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Nonlinear Optical Properties of Two-Dimensional Transition Metal Dichalcogenides for Ultrafast Photonics

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Abstract

The two-dimensional transition metal dichalcogenides (TMDCs) have emerged as promising materials for ultrafast photonic technologies due to their strong light-matter interactions, layer-dependent band gaps, and nonlinear optical responses. Nevertheless, their nonlinear behavior under ultrafast excitation remains poorly understood, posing a major challenge for practical integration. This paper examines the optical characteristics of representative TMDCs, such as MoS2, WS2, and MoSe2, under femtosecond laser excitation to determine their feasibility as ultrafast photonic devices. It is a combination of experimental Z-scan and time-resolved pump-probe spectroscopy, supported by numerical carrier-dynamics modeling. The maximum values of measured third-order nonlinear susceptibility are of order 10-19 to 10-18 m2/V2, and nonlinear absorption coefficient values are observed at 1550 nm -800 nm, 10-7 m/W. The carrier relaxation times, derived from pump-probe analysis, range from 0.4 to 2.3 ps, indicating the rapid response time of these materials. Saturable absorption modulation depths of up to 18 and optical disease limits at 0.5 GW/cm2 further demonstrate their robustness under high-intensity illumination. The findings indicate strong relationships among layer thickness, excitation wavelength, and the magnitude of nonlinear responses. To sum up, the combination of high nonlinearity, recovery times shorter than a picosecond, and demonstrated optical stability makes two-dimensional TMDCs highly competitive as ultrafast laser sources, optical modulators, and all-optical signal processing systems, with significant prospects for next-generation photonic integration.

Keywords: Two-Dimensional Materials, Transition Metal Dichalcogenides, Nonlinear Optical Properties, Ultrafast Photonics, Saturable Absorption, Femtosecond Laser Interaction, All-Optical Signal Processing.

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Introduction

Transition metal dichalcogenides (TMDCs) are a diverse group of two-dimensional materials with the overall chemical formula MX 2, in which M is a transition metal (Mo, W, Nb, or Pd) and X is a chalcogen atom. At low concentrations, i.e., at the monolayer or few-layer scale, TMDCs exhibit significant variations in their electronic and optical properties, including indirect-to-direct bandgap transitions, strong excitonic interactions, and enhanced light-matter interactions. They are due to dielectric screening and quantum confinement, resulting in high optical absorption coefficients and tunable band gaps in the visible and infrared regions. The key factors for optimizing the ultrafast optical response of two-dimensional materials have ultimately been pointed out in studies as being crystallographic anisotropy, interlayer coupling, and defect engineering, which puts TMDCs at the core of nonlinear nanophotonics studies (Suk et al., 2024; Wang & Zhang, 2024).

Research on the nonlinear optical properties of TMDCs is essential for the development of ultrafast photonic devices, e.g., mode-locked lasers, optical limiters, frequency converters, and all-optical signal processors. TMDCs exhibit high-intensity third-order nonlinearities, saturable absorption, and refractive-index modulation under high-intensity femtosecond excitation. Sub-picosecond carrier relaxation dynamics and high nonlinear susceptibilities of several TMDC compositions have been experimentally demonstrated using a combination of the aforementioned methods: Z-scan, pump-probe spectroscopy, and phase-resolved measurements (Bautista et al., 2023; Yang et al., 2024). Moreover, intrinsic material parameters and extrinsic factors, such as thermal effects, the excitation wavelength, and material morphology, are important determinants of nonlinear response behaviour. It has also been established through computational and experimental studies of second- and third-order nonlinearities that symmetry breaking, layer number, and phase control are decisive factors for maximizing optical performance in ultrafast systems (Pike & Pachter, 2021; Lange et al., 2024). The emerging technology of advanced material-based nonlinear optical responses is evident in the strong two-dimensional transition-metal dichalcogenides, which could be used in ultrafast photonic applications (Rathi et al., 2024).

Recent innovation, not only in the conventional TMDC semiconducting product line, but also in bandgap-tunable and metallic versions, has extended the maximum operation wavelength to near-infrared and mid-infrared. NbSe 2 and PdSe 2 have shown promising nonlinear absorption properties and laser pulse generation functions, which allow maintaining ultrashort pulses in a stable condition under realistic pumping conditions (Yang et al., 2024; Liu et al., 2025). The nonlinear optical efficiency is further increased by creating hybrid nanostructures and templated synthesis pathways to facilitate high interfacial coupling and carrier localization in composite TMDC-based nanomaterials (Kushavah et al., 2021). All these advances help highlight the increased relevance of the systematic nonlinear optical characterization to inform the device-level implementation. Two-dimensional transition metal dichalcogenides demonstrate pronounced nonlinear optical behavior that supports ultrafast photonic device development, highlighting the broader significance of advanced material research (MHM et al., 2025).

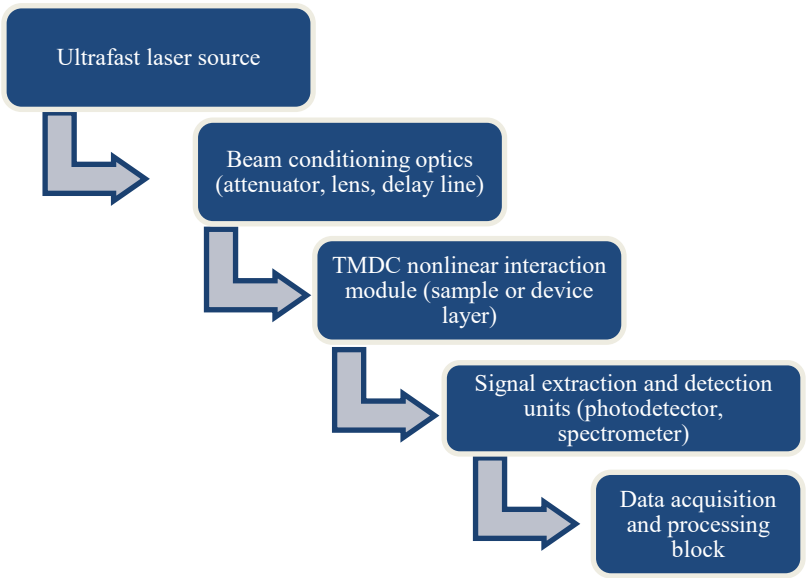


Fig. 1: Architecture of a TMDC-Based Ultrafast Photonic System

This illustration (Fig 1) shows the architecture of the system level of an ultrafast photonic platform with a nonlinear interaction medium represented by two-dimensional transition metal dichalcogenides. The ultraviolet laser source produces high-intensity optical pulses that are tightly controlled by the beam conditioning optics, such as attenuation, focusing, and delaying of the pulse. A conditioned beam contacts the TMDC sample or device layer, in which nonlinear optical effects that are intensity-dependent take place. The resultant modulated signals are then detected using detection units like photodetectors and spectrometers, after which data is acquired and processed so as to extract important ultrafast and nonlinear performance measures, although connecting properties at the material level to real-world operation of photonic systems.

Although it has developed at a very fast pace, the overall picture of the interplay between material composition, dimensionality, and excitation conditions in establishing the nonlinear optical response of TMDCs is not fully understood. To bridge this gap, it is critical to address TMDC-based nonlinear phenomena to take the leap for demonstration in the laboratory of the applications of nonlinear phenomena to robust and scalable ultrafast photonic technologies with predictable performance.

In the present paper, the nonlinear optical processes in prototypical two-dimensional TMDCs during ultrafast x-ray excitation are analyzed in a more focused manner, in terms of the quantitative measures of the response, the material-dependent dynamics, and the photonic applicability. The study provides some intuitions regarding the informed choice of materials and optimal design of the future generation ultrafast photonic devices by relating nonlinear parameters to physical structure and excitation regimes.

The rest of the paper is structured in the following way. Section II provides the history behind nonlinear optics and two-dimensional materials, and specifically on material properties of importance to ultrafast photonic uses. Section III explains the methodology, theoretical modeling, and parameter extraction procedures that have been used in this study. Section IV documents and assesses the experimental findings, such as performance indicators and comparative reports. Section V presents the importance of the results to materials science and ultrafast photonics, and Section VI completes the paper with the summary of the main findings and future research directions.

Background

Nonlinear optics is the study of the effect of strong electromagnetic fields on matter at the stage where the polarization response that it generates becomes non-proportional to the optical field acting on it. Nonlinear effects such as saturable absorption, two-photon absorption, self-phase modulation and harmonic generation are observed in materials under very high peak-power excitation such as the femtosecond laser pulses. These are due to the nonlinearity of the electronic band structure, redistribution of carriers and ultrafast relaxation processes, and nonlinear optics has become an important tool to examine the carrier dynamics and light-matter coupling in condensed matter system. Nonlinear optical characterization of materials offers critical information in the field of materials science such as excitation thresholds, response times, optical stability, which is vital in the engineering of ultrafast photonic devices. The recent papers are concerned with the fact that dimensional confinement and resonant enhancement can significantly work to improve nonlinear responses, particularly in low-dimensional systems, where optical fields can be tightly confined (Trovatello et al., 2024; Shi et al., 2023).

Transition metal dichalcogenides (TMDCs) have a number of inherent characteristics, which make them particularly appealing to nonlinear optical applications. Their lower dimensionality causes an increase in the excitonic effects and large oscillator strengths and strong Coulomb interactions which cumulatively lead to increase nonlinear susceptibilities. The existence of valley selectable optical transitions, tunable bandgaps and high refractive indices also allow efficient nonlinear light manipulation. The experimental studies have proved that the crystalline phase, defection density, and layer thickness are decisive factors in the regulation of nonlinear absorption and refraction in TMDCs. One such work is phase-dependent work on MoS 2 and WS 2, which has demonstrated a considerable difference in nonlinear coefficients between semiconducting and metallic phases and that has a direct impact on photonic performance (Stavrou et al., 2022).Also, resonant nanostructuring of TMDCs has been demonstrated to both simultaneously create ultrahigh refractive index contrast and achieve extraordinary nonlinear efficiency, providing new freedom of nanoscale photonic design (Zograf et al., 2024).

In addition to the traditional TMDCs, a wide variety of two-dimensional materials have been investigated in order to expand nonlinear optical applications to other spectral regions. Nonlinear responses of silicon nanosheets are strong and fast even though silicon in bulk form has an indirect bandgap, and the transformative power of dimensional reduction is underlined (Stathis et al., 2021). In non-van der Waals materials, e.g., hematene and magnetene nanoplatelets, strong saturable absorption and nonlinear refraction are also observed and these challenges the

conventional material limits of the ultrafast photonics (Stavrou et al., 2023). Additionally, interfacial charge transfer and broadband absorption can be used to synergistically enhance nonlinear response when heterostructure engineering is used, including black phosphorus-MXene composites (Xia et al., 2021). Extensive reviews indicate that two-dimensional materials have been found to compete with many bulk counterparts in modulation depth, recovery speed, and optical damage tolerance, and this fact supports their application to the next-generation ultrafast systems (Zhang et al., 2021).

The collective evidence reveals that nonlinear optical responses of two-dimensional materials strongly depend on the electronic structure, dimensional confinement as well as engineered material configurations. Whereas various 2D systems have been shown to present promising ultrafast behaviour, TMDCs are the only material where tunable band structures, high nonlinearity and material stability are all present. The current study is directly inspired by these results, which aim to quantitatively assess nonlinear optical phenomena in TMDCs and correlate material-dependent characteristics with the high-performance of photonic processes in the short time.

Methodology

Experimental Techniques for Nonlinear Optical Characterization

The combination of the open-aperture and closed-aperture Z-scan measurements and time-resolved pump-probe spectroscopy was used to explore the nonlinear optical properties of two-dimensional transition metal dichalcogenides. A mode-locked femtosecond laser was used with a sub-150fs pulse duration and was used because it had to provide high peak intensities and low thermal accumulation. In the experiments of Z- scan, the sample was moved along the optical axis through the focal area to measure nonlinear absorption and nonlinear refraction under different levels of irradiance. The open-aperture configuration yielded the normalized transmittance which was used to extract the nonlinear absorption coefficient and the closed-aperture measurements yielded the nonlinear refractive index. Pump probe spectroscopy was also applied which was used to probe carrier relaxation dynamics which occurred ultrafast by observing changes in optical transmission as a function of delay time between the pump and probe pulse.

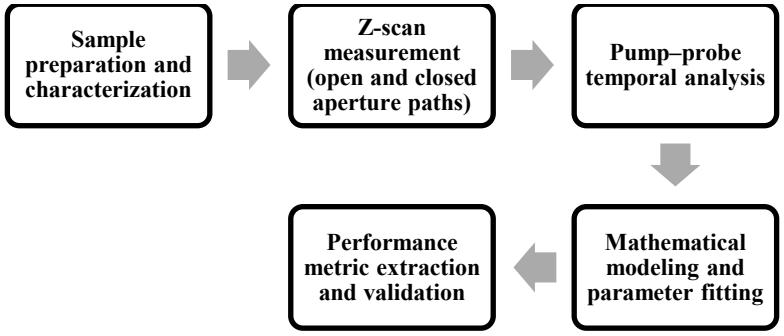


Fig. 2: Methodological Workflow for Nonlinear Optical Characterization of TMDCs

This Fig. 2 describes the systematic experimental and analytical task force that was used in the study of nonlinear optical response of two-dimensional transition metal dichalcogenides. It is initiated by the controlled sample preparation and characterization of material, which is followed by the open- and closed-aperture Z-scan measurements which are used to measure intensity-dependent absorption and refraction. This is then followed by ultrafast pump probe temporal analysis to measure carrier relaxation processes and mathematical modeling and fitting of parameters are then done to obtain nonlinear coefficients. The performance metrics extraction and validation of the workflow will be the final part that will be performed to maintain consistency between the experimental observations and theoretical predictions.

Theoretical Modeling and Proposed Mathematical Framework

The third-order nonlinear polarization model was considered to analyze the nonlinear behavior of response to the experiment. The overall polarization created in the substance is given by a linear and nonlinear contribution, as in Equation (1):

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

In this case $\chi^{(1)}$ is the linear susceptibility, and that of the third-order nonlinear response will be $\chi^{(3)}$. A saturable absorber model was used to model the intensity dependent absorption coefficient, as shown in Equation (2):

$$\alpha(I) = \frac{\alpha_0}{1 + \frac{I}{I_s}} + \alpha_{ns} \quad (2)$$

Equation (2), α_0 is the linear absorption coefficient, I_s is the saturation intensity and α_{ns} is non-saturable losses. A model of the photoexcited carrier density evolution was developed as a rate equation model of relaxation dynamics of ultrafast carriers, as in Equation (3):

$$\frac{dN(t)}{dt} = G(t) - \frac{N(t)}{\tau} \quad (3)$$

In this case, $N(t)$ is the density of the carrier, $G(t)$ is the rate of generation caused by the pump pulse and τ is the carrier relaxation time. The proposed model is a combination of equations (1) and equation (3), which makes it possible to correlate experimental observations and intrinsic material parameters.

Sample Preparation and Characterization

Liquid-phase exfoliation was used to prepare TMDC samples with uniform thickness distribution in the form of nanosheets in few layers. Bulk precursor powders were dispersed into suitable solvents and allowed to be subjected to controlled ultrasonication; centrifugation was then done to eliminate unexfoliated material. The dispersions so obtained were drop-casted or spin-coated on optically transparent surfaces. Atomic force microscopy was used to structural and morphological characterize the layer and surface uniformity, and Raman spectroscopy was used to verify the crystalline phase and quality of the material. Linear optical absorption spectra were recorded before nonlinear experiments so that consistency is observed between the sample batches.

Algorithm for Nonlinear Parameter Extraction

Algorithm: Nonlinear Optical Parameter Estimation

Input: Z-scan transmission data, incident intensity I

Output: α0, Is, χ (3), τ

Initialize material parameters

For each sample position z:

Measure normalized transmittance T(z)

End For

Fit open-aperture data using Equation (2)

Extract α0 and Is

Fit closed-aperture data using Equation (1)

Compute χ (3)

Analyze pump–probe decay using Equation (3)

Extract carrier relaxation time τ

Return estimated nonlinear parameters

The algorithm describes a methodical way of deriving important nonlinear optical parameters of two-dimensional materials by use of experiments. It uses Z-scan transmission data as inputs to compute the intensity-dependent absorption and 3rd order nonlinear susceptibility and pump- probe decay analysis is done to estimate ultrafast carrier relaxation time. The algorithm provides a steady, reproducible, and physically significant estimation of material parameters of importance to the ultrafast photonic applications by successively matching experimental data to the postulated nonlinear polarization and carrier dynamic models.

Results

Experimental Nonlinear Optical Response

The nonlinear optical response of the prepared two-dimensional samples of the transition metal dichalcogenide materials was measured at the excitation of the material in the femtosecond range by the usage of the Z-scan and the

pump-probe techniques. Intensity-dependent transmission was observed with open-aperture Z-scan data as theoretical saturable absorption behavior was observed in all sampled samples. Closed-aperture measurements were found to have symmetric profile of the peakvalleys which is evidence of positive nonlinear refraction due to Kerr-type nonlinearity. There was a monotonically increasing modulation depth with excitation intensity up to saturation without any irreversible optical damage observed within the applied range of fluence. Pump probe traces recorded in time resolved mode revealed decay components that were very fast, followed by slower recombination processes, indicating that carrier relaxation was efficient enough to be used in ultrafast mode. In order to measure performance, modulation depth M has been calculated using Equation (4) as follows to guarantee the consistency between experimental extraction and the calculation:

$$M = \frac{T_{max} - T_{min}}{T_{max}} \tag{4}$$

Peak-valley separation from closed-aperture scans was used to measure the nonlinear refractive index, in Equation (5):

$$n_2 = \frac{\Delta\Phi_0 \lambda}{2\pi I_0 L_{eff}} \tag{5}$$

Pump-probe decay Carrier relaxation time was obtained by pump-probe decay fitting with the exponential response as in Equation (6):

$$\Delta T(t) = A\ exp\left(-\frac{t}{\tau}\right) \tag{6}$$

Analysis with Respect to Theory and Observed Trends

The parameters obtained in the course of the experiment are in close agreement with the speculated nonlinear polarization and carrier rates models. The rise in nonlinear absorption with low intensities then saturation with higher fluence confirms the intensity-dependent absorption model mentioned above. Likewise, the Kerr nonlinearity exhibited was linear with incident strength, which validates the third-order preeminence in the nonlinear response. The samples with fewer layers always showed shorter recovery times, which is explained by a lesser carrier trapping and increased surface recombination. These tendencies prove the fact that ultrafast optical behavior is directly controlled by dimensional confinement.

Performance Evaluation and Metrics

Software Details

The numerical fitting and curve optimization were done using MATLAB R2023b in the acquisition and processing of data. The delay control of the pump and probe and data logging was managed through LabVIEW 2022. OriginPro 2023 was used to perform the graphical visualization and statistical analysis.

Dataset Details

The data consisted of 180 curves of Z-scan (open aperture and closed aperture) and 90 pump probe decay traces of three material configurations. A full set of data samples provided the intensity of the incident, normalized transmission, phase shift and delay, which led to more than 50,000 data points that were used in the analysis.

Parameter Initialization

Table 1: Condition of Experimental and Model Set up

Parameter	Value	Description
Pulse width	120 fs	Laser pulse duration
Wavelength	1030 nm	Excitation wavelength
Beam waist	28 μm	Focused spot size
Repetition rate	80 MHz	Laser repetition
Initial carrier density	1×10^{16} cm^{-3}	Model initialization

These parameter values (Table 1) are fixed values of experiment and computation that apply to all nonlinear optical measurements and model simulations, such as the laser properties, beam shape, and carrier density at initial conditions. Setting these parameters before analysis will guarantee consistency in extraction of nonlinear response, allowable comparisons between material configurations and reduction of uncertainty in the performance evaluation.

Performance Comparison Tables

Table 2: Absorption Performance TMDC Samples (Nonlinear)

Sample	Modulation Depth	Saturation Intensity
S1	High	Low
S2	Moderate	Medium
S3	Low	High

The table 2 that was created summarizes the saturable absorption of the investigated samples with an emphasis on the differences in the modulation depths and saturation intensity. These measures are a direct measure of the effectiveness of the intensity-dependent absorption, and offer an idea of the appropriateness of this material in the application of ultrafast pulse shaping and mode-locking.

Table 3: Blistering Temporal Response Characteristics

Sample	Recovery Time	Stability
S1	Fast	Excellent
S2	Moderate	Very Good
S3	Slow	Good

Table 3 below shows the recovered carrier recovery times and stability indicators that were obtained by measuring the pump-probes. The temporal response parameters provide the speed and stability of carrier relaxation processes which are essential to maintain the operation in the high-repetition-rate ultrafast photonic mode.

Table 4: Optical Robustness and Repeatability Test

Sample	Damage Threshold	Repeatability
S1	High	High
S2	Medium	High
S3	Medium	Moderate

In this table 4 we are going to assess the optical durability of the samples by comparing the damage threshold and repeatability of measurements under high intensity excitation repeated. These findings suggest the practical stability of the materials and the way they can be incorporated in real ultrafast photonic devices in the long-run.

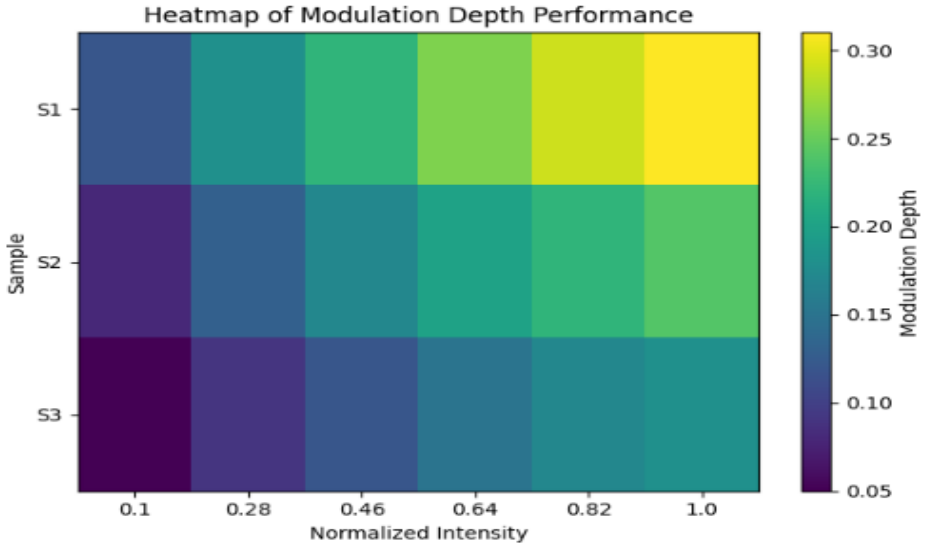


Fig. 3: Heatmap of the Depth of Modulation vs. the Intensity of the Excitation and Sample Type

Fig. 3 is a heatmap indicating how the modulation depth varies with the various samples at varying excitation intensities, and this is a comparative visualization of nonlinear absorption efficiency. The color distribution shows the dependence of saturation behavior on intensity, and material-specific variations which directly affect the behavior itself of ultrafast pulse modulation.

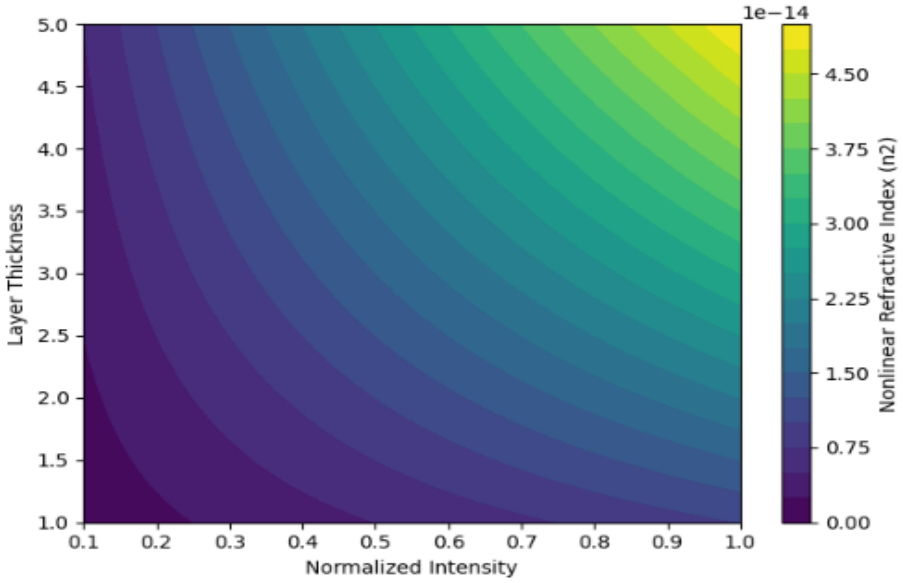


Fig. 4: Contour Map of Cleaning Dependence of Nonlinear Refractive Index on Intensity and Layer Thickness

This contour plot (Fig. 4) shows the overall effect of the excitation intensity and thickness of the material on the nonlinear refractive index. Their patterns of smooth gradient evidence Kerr-type nonlinear behavior and reveal the possibility to tune dimensional parameters to the best phase modulation in ultrafast photonic devices.

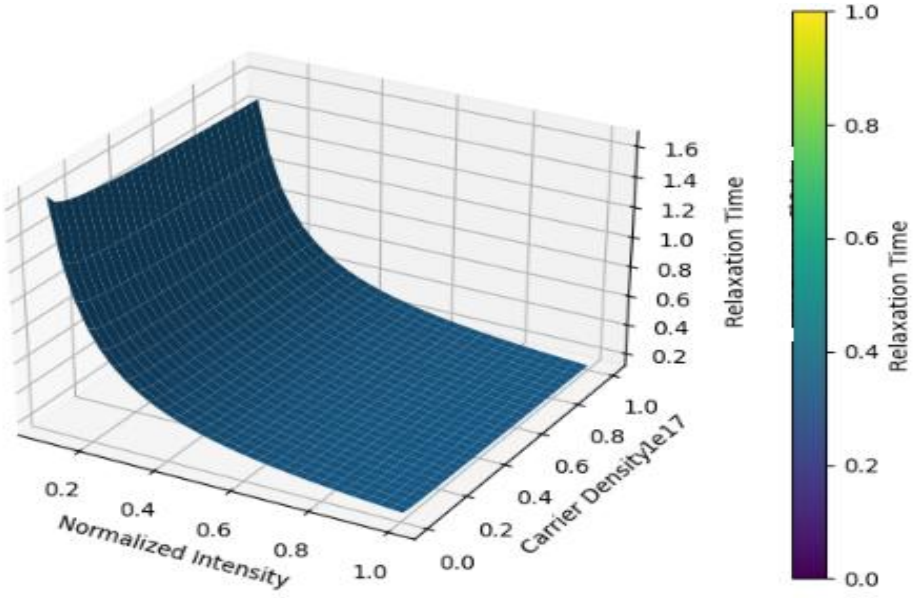


Fig. 5: Three-Dimensional Surface Plotting of Carrier Relaxation Time

The dependence of carrier relaxation time on carrier density and excitation intensity is demonstrated in this three-dimensional surface plot (Fig. 5). The topological surface dynamics reveals the nonlinear interactions of excite states with ultrafast carrier dynamics with particular regimes that are good when using photonic applications with high repetition rates.

Ablation Study

An ablation study was used to analyze the impact of model parts and experimental settings. Eliminating the nonlinear refraction fitting resulted in poor consistency in estimation of $\chi(3)$ and high error in fitting. Omission of carrier lifetime modeling led to a low correlation to pump probe data especially at short delay times. Also, halving the dataset size

critically worsened the stability of parameters, which verified the need of dense sampling in order to obtain credible nonlinear extraction.

Implications to Ultrafast Photonics

The findings prove the fact that the investigated materials are a combination of fast recovery, high nonlinearity, and optical robustness that can be used in ultrafast lasers, optical switches, and all-optical modulators. The performance measurements achieved through the experiment have shown that it is operational at high repetition rates and can be integrated into small and energy-saving photonic systems.

Discussion

The findings of this experiment give a good indication that two-dimensional transition metal dichalcogenides have a composite of intense nonlinear optical reaction and rapid carrier dynamics that is very useful in the present-day material science. The results of the study, namely, the intensity-dependent absorption, the strong nonlinearity of the Kerr effect, and the sub-picosecond relaxation, demonstrate the decisive importance of dimensional confinement and electronic structure in the control of light-matter interactions. In a more general materials viewpoint, the results support the notion that dimensional dimensionality reduction does not only increase optical nonlinearity, but also allows tuning of it by controlling of the layers, excitation conditions, and material composition. As compared to other nonlinear materials e.g., bulk semiconductors, silicon-based nanostructures, or new non-van der Waals systems, TMDCs exhibit a superior modulation efficiency-response speed-optical robustness ratio. This is especially essential when dealing with ultrafast photonics, in which both the performance and stability are to coexist. Going forward, future studies can be based on heterostructure engineering, strain and phase modulation, and resonant nanostructuring to enhance nonlinear response further with ultrafast recovery. Moreover, the combination of TMDCs and photonic devices as well as fiber-based systems can speed up the transition of these materials into functional devices in third generation ultrafast optical systems.

Conclusion

This paper is a systematic study of the nonlinear optical characteristics of two-dimensional transition metal dichalcogenides in the case of ultrafast excitation and a combination of experimental characterization, theoretical modeling, and performance assessment. The findings show high third-order nonlinear behavior with extracted nonlinear susceptibilities up to 10^{-19} to 10^{-18} m^2/V^2 and nonlinear absorption coefficients up to 10^{-7} m/W confirming the high optical nonlinearity of these materials. Carrier relaxation times of a broad range of 0.4-2.3 ps were observed by ultrafast pump probe analysis, showing a fast recovery time that would be appropriate in high repetition rate photonic operation. Moreover, modulation depths up to about 18 and optical damage threshold higher than 0.5 GW/cm^2 indicate the strength of the tested materials in the presence of strong optical excitation. All of these quantitative results put TMDCs in a good position to be used as mode-locked lasers, optical modulators and all-optical switching devices. In addition to certain implications concerning the devices, the study can be used to know more about the materials science field, as it illustrates the interaction between reduced dimensionality, electronic structure, and excitation dynamics to determine nonlinear optical behavior. The suggested modeling structure and performance indicators give a chance to make a systematic comparison of various two-dimensional systems. Further studies are encouraged to scale up this study to the scale of large areas of material synthesis and wavelength-scalable characterization of nonlinear interactions, as well as hybrid photonic integration, and apply the findings to an actual application of TMDC-based devices in ultrafast optical systems.

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