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# Method for measurements of second-order nonlinear optical coefficient based on *Z*-scan

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**Abstract**

The method for measuring second-order nonlinear optical coefficients based on well-known *Z*-scan is presented. The influence of linear absorption coefficients on normalized transmittance is discussed. Using this method, we obtained the second-order nonlinear coefficient $d_{31}(5\%\text{MgO:LiNbO}_3) = 4.5 \times 10^{-12}$ m/v at 1064 nm, which agrees well with theoretical calculations and previous well-known values.


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*Keywords:* Second-order nonlinear; *Z*-scan

## 1. Introduction

The second-order nonlinear optical coefficient plays a key role in the performance of nonlinear optical devices such as frequency doublers and parametric oscillators. In recent years, there has been growing interest in second-order optical nonlinear effects in organic materials [1,2], because a large nonlinear phase shift arises from cascading of second-order processes. Accurate values of second-order nonlinear optical coefficients are also of interests for theoretical considerations since they are important to search for new materials to improve performance of nonlinear optical devices.

Absolute second-order nonlinear optical coefficients can usually be measured by phase matched method (PM) [3] and parametric fluorescence (PF) [4]. The PM is a convenient method but requires precise absolute measurements of the fundamental and second-harmonic (SH) light power. Meanwhile, the signal power generated in the PF may be very weak, and a finite spectral distribution of signal power needs to be measured. The common methods used require relative measurements, the Maker fringe method (MF) [5] as described in detail by Jerphagnon and Kurtz [6] and reviewed by Herman and Michael Hayden [7], has almost exclusively been used to determine second-order nonlinear optical coefficients. However, it requires a well-characterized reference sample that one can trust.

In this paper, a method for measuring second-order nonlinear optical coefficient based on *Z*-scan is presented. The obvious difference from other methods is that the method is to measure fundamental light, not SH light or PF. The influence of the linear absorption coefficients on the normalized transmittance is also discussed. By this method we got second-order nonlinear optical coefficient $d_{31}$ of 5%MgO:$LiNbO_3$ crystal at 1064 nm, which agrees

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well with theoretical calculation. Finally, we compared our result with well-known values, and result shows that our result agrees well with those.

## 2. Theory

The $Z$-scan technique [8,9] becomes very popular for measurements of nonlinear refraction and nonlinear absorption because of its simplicity and high accuracy. It has been extensively used to measure nonlinear optical properties of materials [10,11]. In the $Z$-scan technique, a sample is moved along the optic axis through the focus of a single laser beam, while the energy transmitted through an aperture in the far field is recorded as a function of sample position. The technique includes open aperture $Z$-scan and close aperture $Z$-scan.

Assuming a fundamental $TEM_{00}$ Gaussian beam traveling in the $+z$ direction, we can write $E$ as [9]

$$E(z,r,t) = E_0(t)\frac{\omega_0}{\omega(z)}\exp\left[-\frac{r^2}{\omega^2(z)} - \frac{\mathrm{i}kr^2}{2R(z)}\right]\exp[-\mathrm{i}\phi(z,t)], \tag{1}$$

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

For a paraxial tiny annular element of area, $\mathrm{d}S = 2\pi r\,\mathrm{d}r$, the electric field on this area can be considered as a plane wave. Using the plane wave theory, we can obtain the second-harmonic generation (SHG) conversion efficiency [12] including the effect of linear absorption under nondepleted pump approximation and thin medium limit corresponding to the case that the nonlinear medium length $L$ is much shorter than the diffraction length of beam $z_0$ [8]

$$\eta_{2\omega}(r,z,t) = \frac{\eta^0_{2\omega}(r,t)}{1+\bar{z}^2}\exp[-(\alpha_\omega+\alpha_{2\omega}/2)L]C(\Delta k,\alpha_\omega,\alpha_{2\omega}) \tag{2a}$$

with

$$\eta^0_{2\omega}(r,t) = \frac{8\pi^2 d_{\mathrm{eff}}^2 L^2 I^0_\omega \exp(-2r^2/\omega^2)\exp(-t^2/\tau^2)}{\varepsilon_0 n_\omega^2 n_{2\omega} c\lambda_\omega^2}, \tag{2b}$$

$$C(\Delta k,\alpha_\omega,\alpha_{2\omega}) = \frac{\sin^2(\Delta kL/2)+\sinh^2[(\alpha_\omega-\alpha_{2\omega}/2)L/2]}{(\Delta kL/2)^2+[(\alpha_\omega-\alpha_{2\omega}/2)L/2]^2}, \tag{2c}$$

where $d_{\mathrm{eff}}$ is the effective second-order nonlinear optical coefficient, $L$ is the nonlinear medium length, $I^0_\omega$ is the fundamental on-axis intensity in vacuum, $\tau$ is the pulse width, $\varepsilon_0$ is the electric permittivity of free space, $n_\omega$ and $n_{2\omega}$ are the refractive indexes of fundamental and SH light, $c$ is the velocity of light in vacuum, $\alpha_\omega$ and $\alpha_{2\omega}$ are the linear absorption coefficients of fundamental and SH light, $\Delta k = 2k_\omega - k_{2\omega}$ represents the wave-vector mismatch, and $\bar{z} = z/z_0$.

Then, we can obtain the SHG light intensity through $\mathrm{d}S$ by Eq. (2a) as $\mathrm{d}(I_{2\omega}(\mathrm{SHG})) = \eta_{2\omega}(r,z,t)I_\omega\cdot(\mathrm{d}S)$. Considering the existence of linear absorption for SH light, the total SH light intensity through $\mathrm{d}S$ generated in the nonlinear medium can be written as

$$\mathrm{d}(I_{2\omega}(\mathrm{Total})) = \mathrm{d}(I_{2\omega}(\mathrm{SHG})) + \mathrm{d}(I_{2\omega}(\mathrm{Abs})), \tag{3a}$$

$$\mathrm{d}(I_{2\omega}(\mathrm{Abs})) = \mathrm{d}(I_{2\omega}(\mathrm{Total}))\cdot[1-\exp(-\alpha_{2\omega}L)], \tag{3b}$$

where $\mathrm{d}(I_{2\omega}(\mathrm{Abs}))$ is the absorbed part of SH intensity through $\mathrm{d}S$. Accordingly, the total SHG intensity through $\mathrm{d}S$ can be obtained

$$\mathrm{d}(I_{2\omega}(\mathrm{Total})) = \frac{\mathrm{d}(I_{2\omega}(\mathrm{SHG}))}{\exp(-\alpha_{2\omega}L)}. \tag{4}$$

Therefore, the normalized transmittance $T(z)$ of fundamental light can be written as

$$\begin{aligned} T(z) &= \frac{I_{\omega,\mathrm{out}}}{I_{\omega,\mathrm{in}}}, \\ &= \frac{\int_0^\infty\int_{-\infty}^\infty I_\omega\exp(-\alpha_\omega L)2\pi r\,\mathrm{d}r\,\mathrm{d}t - \int_0^\infty\int_{-\infty}^\infty I_{2\omega}(\mathrm{Total})2\pi r\,\mathrm{d}r\,\mathrm{d}t}{\int_0^\infty\int_{-\infty}^\infty I_\omega\exp(-\alpha_\omega L)2\pi r\,\mathrm{d}r\,\mathrm{d}t}, \\ &= 1-\frac{1}{2\sqrt{2}}\frac{\eta^{00}_{2\omega}}{1+\bar{z}^2}\exp(\alpha_{2\omega}L/2)C(\Delta k,\alpha_\omega,\alpha_{2\omega}) \end{aligned} \tag{5a}$$

with

$$\eta^{00}_{2\omega} = \frac{8\pi^2 d_{\mathrm{eff}}^2 L^2 I^0_\omega}{\varepsilon_0 n_\omega^2 n_{2\omega} c\lambda_\omega^2}. \tag{5b}$$

Fig. 1 gives the $Z$-scan curves for different $L$. It is seen that the nonlinear medium length $L$ is important since the normalized transmittance $T(z)$ is approximately proportional to $L^2$. In order to increase $T(z)$, a weak focusing configuration is suggested since it can permit larger medium length while thin sample approximation is satisfied.

Generally, one cannot radically eliminate the effect of linear absorption coefficient of fundamental light on

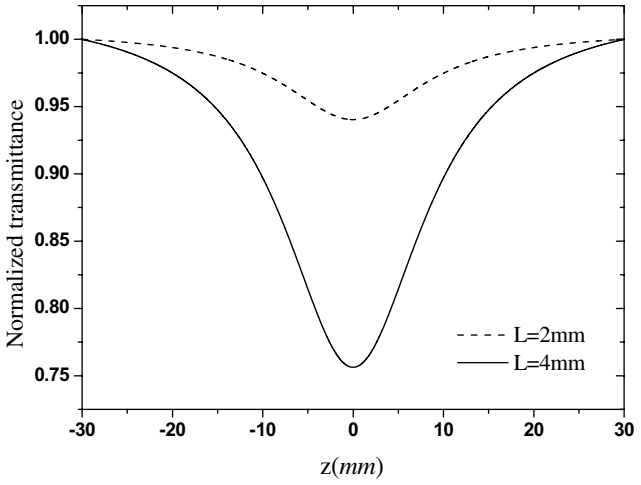


Fig. 1. The $Z$-scan curves for different $L$.

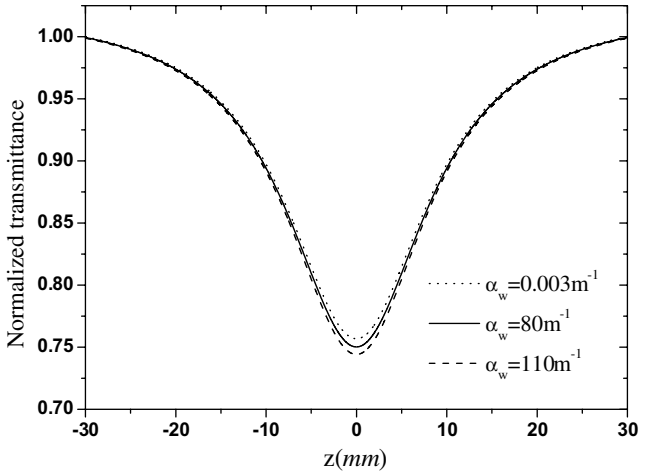


Fig. 2. The $Z$-scan curves for different linear absorption coefficients of fundamental light, where $E_{in} = 40\ \mu J$, $\omega_0 = 38 \mu m$, $L = 4$ mm, and $\Delta kL = 3.8$.

SHG even though it is very small and is often masked by optical scattering. The $Z$-scan curves for different linear absorption coefficients of fundamental light are given in Fig. 2, where linear absorption coefficients of fundamental light are set to be 0.003, 80 and 110 $m^{-1}$, respectively. The results show that linear absorption coefficient of fundamental light almost does not affect the $Z$-scan curves. Therefore, the effect of linear absorption coefficient of fundamental light can be neglected for determinations of second-order nonlinear coefficients. Meanwhile, $Z$-scan curve is also insensitive to linear absorption coefficient $\alpha_{2\omega}$ of SH light. The $Z$-scan curves for different linear absorption coefficients of SH light are given in Fig. 3, where linear absorption coefficients of SH light are set to be 1, 20 and 40 $m^{-1}$, respectively. We can see that shape of $Z$-scan curve changes slightly when linear absorption coefficient of SH light changes greatly. Therefore, linear absorption coefficients of media do not need to be exactly measured when we measure the second-order nonlinear coefficients by this method.

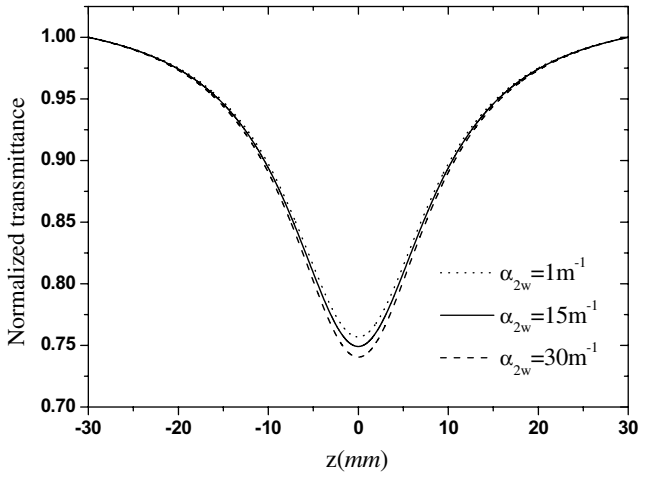


Fig. 3. The $Z$-scan curves for different linear absorption coefficients of SH light, where $E_{in} = 40\ \mu J$, $\omega_0 = 38\ \mu m$, $L = 4$ mm, and $\Delta kL = 3.8$.

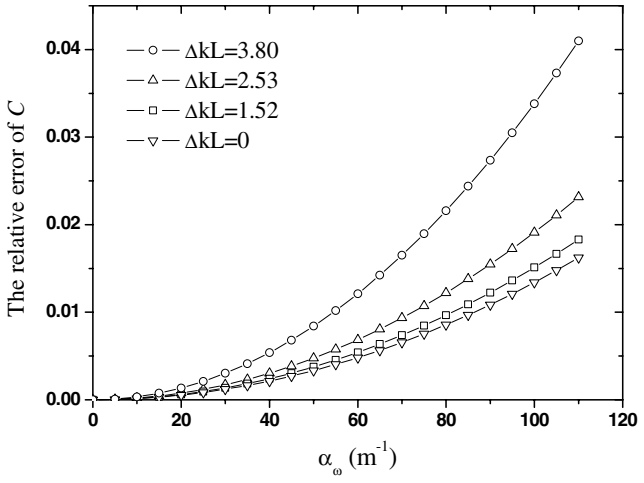


Fig. 4. The relative error curves of $C(\Delta k, \alpha_\omega, \alpha_{2\omega})$ as a function of $\alpha_\omega$ for different $\Delta kL$. $\Delta kL$ are set to be 0, 1.52, 2.53, and 3.80, respectively.

When $\alpha_{2\omega}L$ is little, $\exp(\alpha_{2\omega}L/2) \approx 1$ will exist, and the influence of linear absorption on normalized transmittance is mainly attributed to $C(\Delta k, \alpha_\omega, \alpha_{2\omega})$ in Eq. (5a). Using taylor expansion, $C(\Delta k, \alpha_\omega, \alpha_{2\omega})$ can be reduced to following:

$$C(\Delta k, \alpha_\omega, \alpha_{2\omega}) \approx \sin c^2(\Delta kL/2)\left[1 + \frac{x^2}{\sin^2(\Delta kL/2)} - \frac{x^2}{(\Delta kL/2)^2} + \frac{x^4}{6\sin^2(\Delta kL/2)}\right], \quad (6)$$

where $x = (\alpha_\omega - \alpha_{2\omega}/2)L/2$.

When $\Delta kL = 3.8$, $x^2/\sin^2(\Delta kL/2) - x^2/(\Delta kL/2)^2 \ll 1$ will exist, $T(z)$ depends very little on $\alpha_\omega$ and $\alpha_{2\omega}$. The relative error curves of $C(\Delta k, \alpha_\omega, \alpha_{2\omega})$ as a function of $\alpha_\omega$ for different $\Delta kL$ are given in Fig. 4, where $\Delta kL$ are set to be 0, 1.52, 2.53, and 3.80, respectively. Results also show that the relative error of $C(\Delta k, \alpha_\omega, \alpha_{2\omega})$ is very little and increases with $\alpha_\omega$, but stabilizes gradually as $\Delta kL$ decreases while keep $\alpha_\omega$ unchanged. The values of $\alpha_\omega$ and $\alpha_{2\omega}$ used in Figs. 2 and 3 have far exceeded actual values. In fact, absorption coefficients of practical materials will not change so greatly. Therefore, the error caused by the change of the absorption coefficients should be very little.

## 3. Experiments and discussions

Fig. 5 demonstrates the $Z$-scan experimental setup used in our experiments. A frequency-doubled Continuum Surelite **II**-10 Q-switch Nd:YAG laser with a 6-ns pulse width and a 10-Hz repetition frequency at 1064 nm, was used as fundamental light in our experiments. A half-wave plate placing before a focusing lens was used to adjust the polarization of the incident beam. The fundamental light was focused to produce a beam waist radius $\omega_0$ of 38 μm (HWHM in irradiance). The sample was translated near by the beam waist along $z$ direction. The SH light behind the sample was filtered out from output light by a harmonic separator, and an IR-pass filter was used to elimi-

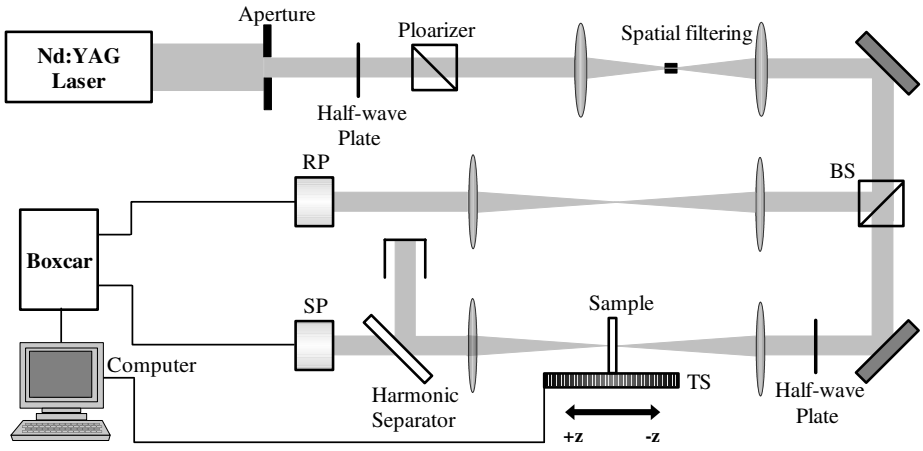


Fig. 5. The $Z$-scan experimental setup. BS, beam splitter; SP, signal probe; and RP, reference probe.

nate the SH light partially transmitted through the harmonic separator. The fundamental light energy was measured by a Molectron J3S-10 energymeter. A small part of input beam obtained by a beam splitter was used to monitor the pulse-to-pulse energy fluctuation. At every sample position, we took an average of 50 transmittance data corresponding to 50 incident pulses with less than $\pm 5\%$ energy fluctuation.

The sample used in our experiments was uncoated (1 1 0) cut 5%MgO:$LiNbO_3$ crystal. The sample was $10 \times 10 \times 4$ mm (length $\times$ width $\times$ thickness), and was polished on two surfaces. The fundamental $s$-polarization beam was incident on the (1 1 0) plane of 5%MgO:$LiNbO_3$ crystal. In this geometry configuration, the effective second-order nonlinear coefficient $d_{\text{eff}}$ reduces to $d_{31}$. By adjusting the angle of incidence and $z$-axis of the samples, maximal SHG conversion efficiency can be obtained. The experimental and theoretical $Z$-scan curves of 5%MgO:$LiNbO_3$ crystal are given for different incident energy $E_{\text{in}}$ in Fig. 6, where $n_{\omega}^{o} = 2.231998$ and $n_{2\omega}^{e} = 2.216402$ calculated from Ref. [13] are used. It is shown that experimental

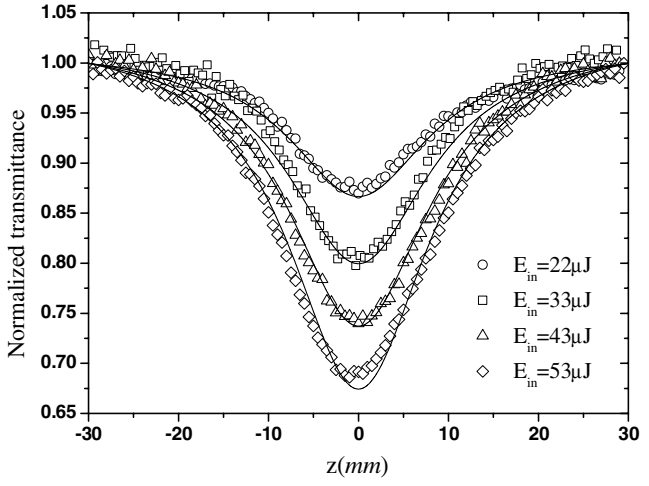


Fig. 6. The experimental and theoretical $Z$-scan curves of 5%MgO:$LiNbO_3$ crystal for different incident energy $E_{\text{in}}$. The solid lines represent the theoretical fittings, where $\alpha_{\omega} = 0.03\ \text{m}^{-1}$, $\alpha_{2\omega} = 2\ \text{m}^{-1}$, and $\Delta kL = 3.8$.

Table 1
Previous reported values and our result of nonlinear optical coefficients $d_{31}$ of 5%MgO:$LiNbO_3$ crystal at 1064 nm

| Crystal | Method | $d_{\text{fl}}$ ($\times 10^{-12}$ m/v) | Reference |
|---|---|---|---|
| 5%MgO:$LiNbO_3$ | Our method | $d_{31} = 4.5$ | This experiment |
| | MF | $d_{31} = 4.4$ | [14] |
| | SHG | $d_{31} = 4.69$ | [15] |

results agree well with theoretical simulations. By fitting data, $d_{31} = 4.5 \times 10^{-12}$ m/v for 5%MgO:$LiNbO_3$ crystal can be obtained. The method presented in this paper is based on well-known open aperture $Z$-scan. In the condition of no aperture, the results of $Z$-scan will only be sensitive to nonlinear absorption, not to nonlinear refraction. Therefore, the existence of nonlinear refraction will not affect measurements of second-order nonlinear coefficient $d_{\text{eff}}$ in our experiments. Meanwhile, we have done the open and closed aperture $Z$-scan experiments at the incident energy $E_{\text{in}} = 53$ μJ when the phase mismatch value exceeds critical one, and the other experimental conditions are the same as those in Fig. 6. The results show that nonlinear refraction and nonlinear absorption were not observable at this incident energy.

Table 1 gives previous reported values [14,15] and our result of second-order nonlinear coefficient $d_{31}$ of 5%MgO:$LiNbO_3$ crystal at 1064 nm. The $d_{31} = 4.4 \times 10^{-12}$ m/v is regarded as the most reliable one, and our result is 2.3% larger than this one.

## 4. Conclusion

In conclusion, we have presented a method for determining second-order nonlinear coefficient based on well-known $Z$-scan method. It has following advantages. First, it does not require measurements of SH power or energy, output light distribution or PF, and only requires measurements of fundamental light power or energy. Second, it does not require a well-characterized reference that one can trust. Third, the influence of linear absorption coefficients of fundamental and SH light on $Z$-scan curves by

this method is very slight and can be neglected. Therefore, we think that this method is simple and effective, and can be extensively applied to determinations of second-order nonlinear coefficients.

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