scan curve. The computing results are plotted in Fig. 3 (just for  $\gamma < 0$ , it is similar for  $\gamma > 0$ ). The parameters used are  $\lambda = 532$  nm,  $\omega_0 = 5.82$   $\mu$ m,  $Z_0 = 0.2$  mm. It can be seen that under the condition of  $|\Delta\phi_0| < \pi$ , with increasing nonlinear phase shifts, the dispersion-like curves (symmetric with respect to focus) broaden and the peak-to-valley separations increase. However, as the nonlinear phase shifts increase larger, the shapes of the Z-scan curves differ entirely from those obtained with the small nonlinear phase shift and are asymmetric. The valley of the transmittance is severely suppressed (nearly to zero of the transmittance) and the peak is greatly enhanced. This result is coincident with that of Kwak's theory<sup>[8]</sup>. In order to get the analytic expression, only the first

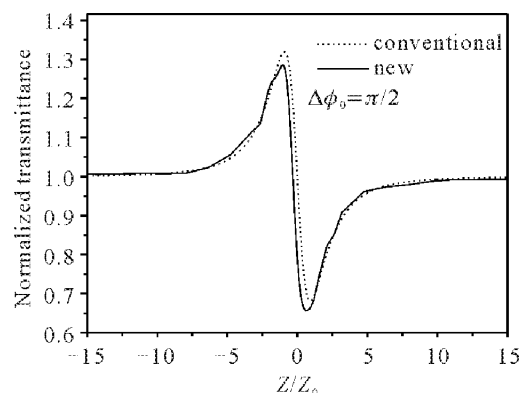


Fig. 2. Comparison of the new Z-scan theory (solid curve) with the conventional theory (dash curve) under small nonlinear phase shift,  $\Delta\phi_0 = -\pi/2$ .

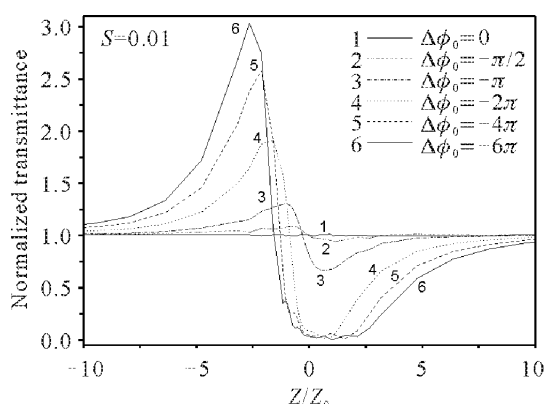


Fig. 3. Theoretical plots of closed-aperture  $Z$ -scan transmittance for purely nonlinear refraction under different nonlinear phase shift.

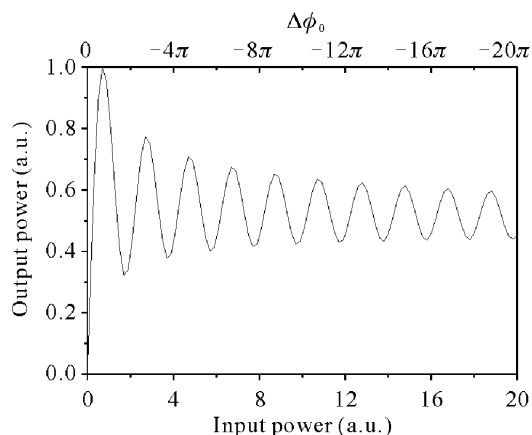


Fig. 4. Theoretical plot of the output power through the aperture versus the input laser power or the corresponding  $\Delta\phi_0$ . Sample is fixed at focus,  $a = 2$  mm,  $Z_2 = 1$  m.

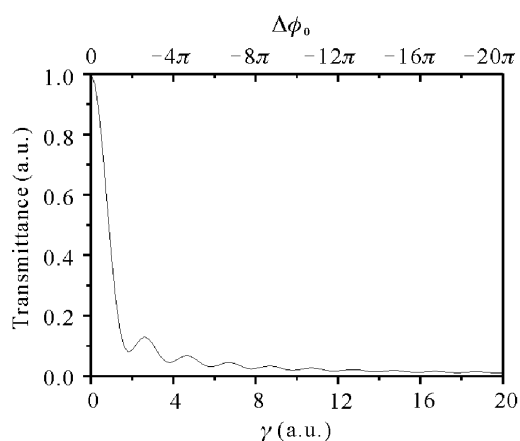


Fig. 5. Theoretical plot of aperture transmittance versus Kerr constant or  $\Delta\phi_0$ . Sample is fixed at focus,  $a = 2$  mm,  $Z_2 = 1$  m.

two terms of Fourier series are retained in Kwak's theory, while our purely numerical theory retains all terms. To some extent, the new  $Z$ -scan theory based on the diffraction model can get more accurate results for both the small and large phase shift.

If the sample is fixed at focus, changing the input laser

power to increase the  $\Delta\phi_0$ , we find an interesting phenomenon. The output power through the aperture will firstly go up to a peak, then fall down to a valley, then go up again, make an oscillation, as shown in Fig. 4. Figure 5 shows the dependence of the aperture transmittance on the Kerr constant or the  $\Delta\phi_0$ . This curve indicates an optical nonlinear medium with larger Kerr constant has much better optical limiting property.

In conclusion, we have set up a diffraction model of nonlinear optical media to Gaussian beam for purely nonlinear refraction based on Fresnel-Kirchhoff diffraction theory. This theory can explain the  $Z$ -scan phenomenon from a new way. Numeric computations indicate the shapes of  $Z$ -scan curves are greatly affected by the values of nonlinear phase shifts. The peak-valley transmittance difference ( $\Delta T_{p-v}$ ) and the distance between peak and valley ( $\Delta Z_{p-v}$ ) become larger with increasing of nonlinear phase shift. We also find the aperture size has influence only on the  $\Delta T_{p-v}$ , not on the  $\Delta Z_{p-v}$  when the aperture size is about 1 – 10 mm, while the distance between the sample and the aperture almost has no effect on the  $Z$ -scan curve as long as the far field condition is satisfied. The output power through the aperture will oscillate with increasing the nonlinear phase shift caused by the input laser power. The aperture transmittance will attenuate and saturate with the Kerr constant increasing, which indicates large Kerr constant materials have much better optical limiting property.

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