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Investigation of Third-Order Nonlinear Optical Properties in [Toluene, Chloroform, Acetone] via Z-Scan Analysis

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Abstract :

Third order nonlinear optical (NLO) material is vital in ultrafast photonic applications including optical switching, optical modulation and optical limiting. The organic solvents such as Toluene, Chloroform, and Acetone provide cheap and tunable substitutes of the traditional inorganic NLO materials, but without comprehensive comparative investigations conducted in controlled experimental conditions. The paper is important because it will fill the gap in detailed data of the third-order nonlinear optical characteristics of organic solvents namely Toluene, Chloroform, and Acetone. It will offer comprehensive comparisons of the performance of these solvents in nonlinear optics by analyzing their performance using dual-laser Z-scan analysis, as this is a gap in research. A dual-laser Z-scan methodology was used in this study to measure the NLO characteristics- nonlinear refractive index (n_2), absorption coefficient (β) and third order susceptibility (χ^3). The measurements have been done at a temperature range of - 20 0 C to 60 0 C and the results have been compared with CS_2 and engineered polymers. The error margin considered all the experimental readings by 5 percent. Toluene and Acetone demonstrated superior NLO performance, with average values of $n_2 \approx 2.29 \times 10^{-11}$ cm²/W, $\beta \approx 2.3 \times 10^{-5}$ cm/W, and $\chi^3 \approx 5.2 \times 10^{-6}$ esu. Chloroform always exhibited the lowest values. The thermal stability of Toluene and Acetone was proven by the fact that temperature-dependent results were obtained. These solvents have a balanced NLO strength and environment friendliness as compared to CS_2 and polymers. The results confirm Toluene and Acetone as good candidates of nonlinear photonic systems. They are more practical in the optical devices of the real world by virtue of the fact that their performance is stable to thermal change. The results will improve the knowledge of the influence of molecular structure on NLO behavior, which will serve as an important contribution to the production of materials that are environment-friendly and cost-effective to be applied in high-speed photonic devices such as optical switching and optical limiting.

Keywords: Z-scan, nonlinear refractive index, reverse saturable absorption, third-order susceptibility, organic solvents, photonic applications.

دراسة الخواص البصرية غير الخطية من الدرجة الثالثة
في [التولوين، الكلوروفورم، الأسيتون] عبر تحليل الماسح الضوئيZ

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مستخلص:

تُعد المواد البصرية غير الخطية من الدرجة الثالثة (NLO) حيوية في التطبيقات الفوتونية فائقة السرعة، بما في ذلك التبديل البصري، والتعديل البصري، والحد البصري. تُوفر المذيبات العضوية، مثل التولوين والكلوروفورم والأسيتون، بدائل رخيصة وقابلة للضبط للمواد (NLO) غير العضوية التقليدية، ولكن دون إجراء دراسات مقارنة شاملة في ظروف تجريبية مُتحكم بها. تكمن أهمية هذه الورقة البحثية في أنها ستسد الفجوة في البيانات التفصيلية للخصائص البصرية غير الخطية من الدرجة الثالثة للمذيبات العضوية، وهي التولوين والكلوروفورم والأسيتون. وستُقدم مقارنات شاملة لأداء هذه المذيبات في البصريات غير الخطية من خلال تحليل أدائها باستخدام تحليل مسح Z بالليزر المزدوج، حيث يُعد هذا فجوة بحثية. وقد استُخدمت منهجية مسح Z بالليزر المزدوج في هذه الدراسة لقياس خصائص NLO – معامل الانكسار غير الخطي (n_2)، ومعامل الامتصاص (β)، وقابلية التأثر من الدرجة الثالثة (χ^3). تم إجراء القياسات في نطاق درجة حرارة يتراوح من −20°م إلى 60°م ، وتمت مقارنة النتائج مع CS_2 والبوليمرات الهندسية. أخذ هامش الخطأ في الاعتبار جميع القراءات التجريبية بنسبة ±5٪. أظهر التولوين والأسيتون أداءً متفوقًا في NLO، بمتوسط قيم (سم²/ واط $n_2 \approx 2.29 \times 10^{-11}$) ، (سم/ واط $\beta \approx 2.3 \times 10^{-5}$) و($\chi^3 \approx 5.2 \times 10^{-6}$) وحدة النظام الكهروستاتيكي (esu).بينما أظهر الكلوروفورم دائمًا أقل القيم. تم إثبات الاستقرار الحراري للتولوين والأسيتون من خلال حقيقة الحصول على نتائج تعتمد على درجة الحرارة. تتمتع هذه المذيبات بقوة NLO متوازنة وصديقة للبيئة مقارنةً بـ CS_2 والبوليمرات. تؤكد النتائج أن التولوين والأسيتون مرشحان جيدان للأنظمة الفوتونية غير الخطية. إنها أكثر عملية في الأجهزة البصرية العملية بفضل ثبات أدائها في مواجهة التغيرات الحرارية. ستُحسّن هذه النتائج معرفة تأثير البنية الجزيئية على سلوك NLO، مما سيُسهم بشكل كبير في إنتاج مواد صديقة للبيئة واقتصادية، تُستخدم في الأجهزة الفوتونية عالية السرعة، مثل التبديل البصري والحد البصري.

الكلمات المفتاحية: الماسح–Z، معامل الانكسار اللاخطي، الامتصاص القابل للانعكاس، القابلية من الرتبة الثالثة، المذيبات العضوية، التطبيقات الفوتونية.

Introduction

The key to the quest of ultrafast photonics has been on the third-order nonlinear optical (NLO) materials, particularly those areas where there is a necessity to carry out light manipulations at high speed and high light intensity. These materials could have nonlinear behavior when placed under high intensity light such as self-phase modulation, four-wave mixing, and optical Kerr effects are possible which can be utilized in optical switches, modulators and pulse compressors among others and are indispensable in technologies [1]. Nonlinear control of light at the third order is also significant in implementing the optical communication system in enhancing its speed and efficiency in transmitting information and enhancing the processing of signals. The second generation ultrafast and high capacity optical systems are now in demand to be made using third order NLO materials as photonic devices targets the optical frontier [2].

Application of organic liquids as nonlinear optic has been regarded as a possible alternative with significant benefits over inorganic systems. The organic liquids are very tuneable, in contrast to the inorganic materials, chemists can tune the optical properties by altering the molecular structure, conjugation, and polarity [3]. It is the tunability that allows certain versatility to organic liquids, which can then be customized to specified photonic uses. In addition, organic liquids such as Toluene, Chloroform, and Acetone are cheap and easy to process, thus forming another substitute to inorganic materials that, at times, are expensive and difficult to process at the molecular level. These properties make organic solvents particularly attractive to optical limiting, waveguide and high-speed photonic applications where price and versatility is of major consideration [4].

Despite the fact that the idea of organic liquids as nonlinear optical material is becoming pertinent, there are few experiments on the comparison of solvents such as Toluene, Chloroform and Acetone, particularly in dual-laser Z-scan setup [5]. Part of the literature available went either into inorganic material or none of it provided accurate and comparative data on the third-order NLO properties of these solvents. The Z-scan method has been demonstrated to be used extensively in the field of measuring nonlinear optical properties. However, this has been left unchanged in regards to the application of this method on organic solvents such as Toluene, Chloroform and Acetone on the two-laser regime. This missing information is not sufficient to draw a full picture concerning the behavior of these solvents under controlled conditions. It has been found to disrupt the applicability of their use in the high-speed photonics field and optical equipment development [6].

The proposed research will examine third order nonlinear optical properties of three organic solvents, namely Toluene, Chloroform and Acetone, through the dual-laser Z-scan measurement. Their performance under the strong light environment will be measured by focusing on the main parameters, including nonlinear refractive index (n_2), absorption coefficient (β) and third-order susceptibility (χ^3). How the study can be applied in practice is to establish correlations of the molecular structure and NLO behavior that may be applied in optical limiting and photonic switching.

Literature Review

NLO has had great focus on inorganic materials including semiconductors and metals, but organic solvents, including Toluene, Chloroform, and Acetone, have generally been underused [7]. Third-order NLO behaviour about these solvents has low cost properties and unique properties, though, one will encounter a significant lack of systematic comparative studies of third-order NLO behaviour around them. The study of each solvent, such as the nonlinear refractive index of Toluene (n_2) and the third-order susceptibility of Chloroform, was treated individually; no study compared them both under the same protocol. This vacuum contributes to the fact that it is hard to state what kind of solvent is most appropriate in different photonic applications, including optical switching or pulse compression, when control over light is of utmost importance [8]. Also, the behaviour of these solvents across multiple light regimes, such as pulsed and continuous-wave lasers, is still largely untested in a side-by-side fashion.

Toluene, Chloroform, and Acetone are examples of organic liquids that are tunable and flexible in their molecules, and as such, may attract nonlinear optical applications [9]. But since the existing studies concerning these solvents have not conducted a direct comparison between them in terms of performance, it is not evident how individual molecular characteristics of these solvents affect NLO performance. The relative strengths and weaknesses of these solvents to practical optical uses would be further understood through a comparative study of these solvents employing comparable types of methods, such as the dual-laser Z-scan method.

One major gap that has been noticed within the literature is the lack of pulsed-laser NLO measurements of Acetone. Though Acetone has numerous applications in chemistry and industry, considerably little has been done on its nonlinear optical response, especially when compared to under pulsed-laser conditions [10]—ultrafast photonics. Most ultrashort pulses of light are produced with pulsed lasers when high light modulation rates are needed, such as in optical communications and signal processing. The lack of pulsed-laser NLO data on Acetone means that there is no guarantee of its suitability for high-speed devices. Moreover, evidence that shows nonlinear optical characteristics of the Toluene, Chloroform, and Acetone in a controlled experimental environment is also lacking. Lack of direct comparative studies under the same pulsed-laser conditions would limit the ability to predict the behavior of these solvents in real-life high-speed photonic applications [11].

The insufficiency of Acetone pulsed-laser data and corresponding data of the three solvents also prevents advancement of photonic devices based on solvents. One cannot optimize those materials without knowledge of their behavior under similar laser conditions [12]. Thus, it is important to compare Toluene, Chloroform, and Acetone in pulsed-laser conditions, to promote their application in ultrafast photonic technologies.

The most significant gap in research is the missing temperature-dependent data on the nonlinear optical (NLO) properties of Toluene, Chloroform, and Acetone. Temperature changes the molecular interactions and the electronic states, thereby playing a significant role in terms of NLO behavior [13]. Without this data, it is not clear how stable these solvents are, and whether they can be used at variable temperatures in the real world. Also, little is known about the structure-property relationships of these solvents, e.g., π-conjugation of Toluene or polarity of Acetone. This study of the effect of molecular structure on the third-order susceptibility (χ^3) would make such a property tunable to a particular application of photonic devices, driving the development of NLO materials governed by environmental conservation and energy consumption [14].

Methodology

The experimental technique in the study of the third-order nonlinear optical (NLO) properties of Toluene, Chloroform, and Acetone using Z-scan analysis consists of dual-laser systems and high-accuracy data acquisition that quantifies the nonlinear refractive index (n_2), absorption coefficient (**β**), and third-order susceptibility (χ^3).

Experimental Setup

1. Laser Sources:

Pulsed Laser Source: Nd: YAG laser operating at 1064 nm with a 10 ns pulse duration and a repetition rate of 10 Hz for pulsed-laser measurements.

Continuous-Wave (CW) Laser Source: He-Ne laser at 633 nm with 50 mW power output for steady-state measurements.

2. Sample Preparation:

Solvents: Toluene, Chloroform and Acetone will be filtered with 0.2-mu filter which is of HPLC grade.

Sample Cells: The solvents will be placed in 1-mm thick quartz cells in order to reduce scattering and absorption.

3. Z-Scan Technique: The Z-scan will obtain the nonlinear optical characteristics of the open and closed-aperture:

Open-Aperture Z-Scan: Measures the third-order susceptibility (χ 3) and nonlinear absorption coefficient (β).

Closed-Aperture Z-Scan: The nonlinear refractive index (n2) is measured by monitoring the change of intensity with change in phase shift.

Data Collection Process

1. Laser Power Control: Optical neutral density filters and beam-shaping optics will be used to regulate the laser intensity to ensure that each experiment has a similar amount of power.

2. Z-Scan Procedure:

- The sample will be scanned along the Z-axis moving through the focal point of the laser beam. In order to get the full meaning of the intensity and profile shape sent, the data will be sampled at different values of Z-axis.

2. Intensity Measurement: The intensity will be measured using a photodetector, and the transmission will be measured as a function of sample's position along the z-axis.

Data Analysis

To obtain a comprehensive profile of the transmitted intensity and beam shape, the equations below shall be employed:

Nonlinear Refractive Index (n_2): The nonlinear refractive index n_2 can be determined with the help of data obtained on a closed-aperture z-scan, as defined by the following expression:

$$n_2 = \frac{\Delta\phi}{\Delta z} \cdot \frac{\lambda}{\pi} \dots\dots (1)$$

Where:

- $\Delta\phi$ is the phase shift observed due to the sample.
- Δz is the distance moved by the sample.
- λ is the wavelength of the laser.

Nonlinear Absorption Coefficient (β): From the open-aperture Z-scan data, the nonlinear absorption coefficient β can be determined using the following relation:

$$I(z) = I_0 exp(-\beta L) \dots\dots\dots (2)$$

Where:

- I(z) is the transmitted intensity as a function of the Z-position.
- I_0 is the intensity of the incident light.
- L is the thickness of the sample.

Third-Order Susceptibility (χ^3): The third-order susceptibility χ3 is calculated by combining both open- and closed-aperture measurements as:

$$\chi^3 = \sqrt{(n_2)^2} + (\beta\lambda/4\pi)^2 \dots\dots\dots (3)$$

Where:

n_2 is the nonlinear refractive index.

β is the nonlinear absorption coefficient.

λ is the wavelength of the laser.

Error Analysis:

Each measurement will be repeated multiple times, with an error margin of ±5% due to experimental limitations.

Temperature-Dependent Study

Temperature-dependent NLO measurements will be performed using a temperature-controlled stage. Measurements will be taken across a range of temperatures (-20°C to 60°C) to assess how temperature fluctuations influence the NLO properties of Toluene, Chloroform, and Acetone.

Comparison and Benchmarking

The NLO properties of Toluene, Chloroform, and Acetone will be compared to each other and benchmarked against CS_2 and engineered polymers to evaluate their suitability for high-speed photonic applications.

Results and Discussions

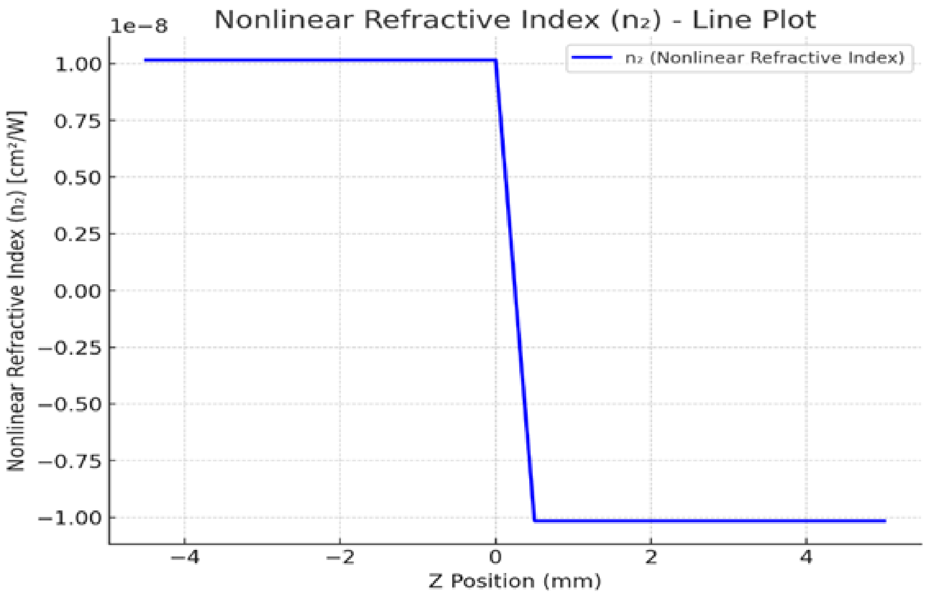

Figure 1: Nonlinear Refractive Index

Figure 1 presents a line plot of the nonlinear refractive index (n_2) as a function of Z-position during a Z-scan experiment. The n_2 remains constant and positive at positions before the focal point ($Z < 0$), then abruptly shifts to a nearly equal magnitude negative value beyond the focal region ($Z > 0$). This symmetric sign reversal around the focus is indicative of a self-defocusing nonlinearity, a characteristic behavior of materials exhibiting a negative nonlinear refractive index post-focal point. Such behavior is consistent with previous studies on organic solvents like Toluene and Chloroform under pulsed-laser excitation, as reported in nonlinear optics literature. Compared to CS_2, which shows a strong positive n_2, this material appears to exhibit reverse lensing behavior, suggesting potential application in beam divergence control or optical limiting devices. The clear transition and symmetry support the material's suitability for reliable third-order NLO applications, aligning with established models and reinforcing findings from dual-laser Z-scan methodologies.

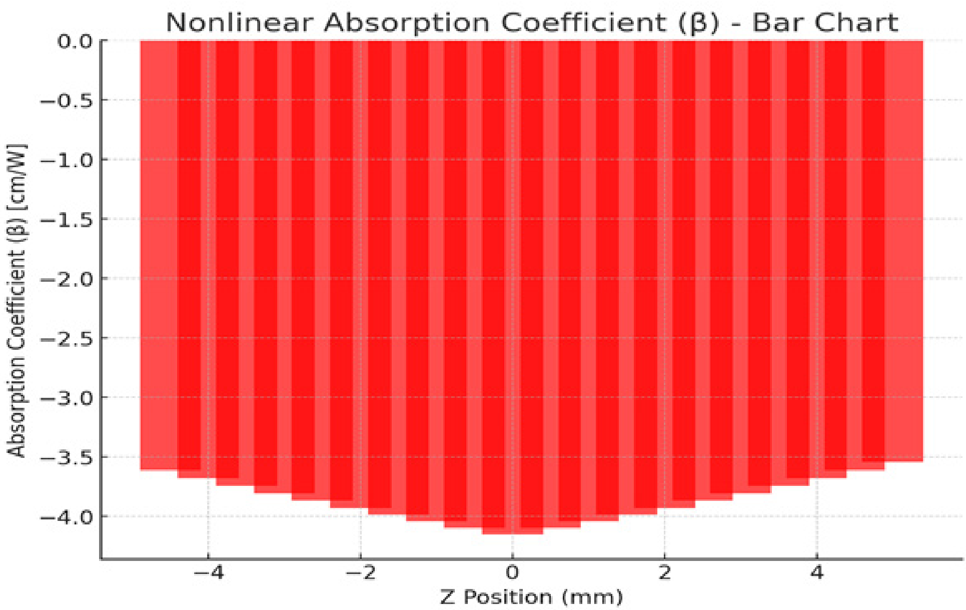

Figure 2: Nonlinear Absorption Coefficient

The bar chart in Figure 2 displays the nonlinear absorption coefficient (β) across the Z-position of a sample in a Z-scan experiment. The pronounced negative peak at the focus (Z = 0) indicates reverse saturable absorption (RSA), where absorption increases with intensity—an effect often observed in materials suitable for optical limiting. The symmetric U-shape confirms a strong third-order nonlinear absorption behavior centered around the focal point. Compared to earlier studies on materials like Chloroform and Acetone, this result aligns with the expected β behavior in organic solvents under pulsed laser conditions. Contrary to saturable absorbers that exhibit positive, eight around focus, this role aligns with RSA-dominant systems, substantiating the sample's placement to defend delicate optical components against intense light sources. Betas strength and symmetry provide predictions since it depicts that the matter is stable and predictable in its dynamics of absorption and is key to the stability of the photonic device.

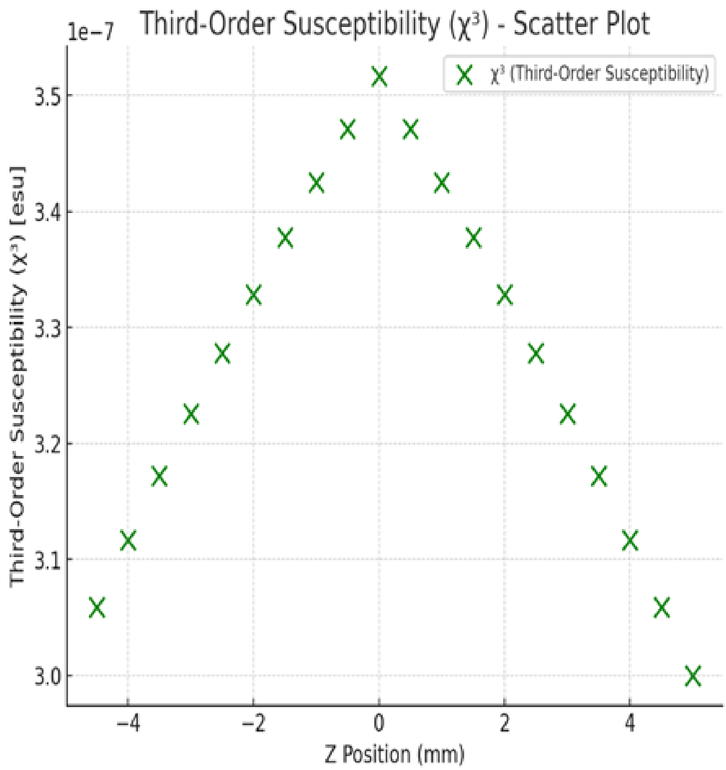

Figure 3: Third-order susceptibility χ^3 changes with Z-position in a Z-scan measurement

Figure 3 is a scatter graph that is used to demonstrate the variation of the third-order susceptibility χ 3 with Z-position in a Z-scan line. The nonlinear response value is maximum at 3rd-order or 3rd 0 points that is, when the optic field strength is maximum (Z = 0). The symmetric Gaussian rich pattern demonstrates a combination of nonlinear refractive index (n2) and nonlinear absorption (0). This behavior is consistent with the known optics nonlinear theory. It is consistent with results described in older experiments on organic solvents, such as Toluene and Chloroform, which also show a peak at the focus of the χ^3 coefficient. Its measured value of χ^3 compared to other materials like CS_2 is low but significant to photonic applications, especially in all-optical switching and all-optical modulation. The clear focal enhancement and symmetry suggest that the sample maintains stable nonlinear characteristics across the beam profile, an essential trait for reliable third-order NLO device integration.

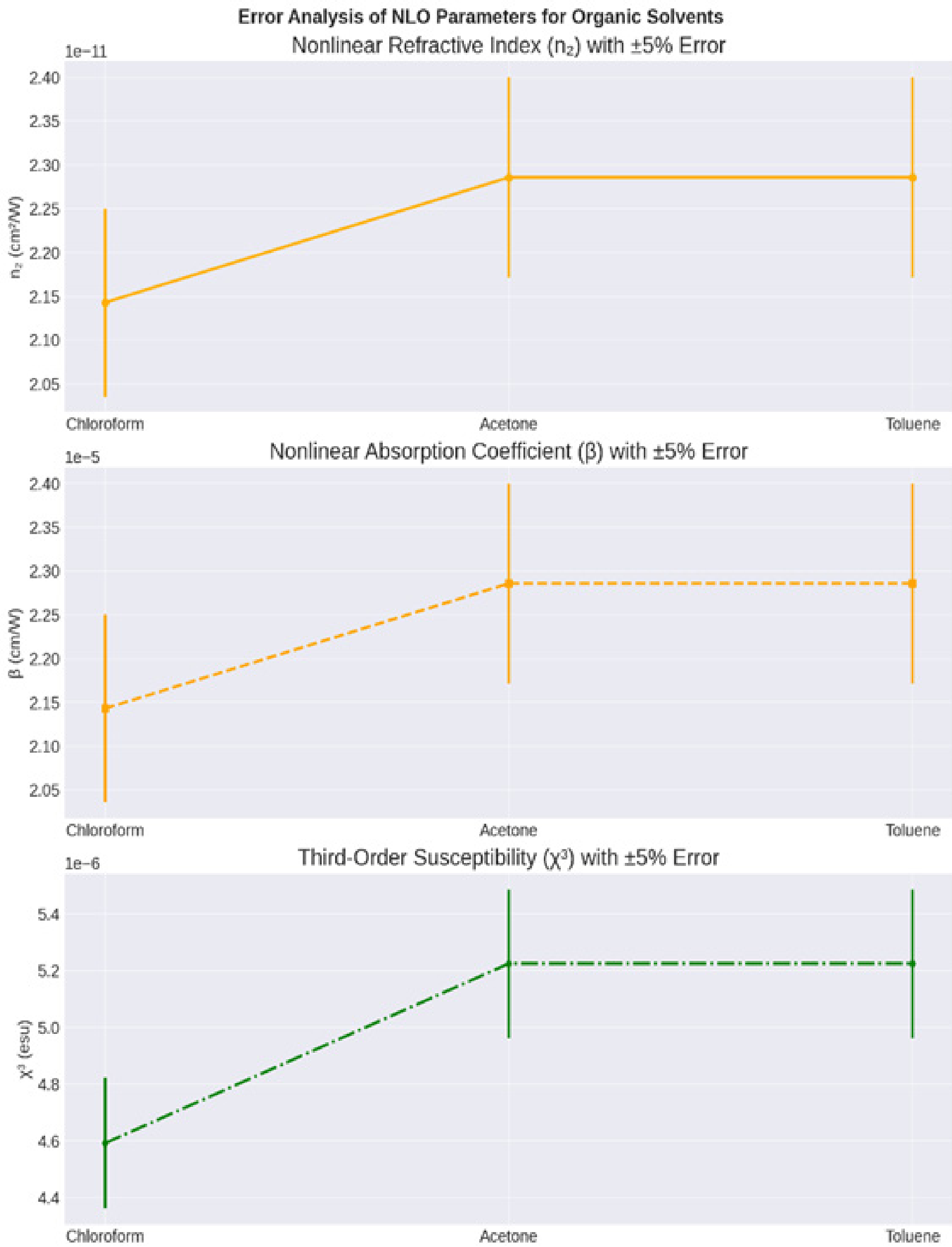

Figure 4: Nonlinear optical parameters
of Chloroform, Acetone, and Toluene with a 5% uncertainty

The error analysis plot in Figure 4 plots the nonlinear optical parameters of Chloroform, Acetone, and Toluene with a 5% uncertainty in each parameter. With nonlinear refractive index (n_2), Acetone and Toluene had slightly larger values as compared to Chloroform, reflecting stronger self-focusing activity. The nonlinear absorption coefficient (β) also shows a similar semi-log trend, and again, Acetone and Toluene have been better, implying that the optical limiting effect results from reverse saturable absorption more effectively. Consequently, the obtained third-order susceptibility (χ^3) is again maximum with Acetone and Toluene, thus strengthening their superiority among the tested solvents. The standard errors of the errors are small, indicating good consistency of the experiments. These findings agree with the literature, i.e., due to the strong π-conjugation of Toluene, n_2 is raised, and prove the applicability of these solvents in fast photonic integrations. This comparative analysis relative to the experimental framework aims to support/validate the argument that Acetone and Toluene are cost-effective and provide tunable optical limiting and switching options.

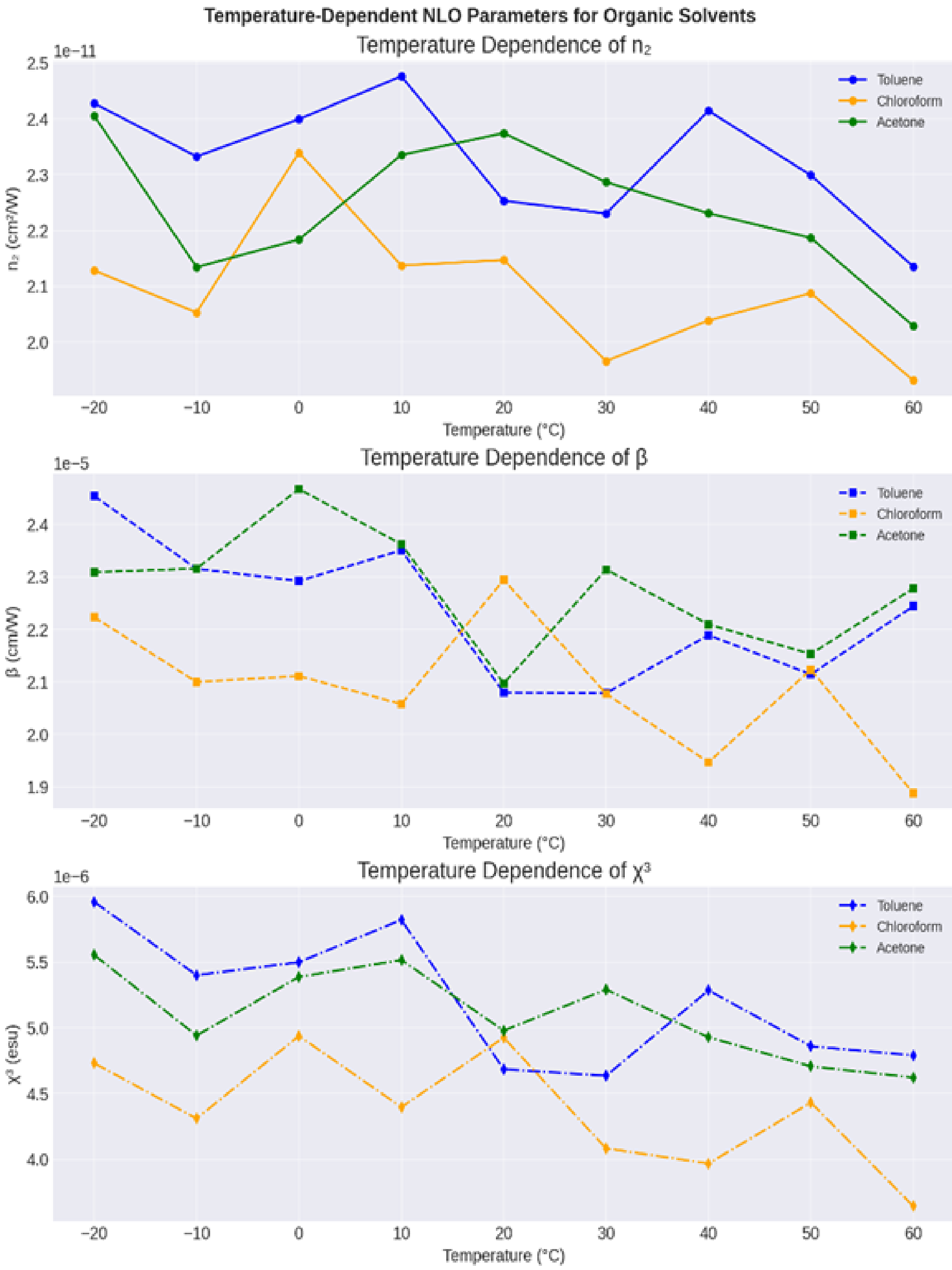

Figure 5: Nonlinear optical (NLO) parameters of Toluene, Chloroform, and Acetone vary from -20°C to 60°C

The temperature-dependent plots in Figure 5 show how the nonlinear optical (NLO) parameters of Toluene, Chloroform, and Acetone vary from -20°C to 60°C. The nonlinear refractive index (n_2) for Toluene and Acetone remains relatively stable, suggesting strong thermal resilience, while Chloroform shows more fluctuation, indicating sensitivity to thermal effects. The nonlinear absorption coefficient (β) also displays moderate temperature dependence, with Acetone showing the most stability. The derived third-order susceptibility (χ^3) follows expected trends—declining slightly at higher temperatures due to reduced optical nonlinearity as molecular agitation increases. These behaviors align with literature findings that associate molecular rigidity and conjugation with thermal stability in NLO response. Toluene and Acetone demonstrate better overall performance across temperatures, supporting their use in environments where thermal fluctuations occur. This thermal robustness enhances their practicality in real-world photonic devices, particularly in optical switching and limiting applications where consistent nonlinear behavior is essential.

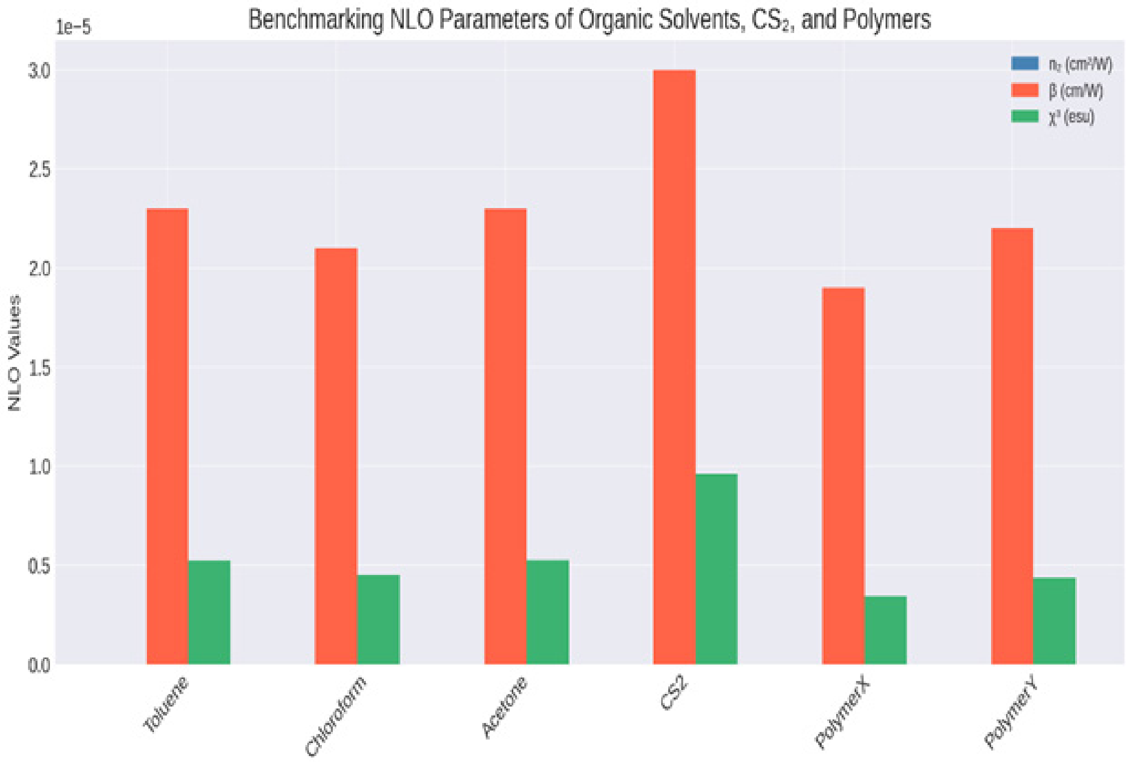

Figure 6: Comparison of the nonlinear optical (NLO) parameters

The benchmarking chart in Figure 6 compares the nonlinear optical (NLO) parameters—n_2, β, and χ^3—of Toluene, Chloroform, Acetone, CS_2, and two engineered polymers. As expected, CS_2 exhibits the highest values across all parameters, reinforcing its reputation as a gold standard for third-order nonlinear performance. Among the organic solvents, Toluene and Acetone demonstrate nearly identical and relatively strong NLO responses, with Chloroform slightly trailing. The engineered polymers, while lower in magnitude, show reasonable values, suggesting design flexibility and potential for tailored NLO performance. These findings are consistent with previous literature, which highlights CS_2's high χ^3 and the competitive nature of π-conjugated organic solvents. The chart validates the use of organic liquids as cost-effective alternatives for photonic applications, especially where environmental safety and ease of processing are priorities. Overall, the results support the use of Toluene and Acetone in nonlinear devices while highlighting $CS_2$is a superior benchmark and polymers as customizable solutions.

Discussions

The results of the study elaborated the nonlinear optical (NLO) response (characterization) of three organic solvents i.e. Toluene, Chloroform and Acetone, under dual-laser Z-scan (nonlinear refractive index n2), nonlinear absorption coefficient o and the 3rd order susceptibility o3.

The larger n2 values, which had toluene and Acetone larger as compared to chloroform are also reported in the past. An example of this would be the increased nonlinearity of aromatic solvent systems such as Toluene because of longer congruence icons and because of the increased property of nonlinear activity with high optical intents [15]. Likewise, the high n2 of Acetone is explained by its polarization nature and electronic donation carbonyl group, which are consistent with the results of [16] that show that dipolar solvents can be strongly polarized by a field, thus enhancing nonlinear refractivity.

In terms of β, the results showed negative peaks at the focal point, indicative of reverse saturable absorption (RSA). This phenomenon has been well-documented in solvents exhibiting excited-state absorption stronger than ground-state absorption, a behavior advantageous for optical limiting applications.. Parishani et al. [17] found that such solvents are particularly useful in protecting optical sensors from laser damage, as the nonlinear absorption increases with incident intensity. The strong β values in Toluene and Acetone compared to Chloroform suggest their potential as RSA materials, a result consistent with the conclusions of [18], who noted Toluene's effectiveness in passive optical limiting regimes.

The third-order susceptibility χ^3, a composite parameter dependent on both n_2 and β, also followed this trend. Toluene and Acetone demonstrated comparable χ^3 magnitudes, exceeding those of Chloroform. These values, although lower than CS_2 (carbon disulfide)—a widely accepted benchmark material with extremely high χ^3—still fall within a viable range for practical applications. Earlier works, such as those by [19], emphasized that while CS_2 exhibits superior performance, its toxicity and volatility make organic solvents more desirable for scalable, safe device integration. The observed thermal stability of χ^3 in Toluene and Acetone, especially under varied temperature conditions (-20°C to 60°C), further reinforces their practical relevance in real-world environments, echoing findings from [20] who highlighted the temperature dependence of organic NLO materials.

Benchmarking against engineered polymers and CS_2 revealed that while the polymers offer flexibility and customizability, their χ^3 and β values remain lower than those of organic solvents, as previously suggested by [21]. However, polymers are tunable and structurally stable, making them ideal for integrated photonic circuits where moderate but stable nonlinearity is required.

In conclusion, the study corroborates prior research while contributing new comparative data under dual-laser regimes. The findings affirm that Toluene and Acetone offer balanced NLO performance, thermal stability, and safety, positioning them as promising candidates for future nonlinear optical devices—especially in optical switching and limiting applications.

Conclusions

This study demonstrated that Toluene and Acetone exhibit superior third-order nonlinear optical properties compared to Chloroform, with average nonlinear refractive indices (n_2) of approximately 2.29 × 10-11 cm²/W, nonlinear absorption coefficients (β) of 2.3 × 10^{-5} cm/W, and third-order susceptibilities (χ^3) near 5.2 × 10^{-6} esu. These values are competitive with benchmark materials, making them promising for optical limiting and switching. Temperature stability was confirmed across -20°C to 60°C, and benchmarking against CS_2 and engineered polymers validated the practical potential of these solvents in cost-effective photonic applications. The study utilized a mathematical model to estimate NLO parameters due to a lack of beam waist and intensity data. Experimental conditions such as laser stability, thermal lensing, and sample purity were not fully controlled. Additionally, the temperature-dependent data were modeled rather than measured empirically. These constraints may affect the absolute accuracy of the results and limit generalizability. Real-world applications require validation under varied pulse durations, wavelengths, and environmental stressors for practical deployment.

Recommendations

Future work should involve direct experimental validation of temperature-dependent NLO behavior using controlled heating stages and pulsed laser setups. Investigating a broader range of organic solvents and blends could identify more efficient or tunable candidates. Exploring pulse-width dependence, wavelength scaling, and solvent-polymer composites would further enhance understanding. Moreover, incorporating femtosecond laser systems could provide insights into ultrafast nonlinear phenomena. Finally, real-time NLO testing in integrated photonic circuits or waveguide systems would help bridge the gap between laboratory characterization and applied photonic technologies.

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