Characterization of saturable absorbers using an open-aperture Gaussian-beam *Z* scan

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We present a theoretical investigation on the open-aperture Gaussian-beam *Z*-scan technique for two dominant saturable absorption models, by using the Adomian decomposition method. Analytic expressions of the *Z*-scan traces are deduced for a cw laser and a pulsed laser, respectively; in particular, for performable simulations, the optimal sum upper limit is found. We give an experimental demonstration, in which a side-chain azobenzene polymer poly [6-[1-(4-(4-nitrophenylazo)phenyl) piperazine] hexyl methacrylate] (Pda) film behaves as a test sample. The results manifest that our theory is reasonable and the Pda film is a good saturable absorber at the wavelength of 532 nm.

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I. INTRODUCTION

An intense radiation can induce a profound change on the absorption property of a material, resulting in the intensity-dependent transmittance, which is the so-called nonlinear absorption [1]. Nonlinear absorption can be classified into two types: (i) transmittance increases with increasing optical intensity; this case corresponds to the well-known saturable absorption (SA); (ii) transmittance reduces with increasing optical intensity; this type includes two-photon absorption, multiphoton absorption, and reverse SA. Different effects originating from different physical mechanisms could lead to a variety of different applications. For instance, SA materials have been used extensively in short-pulsed laser generations [2] as crucial passive mode-locking or *Q*-switching elements. Thus, it is paramount to fully characterize saturable performance, in which a typical figure of merit is the saturable intensity. SA characteristics depend on the inherent properties of a material and the parameters (such as wavelength, intensity, and pulse duration) of the laser used. To characterize nonlinear absorption, the open-aperture (OA) *Z*-scan technique, which was first pioneered by Sheik-Bahae *et al.* [3], has been extensively used. Recently, we developed an OA *Z*-scan theory for the materials with simultaneous two- and three-photon absorption, which allows us to identify and determine the two- and three-photon absorption coefficients from a single OA *Z*-scan trace [4]. The SA properties of some materials were observed experimentally and analyzed theoretically [5–11].

Here we develop the OA *Z*-scan theory for SA with the use of the Adomian decomposition (AD) method [12], which has been introduced in nonlinear optics [13] and its convergence was also discussed [14]. We obtain the analytical expressions of the OA *Z*-scan traces for two dominant SA models. The theory allows a straightforward estimation of the saturable intensity and the determination of the SA model for a material by fitting the experimental data. As a test, we explore the SA property of a recently synthesized side-chain azobenzene polymer with π-electron system, poly [6-[1-(4-(4-nitrophenylazo)phenyl) piperazine] hexyl methacrylate] (Pda). The synthesis, chemical structure, linear absorption, and quadric nonlinearity induced by all-optical poling have been described in Ref. [15]. The reason we chose the π-electron system is due to its large nonlinearity [1,16], and its good flexibility for the structural modifications. We also reveal the possible mechanism of SA.

II. THEORY

It is well known that there are two dominant SA models as mentioned below. Now we give a theoretical investigation. Let us consider the beam propagation in a thin saturable absorber; the optical intensity loss is governed by the following differential equation:

$$dI/dz' = -f_j(I) = -\alpha_j(I)I \tag{1}$$

where z' and I are the propagation distance and the optical intensity inside the saturable absorption sample, respectively. $\alpha_j(I)$ is the intensity-dependent absorption coefficient. The suffix $j=1$ and 2 indicates the two different SA models as described below, respectively,

$$\alpha_1(I) = \alpha_0/(1 + I/I_S), \quad \text{for model I} \tag{2}$$

$$\alpha_2(I) = \alpha_0/\sqrt{1 + I/I_S}, \quad \text{for model II}. \tag{3}$$

Models I and II are often referred to SA in homogeneous [5] and inhomogeneous [6] broadening systems. In the above two expressions, α_0 and I_S are the linear absorption coefficient under the low-intensity approximation and the saturable intensity, respectively.

In general, it is difficult to give explicit analytical solutions of the optical intensity transmitted though the saturable

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absorber with a length of L, I_j^{out}, for this two SA models. Equation (1) can be integrated formally

$$I_j^{out} = I^{in} - \int_0^L f_j(I)dz', \tag{4}$$

where I^{in} is the input optical intensity at the entrance face of the saturable absorber.

On the basis of the AD method [12], however, the solutions of I_j^{out} can be always expressed in terms of the polynomial, as follows

$$I_j^{out} = \sum_{m=0}^{\infty} u_{jm} \tag{5}$$

and the nonlinear term by an infinite series of Adomian's polynomials

$$f_j(I) = \sum_{m=0}^{\infty} A_{jm}(u_{j0}, u_{j1}, \ldots, u_{jm},), \tag{6}$$

where A_{jm} polynomials is defined by

$$A_{jm} = \frac{1}{m!} \left[\frac{d^m}{d\zeta^m} f_j\left(\sum_{k=0}^{m} \zeta^k u_{jk} \right) \right] \Bigg|_{\zeta=0}, \tag{7}$$

where ζ is an intermediate variable.

Substituting Eqs. (5) and (6) into (4) gives

$$\sum_{m=0}^{\infty} u_{jm} = I^{in} - \int_0^L \sum_{m=0}^{\infty} A_{jm} dz'. \tag{8}$$

Identifying the zeroth component $u_{j0}=I^{in}$, one gets u_{jm} by the recurrence formula as follows:

$$\begin{aligned} u_{j(m+1)} &= -\int_0^L A_{jm} dz' \\ &= -\frac{L}{(m+1)!} \left[\frac{d^m}{d\zeta^m} f_j\left(\sum_{k=0}^{m} \zeta^k u_{jk} \right) \right] \Bigg|_{\zeta=0}. \end{aligned} \tag{9}$$

It is obvious that once u_{j0} is known, all u_{jm} can be given by Eq. (9). With this procedure, one can easily build the exact solution of the problem $I_j^{out}=\Sigma_{m=0}^{\infty} u_{jm}$.

According to Eq. (9), the exact solution of I_1^{out} for model I in a series form is given by

$$I_1^{out} = I^{in}\left[1 + \sum_{m=1}^{\infty} \frac{(-\alpha_0 L)^m}{m!} \frac{g_{1m}(\eta)}{(1+\eta)^{2m-1}} \right], \tag{10}$$

where $\eta=I^{in}/I_S$. The first five terms of $g_{1m}(\eta)$ are obtained as follows:

$$g_{11}(\eta) = 1,$$

$$g_{12}(\eta) = 1,$$

$$g_{13}(\eta) = 1 - 2\eta,$$

$$g_{14}(\eta) = 1 - 8\eta + 6\eta^2,$$

$$g_{15}(\eta) = 1 - 22\eta + 58\eta^2 - 24\eta^3. \tag{11}$$

Similarly, the exact solution of I_2^{out} for model II can be expressed as

$$I_2^{out} = I^{in}\left[1 + \sum_{m=1}^{\infty} \frac{(-\alpha_0 L)^m}{m!} \frac{g_{2m}(\eta)}{(1+\eta)^{(3m-2)/2}} \right] \tag{12}$$

and the first five terms of $g_{2m}(\eta)$ are given as follows:

$$g_{21}(\eta) = 1,$$

$$g_{22}(\eta) = 1 + \eta/2,$$

$$g_{23}(\eta) = 1,$$

$$g_{24}(\eta) = 1 - 5\eta/2,$$

$$g_{25}(\eta) = (2 - 18\eta + 15\eta^2)/2. \tag{13}$$

To characterize the SA property of a material, the OA Z-scan technique is often used, in which a tightly focused TEM_{00} Gaussian beam propagates through a sample along the $+z$ direction, and a saturable absorber moves along the z axis near the focus of the Gaussian beam (the Gaussian-beam waist is defined as the coordinate origin $z=0$). In this case, the optical intensity at the incident plane of the sample I^{in} should be replaced by the following expression [4]:

$$I^{in}(x,t) = [I_0(t)/(1+x^2)]\exp[-2r^2/\omega^2(x)], \tag{14}$$

where $\omega^2(x)=\omega_0^2(1+x^2)$, $x=z/z_0$ is the sample relative position, z is the sample position, ω_0 and $z_0=\pi\omega_0^2/\lambda$ are the radius of the beam waist and the Rayleigh length, respectively, and λ is the wavelength of the used laser in vacuum. $I_0(t)$ is the instantaneous on-axis intensity of the Gaussian beam at the waist plane.

Substituting $I^{in}(x,t)$ in Eq. (14) into Eqs. (10) and (12), the intensity $I_j^{out}(x,t)$ transmitted though the saturable absorber can be easily yielded. Thus the analytic solution of the normalized transmittance as a function of the sample relative position x is given by

$$T_j(x,t) = \frac{\int_0^{\infty} I_j^{out}(x,t) r\, dr}{e^{-\alpha_0 L}\int_0^{\infty} I^{in} r\, dr} = e^{\alpha_0 L}\left[1 + \sum_{m=1}^{\infty} \frac{(-\alpha_0 L)^m}{m!} q_{jm}(\rho) \right], \tag{15}$$

where ρ is time dependent, $\rho(t)=I_0(t)/[I_S(1+x^2)]$. It is obvious that the Z-scan traces of the saturable absorber depend only on parameters $\alpha_0 L$ and ρ.

For the purpose of the convenience in applying our theory, we give the first five terms of $q_{1m}(\rho)$ for model I

$$q_{11}(\rho) = \frac{\ln(1+\rho)}{\rho},$$

$$q_{12}(\rho) = \frac{1}{2\rho}\left[1 - \frac{1}{(1+\rho)^2} \right],$$

$$q_{13}(\rho)=\frac{1}{12\rho}\left[1-\frac{1-8\rho}{(1+\rho)^4}\right],$$

$$q_{14}(\rho)=\frac{2-3\rho}{2(1+\rho)^6},$$

$$q_{15}(\rho)=\frac{1}{120\rho}\left[\frac{1+128\rho-872\rho^2+576\rho^3}{(1+\rho)^8}-1\right] \quad (16)$$

and the first five terms of $q_{2m}(\rho)$ for model II are also given as

$$q_{21}(\rho)=\frac{2}{\rho}[(1+\rho)^{1/2}-1],$$

$$q_{22}(\rho)=\frac{1}{2(1+\rho)}+\frac{\ln(1+\rho)}{2\rho},$$

$$q_{23}(\rho)=\frac{2}{5\rho}\left[1-\frac{1}{(1+\rho)^{5/2}}\right],$$

$$q_{24}(\rho)=\frac{1}{24\rho}\left[1-\frac{1-20\rho}{(1+\rho)^4}\right],$$

$$q_{25}(\rho)=\frac{1}{231\rho}\left[-2+\frac{2+242\rho-495\rho^2}{(1+\rho)^{11/2}}\right]. \quad (17)$$

For the practically performable simulations, however, the sum upper limit in Eq. (15) can never be infinite. We can find from Eq. (15) that the truncated error $\varepsilon(M)$ depends only on two parameters $\alpha_0 L$ and $I_0(t)/I_S$, where M is defined as the optimum sum upper limit. In particular, we have manifested that the maximum value of $\varepsilon(M)$, $\varepsilon_{\max}(M)$, is independent of $I_0(t)/I_S$ or $I_0(t)$

$$\varepsilon_{\max}=e^{\alpha_0 L}\left|e^{\alpha_0 L}-1-\sum_{m=1}^{M}\frac{(\alpha_0 L)^m}{m!}\right|; \quad (18)$$

this criterion is of great importance since we do not have to care for the intensity of the laser used in experiment, because the optimum sum upper limit M is determined by the linear loss parameter $\alpha_0 L$ of a measured material. In fact, $\alpha_0 L$ can be easily measured. Under the restriction of $\varepsilon_{\max}(M)\leqslant 1\%$ (this has been in fact a high precision enough), the dependence of the optimum sum upper limit M on the parameter $\alpha_0 L$ is investigated. The calculated results (open circles) in Fig. 1 shows that the optimum sum upper limit M has a nearly linear relationship with of $\alpha_0 L$. Inasmuch as M must be an integer, the choice of M should obey the solid step line in Fig. 1.

One can easily yield the general features of the OA Z scan using cw laser (or the steady-state case), implying that $I_0(t)=I_0$ and $T_j(x,t)=T_j(x)$.

When the experiment of characterizing the saturable absorption is performed by using the pulsed laser, if the nonlinear response time of the samples is much shorter than the laser pulsed width, one can assume that the nonlinear effect

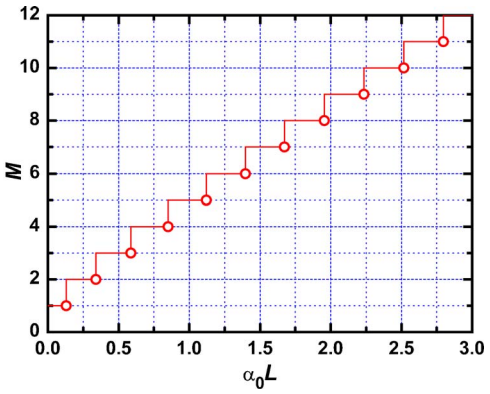

FIG. 1. (Color online) The optimal sum upper limit M as a function of $\alpha_0 L$ when the truncated error is below 1%.

depends on the instantaneous intensity inside the sample. Thus $I_0(t)=I_{00}h(t)$, where I_{00} is the on-axis peak intensity of the pulses and $h(t)$ describes the temporal profile of the laser pulse such as $h(t)=\exp[-(t/t_0)^2]$ for Gaussian and $h(t)=\mathrm{sech}^2(t/t_0)$ for hyperbolic secant squared pulses. The Z-scan expression for a pulsed laser can be obtained by

$$T_j(x)=\frac{\int_{-\infty}^{\infty}T_j(x,t)h(t)dt}{\int_{-\infty}^{\infty}h(t)dt}. \quad (19)$$

It is difficult to obtain the analytic expression of Eq. (19). However, one could finish the time integral by highly efficient Simpson arithmetic.

III. EXPERIMENT

As a test, here we explore the nonlinear absorption property of an organic sample with π-electron system, which is a side-chain azobenzene polymer (Pda) (was synthesized as described in Ref. [15]), by the OA Z-scan technique. In our experiment, Pda was a thin film sample. The thickness and linear transmission were measured to be 850 nm and 20.66% (the linear absorption factor of $\alpha_0 L=1.577$) at the wavelength of 532 nm, respectively. The strong one-photon absorption may often be expected to exhibit the SA phenomenon. The laser source is a frequency-doubled Nd:YAG (yttrium aluminum garnet) laser (a wavelength of 532 nm, a repetition rate of 10 Hz, and a pulse duration of 35 ps). The transverse distribution of the laser beam has a nearly Gaussian profile and the temporal profile is also nearly Gaussian shape. The beam was focused by a lens with a focal length of 300 mm, and the focused beam waist was estimated to be 18.5 μm (the Rayleigh range $z_0=2$ mm). Our experiments were performed at different levels of $I_{00}=1.90$, 2.50, 3.75, 5.90, and 7.35 GW/cm^2. As an example, Fig. 2 shows a Z-scan trace (open circles) measured in an experiment at $I_{00}=2.50$ GW/cm^2. The experimental results have demonstrated that Pda could indeed behave as a saturable absorber.

Now we will evaluate the important parameter of the Pda film (the saturable intensity I_S) and attempt to interpret its SA behavior. The high efficient yet feasible method is to fit the

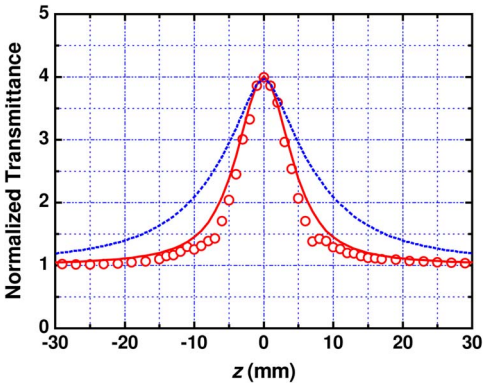

FIG. 2. (Color online) An example of the OA *Z*-scan trace (open circles) of the Pda film at $I_{00}=2.50$ GW/cm^2. The solid and dotted curves are the theoretical fittings for models I and II, respectively.

OA *Z*-scan traces we measured in experiments by Eq. (19). Since the linear loss factor of the Pda film is $\alpha_0 L=1.577$, we can determine from Fig. 1 that if we take the sum upper limit $M=7$ in Eq. (19) for fitting, the error is certainly lower than 1%. We use the two SA models described in Eqs. (2) and (3) to fit our experimental OA *Z*-scan trace (open circles) displayed in Fig. 2, with only one adjustable parameter (I_S). We give a comparison of the profiles of the theoretical OA *Z*-scan traces fitted by models I, and II in Fig. 2, as the solid and dotted curves, respectively. The fittings give the respective I_S to be $I_S=51.6$ and 5.82 MW/cm^2, for models I and II, respectively. It is very clear that the profile of the OA *Z*-scan trace fitted by model I is in good arrangement with the experimental data. In addition, all the OA *Z*-scan traces measured at the other different levels of I_{00} ranging from 1.90 to 7.35 GW/cm^2 can be also well reproduced by use of model I, the resulting values of I_S are within a range of $I_S=51.6\pm0.2$ MW/cm^2. The results imply certainly that the SA behavior of the Pda film obeys model I, instead of model II, and no reverse saturable absorption phenomena are observed. We know that model II can describe the SA behavior in the inhomogeneous broadening two-level systems well, such as the gas atom and molecule systems with the Doppler effect. However, for the solid Pda film, the Doppler effect is never significant, so we easily understand the reason why the SA behavior disobeys model II. It is well known that model I could describe the SA effect in an homogeneous broadening two-level system very well. However, we also know that for the organic Pda molecule as a polyatomic molecule system, its level has a complicated structure (a simplified five-level model [1], including a ground singlet state S_0, a first excited singlet state S_1, a higher-lying singlet state S_2, a first excited triplet state T_1, and a higher-lying triplet state T_2, is extensively accepted) instead of a simple two-level system. In the Pda film, the strong SA and no reverse SA implies that the absorption cross section of S_0 is much larger than both absorption cross sections of S_1 and T_1; that is to say, the dominant absorption transition happens from S_0 to S_1.

IV. CONCLUSION

On the basis of the AD method, for the two SA models, we have given the analytic formula in the terms of the infinite polynomial for the OA Gaussian-beam *Z*-scan trace. For performable simulations, we found the optimum sum upper limit, which allows us to straightforwardly fit the measured *Z*-scan traces in experiments and then to extract the saturable intensity from a single OA *Z*-scan trace. As an example, we explore the SA property of a polyatomic molecule system (Pda). Our experimental results indicate the Pda film to behave as a saturable absorber at the wavelength of 532 nm.

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