

Polarization-Resolved Heterodyne Z-Scan for Nonlinear Refractive Index Discrimination Using Ultrafast-High-Repetition Laser Pulses

Publisher: IEEE[Cite This](#)[PDF](#)Renato M. Moysés ; Lino Misoguti [All Authors](#)

Abstract

Document Sections

[I. Introduction](#)[II. Nonlinear Refractive Effects](#)[III. Polarization-Resolved Heterodyne Z-Scan](#)[IV. Experimental Results](#)[Conclusions](#)[Authors](#)[Figures](#)[References](#)[Keywords](#)[More Like This](#)

Abstract:

This work presents a new technique, Polarization-Resolved Heterodyne Z-Scan (PR-HZS), which is capable of discriminating and quantifying three different nonlinear refractive indices: ultrafast nonresonant electronic, molecular orientation (or nuclear) and thermal. This technique explores laser polarization and phase delay introduced by noninstantaneous nonlinearities to separate these nonlinear effects. Femtosecond pulses at megahertz repetition rate are used to induce simultaneously these nonlinear effects in different materials. We characterized Carbon Disulfide (CS_2), SF6 and LaSF-N30 optical glasses, and a Neutral Density Filter AND-30C-40, which are known to present more than one nonlinearity.

Published in: [2025 SBFoton International Optics and Photonics Conference \(SBFoton IOPC\)](#)**Date of Conference:** 21-24 September 2025**Date Added to IEEE Xplore:** 31 October 2025

ISBN Information:

Electronic ISBN: 979-8-3315-9497-8**Print on Demand(PoD) ISBN:** 979-8-3315-9498-5

ISSN Information:

Electronic ISSN: 2837-4967**Print on Demand(PoD) ISSN:** 2837-4959**DOI:** [10.1109/SBFotonIOPC66433.2025.11218418](#)**Publisher:** IEEE**Conference Location:** São Pedro/SP, Brazil**Funding Agency:**

Polarization-Resolved Heterodyne Z-Scan for nonlinear refractive index discrimination using ultrafast-high-repetition laser pulses

Renato M. Moysés
 Sao Carlos Institute of Physics
 University of Sao Paulo, IFSC-USP
 São Carlos, Brazil
 renotamaframoyes@usp.br

Lino Misoguti
 Sao Carlos Institute of Physics
 University of Sao Paulo, IFSC-USP
 São Carlos, Brazil
 misoguti@ifsc.usp.br

Abstract— This work presents a new technique, Polarization-Resolved Heterodyne Z-Scan (PR-HZS), which is capable of discriminating and quantifying three different nonlinear refractive indices: ultrafast nonresonant electronic, molecular orientation (or nuclear) and thermal. This technique explores laser polarization and phase delay introduced by non-instantaneous nonlinearities to separate these nonlinear effects. Femtosecond pulses at megahertz repetition rate are used to induce simultaneously these nonlinear effects in different materials. We characterized Carbon Disulfide (CS₂), SF₆ and LaSF-N30 optical glasses, and a Neutral Density Filter AND-30C-40, which are known to present more than one nonlinearity.

Keywords—Nonlinear Optics, Ultrafast Lasers, Z-Scan.

I. INTRODUCTION

Different nonlinear refractions (NLR) can take place simultaneously contributing to a single nonlinear optical signal. Here, we considered three important ones: the ultrafast nonresonant electronic, molecular orientational (or nuclear) and thermal effects. Photonic devices applications, such as all-optical switches (AOS) [1], usually require high pure electronic nonlinearity and, in this way, it is interesting to discriminate it from nuclear and thermal contributions [2].

Recently, we developed a Polarization-Resolved Z-Scan (PRZS) [3], based on the traditional Z-Scan [4] to discriminate different NLR by employing different laser polarizations [5] and pulse repetition rates [6], taking into account a Thermal Lens (TL) model [7]. Using the PRZS, we could discriminate simultaneously these three important NLR. However, this technique requires a tunable repetition rate laser system.

Here, we propose a new method called Polarization-Resolved Heterodyne Z-Scan (PR-HZS) technique, which uses a heterodyne detection method [8] to separate the thermal refraction, working with fixed high-repetition rate ultrafast laser and a simple optical chopper. Using different laser polarizations, PR-HZS can discriminate and quantify a mixture of the three NLR, while also obtaining the thermal temporal response by varying the chopper frequency.

II. NONLINEAR REFRACTIVE EFFECTS

The most important nonlinear refractive index (n_2) for photonic devices is the pure electronic and is described as $n = n_0 + n_2 I$, where n_0 is the linear refractive index, and I is the laser irradiance. However, when NLR effects are mixed in the optical signal, an effective change Δn replaces n_2 . Despite not originating from ultrashort pulse excitation, thermal NLR can arise due to inter-pulse heating from high repetition rates.

In general, to evaluate the magnitude of the effects we analyze the NLR as terms of the third-order nonlinear susceptibility tensor $\chi^{(3)}_{ijkl}$, which in the case of isotropic

liquids and glasses can be reduced to the coefficients $A (= 6\chi^{(3)}_{1122})$ and $B (= 6\chi^{(3)}_{1221})$. Furthermore, Δn can be written, for linear (lin) and circular (circ) polarization, as [2]:

$$\Delta n_{lin} = \frac{I}{4 n_0^2 \epsilon_0 c} \left(A + \frac{B}{2} \right), \quad (1)$$

$$\Delta n_{circ} = \frac{I}{4 n_0^2 \epsilon_0 c} A, \quad (2)$$

where ϵ_0 is the vacuum electrical permittivity and c is the speed of light. The ratio between these coefficients is different for each nonlinear process: $B/A=1$ (electronic), $B/A=6$ (molecular orientation), and $B/A=0$ (thermal). For liquids it was already verified [3], including the difference between heating originated from linear and nonlinear absorption (NLA). For glasses we do not expect molecular orientation. In this way, it is possible to predict how the Z-Scan signal will be affected by the nonlinearities, since $\Delta n_{lin}/\Delta n_{circ} = 1+B/(2A) = 1.5$ for electronic (ele), 4 for orientational (ori) and nuclear (nuc), and 1 for thermal (ther) nonlinearities. Moreover, in the case of a mixture Δn (or the Z-Scan signature ΔT_{pv}) can be expressed as a sum of independent terms.

III. POLARIZATION-RESOLVED HETERODYNE Z-SCAN

Our new PR-HZS (Fig. 1) consists of an ultrashort pulsed laser (Light Conversion Carbide, 515 nm, ~192 fs) beam of high repetition rate (1 MHz), with linear or circular polarization (via quarter-waveplate), passing through an optical chopper (20-1000 Hz, 50% duty cycle) and being focused by a lens ($f \sim 13$ cm) into the sample. As the sample is translated in the z -axis there is the formation of the NLR, which cause the beam to be self-focused (or self-defocused) due to effective Kerr effect (electronic-nuclear) and TL. The transmitted light in a small aperture reaches a photodetector and is analyzed via dual-phase lock-in amplifier (LIA).

Differently from the HZS [10], which uses sinusoidal modulation, the PR-HZS uses an optical chopper, which implies in a square waveform signal that reaches the sample (Fig. 1). Since it distorts as the laser beam passes through the sample (due to the slow TL), the LIA detects it as phase shifts and magnitude change as a function of z -axis, which will result, respectively in $\varphi(z)$ and $T(z)$, defined as [3,8]:

$$T(z) = 1 + \frac{4 \langle \Delta \Phi \rangle \left(\frac{z}{z_0} \right)^2}{\left[1 + \left(\frac{z}{z_0} \right)^2 \right] \left[9 + \left(\frac{z}{z_0} \right)^2 \right]} + \frac{\theta}{2} \tan^{-1} \left[\frac{2 \left(\frac{z}{z_0} \right)^2}{3 + \left(\frac{z}{z_0} \right)^2} \right], \quad (3)$$

$$\varphi(z) = -A \left(c_1 \frac{\left(\frac{z}{z_0} \right)^2}{\left[1 + \left(\frac{z}{z_0} \right)^2 \right] \left[9 + \left(\frac{z}{z_0} \right)^2 \right]} + c_2 \tan^{-1} \left[\frac{2 \left(\frac{z}{z_0} \right)^2}{3 + \left(\frac{z}{z_0} \right)^2} \right] \right), \quad (4)$$

where z_0 is the Rayleigh range, $\langle \Delta \Phi \rangle$ is the time-averaged nonlinear induced Kerr phase shift [4] and θ is the phase shift induced by the TL effect according to the model in Ref. [7]. Both are Z-Scan signatures [4] and thus proportional to Δn , in

We thank the founding agencies CNPq and FAPESP.

which positive NLR appear as peak-valley for $T(z)$ and valley-peak for $\varphi(z)$. However, $\varphi(z)$ only carry information about slow cumulative nonlinear effect such as the TL. The factors A (phase shift amplitude), c_1 , and c_2 (constants) are not yet fully understood; however, taking the peak-valley difference in experimental $\varphi(z)$ curves is sufficient for the present work.

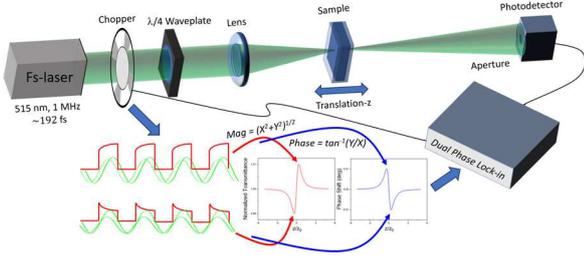


Figure 1 - Scheme of the experimental setup for the new PR-HZS.

Considering the model developed for the PRZS [3] and the traditional HZS [8] equations, we proposed new relations:

$$\Delta T_{pv}^{lin}(\delta) = a(\Delta T_{pv}^{ele} + \Delta T_{pv}^{ori}) + \frac{b}{2} \left(\frac{2+\delta^2}{1+\delta^2} \right) f \Delta T_{pv}^{ther}|_{lin}, \quad (5)$$

$$\Delta T_{pv}^{circ}(\delta) = a \left(\frac{\Delta T_{pv}^{ele}}{1.5} + \frac{\Delta T_{pv}^{ori}}{4} \right) + \frac{b}{2} \left(\frac{2+\delta^2}{1+\delta^2} \right) f \Delta T_{pv}^{ther}|_{circ}, \quad (6)$$

$$\Delta \varphi_{pv}^{lin,circ}(\delta) = \frac{b}{2} \left(\frac{\delta}{1+\delta^2} \right) f \Delta T_{pv}^{ther}|_{lin,circ}, \quad (7)$$

where ΔT_{pv} and $\Delta \varphi_{pv}$ are, respectively, the peak-valley change in transmittance and phase, f is the laser pulse repetition rate and the parameter δ is defined by $\delta = \tau 2\pi F$ [8], with F being the optical chopper frequency and τ is the thermal response time. From Eq. (7), when $\Delta \varphi_{pv}$ is maximum $\delta \rightarrow 1$, so τ can be obtained by $\tau = 1/(2\pi F_p)$ with F_p being the frequency when $\Delta \varphi_{pv}$ is maximum. Furthermore, the factor 1/2 is due to the difference in Δz_{pv} between Kerr and TL [3,7] signatures, with $a=1$ and $b=0.82$ if the electronic-nuclear effect is dominant and $a=0.71$ and $b=1$ if the thermal is the most relevant signal.

Hence, from $\Delta T_{pv}(\delta)$ and $\Delta \varphi_{pv}(\delta)$ it is possible to obtain ΔT_{pv}^{ther} , ΔT_{pv}^{ele} and ΔT_{pv}^{ori} (or ΔT_{pv}^{nuc}) and convert into $n_2^{ele,ori}$. Here, to test our technique, we first perform PRZS [3] varying f . The chopper is used after the sample only for detection purpose. In this case, $\delta=0$ and Eqs. (5,6) turn into:

$$\Delta T_{pv}^{lin}(f)|_{\delta=0} = a(\Delta T_{pv}^{ele} + \Delta T_{pv}^{ori}) + bf \Delta T_{pv}^{ther}|_{lin}, \quad (8)$$

$$\Delta T_{pv}^{circ}(f)|_{\delta=0} = a \left(\frac{\Delta T_{pv}^{ele}}{1.5} + \frac{\Delta T_{pv}^{ori}}{4} \right) + bf \Delta T_{pv}^{ther}|_{circ}, \quad (9)$$

whereas no phase shift signals are observed. Therefore, we can obtain reference values to be compared with the PR-HZS.

Using the PR-HZS and PRZS, we characterize the solvent Carbon Disulfide (CS_2), in a silica cuvette of 2 mm, well-known for having high electronic-orientational NLR, and three optical glasses: SCHOTT LaSF-N30 and SF6, both of 2.1 mm, and a 2.3 mm thick Neutral Density Filter (NDF) AND-30C-40 from Opto-sigma. For SF6 we also performed open-aperture Z-Scan, since it presents high two-photon absorption (2PA) at 515 nm. In contrast, CS_2 and LaSF-N30 present low NLA and the NDF presents linear absorption.

IV. EXPERIMENTAL RESULTS

Firstly, we did PRZS measurements in CS_2 , where the laser pulse repetition rate was changed from 1 kHz up to 1 MHz. The pulse energy was kept constant at ~5.6 nJ (laser power $P = 5.6$ mW at 1 MHz). The results are shown at Fig.

2(a), in which it is possible to observe a negative thermal contribution, since ΔT_{pv} decreases as f increases.

Based on Eqs. (8,9) we obtain for CS_2 : $\Delta T_{pv}^{ele} = (0.087 \pm 0.004)$ and $\Delta T_{pv}^{ori} = (0.071 \pm 0.005)$, respectively, resulting in $n_2^{ele} = (3.8 \pm 0.6) \times 10^{-19}$ m²/W and $n_2^{ori} = (3.1 \pm 0.5) \times 10^{-19}$ m²/W, similar to Ref.[9]. We also find $\Delta T_{pv}^{ther}|_{lin} = -(0.090 \pm 0.008)$ and $\Delta T_{pv}^{ther}|_{circ} = -(0.062 \pm 0.005)$ for 1 MHz. The ratio $\Delta T_{pv}^{ther}|_{lin}/\Delta T_{pv}^{ther}|_{circ}$ is equal to $1.45 \pm 10\%$, close to 1.5, indicating heating originated from 2PA, as expected [6].

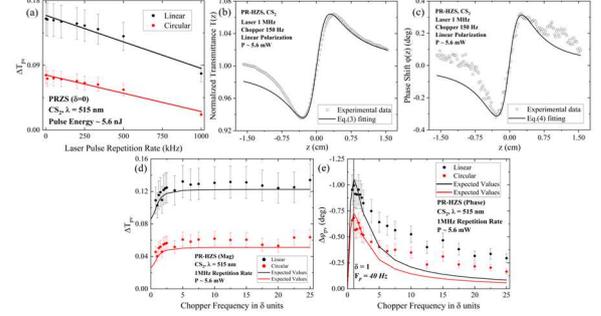


Figure 2: (a) PRZS measurements in CS_2 . (b,c) $T(z)$ and $\varphi(z)$, respectively, obtained by PR-HZS for linear polarization and $F = 150$ Hz. (d,e) Values of ΔT_{pv} and $\Delta \varphi_{pv}$, respectively, obtained by PR-HZS.

Then, we perform PR-HZS in CS_2 with fixed laser rate (1 MHz) and variable chopper frequency (20-1000 Hz). In this case, we put the chopper before the sample (Fig. 1). Fig. 2(b,c) shows one of the measurements, for linear polarization and $F = 150$ Hz, from which $\Delta T_{pv} > 0$ and $\Delta \varphi_{pv} < 0$, in accord to PRZS. By calculating ΔT_{pv} and $\Delta \varphi_{pv}$ for all δ , we get Fig. 2(d,e), respectively. For ΔT_{pv} , we can observe good agreement with the model proposed (Eqs. (5-7) with PRZS results). For $\Delta \varphi_{pv}$, the peak position at 40 Hz results in $\tau = (4.0 \pm 0.1)$ ms, reasonable for liquids [10]. On the other hand, the expected magnitude for $\Delta \varphi_{pv}$ do not match very well for high δ . This can be an indication that CS_2 can have thermal effects related to different physical processes with different response times.

Now, for the glasses, we have used pulse energies of 44.9 nJ for LaSF-N30, 8.2 nJ for SF6 and 23.3 nJ for the NDF. Firstly, the results of PRZS ($f = 1-1000$ kHz) in LaSF-N30, SF6 and the NDF, respectively, can be observed in Fig. 3(a-c) in which, all samples, present positive thermal contributions.

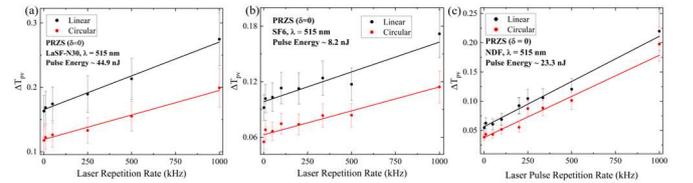


Figure 3: PRZS in (a) LaSF-N30, (b) SF6, and (c) NDF NE03A-B.

From this, using Eqs. (8,9) we summarize the results obtained with PRZS in Table 1, where the values of n_2^{ele} are consistent to ones of similar glasses [11], but all ΔT_{pv}^{nuc} obtained are small and inconclusive. In fact, we do not expect significant nuclear contribution for glasses. Furthermore, the obtained ΔT_{pv}^{ther} values are novel. Although most counterparts are not yet available in literature for direct comparison, the present results exhibit reasonable orders of magnitude [2].

Moreover, the ratio between $(\Delta T_{pv}^{ther})_{lin}$ and $(\Delta T_{pv}^{ther})_{cir}$ for LaSF-N30 and SF6 is, respectively, 1.38 and 1.25, with about 10% error. These values could indicate that the thermal process is excited partially by 2PA, since it is close to 1.5. On

the other hand, the NDF ratio is 1.1 ± 0.1 , which is a clear indication of linear absorption heating, since it is close to 1.0.

TABLE I. PRZS RESULTS FOR THE OPTICAL GLASSES

Parameters	Materials		
	LaSF-N30	SF6	NDF
ΔT_{pv}^{ele}	0.186 ± 0.005	0.092 ± 0.007	0.081 ± 0.009
$n_2^{ele} (\times 10^{-19} \text{ m}^2/\text{W})$	1.0 ± 0.2	2.6 ± 0.4	1.1 ± 0.3
ΔT_{pv}^{nuc}	-0.021 ± 0.005	0.007 ± 0.008	-0.001 ± 0.009
$(\Delta T_{pv}^{ther})_{lin}$	0.128 ± 0.007	0.08 ± 0.01	0.15 ± 0.01
$(\Delta T_{pv}^{ther})_{cir}$	0.093 ± 0.009	0.06 ± 0.01	0.14 ± 0.01

Then, PR-HZS ($f = 1 \text{ MHz}$, $F = 20\text{-}100 \text{ Hz}$) is performed in glasses and Fig. 4(a-c) illustrates some of the measurements acquired for $T(z)$ and Fig. 4(d-f) for $\varphi(z)$. It is possible to observe positive ΔT_{pv} and $\Delta\varphi_{pv}$, in accord to PRZS, and the difference in Δz_{pv} between LaSF-N30 and SF6 (strong electronic-nuclear NLR) and the NDF (strong thermal NLR).

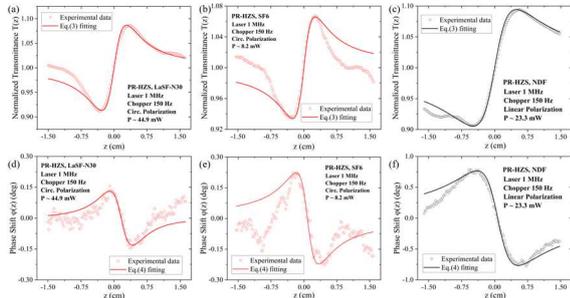


Figure 4: $T(z)$ and $\varphi(z)$ obtained by PR-HZS using $F = 150 \text{ Hz}$ in (a,d) LaSF-N30, (b,e) SF6, and (c,f) NDF NE03A-B.

By taking ΔT_{pv} and $\Delta\varphi_{pv}$ values for all δ and comparing to the model (Eqs. (5-7)) according to the PRZS results, it can be noticed, from Fig. 5(a-c), that ΔT_{pv} is higher than expected for high δ . On the other hand, from Fig. 5(d-f), $\Delta\varphi_{pv}$ obtained is lower than expected for low δ . These behaviors require further investigation for possible model improvement. Nevertheless, we observed a peak at $F = 150 \text{ Hz}$ for all samples, resulting in $\tau = (1.1 \pm 0.1) \text{ ms}$, which is reasonable for glasses [7].

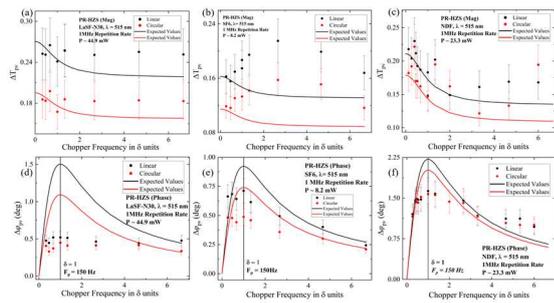


Figure 5: Values of ΔT_{pv} and $\Delta\varphi_{pv}$ as a function of δ obtained by PR-HZS in (a,d) LaSF-N30, (b,e) SF6, and (c,f) NDF NE03A-B.

The experimental results until now are promising and show the potential of the PR-HZS to discriminate three NLR effects using ultrafast high-fixed-repetition laser pulses.

CONCLUSIONS

The PR-HZS is a promising new method to discriminate and quantify three nonlinear refractive effects (electronic,

nuclear and thermal) from solvents and glasses. It discriminates the thermal lens from other nonlinearities when ultrafast laser pulses of fixed high repetition rate are applied. We could see good agreement between PRZS and PR-HZS.

ACKNOWLEDGMENT

We thank the founding agencies CNPq and FAPESP.

REFERENCES

- [1] Z. Chai, X. Hu, F. Wang, X. Niu, J. Xie, and Q. Gong, "Ultrafast All-Optical Switching," *Adv. Opt. Mater.*, vol. 5, no. 7, Apr. 2017, doi: 10.1002/adom.201600665.
- [2] R. W. Boyd, "The Intensity-Dependent Refractive Index," in *Nonlinear Optics*, Elsevier, 1992, pp. 159–190. doi: 10.1016/B978-0-12-121680-1.50008-4.
- [3] R. M. Moysés, E. C. Barbano, and L. Misoguti, "Discrimination of thermal, molecular orientation, and pure electronic refractive nonlinearities using the polarization-resolved Z-scan technique," *J. Opt. Soc. Am. B*, vol. 40, no. 4, p. C60, Apr. 2023, doi: 10.1364/JOSAB.482486.
- [4] M. Sheik-Bahae, A. A. Said, T.-H. Wei, D. J. Hagan, and E. W. Van Stryland, "Sensitive measurement of optical nonlinearities using a single beam," *IEEE J. Quantum Electron.*, vol. 26, no. 4, pp. 760–769, Apr. 1990, doi: 10.1109/3.53394.
- [5] A. Bundulis, I. Mihailovs, and M. Rutkis, "Origin of the Kerr effect: investigation of solutions by polarization-dependent Z-scan," *J. Opt. Soc. Am. B*, vol. 37, no. 6, p. 1806, Jun. 2020, doi: 10.1364/JOSAB.389520.
- [6] M. Falconieri and G. Salvetti, "Simultaneous measurement of pure-optical and thermo-optical nonlinearities induced by high-repetition-rate, femtosecond laser pulses: application to CS 2," *Appl. Phys. B Lasers Opt.*, vol. 69, no. 2, pp. 133–136, Aug. 1999, doi: 10.1007/s003400050785.
- [7] V. S. Zanuto, J. F. M. dos Santos, M. L. Baesso, and T. Catunda, "Single-beam time-resolved cw thermal Z-scan analysis applied in solids," *Opt. Laser Technol.*, vol. 142, no. March, p. 107248, Oct. 2021, doi: 10.1016/j.optlastec.2021.107248.
- [8] I. Guedes, L. Misoguti, L. de Boni, and S. C. Zilio, "Measuring optical nonlinearities using a heterodyne Z-Scan technique," in *2006 Conference on Lasers and Electro-Optics and 2006 Quantum Electronics and Laser Science Conference*, IEEE, 2006, pp. 1–2. doi: 10.1109/CLEO.2006.4628510.
- [9] R. A. Ganeev *et al.*, "Nonlinear refraction in CS₂," *Appl. Phys. B*, vol. 78, no. 3–4, pp. 433–438, Feb. 2004, doi: 10.1007/s00340-003-1389-y.
- [10] R. A. Cruz, M. C. Filadelpho, M. P. P. Castro, A. A. Andrade, C. M. M. Souza, and T. Catunda, "Very low optical absorptions and analyte concentrations in water measured by Optimized Thermal Lens Spectrometry," *Talanta*, vol. 85, no. 2, pp. 850–858, Aug. 2011, doi: 10.1016/j.talanta.2011.04.015.
- [11] J. Cimek, N. Liaros, S. Couris, R. Stępień, M. Klimczak, and R. Buczyński, "Experimental investigation of the nonlinear refractive index of various soft glasses dedicated for development of nonlinear photonic crystal fibers," *Opt. Mater. Express*, vol. 7, no. 10, p. 3471, Oct. 2017, doi: 10.1364/OME.7.003471.