Quantitative deviation of the two-photon absorption coefficient based on three laser pulse models

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The pulse profile influence of excitation light on the two-photon absorption coefficient β is theoretically and numerically studied. Based on Gaussian spatial and temporal laser, we obtain an expansion formula of energy transmission. As compared with a plain beam and a pulse beam that is rectangular in time but Gaussian in space, the relative deviations of β turn out to be about 214% and 47%, respectively. These differences indicate that a smaller β may be obtained than the real one in usual nonlinear transmission. Our result suggests that by taking real pulse profile into account, a more exact β can be derived in energy transmission measurement.

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The two-photon-induced behavior of organic materials has received increasing attention in recent years due to its applications in two-photon fluorescent microscopy and imaging[1−3], optical limiting of two-photon absorption (TPA)[4−9], three-dimensional (3D) optical data storage[10−12], fabrication of photonic crystal[13], and two-photon photodynamic therapy[14,15]. The wavelength used for two-photon excitation is roughly twice that for one-photon excitation, so the influences of scattering on the beam intensity $(1/\lambda^4)$ and photodamage to a healthy tissue can be greatly reduced. In the usual TPA process, ultra-short pulse lasers are often used as the two-photon excitation source because of their high peak power, which gives rise to the high transition probability of TPA with a comparatively small average power[1,16].

The material properties of TPA can be described partly but very importantly by the TPA cross-section of a molecule (σ_2). Therefore, a lot of efforts[17−37] have been exerted to examine it experimentally and theoretically. There are many methods, such as nonlinear transmission, Z-scan, and two-photon-induced fluorescent comparison, to obtain molecular absorption cross-section experimentally. In these measurements, nonlinear energy transmission is often performed mainly for its easier setup.

Boggess *et al.*[4,38] presented the experimental principle of nonlinear transmission. According to the principle of the TPA process, the beam intensity change along the propagation direction (z axis) can be described as

$$\mathrm{d}I/\mathrm{d}z + \alpha I + \beta I^2 = 0, \tag{1}$$

where α is the attenuation coefficient due to linear absorption and scattering, and β is the nonlinear absorption coefficient due to TPA. The excitation irradiance $I = I(r, z, t)$ is a function of time t and the transverse position r as well as z.

Assume that there is a uniform transverse intensity distribution in the beam section, one of which is a plain beam; for example, in the case of $\alpha z \ll 1$, the transmission solution of Eq. (1) is[39]

$$T(z) = I(z)/I(0) = \frac{\mathrm{e}^{-\alpha z}}{1 + \beta z I_0} = \frac{T_{\mathrm{lin}}}{1 + \beta z I_0}$$
$$= T_{\mathrm{lin}} \cdot T^p_{\mathrm{nonlin}}, \tag{2}$$

where

$$T^p_{\mathrm{nonlin}} = \frac{1}{1 + \beta z I_0}, \tag{3}$$

$I_0 = I(0)$ is the initial intensity, T_{lin} is the linear transmittivity independent of I_0, and T^p_{nonlin} is the nonlinear transmission dependent on I_0.

A focused pulse laser is more frequently used to reach a high-intensity level for nonlinear absorption measurements. In this case, a Gaussian transverse distribution in the nonlinear medium can be assumed, and Eq. (3) should be modified. For a pulse laser that is rectangular in time but Gaussian in space, Eq. (3) can be rewritten as[4]

$$T^G_{\mathrm{nonlin}} = \frac{\ln(1 + \beta z I_0)}{\beta z I_0}. \tag{4}$$

Based on Eq. (4), the frequency-dependent TPA coefficient β (in units of cm/GW) of the sample can be determined by measuring the nonlinear transmissivity T^G_{nonlin} for a given input intensity I_0 and a given sample thickness $z = L$.

Equation (4) is often used as the basic formula to obtain the TPA coefficient of some materials in nonlinear transmission measurements. However, to enhance nonlinear absorption, ultra-short pulse lasers are often used nowadays as the excitation sources. In this situation, the temporal profile of a real laser pulse is usually not rectangular. On the other hand, Oulianov *et al.*[40] indicate that under certain conditions, the nonlinear transmission method, without consideration of other nonlinear effects, results in erroneous values for the TPA cross-sections.

Therefore, we believe that the detailed deviations of β based on different laser models may be useful in understanding the problem.

In this letter, we attempt to give a quantitative description of this deviation based on the assumption of a Gaussian spatial and temporal profile for an excitation pulse laser beam. The energy transmission is derived and discussed in a typical nonlinear measurement. For comparison with a plain beam and a Gaussian spatial and rectangular temporal pulse beam, the absolute and relative deviations of β are theoretically estimated based on linear approximation. Further numerical simulations are also performed in general. Finally, a proposed way to obtain an exact β is also discussed in nonlinear transmission measurement.

We initially assume α to be a constant for a given wavelength λ. Equation (1) can be solved exactly to give the transmitted irradiance $I(r, L, t)$ in terms of the incident irradiance $I(r, 0, t)$[38]:

$$I\left(r,L,t\right) = T_0 I\left(r,0,t\right) / \left[1 + I\left(r,0,t\right)/I_{c2}\right], \quad (5)$$

where

$$T_0 = \left(1 - R_1\right)\left(1 - R_2\right) e^{-\alpha L} \quad (6)$$

is the linear transmission of a sample with a thickness L and a front (rear) surface reflectivity R_1 (R_2), I_{c2} is defined as

$$I_{c2} = (\alpha/\beta) / \left[\left(1 - R_1\right)\left(1 - e^{-\alpha L}\right)\right], \quad (7)$$

the factor $(1-R_1)$ accounts for the front surface reflection and $(1-e^{-\alpha L})$ is the fractional linear absorption for a sample with thickness L. For irradiance approaching the critical irradiance for TPA, the transmission deviates from linear absorption to TPA and the nonlinear absorption becomes comparable to a linear one.

However, the actual quantity typically measured in a pulsed experiment is the transmitted energy, not the irradiance. In this case, through the spatial and temporal integral, the transmitted energy can be expressed as

$$E_l = \int_0^\infty \int_{-\infty}^\infty 2\pi r \mathrm{d}r \mathrm{d}t I\left(r, L, t\right). \quad (8)$$

Similarly, the initial energy can also be expressed as

$$E_0 = \int_0^\infty \int_{-\infty}^\infty 2\pi r \mathrm{d}r \mathrm{d}t I\left(r, 0, t\right). \quad (9)$$

The spatial integral over the transverse coordinates and the temporal integral over the pulse time can be performed for specific spatial and temporal profiles. For a Gaussian spatial and temporal profile where

$$I\left(r,0,t\right) = I_0 e^{-\left(\frac{r}{r_0}\right)^2} e^{-\left(\frac{t}{t_0}\right)^2}, \quad (10)$$

after the spatial integral, the energy transmission can be written formally as

$$T = \frac{E_l}{E_0} = \frac{T_0 I_{c2}}{I_0 t_0 \sqrt{\pi}} \int_{-\infty}^\infty \ln\left(1 + \frac{I_0}{I_{c2}} e^{-\left(\frac{t}{t_0}\right)^2}\right) \mathrm{d}t, \quad (11)$$

where r_0 is the l/e radius and t_0 is the l/e half-width of the spatial and temporal irradiance profiles, respectively. Physically, I_0/I_{c2} grossly compares the linear absorption coefficient α to the TPA coefficient β.

According to Tayler's expansion, after the temporal integral of Eq. (11), we can obtain the energy transmission

$$T = T_0 \left\{\sum_{k=0} \frac{\left(-I_0/I_{c2}\right)^k}{(k+1)\sqrt{(k+1)}}\right\}. \quad (12)$$

Equation (12) may be used as an available formula to obtain β under the excitation of a Gaussian pulse laser, if I_{c2} can be expressed in terms of βL in a dedicated experiment of nonlinear transmittance.

A typical setup for the measurement of two-photon-induced nonlinear transmittance is shown in Fig. 1[36,37]. The input intensity of the beam can be changed by turning the left Nicol's prism in Fig. 1. The laser beam was split by a beam splitter (BS) into two beams. The weaker one was a reference measured by detector D_1 of a two-channel laser energy meter in order to monitor the intensity of the excitation beam. The stronger one was focused on the sample (S) by the lens (L). The transmitted energy was recorded using the other detector (D_2) of the same energy meter. The influence of the solvent and cell walls was eliminated by alternative measurement of the transmitted beam energy (E_l) through the sample and the energy through the pure solvent (E_0) in the same cell. Furthermore, if the irradiance wavelength is far away from the linear absorption band of the measured material, where $\alpha L \ll 1$, we can assume that $R_1 \approx 0 \approx R_2$ and $I_{c2} \approx 1/\beta L$.

If the measurement of β is based on the way shown in Fig. 1, Eqs. (11) and (12) can be formally rewritten, respectively, as the following expressions:

$$T = \frac{E_l}{E_0} = \frac{T_0}{I_0 \beta L t_0 \sqrt{\pi}}$$
$$\int_{-\infty}^\infty \ln\left(1 + \beta L I_0 e^{-\left(\frac{t}{t_0}\right)^2}\right) \mathrm{d}t, \quad (13)$$

and

$$T_n = T_0 \left\{\sum_{k=0}^n \frac{\left(-\beta L I_0\right)^k}{(k+1)\sqrt{(k+1)}}\right\}$$
$$= T_0 \left\{1 - \frac{1}{2\sqrt{2}}\beta L I_0 + \frac{1}{3\sqrt{3}}(\beta L I_0)^2\right.$$
$$\left. - \frac{1}{4\sqrt{4}}(\beta L I_0)^3 + \frac{1}{5\sqrt{5}}(\beta L I_0)^4 + \cdots\right\}, \quad (14)$$

where

$$T_0 = \left(1 - R_1\right)\left(1 - R_2\right) e^{-\alpha L} \cong e^{-\alpha L} = T_{\text{lin}}. \quad (14a)$$

For further discussion, we define

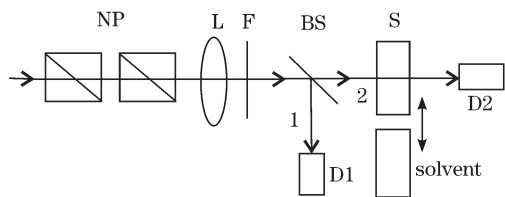$$T_1 \approx T_0 (1 - \frac{1}{2\sqrt{2}}\beta L I_0), \quad (14b)$$

Fig. 1. Experimental setup for TPA transmission measurement. NP: Nicol's prism; L: lens; F: filter; BS: beam splitter; S: sample; D1, D2: detectors of a two-channel laser energy meter.

$$T_2 \approx T_0 \left\{ 1 - \frac{1}{2\sqrt{2}} \beta L I_0 + \frac{1}{3\sqrt{3}} (\beta L I_0)^2 \right\}, (14c)$$

$$T_3 \approx T_0 \left\{ 1 - \frac{1}{2\sqrt{2}} \beta L I_0 + \frac{1}{3\sqrt{3}} (\beta L I_0)^2 \right.$$
$$\left. - \frac{1}{4\sqrt{4}} (\beta L I_0)^3 \right\}, \tag{14d}$$

$$T_4 \approx T_0 \left\{ 1 - \frac{1}{2\sqrt{2}} \beta L I_0 + \frac{1}{3\sqrt{3}} (\beta L I_0)^2 \right.$$
$$\left. - \frac{1}{4\sqrt{4}} (\beta L I_0)^3 + \frac{1}{5\sqrt{5}} (\beta L I_0)^4 \right\},$$
$$\cdots , \tag{14e}$$

respectively, where T_1 denotes the linear approximation of energy transmission, T_n ($n \geq 2$) represents the nonlinear term of T.

Furthermore, if the concentration C_0 (in units of mol/L) of the solute or dopant is known, the molecular TPA cross-section σ_2 (in units of cm^4/GW) or σ'_2 (in units of cm$^4 \cdot$s/photon) can be determined by[36,37]

$$\beta = \sigma_2 N_A C_0 \times 10^{-3}, \tag{15}$$

and

$$\sigma'_2 = h\nu\sigma_2 , \tag{16}$$

where $N_A = 6.023 \times 10^{23}$ is the Avogadro number, and ν is the frequency of excitation light.

Equation (14) may become a reference formula, based on which β can be obtained exactly in usual nonlinear energy transmission under the excitation of a Gaussian spatial and temporal pulse laser.

For comparison, we outline the curves of Eqs. (13) and (14) in Fig. 2, where $L = 1$ cm and $n = 50$. Figure 2 indicates that in the case of $\beta L I_0 < 1$, there is no difference between Eqs. (13) and (14). It also shows that energy transmission is approximately a linear function of the incidence intensity I_0 in the range of $\beta L I_0 < 0.1$.

According to Eqs. (8) and (9), energy transmission depends not only on the spatial profile but also on the temporal profile of an excitation laser. There are certain differences in the energy transmission (T) of different excitation pulse lasers. In other wards, regarding β for a given nonlinear transmission, very different values
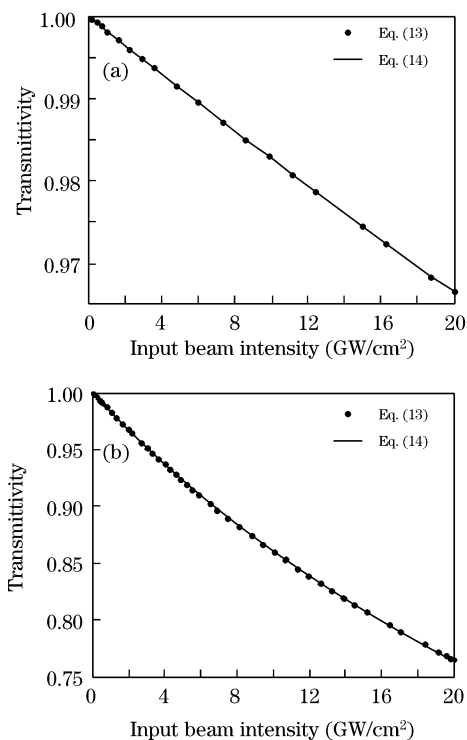

Fig. 2. Curves of Eqs. (12) and (13) with $L = 1$ cm and (a) $\beta = 0.005$ cm/GW, (b) $\beta = 0.05$ cm/GW.
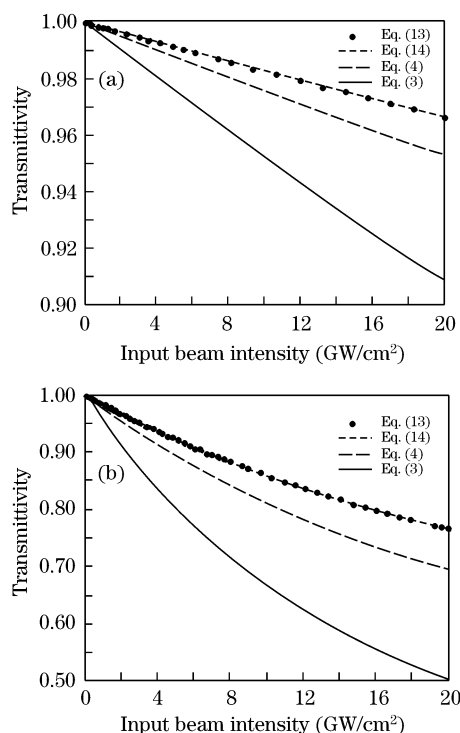

Fig. 3. Curves of Eqs. (3), (4), (13), and (14) at (a) $\beta = 0.005$ cm/GW and (b) $\beta = 0.05$ cm/GW.

of β may be obtained on the assumptions of the excitation laser as a plain beam, a Gaussian spatial and rectangular temporal beam, and a Gaussian spatial and temporal beam. The quantitative differences in the TPA coefficient are further described in the follows.

Notice that in Eq. (14) for a Gaussian spatial and

temporal pulse beam, Eq. (4) for a pulse beam that is rectangular in time but Gaussian in space, and Eq. (3) for a plain beam, there are clear differences in β from the three equations, as shown in Fig. 3. The outlines in Fig. 3 are based on Eqs. (14), (4), and (3), respectively, for a given β of energy transmission. These differences can tell us that if Eqs. (3), (4), and (14) (or (13)), respectively, are employed to fit a set of experimental data on nonlinear energy transmission, very different values of β (or σ_2) would be obtained. The detailed differences can also be approximately estimated in theory.

In the range of $\beta L I_0 \leq 0.1$, energy transmission is approximately a linear function of $\beta L I_0$ which is similar to the upper curves shown in Fig. 3. Based on the assumption of a linear relationship, we estimate the differences resulting from Eqs. (14), (4), and (3) as follows.

Based on linear approximation, Eqs. (14), (4), and (3) can be expressed, respectively, as

$$T_1 \approx T_0(1 - \frac{1}{2\sqrt{2}}\beta_0 L I_0), \tag{17}$$

$$T_0 \frac{\ln(1 + I_0 z \beta_1)}{I_0 z \beta_1} \approx T_0(1 - \frac{1}{2}\beta_1 L I_0), \tag{18}$$

$$T_0 \frac{1}{1 + \beta_2 L I_0} \approx T_0(1 - \beta_2 L I_0). \tag{19}$$

Comparing Eqs. (17), (18), and (19), if we order

$$\beta_1 = \beta_0/\sqrt{2} \quad , \tag{20}$$

and

$$\beta_2 = \beta_0/(2\sqrt{2}), \tag{21}$$

the curves of Eqs. (17)−(19) will become the same line. This means that if Eq. (18) is used to fit an experimental energy transmission result, the value of β_1 obtained is about 0.707 times that of β_0, and the value of β_2 obtained using Eq. (19) is only 0.354 times that of β_0. These differences indicate that the TPA coefficient value based on Eq. (14) is larger than those based on Eqs. (4) and (3) for the same energy transmission of a nonlinear measurement.

The absolute and relative deviations between Eqs. (17) and (18) are

$$\alpha_1 = \beta_0 - \beta_1 \approx 0.293\beta_0 \quad , \tag{22}$$

and

$$\delta_1 = \frac{\beta_0 - \beta_1}{\beta_1} \approx 41.4\%. \tag{23}$$

The related deviations between Eqs. (17) and (19) are

$$\alpha_2 = \beta_0 - \beta_2 \approx 0.646\beta_0 \quad , \tag{24}$$

and

$$\delta_2 = \frac{\beta_0 - \beta_2}{\beta_2} \approx 182.8\%. \tag{25}$$

According to Eqs. (15) and (16), the relative deviations of the molecular TPA cross-section σ_2 and σ_2' are 41.4% and 182.8%, respectively.

Moreover, if we take the high order terms of energy transmission into account, after numerical simulation, we can find that the relative differences are even larger. The detailed numerical results are shown in Figs. 4, 5, and Table 1, respectively.

In our numerical performance analysis, we set the curve of Eq. (14) as reference. That is, the energy transmission data of some materials are assumed to be closely fitted to Eq. (14). In this way, we can obtain the difference in detail between Eqs. (14) and (4) (or Eq. (3)).

Figure 4 and Table 1 present the numerical simulation results between Eqs. (14) and (4). Including the high order terms of energy transmission, the simulating lines show that the relative deviation between Eqs. (14) and (4) becomes larger with the increase in nonlinear term contribution (or input beam intensity). This result means that the pulse profile influence of an excitation laser on β becomes considerably large with the increase in nonlinear energy transmission. The difference in data between Eqs (14) and (4) in Table 1 indicates that if Eq. (4) is used to obtain the TPA coefficient, the deviation
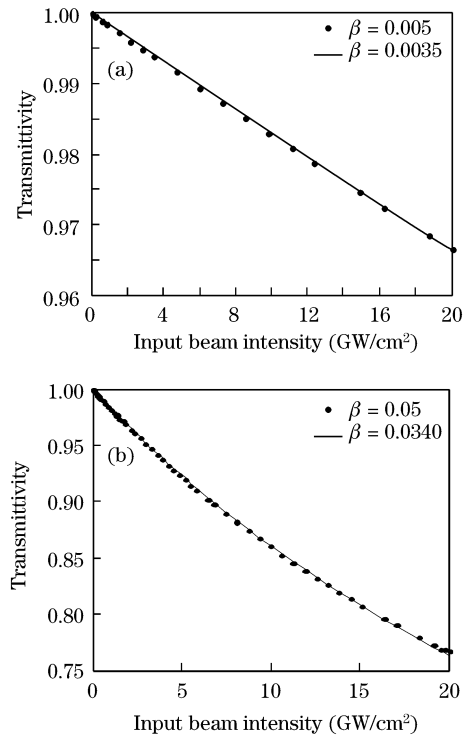

Fig. 4. Typical simulated curves of Eq. (14) using Eq. (4).

Table 1. Relative Deviations of Eqs. (4) and (3) Based on Eq. (14)

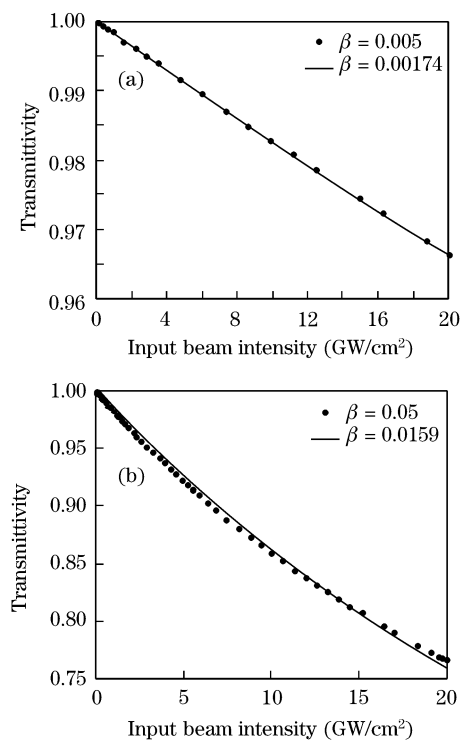| β_0 (×10⁻³ cm/GW)	1.0	3.0	5.0	8.0
$\delta_1(\%)$	41.64	41.84	42.17	42.58
$\delta_2(\%)$	183.29	185.17	186.70	188.91
β_0 (×10⁻³ cm/GW)	10.0	20.0	40.0	50.0
$\delta_1(\%)$	42.84	44.06	46.11	47.01
$\delta_2(\%)$	190.36	197.13	208.88	214.07

Fig. 5. Typical simulated curves of Eq. (14) using Eq. (3).

of β is about 47% relative to Eq. (14).

The same situation exists between Eqs. (14) and (3), as shown in Fig. 5 and Table 1. The simulation results indicate that the absolute and relative deviations are larger than those of Eqs. (22) and (23). The value of β obtained based on Eq. (3) would deviate largely from the true one if the excitation source is a pulse laser.

Generally, for the purpose of enhancing nonlinear absorption and reducing the excited-state absorption of some nonlinear media, a good choice is to use an ultra-short pulse laser as the excitation source. In usual TPA performance analysis, ultra-short pulse beams are used with the focus of the lens. In this case, the temporal profile of a real pulse may often not be rectangular. Therefore, the pulse profile influence on β should be taken into account when dealing with the TPA of nonlinear energy transmission. In other words, it is important to give the exact description of the pulse profile aside from the space profile in usual TPA performance analysis for β.

Although a real pulse may not be exactly Gaussian, the Gaussian model is a well-known approximation for a real pulse. In this situation, Eq. (14) may be a suitable equation, based on which a more exact TPA coefficient β can be obtained for a given experimental result of nonlinear energy transmission measurement. In this way, the real TPA cross-section of some molecule may be determined by way of nonlinear transmission performance.

There are some typical profiles of laser pulse, such as Gaussian monocycle, Scholtz's monocycle, and Gaussian doublet-pulse. These pulse profiles' influences on β are an interesting topic, and relative research in this area is in progress.

In conclusion, it is found that there are clear differences in β based on the assumptions of a plain beam, a Gaussian spatial and rectangular temporal pulse beam, and a Gaussian spatial and temporal pulse beam for an excitation light. The pulse profile influence on the TPA coefficient cannot be neglected in the usual process of energy transmission measurements. Since the Gaussian temporal profile, relative to other profiles, is closer to a real pulse, Eq. (14) may be a suitable formula, through which a more exact value of the TPA cross-section (σ_2) of the molecule can be obtained. Moreover, the results also suggest that greater importance should be given on the pulse profile of excitation light in usual nonlinear performance analysis. By taking both spatial and temporal influences of an excitation laser into account, the real β value of some materials in a designed nonlinear transmission measurement can be determined.

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