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Nonlinear optical properties of semiconductor materials by the Z-scan technique: review article

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Abstract

The nonlinear optical (NLO) properties of semiconductor nanomaterials have a significant role in opto-electronic devices. In this review, recent results on the NLO of semiconductor materials (including zinc sulphide, Indium Oxide, birnessite-type manganese oxide, GaAs doped with Bismuth and ZnO doped with transition metals) have been discussed. Z-scan has been used to study the nonlinear optical properties of these materials. The NLO properties and the optical limiting for different laser sources either pulsed or continuous are studied experimentally.

Keywords: Nonlinear optics; semiconductor; opto-electronic; Z-scan

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1. Introduction

Nonlinear optical (NLO) materials have recently received much attention, specifically developments in ultrashort pulsed lasers that are enabling new phenomena within their NLO properties to be explored. This is due to their promising applications such as optical computing (Li et al., 2002; Yeh et al., 1989), laser technology, optical communications (Correa et al., 2011; Li et al., 2002; Yeh et al., 1989), and optical limiters (Fan et al., 2010). Moreover, the research in this field is significant for the rapid development in many fields such as nonlinear spectroscopy (Mukamel, 1999), femtochemistry (Asobe et al., 1992; Auston & Applications, 1988), and medical imaging (Barbara et al., 1991; Squier & Müller, 2001). The development of devices either optical or electro-optical depends heavily on NLO materials with substantial optical nonlinearity (Rashed et al., 2019; Singh & Singh, 2007). Additionally, by modifying the thickness and free carrier concentrations of optical materials, NLO properties can be enhanced, opening up the possibility of employing these materials in new applications (Elim et al., 2006).

NLO is the branch of optics that studies the optical properties of matter when interacting with intense light, which induces NLO mechanisms. Electric polarization has a significant role in the NLO phenomena. Owing to the interaction of the material's inherent charges with the high incident electric field, the phase, frequency and electric polarization of the material are altered (Franken et al., 1961; S. J. M. U. Pramodini, 2015). The second harmonic generation (SHG) is the first NLO phenomenon that was studied theoretically by Maria Goeppert in 1931 (Göppert-Mayer, 1931). The beginning of NLO field was in 1961 by Peter Franken et al when they discovered the SHG phenomena experimentally using the ruby laser. When a high-power red light with wavelength 6493A˚ from a ruby laser focused onto a quartz crystal, the transmitted light was in the ultraviolet region due to SHG, where the frequency was double (i.e., the wavelength was $\lambda/2$). The field of NLO has significant development across the years after the discovery of SHG, thus many types of NLO phenomena were discovered including high harmonic generation (Terhune et al., 1962), sum frequency generation (Guyot-Sionnest et al., 1987), optical

Kerr effect (Nurhuda et al., 2008; Wijayan, 2019), optical parametric amplifications (Baumgartner & Byer, 1979), optical solitons) Taylor & Taylor, 1992 (and self-phase modulation (Stolen & Lin, 1978). Such of these nonlinear optical phenomena made it suitable for promising applications in different fields (Caspani et al., 2011; Cotter et al., 1999; Henari & Henari, 2016; Mukamel, 1995; Ni & Zhuo, 2015; Schneider, 2004; Seidel et al., 2017).

1.1. Nonlinear Wave Equation

When a low or moderate electric field irradiates matter that has a group of positively charged molecules or atoms encircled by electrons with negative charges. Matter behaves in a linear manner, thus the induced polarization responds linearly to the electric field; the inducing polarization is provided by (Li & Applications, 2017):

$$P = \varepsilon_0 \chi^{(1)} E \tag{1}$$

where ε_0 is the permittivity of free space and $\chi^{(1)}$ is the linear susceptibility. On the other hand, when an intense electric field radiates the matter (10^7 to $10^{10}\ V/cm$), the induced electric polarization responds nonlinearly to the electric field, thus the nonlinear polarization can be defined as follows (Laud, 1986; Sutherland, 2003):

$$P = \varepsilon_0 \left[\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + ..\right] \tag{2}$$

Where $\chi^{(2)}$is the second-order nonlinear optical susceptibility, $\chi^{(3)}$is the third-order nonlinear optical susceptibility.

In the right side of Eq. (2), the first item represents the linear polarization, while the second, third and fourth items represent the high-order nonlinear polarizations, thus Eq. (2) can be interpreted by electric polarizations as:

$$P = P^{(1)} + P^{(2)} + P^{(3)} + \cdots \quad = P_L + P_{NL}, \tag{3}$$

Where $P_L = P^{(1)}$ and P_L represents the linear polarization terms, and $P_{NL} = P^{(2)} + P^{(3)}$, P_{NL} is the nonlinear polarization term.

The polarization of medium depends on the strength of the field for linear polarization whereas for second order NL polarization, it is dependent on the square of the strength of the field; and for third-order NL polarization, it is dependent on the triple of the field strength.

1.2. Third order nonlinear process

The NLO processes generated in the NL medium depended on the third-order nonlinearity $\chi^{(3)}$such as two-photon absorption (Laud, 1986; Li & Applications, 2017; Sutherland, 2003), nonlinear refraction (Laud, 1986; Li & Applications, 2017; Sutherland, 2003), optical Kerr effect, third-harmonic generation and degenerate four-wave mixing

2. Z-scan Technique

There are several characterization techniques used for measuring NLO properties including nonlinear interferometry (Moran et al., 1975), degenerate four-wave mixing (Friberg & Smith, 1987), nearly degenerate three-wave mixing (Adair et al., 1987), and ellipse rotation (Owyoung, 1973). These techniques are sensitive, but their setup is complicated. The Z-scan method developed by Sheik-Bahae (Sheik-Bahae et al., 1990) is considered a simple and sensitive technique for measuring NLO properties of materials such as nonlinear absorption coefficient and nonlinear refractive index.This technique involves moving the sample along a Gaussian laser beam. Due to the dependent positioning of the sample with respect to the focus, the laser intensity changes as it is moved. The name "Z-scan" refers to the process of moving a sample across the beam waist of a focused beam to measure the power passed through it. The experimental setup for the Z-scan technique is shown in Fig. 1.

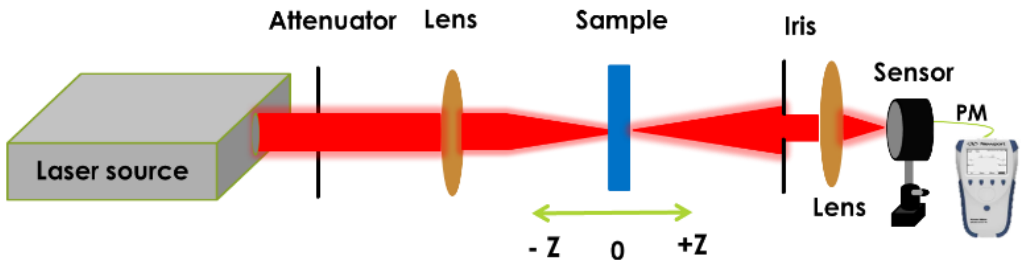

Fig. 1. Z-scan experimental setup: A; Attenuator, L; Convex Lens, S; Sample, I; Iris and PM; Power meter.

2.1. Closed Aperture Z-scan

A closed aperture Z-scan experiment is carried out to determine the nonlinear index of refraction n_2. The refractive index of the medium changes rapidly at strong intensity. If the material has a positive nonlinearity ($n_2 > 0$), The T(z) graph exhibits a valley followed by a peak as shown in Fig. 2(a). The graph is exactly the opposite (exhibits a peak followed by a valley) for the sample where $n_2 < 0$ as shown in Fig. 2(b). When the sample self-focuses, the beam tends to collimate and narrow at the aperture, increasing the measured transmittance; conversely, when the sample self-defocuses, the beam broadens at the aperture, lowering the transmittance. The scan is complete once the transmittance returns to linear.

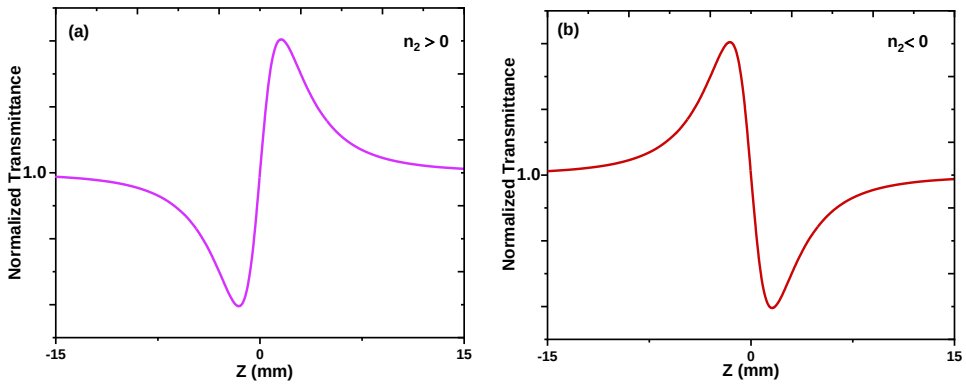

Fig. 2. The sign of n_2 can be determined from the transmittance T(z) graph.

Not only the nonlinear refractive index sign n_2 can be estimated from the Z-scan technique, but also its magnitude can be calculated as shown in the following measurement analysis. When Gaussian laser beam travels in Z-direction, the

electric field E magnitude is expressed as (Sheik-Bahae et al., 1989; Sheik-Bahae et al., 1990):

$$E(r,z,t) = E_0(t)\frac{w_0}{w(z)}\left\{\exp\left[-\frac{r^2}{w^2(z)} - \frac{ikr^2}{2R(z)}\right]\right\}.\{\exp[-i\varphi(z,t)]\} \quad (4)$$

where E_0 is the electric field at the focus, $w^2(z) = w_0^2(1 + z^2/z_0^2)^2$ is the beam radius at z; $z_0 = kw_0^2/2$ is the Rayleigh length, $k = 2\pi/\lambda$ is the wave vector, λ is the wavelength of laser beam, $R(z) = z(1 + z^2/z_0^2)$, is the wave front's curvature radius at position z, the term $\exp[-i\varphi(z,t)]$ contains all the radially uniform phase variations. If the sample length (L) is small enough to ignore changes in the beam diameter within the sample caused by diffraction or nonlinear refraction and the medium is regarded as "thin". The thin medium is defined as $L \ll n_0 z_0$, where L is the medium's thickness, n_0 is medium linear refractive index. If cubic nonlinearity and negligible nonlinear absorption are assumed, the problem is greatly simplified. The phase shift $(\Delta\varphi)$ at the sample's exit surface is represented as follows (Sheik-Bahae et al., 1989; Sheik-Bahae et al., 1990)

$$\Delta\varphi(z,r,t) = \frac{\Delta\varphi_0(t)}{1+z^2/z_0^2}\exp\left[-\frac{2r^2}{w^2(z)}\right] \quad (5)$$

with

$$\Delta\varphi_0(z,t) = \frac{\Delta\varphi_0(t)}{1+(z^2/z_0^2)} \quad (6)$$

where $\Delta\varphi_0(t)$ the on-axis phase shift at the focus (Z = 0), is expressed as

$$\Delta\varphi_0(t) = k\Delta n_0(t)L_{eff,} \quad (7)$$

where $L_{eff} = \frac{(1-e^{(-\alpha_0 L)})}{\alpha_0}$ is the sample's effective length, L is the sample length, α_0 is the linear absorption coefficient, Δn_0 is the change in refractive index at focus $(\Delta n_0 = n_2 I_0)$ with I_0 is the on-axis irradiance at focus. At the exit of the sample there is the complex electric field E_e that contains the nonlinear phase distortion. By use of a Taylor series expansion of the NL phase term, the complex electric field at the exit plane of the sample is divided into a sum of Gaussian beams using the Gaussian decomposition (GD) technique (Falconieri & Optics,

1999). All these beams are once more combined into a single beam when they come to the aperture plane. We obtain the electric field at the aperture, $E_a(r,t)$, which is a function of $\Delta\varphi_0$. The transmitted power $P_T(\Delta\varphi_0(t))$ is obtained by spatially integrating up to the aperture radius r_a. Now that the normalised Z-scan transmittance $T(z)$ has been calculated (Sheik-Bahae et al., 1989; Sheik-Bahae et al., 1990):

$$T(z) = \frac{\int_{-\infty}^{\infty} P_T(\Delta\varphi_0(t))dt}{S\int_{-\infty}^{\infty} P_i(t)dt} \tag{8}$$

where S is the aperture linear transmittance and $P_i(t)$ is the instantaneous input power within the sample, with $S = 1 - \exp\left(-\frac{2r_a^2}{w_a^2}\right)$ (w_a is the beam radius at the aperture). The aperture linear transmittance S is a major factor because a large aperture minimises the variations in $T(z)$ (Sheik-Bahae et al., 1989; Sheik-Bahae et al., 1990). The normalized transmittance T_{CA} of CA Z-scan was fitted using the following equation, which related directly to phase shift (Sheik-Bahae et al., 1990):

$$\Delta T_{CA} = 1 \pm \frac{4x\Delta\varphi_0}{(9+x^2)(1+x^2)} \tag{9}$$

Where $x = \frac{z}{z_0}$, which is + for positive n_2, and – for negative n_2.

Usually, the most significant quantity is ΔT_{p-v}, which is the difference between the peak and valley values of transmittance T_{p-v} as shown in Fig. 3. The relation between T_{p-v} and $\Delta\varphi_0$ is dependent on numerical fitting (which is a function of n_2) (S. Pramodini, 2015; Sheik-Bahae et al., 1990).

$$\Delta T_{p-v} \approx 0.406(1-S)^{0.25}|\Delta\varphi_0| \tag{10}$$

For $|\Delta\varphi_0| \leq \pi$. we can calculate the nonlinear refractive index n_2 using Eq. (7)

$$n_2 = \frac{\Delta\varphi_0}{kI_0L_{eff}} \tag{11}$$

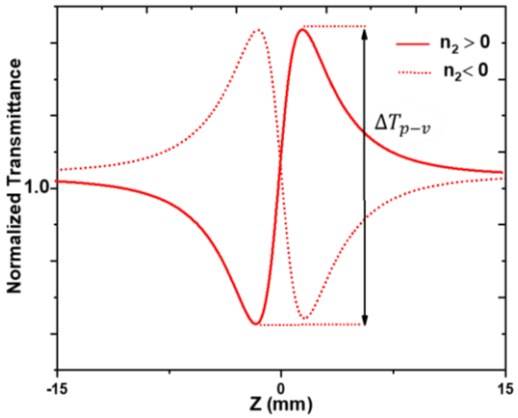

Fig. 3. The normalized transmittance for a closed Z-scan aperture as ΔT_{p-v}.

2.2. Open Aperture Z-scan

Z-scan technique is also utilized to calculate the nonlinear absorption coefficient β. In the case of OA Z-scan experiment, the transmitted light from the sample measured directly by power meter. The aperture (S = 1) in an OA z-scan is not sensitive to nonlinear refraction. It is assumed that the Z-scan traces acquired without an aperture are symmetric with regard to the focus ($Z \backsimeq 0$), where the transmittance is low. Samples for this experiment are made to move from one end of the far field to the other ($Z << 0$), passing through the focus. Since the intensity is low in the far field, linear absorption occurs. The sample position is plotted along the x-axis of a graph with normalised transmittance along the y-axis. The measured transmittance in the detector either reduces or increases as the sample approaches the focus, creating a valley or peak at the focus. Figure 4(a) depicts the RSA process, which exhibits the measured transmittance to form a valley at the focus. Figure 4(b) shows the SA process, which induces the measured transmittance to form a peak at the focus. RSA is referred to a positive type of absorption nonlinearity$(+\beta)$, whereas SA is referred to a negative type of absorption nonlinearity $(-\beta)$. The nonlinear absorption coefficient is calculated using the OA Z-scan traces and is related to the imaginary part of the third-order NLO susceptibility $\chi^{(3)}$.

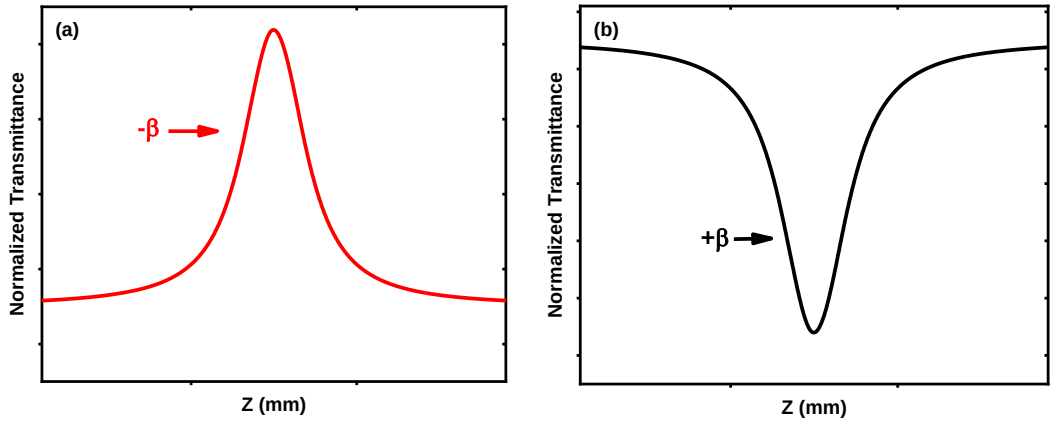

Fig. 4. OA z-scan traces of (a) saturable absorption and (b) reverse saturable absorption.

The total absorption is defined by (Sheik-Bahae et al., 1989; Sheik-Bahae et al., 1990):

$$\alpha(I) = \alpha_0 + \beta I \tag{12}$$

where α_0 is linear absorption and I is the intensity of laser beam. The normalized transmittance can be expressed by (Sheik-Bahae et al., 1990) as follows:

$$\Delta T_{OA} = 1 \pm \frac{\beta I_0 L_{eff}}{n^{\left(\frac{3}{2}\right)}[1+(Z/Z_0)^2]} \tag{13}$$

where ΔT_{OA} is the normalized transmission, n is the number of photons ($n = 2$), I_0 is the laser peak intensity at focus, L_{eff} is the sample's effective length, Z is the relative displacement of the sample from the focus, Z_0 is the Rayleigh length ($Z_0 = \frac{n\pi\omega_0^2}{\lambda}$).

2.3. Optical Limiter

The development of lasers signified significant developments in science and technology and gave rise to an extensive variety of applications in industry, research, the military and medical fields (S. Pramodini, 2015). The protection of sensors, optical components, and human eyes from high intensity is essential due to the recent development of powerful and wide wavelength range laser sources. The main aim of the protective device is to prevent the sensor from being

exposed to hazardous intensity while being transparent. In order for light to pass through the material during low intensity light, it needs to be linearly transmissive [106]. The optical limiter is the device that exhibits a decline in transmittance with increasing intensity. Below a limiting threshold value, an ideal optical limiter demonstrates linear transmission; above that value, it clamps the output to a fixed value to protect the sensors. Figure 5 depicts the operation of an ideal optical power limiter.

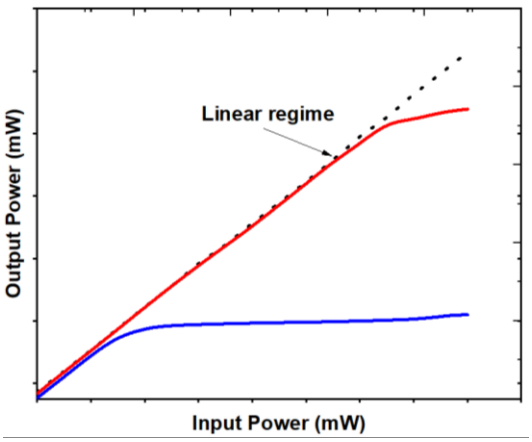

Fig. 5. depicts the operation of an ideal optical power limiter.

3. Criteria for Selecting Applicable NLO Materials

For use in photonic and electrical devices, a thorough search for materials with NLO properties has been carried out in recent years. Such nonlinear materials used in optical device applications must have some essential properties such as high nonlinearity, broadband spectrum response, low absorption, quick response time, and a wide dynamic range (Kivshar, 2008). Many photonic materials including semiconductor thin films, liquid crystals, organic and inorganic crystals, polymers, dyes, porphyrins, quantum dots, phthalocyanines, and nanomaterials either semiconductors or insulators, have recently been explored (S. Pramodini, 2015). Typically, the NLO characteristics of materials in their different forms are studied by different laser sources either pulsed or continuous wave (CW).

4. NLO Materials of Recent Interest

Here are some nonlinear semiconductor materials that have been reported in the literature:

As semiconductor nanoparticles have significant applications for various opto-electronic devices, they have been thoroughly studied (Bunning et al., 1991; Unnikrishnan et al., 2003). Dehghani et al. (Dehghani et al., 2011) used poly vinyl pyrrolidone as a capping agent and a simple chemical method to study the zinc sulphide nanoparticles optical nonlinearity. The sample was analyzed using a 632.8 nm CW Helium-Neon (He-Ne) laser and the Z-scan method. They revealed that the nonlinear refractive index of the sample was found to be negative ($-n_2$) that is generated by self-defocusing characteristic, which was the result of their research. They reported that both nonlinear absorption coefficient β and nonlinear refractive index n2 values as 10^{-3} cm/W and $-10^{-8}\text{cm}^2/\text{W}$, respectively.

The optically nonlinear and optically limiting (OL) behavior of Indium Oxide nanoparticles was studied by Yu et al. (Yu et al., 1997). For the first time, they found that the nonlinear coefficient n_2 is effective intensity dependent. According to their results, coated Indium Oxide nanoparticles exhibit strong thermally induced lensing effects and high effective nonlinearities. The limiting thresholds for saturation and nonlinear refractive index (n_2) were observed to be $780\ \text{W/cm}^2$ and $-10^{-7}\text{cm}^2/\text{W}$, respectively.

The nanoparticles of birnessite-type manganese oxide (γ-MnO2) were also studied and synthesized by Naderi et al. (Naderi et al., 2013) using the gelation routes technique. They first used an optical limiting setup to calculate the linear absorption for these nanoparticles. Then, they used a laser of He-Ne with 632.8 nm wavelength to measure the NLO parameters of γ -MnO2 nanoparticles utilizing open and close-apertures Z-scan at various incident powers. It was observed that γ -MnO2 nanoparticles have nonlinear absorption coefficients β and nonlinear refractive indices n2 of 10^{-2} cm/W and $10^{-7}\text{cm}^2/\text{W}$, respectively.

In (Auyang & Wolff, 1989; Dehghani et al., 2011; Xu et al., 2018; Zhang et al., 2014), the contribution of free carriers on the NLO of semiconductor materials has been studied at different laser intensities, wavelengths, and pulse widths. Using nano- and femtosecond lasers, the NLO parameters of Gallium Arsenide (GaAs) and GaAs doped with Bismuth (Bi) were investigated at 1064 nm (Xu et al., 2018). The results indicate that the GaAs doped with Bi show better NLO performances than GaAs and a stronger optically limiting response (Xu et al., 2018) after determining the NLO parameters of saturable absorption in the GaAs and GaAs doped with Bi, including two-photon absorption (2PA) coefficient. The nonlinear optical parameters of graphite oxide (GO) thin film were also investigated using 532 nm Q-switching Nd: YAG (Sreeja et al., 2016). They found that GO is suitable for both laser pulse narrowing and optical switching due to its saturable absorption behavior (Sreeja et al., 2016).

Recently, several studies had shown that ZnO doped with transition metals such as Sn, F, Al, Co, Zr, Ce, Mn, Er or Ni (Hu et al., 2012; Nagaraja et al., 2013) enhances its electric, magnetic and optical properties either linear or nonlinear (Bao et al., 1998; Fujihara et al., 2001; Guo et al., 2000; Rana et al., 2015; Shehata et al., 2020; Tsay & Fan, 2008). The NLO parameters of pure Zinc Oxide (ZnO) and ZnO doped with Cobalt (Co) thin films were evaluated utilizing the Z-scan method at various input laser intensities and 950 nm excitation wavelength by 100 fs laser pulses (Shehata et al., 2020). The nonlinear absorption coefficient β and nonlinear refractive index n2 of Co-doped ZnO thin film were found to be input laser intensity dependent. An enhancement of 100 times in the n2 was demonstrated for the Co-doped ZnO thin film compared to pure ZnO thin film. The optical limiting behavior of pure ZnO and Co-doped ZnO thin films was also investigated, and the data demonstrated that Co-doped ZnO thin film is a good candidate for optical limiters applications due to the presence of strong revers saturable absorption (RSA) (Shehata et al., 2020). The NLO properties of both pure ZnO and ZnO doped with Nickel (Ni) using Z-scan technique had been studied also in (Dar et al., 2016; Rahulan et al., 2019; Rana et al., 2015; Shehata et al., 2020) by different laser sources either pulsed or continuous. These studies showed that the NLO properties of pure ZnO and Ni-

doped ZnO depend on the laser parameters, sample preparation method, sample parameters, dopant material and its concentration.

5. Conclusion

NLO is the branch of optics that studies the optical properties of matter when interacting with intense light, which induces NLO mechanisms. The SHG is the first NLO phenomenon that was studied theoretically in 1931. The beginning of NLO field was in 1961. NLO materials have recently received much attention, especially developments in ultrashort pulsed lasers. This is due to their promising applications. Among several techniques that are used for measuring NLO properties, Z-scan is considered one of the simplest and most sensitive techniques. Semiconductor nanomaterials are one of the most important NLO materials. The NLO characteristics of many semiconductor materials are studied by different laser sources either pulsed or continuous.

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.**Data availability** No datasets were generated or analysed during the current study.

Declarations

Conflict of interest The authors declare no conflict of interest.

Competing interests The authors declare no competing interests.

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