Z-scan technique for investigation of the noninstantaneous optical Kerr nonlinearity

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By treating laser-induced optical Kerr nonlinearity as a noninstantaneous decaying process, we present the pulse-duration-dependent Z-scan analytical expressions for an arbitrary aperture and an arbitrary nonlinear magnitude. This theory has the capacity to characterize the third-order nonlinear refraction induced by a laser pulse with its temporal duration being much longer than or comparable to the recovery time of the nonlinear effect. Through Z-scan measurements at different pulse durations, the nonlinear refractive coefficient and the recovery time could be determined unambiguously and simultaneously. Furthermore, the theory can be utilized to confirm whether the measured optical Kerr nonlinearity is instantaneous or noninstantaneous with respect to the given pulse duration. © 2009 Optical Society of America

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Over the past few decades, optical Kerr materials have received extensive attention. This is because these materials, exhibiting excellent nonlinear-optical (NLO) properties in terms of ultrafast response times and high nonlinear figures of merit, are greatly desirable for ultrafast all-optical communications and signal processing. As such, it is imperative to have accurate assessments on their NLO parameters, in particular, the optical Kerr nonlinear coefficient ($\gamma$) and the recovery time ($\tau_R$), which could be obtained with spectrally resolved two-beam coupling [1,2]. As is well known, the NLO response depends strongly on the pulse duration ($\tau$) [3]. If $\tau_R \ll \tau$ (the so-called steady-state condition), the nonlinearity can be regarded justifiably as responding instantaneously to optical pulses. When $\tau_R \gg \tau$, the nonlinear effect becomes insignificant to the overall NLO process, as it is too slow to respond. In the regime of $\tau \sim \tau_R$, the nonlinearity responds noninstantaneously to laser pulses. Such a scenario has been noticed by a few experimental investigations [4,5].

To investigate the optical nonlinearities of materials, a time-averaging technique has been exploited in Z-scan measurements owing to its high sensitivity and experimental simplicity [6]. During the past decade, the conventional Z-scan technique has been modified for characterizing higher-order nonlinearities [7], instantaneous nonlinearity [8], the thermooptical effect [9], and so on. Besides, in the Z-scan experiments, the laser parameters (e.g., wavelength [10] and peak irradiance [11]) have been exploited. Very recently, the dependence of optical nonlinearities on the pulse duration has been experimentally investigated [12,13]. It should be noted that pulse-duration dependence has not been fully addressed by the Z-scan theory. To perform the pulse-duration-dependent Z-scan measurements, variable pulse duration can be obtained by stimulated Brillouin scattering in liquid [12,13]; a double-pass, single-grating pulse stretcher; or a prism compressor.

In this Letter, we present the Z-scan theory for characterizing the noninstantaneous optical Kerr nonlinearity. Both the values of $\gamma$ and $\tau_R$ can be unambiguously determined, and the pulse-duration-dependent Z-scan theory permits also to identify the instantaneous or noninstantaneous optical nonlinearity with respect to the given pulse duration.

In the following analysis, we assume that the spatiotemporal profiles of incident laser irradiance exhibit the Gaussian distribution. For the Z-scan experiments, a TEM00 Gaussian beam propagating along the $+z$ direction with the coordinate origin being at the Gaussian beam waist, the electric field can be written as

$$E(r,z,t) = \frac{E_0 \omega_0}{\omega(z)} \exp \left[ \frac{-r^2}{\omega^2(z)} + \frac{-ikr^2}{2R(z)} \right] \exp \left[ \frac{-t^2}{2\tau^2} \right],$$

where $\omega^2(z) = \omega_0^2(1+z^2/z_0^2)$ and $R(z) = z(1+z^2/z_0^2)$. $E_0 = E(0,0,0)$ denotes the on-axis peak electric field amplitude at the focus. $\omega_0$ and $k = 2\pi/\lambda$, $z_0 = k\omega_0^2/2$, and $\tau$ are the waist radius, the laser wavelength, the wave vector, the Rayleigh length of the Gaussian beam, and the half-width at $e^{-1}$ of the maximum for the pulse duration, respectively. Its irradiance $I(r,z,t)$ can be expressed as $I(r,z,t) = I_0(z)x(t)^2$, where $I_0(z) = I_0/(1+z^2/z_0^2) \cdot \exp[-2z^2/\omega^2(z)]$, $I_0 = I(0,0,0)$ is the on-axis peak irradiance at the focus.
governed by the Debye relaxation equation as follows:

\[ \frac{d\Delta n(r,z;t)}{dz'} = k\Delta n(r,z;t), \]  

(2)

\[ \frac{dI(r,z;t)}{dz'} = -a_0 I(r,z;t). \]  

(3)

Here \( z' \) is the propagation distance inside the sample. The refractive-index variation, \( \Delta n(r,z;t) \), is governed by the Debye relaxation equation as follows [5]:

\[ \frac{d\Delta n(r,z;t)}{dt} + \Delta n(r,z;t) = \gamma I(r,z;t). \]  

(4)

Solving Eq. (4), the refractive-index variation is given by

\[ \Delta n(r,z;t) = \gamma I(r,z) f(t), \]  

(5)

where

\[ f(t) = \frac{1}{\tau_R} \int_{-\infty}^{t} \exp\left(-\frac{t'^2}{\tau_R^2}\right) \exp\left(\frac{t'-t}{\tau_R}\right) dt'. \]  

(6)

Equation (5) predicts that the magnitude of the refractive-index variation depends strongly on both the pulse duration and the recovery time of the nonlinearity. Figure 1 shows \( \Delta n(0,0;t) \) at different \( \tau \) for \( \tau_R=2 \) ps. It can be seen that \( \Delta n(0,0;t) \) builds up in time over the laser pulse and the magnitude of \( \Delta n(0,0;t) \) increases as the pulse duration \( \tau \) increases. As shown by the squares in Fig. 1, the nonlinearity almost responds instantaneously to optical pulses for \( \tau/\tau_R=10 \). When \( \tau/\tau_R=0.1 \) (see the triangles in Fig. 1), the nonlinear effect becomes insignificant, as it is too slow to respond. Under the extreme condition of \( \tau_R \ll \tau \), Eq. (5) becomes \( \Delta n(r,z;t) = \gamma I(r,z;t) \). On the opposite extreme, we find \( \Delta n(r,z;t) = 0 \). Under the excitation of cw laser, Eq. (5) gives \( \Delta n(r,z) = \gamma I(r,z) \).

By combining Eqs. (1)–(4), the complex field at the exit plane of the sample is given by

\[ E_e(r,z;t) = E_e(r,z) e^{-2\alpha_0 J_0^2} \exp[i\Delta\varphi(r,z;t)], \]  

(7)

where

\[ \Delta n(r,z;t) = \frac{\Phi_0}{1 + z^2/2z_0^2} \exp\left[-2r^2/\omega_0^2(z)\right] f(t). \]  

(8)

Here \( \Phi_0 = k \gamma I_0 (1-R) \) is the on-axis peak phase shift due to the refractive nonlinearity. \( R \) is the Fresnel reflectivity coefficient at the interface of the material with air. \( L \) and \( L_{\text{eff}} = (1-e^{-\alpha_0 z})/\alpha_0 \) are the physical length and the effective length of the sample, respectively.

Based on the Gaussian decomposition method, similar to the investigations in [6–8], the complex electric field at the aperture plane can be easily yielded. Under the far-field condition, we find the pulse-duration-dependent normalized Z-scan transmittance as

\[ T(x) = \left[ 1 - \sum_{u,v=0}^{M} u!v![(u+v+1)(x^2+1)^{u+v}] \right]^{1/2}, \]  

(9)

where

\[ \lambda_{uv} = \frac{(u+v+1)(x^2+1)[x^2+(2u+1)(2v+1)]}{[x^2+(2u+1)^2][x^2+(2v+1)^2]}, \]  

(10)

\[ \psi_{uv} = \left( u - v \right) \left( \frac{\pi}{2} - \frac{2(u+v+1)x(x^2+1)\ln(1-s)}{[x^2+(2u+1)^2][x^2+(2v+1)^2]} \right), \]  

(11)

\[ \lambda_{uv} = \frac{1}{\pi^{1/2}} \int_{-\infty}^{\infty} f(t)^{u+v} \exp\left(-\frac{t^2}{\tau_R^2}\right) dt. \]  

(12)

Here \( x = z/\omega_0 \) is the relative sample position and \( s \) is the linear transmittance of aperture. The optimum sum upper limit \( M \) in Eq. (9) should satisfy \( |\Phi_0^2/M| < 0.5 \) for a given phase shift \( \Phi_0 \).

It should be noted that \( A_{uv} \) represents the contribution of both the pulse temporal variation and the nonlinear response of the material to the Z-scan trace. Under the extreme condition of \( \tau_R \ll \tau \), Eq. (6) becomes \( f(t) = \exp(-t^2/\tau_R^2) \). Substituting \( f(t) \) into Eq. (12), we easily obtain \( A_{uv} = (u+v+1)^{-1/2} \). We ultimately yield the Z-scan expression for characterizing the instantaneous nonlinearity. In the opposite limit of \( \tau_R \gg \tau \), we find \( T(x) = 1 \), because \( A_{uv} = 0 \). If the Z-scan measurement is induced by the cw laser beam, one takes \( A_{uv} = 1 \), as we reported previously [7].

Figure 2 illustrates the Z-scan traces excited by a pulse laser with different pulse durations for \( \tau_R = 2 \) ps. The other numerical parameters are \( \Phi_0 = 2 \),
parameters. With the known other practical procedure to evaluate the nonlinear experimentally measured theory for an instantaneous nonlinearity to fit $Z$ nonlinearity. Inset, $\Delta T_{PV}$ as a function of $\tau/\tau_R$. In the inset, the circles are numerical results, while the solid curve is obtained by Eq. (13).

$s=0.2$, and $M=10$ in Eq. (9). For comparison, the Z-scan trace induced by an instantaneous nonlinearity (solid curve) is also displayed in Fig. 2. As shown by the inset in Fig. 2, the peak–valley normalized transmittance difference $\Delta T_{PV}=T_P-T_V$ (where $T_P$ and $T_V$ are the peak and valley normalized transmittances in the Z-scan trace, respectively) depends strongly on the pulse duration for a given nonlinear effect (for instance, molecular reorientation in Kerr liquids [3,5]). When $\tau/\tau_R \geq 10$, the obtained $\Delta T_{PV}$ value is independent of the pulse duration. For $0.1 < \tau/\tau_R < 10$, the $\Delta T_{PV}$ value dramatically decreases with decreasing pulse duration. When $\tau/\tau_R \approx 0.1$, the noninstantaneous effect becomes insignificant. In addition, the determination of the nonlinear coefficient will give rise to large errors if one uses the Z-scan theory for an instantaneous nonlinearity to fit Z-scan experimental results induced by a noninstantaneous nonlinearity.

In the Z-scan measurements, an useful experimental parameter is $\Delta T_{PV}$. We find an empirical expression

$$\Delta T_{PV} = 0.406(1-s)^{0.25} |\Phi_0| \sqrt[2]{\frac{0.76(\pi/\tau_R)^3}{1 + 0.76(\pi/\tau_R)^3}}.$$  \hfill (13)

The above expression has only a ±2.3% error for $|\Phi_0| = \pi$, $s < 0.5$, and $\tau > 0.1 \tau_R$. If $\tau > \tau_R$, Eq. (13) degenerates into the result reported previously [6].

To determine both $\gamma$ and $\tau_R$, one can perform the Z-scan experiments at different levels of $\tau$. In Eq. (9), two free parameters (namely, $\gamma$ and $\tau_R$) can be evaluated from the best fittings between Eq. (9) and the experimentally measured Z-scan traces. There is another practical procedure to evaluate the nonlinear parameters. With the known $\Delta T_{PV}$ and $s$ measured in the Z-scan experiments at different $\tau$, one can obtain $\gamma$ and $\tau_R$ by the use of Eq. (13).

The instantaneous nonlinear process is independent of the pulse duration, whereas the noninstantaneous nonlinear effect depends strongly on the pulse duration. Accordingly, to identify whether the Kerr nonlinearity is instantaneous or noninstantaneous, one should carry out Z-scan measurements with different pulse durations [12]. The further studies beyond our scope are required to quantitatively separate the contributions of instantaneous and noninstantaneous Kerr nonlinearities with respect to the given pulse duration.

In summary, we have presented the Z-scan theory for investigation of the noninstantaneous optical Kerr nonlinearity. We have demonstrated that the laser-induced refractive-index variation depends strongly on the pulse duration. The pulse-duration-dependent Z-scan theory gives quick determinations of not only the nonlinear refractive coefficient but also the recovery time. The results also provide theoretical justifications for a practical way to identify the instantaneous or noninstantaneous optical Kerr nonlinearities by performing Z-scan measurements at different laser pulse durations.

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