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R. DE SAINT DENIS
M. FROMAGER
E. CAGNIOT
K. AIT-AMEUR✉

# *Z*-scan sensitivity enhancement using a binary diffractive optics

Centre Interdisciplinaire de Recherche Ions Lasers, UMR6637, CEA-CNRS-ENSICAEN
et Université de Caen, CIRIL-ENSICAEN 6, Boulevard Maréchal Juin, 14050 Caen Cedex, France

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**ABSTRACT** We report a theoretical study of the sensitivity enhancement of $Z$-scan technique, using a simple binary diffractive optical element. The latter is a phase aperture made from a transparent plate that has a circular relief introducing a $\pi$ phase shift in the central region of the incident beam. We demonstrate that the phase aperture acts like a divergence multiplier allowing the $Z$-scan sensitivity to be increased by a factor of several hundred.

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## 1 Introduction

There is considerable interest in characterising nonlinear optical properties of materials, both from fundamental and applied points of view. A wide range of techniques has been developed over many years for this purpose. One of the most successful experimental methods is the $Z$-scan technique, which allows one to rapidly measure both nonlinear absorption and nonlinear refraction. The nonlinear refraction can originate, for instance, from the optical Kerr effect [1] or population lens effect [2, 3].

The $Z$-scan technique exploits the light–matter interaction between a laser beam and a material inducing a spatially dependent refractive index change giving rise to a lensing effect that causes the beam to focus or defocus. As the sample is moved with respect to the focal plane of an input lens, the transmittance variation of a diaphragm (of small diameter) set in the far-field region is represented by a typical peak–valley normalised transmittance curve (Fig. 1). Nonlinear refraction is then easily deduced [1] from the difference between normalised peak and valley transmittances $\Delta T_{pv} = T_p - T_v$.

The ultimate sensitivity of the $Z$-scan is determined by how the smallest a transmittance change can be measured. Consequently, sensitivity can be enhanced only by resorting to refinements of the $Z$-scan technique that provide an increase in the difference $\Delta T_{pv} = T_p - T_v$, for a given non-linear phase shift. Phase shifts as small as $\lambda/300$ can be measured by performing classical $Z$-scan technique [1]. A further improvement in the sensitivity has been demonstrated by replacing the diaphragm by an opaque disk (i.e., a stop) to determine power variation in the wings of the beam which is more important than in its central part. This method named eclipsing $Z$-scan (EZ-scan) improves sensitivity by a ratio of about 30 [4]. Another improvement of the $Z$-scan sensitivity by a ratio of 2.5 has been obtained with the top-hat beam $Z$-scan technique [5]. This method consists in carrying out $Z$-scan measurements with a top-hat beam rather than a Gaussian beam.

Before proceeding, let us recall that the classical $Z$-scan technique is a kind of divergence diagnostic. Indeed, the nonlinear lensing effect induced in the sample increases or decreases the angular divergence $\theta$ of the emerging beam. As a consequence, the sensitivity of $Z$-scan technique can be enhanced if the divergence variation $\delta\theta$ could be "amplified". One can expect that $\Delta T_{pv}$ should increase as $\delta\theta$ is multiplied.

## 2 The divergence multiplier

The main point is now to find an optical component that fulfils the function of divergence multiplication. One solution could be a phase aperture the optical function of which is to introduce a $\pi$ phase shift in the central region of the incident beam (Fig. 2a).

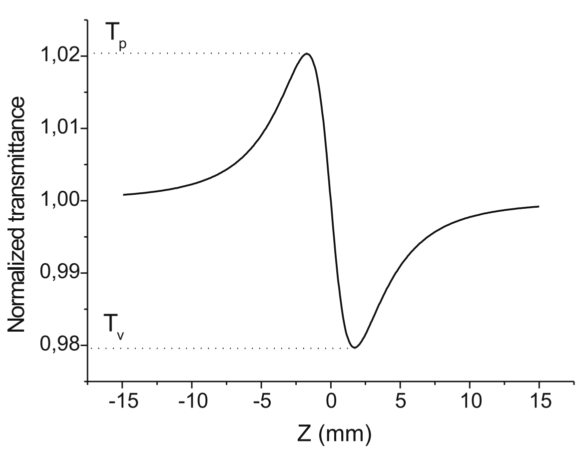


**FIGURE 1** Typical normalized $Z$-scan peak–valley curve

✉ Fax: +33 231 45 25 57, E-mail: kamel.aitameur@ensicaen.fr

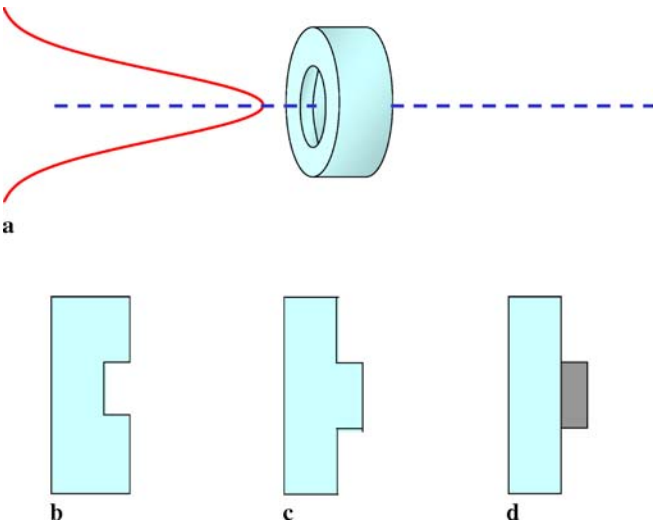


**FIGURE 2** The phase aperture

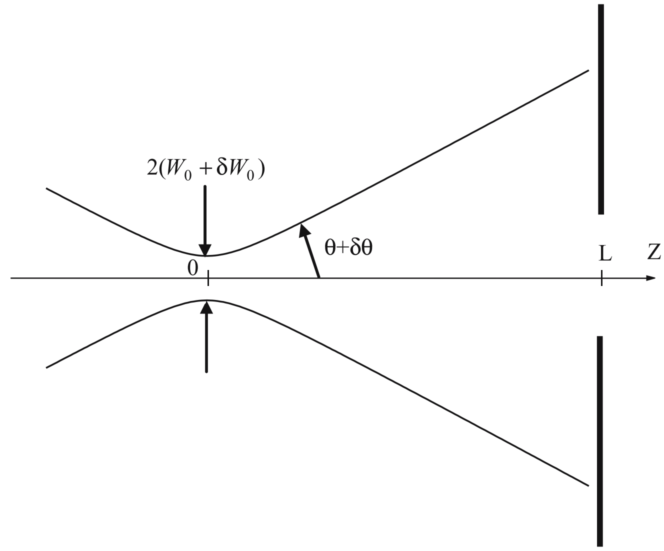


**FIGURE 3** Schematic of the beam divergence diagnostic

For instance, the phase aperture can be made from a transparent plate, with a refractive index $n$, having a relief of depth (Fig. 2b) or height (Fig. 2c) $e = \lambda/[2(n-1)]$, and of diameter $2\varrho_{PI}$, where $\lambda = 1.06\ \mu\mathrm{m}$ being the wavelength of the incident beam. Figure 2d shows the possible realisation of the phase aperture with two different materials by:

1. First, coating the transparent plate with a thin film with adequate thickness
2. Second, etching a plot of diameter $2\varrho_{PI}$.

The phase aperture is, thus, a simple binary diffractive optical element characterised by its transmittance, given by the following:

$$\tau_{PI} = \begin{cases} -1 \text{ for } \varrho \le \varrho_{PI} \\ +1 \text{ for } \varrho > \varrho_{PI} \end{cases} , \tag{1}$$

where $\varrho$ is the radial coordinate.

The phase aperture can be used for beam tailoring in a single-pass setup. It is able to convert a Gaussian beam into a super-Gaussian, a ring-shaped or a doughnut profile [6], and an elliptic beam into a circular one [7]. In the multi-pass case the phase aperture is inserted inside an optical laser cavity in order to enhance some of its properties: whether transverse mode discrimination [8], or fundamental mode volume [9] or even beam brightness [10, 11].

Some of these applications [7, 10, 11] are based on the fact that the divergence of diffracted beam emerging from the phase aperture is larger than the incident one [7] in a ratio which can be as high as 4.5, depending on parameter $Y_{PI}$ given by

$$Y_{PI} = \frac{\varrho_{PI}}{W} , \tag{2}$$

where $W$ is the incident Gaussian beam width. The latter is characterised by its unit-amplitude distribution $E_{\mathrm{in}}$ expressed in the cylindrical coordinates system $(\varrho, z)$:

$$E_{\mathrm{in}}(\varrho, z) = \frac{W_0}{W} \exp\left(-\frac{\varrho^2}{W^2}\right) \exp\left[-\mathrm{i}\left(kz - \varphi + \frac{k\varrho^2}{2R_{\mathrm{c}}}\right)\right], \tag{3}$$

where $R_{\mathrm{c}}$ represent the Gaussian beam radius of curvature at point $z$. These quantities, as well as the Gouy phase shift $\varphi$, are $z$ dependent and are expressed by the usual formulas:

$$W^2(z) = W_0^2\left[1 + (z/z_{\mathrm{R}})^2\right], \tag{4}$$

$$R_{\mathrm{c}}(z) = z\left[1 + (z_{\mathrm{R}}/z)^2\right], \tag{5}$$

$$\varphi(z) = \arctan(z/z_{\mathrm{R}}) , \tag{6}$$

where $z_{\mathrm{R}} = \pi W_0^2/\lambda$ is the Rayleigh range, and $W_0$ the beam-waist radius. The framework of the beam divergence diagnostic under consideration is illustrated by the schematic layout in Fig. 3. The idea is that a beam-waist radius change by a quantity $\delta W_0$ results in a beam divergence $\theta = \lambda/(\pi W_0)$ variation by a quantity $\delta\theta$, so that $\delta\theta/\theta = -\delta W_0/W_0$. The latter can be inferred from the resulting variation of the diaphragm transmission. It is assumed that diaphragm diameter is so small that the detected signal is proportional to the on-axis intensity $I_1(0, L) = E_{\mathrm{in}}(0, L)E_{\mathrm{in}}^*(0, L)$. It is important to note, that the diaphragm is placed in the far field so that distance $L$ is larger than the beam Rayleigh distance, i.e., $L \gg z_{\mathrm{R}}$, allowing on-axis intensity in the diaphragm plane to be deduced from (3) as follows:

$$I_1 = \left(\frac{\pi W_0^2}{\lambda L}\right)^2 . \tag{7}$$

In the following, we note by $W_0^{\mathrm{i}}$ the initial value of $W_0$ and by $W_0 = W_0^{\mathrm{i}} + \delta W_0$ the perturbed value of the beam-waist radius, where $\delta W_0 \ll W_0^{\mathrm{i}}$. The normalised transmission of the diaphragm is given by the following:

$$T_1\left(W_0^{\mathrm{i}}, \delta W_0\right) = \frac{I_1(W_0)}{I_1\left(W_0^{\mathrm{i}}\right)} = \left(1 + \frac{\delta W_0}{W_0^{\mathrm{i}}}\right)^4 = \left(1 - \frac{\delta\theta}{\theta}\right)^4 . \tag{8}$$

In practice, the variation of the beam divergence is small enough to allow expansion of (8) as follows:

$$T_1\left(W_0^{\mathrm{i}}, \delta W_0\right) \approx \left(1 + 4\frac{\delta W_0}{W_0^{\mathrm{i}}}\right) = \left(1 - 4\frac{\delta\theta}{\theta}\right) . \tag{9}$$

In a $Z$-scan experiment, the quantity $\delta W_0$ is positive or negative depending on the position of the sample with respect to the focal plane of the input lens, and also on the sign of the nonlinearity. Our objective in this paper is to propose a new setup which is able to increase the variation $\Delta T_{\mathrm{pv}}$ of the diaphragm transmission for a given $\delta W_0$. For that, we will assume an arbitrary variation $\delta W_0$ not in relation with real $Z$-scan measurements but in order to prove the efficiency of the "divergence multiplier" that will be considered later on. For convenience, we will consider the case where $\delta W_0 = W_0 - W_0^{\mathrm{i}}$ follows a sinus function:

$$W_0 = W_0^{\mathrm{i}} + \Delta W_0 \sin(\Omega)\,. \tag{10}$$

Now, let us consider a phase aperture set in plane $z = 0$, i.e., the beam-waist plane of the incident Gaussian beam. The far-field on-axis intensity of the diffracted beam emerging from the phase aperture is written as follows [see Appendix]:

$$I_2 = \left(\frac{\pi W_0^2}{\lambda L}\right)^2 \left[1 + 4\exp\left(-2Y_{PI}^2\right) - 4\exp\left(-Y_{PI}^2\right)\right], \tag{11}$$

where $Y_{PI} = \varrho_{PI}/W_0^{\mathrm{i}}$.

The normalised diaphragm transmission $T_2(W_0^{\mathrm{i}}, \delta W_0) = I_2(W_0)/I_2(W_0^{\mathrm{i}})$ in the presence of phase aperture is given by the following:

$$\begin{aligned} T_2\left(W_0^{\mathrm{i}}, \delta W_0\right) = A\left[1 + \frac{\delta W_0}{W_0^{\mathrm{i}}}\right]^4 \\ \times\left\{1 + 4\exp\left(-\frac{2\varrho_\pi^2}{\left(W_0^{\mathrm{i}} + \delta W_0\right)^2}\right)\right. \\ \left. -4\exp\left(-\frac{\varrho_\pi^2}{\left(W_0^{\mathrm{i}} + \delta W_0\right)^2}\right)\right\}, \end{aligned} \tag{12}$$

where $A = [1 + 4\exp(-2Y_{PI}^2) - 4\exp(-Y_{PI}^2)]^{-1}$.

Figure 4 shows the comparison of the variations of normalised diaphragm transmission with and without a phase aperture for $\Delta W_0 = 0.01$ mm, $W_0^{\mathrm{i}} = 0.1$ mm and $Y_{PI} = 0.7$. Undoubtedly, it is seen that such a phase aperture increases the quantity $\Delta T_{\mathrm{pv}} = T_{\mathrm{p}} - T_{\mathrm{v}}$ for a given $\Delta W_0$. Now, we have to optimise values of setup parameters in order to achieve the highest value of $\Delta T_{\mathrm{pv}}$, thus the highest sensitivity enhancement in the $Z$-scan measurements. A pertinent quantity which can express the multiplying factor, by which the sensitivity of the setup is multiplied, is the ratio $\eta$ of $T_1$ and $T_2$ derivatives:

$$\eta = \left.\frac{\mathrm{d}T_2/\mathrm{d}(\delta W_0)}{\mathrm{d}T_1/\mathrm{d}(\delta W_0)}\right|_{\delta W_0 = 0}\,. \tag{13}$$

The calculus is quite simple, and here is the following expression for the multiplying factor:

$$\eta = 1 + \frac{2Y_{PI}^2 \exp\left(-Y_{PI}^2\right)\left[2\exp\left(-Y_{PI}^2\right) - 1\right]}{1 + 4\exp\left(-Y_{PI}^2\right)\left[\exp\left(-Y_{PI}^2\right) - 1\right]}\,. \tag{14}$$

Figure 5 displays the variations of $\eta$ as a function of parameter $Y_{PI}$. It is clear from the graph that a huge enhancement in the multiplying factor $\eta$, as high as several hundreds, occurs for $Y_{PI}$ close to 0.83. Otherwise, the multiplying factor is close to unity. This particular value of $Y_{PI}$ is the one which nullifies the denominator in (14). A particular feature appears in Fig. 5 since the sign of $\eta$ changes when $Y_{PI}$ crossed the value 0.83, as if the sign of the studied nonlinear lensing effect has been have changed. If we take into account that the ratio of sensitivities of classical $Z$-scan and EZ-scan is about 30 [4], it can be concluded that $Z$-scan setup based on combination diaphragm-phase aperture should be superior to EZ-scan from a sensitivity point of view.

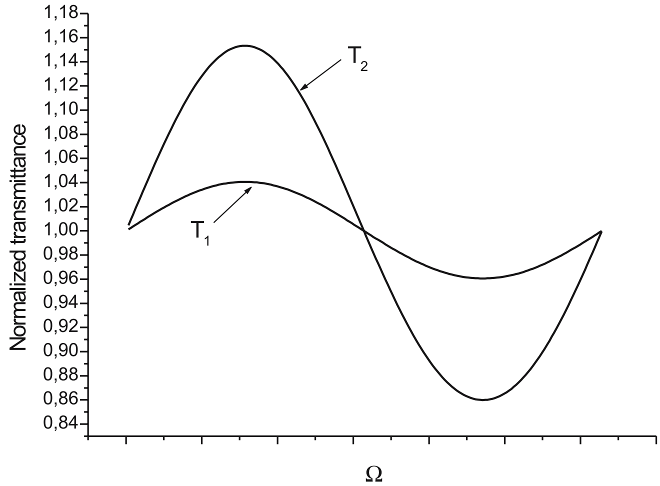


**FIGURE 4** Variations of diaphragm normalized transmittance without ($T_1$) and with ($T_2$) the phase aperture for $\Delta W_0 = 0.01$ mm, $W_0^{\mathrm{i}} = 0.1$ mm, and $Y_{PI} = 0.7$

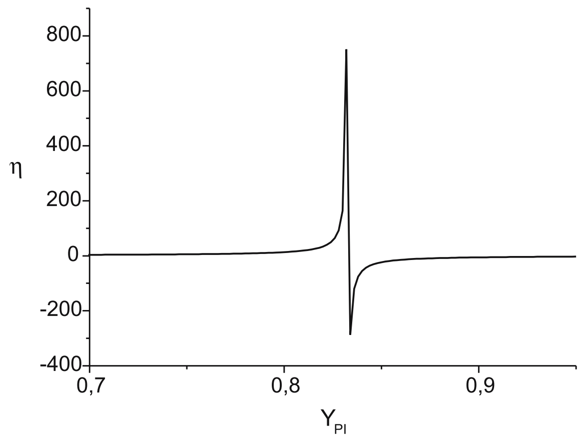


**FIGURE 5** Variations of $\eta$ the multiplying factor as a function of parameter $Y_{PI}$

## 3 Discussion and conclusion

Let us now consider from a practical point of view how the concept of divergence multiplier previously described can be implemented in order to enhance sensitivity of a $Z$-scan experiment. Rearrangement of $Z$-scan setup concerns only the postsample optics since a phase aperture between sample and diaphragm as shown in Fig. 6 has to be set. The reference arm must be as similar as possible to the main arm in order to measure the $Z$-scan normalised diaphragm transmittance. Photodetector $\mathrm{PD}_1$ delivers a signal $\mathrm{V}_1$ which is proportional to the unperturbed on axis intensity. Detector

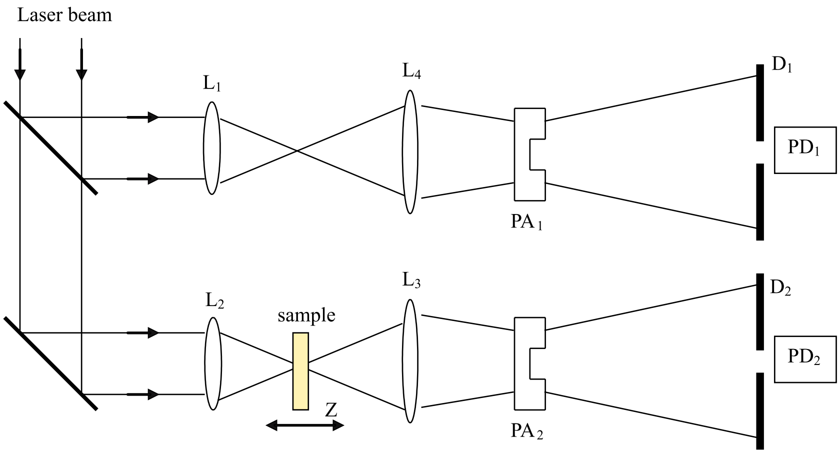


**FIGURE 6** Experimental setup involving a phase aperture (PA) as a divergence multiplier in a $Z$-scan experiment

$PD_2$ monitors the axial intensity perturbed by the sample nonlinearity and the divergence multiplication; it delivers a signal $V_2$. The measured aperture transmission is defined by ratio $T = V_2/V_1$, and its normalisation is such that $T \approx 1$ for the sample far from lens $L_2$ focus, i.e., when the nonlinearity is negligible. The role of lens $L_3$ is double:

1. First, it is important to note that diffraction properties of the phase aperture depend on $Y_{PI}$, but also on the incident wavefront radius of curvature of the [6]. Consequently, if an experimental investigation from this theoretical study has to be made, the phase aperture has to be set in the waist plane of the beam emerging from the sample. This can be achieved by setting the phase aperture in the focal plane of $L_3$ and $L_4$. In fact, it is not so simple to do since, when dealing with focused Gaussian beam, the point where the wavefront is plane is not at the geometric focus but is displaced [12]. This effect, known as focal shift, has a magnitude which is governed by the width and wavefront curvature of the beam incident on the focusing lens. One can expect that the focal shift of lens $L_3$ to slightly change as the sample moves due to the nonlinear lensing effect.
2. In the case where the phase aperture is patterned in a transparent glass plate by standard UV photolithography and chemical or ion beam etching, we are faced with the problem of lateral phase uniformity particularly for large diameters. Consequently, one can expect a higher quality in phase uniformity for a relatively small phase aperture diameter, for instance 0.1 mm, and the role of the lens is to reduce the beam width in this range. Moreover, one can expect multiplying factor $\eta$ to be unaffected by lens $L_3$, since it depends only on ratio $Y_{PI}$ as shown in (14).

In the introduction it has been recalled that improvement of $Z$-scan technique can be achieved by replacing the diaphragm by a stop [4]. Is it still the case if diaphragm $D_2$ is replaced in Fig. 6 by a stop? For that, one has to numerically solve integral (A.1) for getting the characteristic $Z$-scan normalised stop transmission curve. We have observed that the value of $\Delta T_{\mathrm{pv}}$ for the EZ-scan is affected little by the presence of the phase aperture, and consequently we do not observe the dramatic improvement demonstrated with the combination diaphragm-phase aperture. This can be understood by considering the phase aperture action as a transverse reshaping of the far-field beam pattern which is more pronounced in the beam centre than in its periphery.

In summary, the characteristic peak to valley $\Delta T_{\mathrm{pv}}$ measured by using a $Z$-scan technique can considerably be increased by resorting to a divergence multiplier. The latter is a simple binary diffractive optical element called a phase aperture which has been found to increase sensitivity of the $Z$-scan by a factor of several hundred. One can benefit from this sensitivity enhancement to determine nonlinearities in thin films or to reduce the peak power of laser sources involved in the $Z$-scan.

## Appendix

The intensity distribution in the diaphragm plane of light diffracted by the phase aperture is computed within the framework of the Huygens–Fresnel diffraction theory:

$$I(r, L) = \left| \frac{2\pi}{\lambda L} \int_0^\infty \tau_{PI}(\varrho) E_{\mathrm{in}}(\varrho) J_0\left(\frac{2\pi\varrho}{\lambda L} r\right) \varrho \,\mathrm{d}\varrho \right|^2 , \qquad \text{(A.1)}$$

where $r$ is the radial coordinate in the diaphragm plane, $L$ the distance between phase aperture and diaphragm, and $J_0$ the zero-order Bessel function.

For off-axis points ($r \neq 0$), solving (A.1) requests a numerical calculation. However, for on-axis points ($r = 0$), $J_0(0) = 1$ and (A.1) reduces to:

$$I(0, L) = \left(\frac{2\pi W_0^2}{\lambda L}\right)^2 \times \left| -\int_0^{Y_{PI}} \exp(-X^2) X \,\mathrm{d}X + \int_{Y_{PI}}^\infty \exp(-X^2) X \,\mathrm{d}X \right|^2 , \qquad \text{(A.2)}$$

where the reduced transverse coordinate $X$ is defined as $X = \varrho/W_0$. The phase aperture is assumed to be located in the waist plane, of width $W_0$, of the beam emerging from lens $L_3$ (Fig. 6). Integrals in (A.2) can be analytically solved and yields

$$I(0,\,L)=\left(\frac{\pi W_0^2}{\lambda L}\right)^2\left\{1+4\exp\left(-2Y_{PI}^2\right)-4\exp\left(-Y_{PI}^2\right)\right\}. \tag{A.3}$$

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