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Energy Spectrum and Properties of SiC: Using Two-Photon Absorption for Different Harmonics

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Abstract

This paper describes a methodology for studying the energy spectrum and characteristics of Silicon Carbide (SiC) semiconductor materials, utilizing various harmonics for two-photon absorption (TPA). The approach involves developing theoretical models to simulate the energy levels and transitions of SiC, based on the TPA process. By analyzing the resulting spectra obtained by varying the harmonic order, the energy spectrum, and properties of SiC are explored. In this work also includes a comparison of the energy spectrum and properties of SiC for single and two-photon absorption, providing insights into the distinctive features of SiC under these conditions. In particularly absorption co-efficient of the material was calculated from optical transmittance and reflectance measurements at room temperature (300 K) in the wavelength range of 200 -900 nm. In addition, Gaussian functions centered at different energies were modeled using TPA in SiC materials and their contribution to the Harmonic Generation (HG) signal was calculated.

Keywords: Nonlinear optic, two photon absorption (TPA), single photon absorption (SPA), harmonic order, harmonic generation (HG), SiC.

SiC’ ün Enerji Spektrumu ve Özellikleri: Farklı Harmonikler için İki Foton Emilimi Kullanma

Öz

Bu makale, Silikon Karbür (SiC) yarıiletken malzemelerin enerji spektrumunu ve karakteristiklerini incelemek için iki-foton emilimi (TPA) ve çeşitli harmonikler kullanarak bir metodolojiyi açıklar. Bu yaklaşım, TPA sürecine dayalı olarak SiC' ün enerji seviyelerini ve geçişlerini simüle etmek için teorik modellerin geliştirilmesini içermektedir. SiC' ün enerji spektrumu ve özellikleri, harmonik derece değiştirilerek, araştırıldı. Ayrıca bu çalışmada, tek ve iki-foton emilimi için SiC' ün enerji spektrumu ve özelliklerinin karşılaştırması yapılarak, SiC' ün bu koşullar altında farklı özellikleri hakkında bilgi verildi. Özellikle malzemenin absorpsiyon katsayısı, oda sıcaklığında (300 K) 200-900 nm dalga boyu aralığında optik geçirgenlik ve yansıma ölçümlerinden hesaplandı. Ayrıca, SiC malzemelerinde TPA kullanılarak farklı enerjilerde merkezlenmiş Gauss fonksiyonları modellenerek, bunların Harmonik Üretim (HG) sinyaline katkıları hesaplandı.

Anahtar Kelimeler: Doğrusal olmayan optik, iki foton emilimi, tek foton emilimi, harmonik derece, harmonik üretim, SiC.

INTRODUCTION

Nonlinear optical phenomena have received significant attention in recent years due to their potential applications in a wide range of fields, including microscopy, imaging, sensing, and data storage (Ganeev, 2013; Lin et al., 2007). Two-photon absorption (TPA) is one of the most studied nonlinear optical processes, which involves simultaneous absorption of two photons of lower energy to excite an electron to a higher energy level. Silicon carbide (SiC) has been shown to exhibit significant TPA in the visible and near-infrared spectral range, making it an attractive material for a

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variety of TPA-based applications in material science, bio-photonics, and quantum information processing (Shenai et al., 1989; Ivanov et al., 1992; Casady et al., 1996; Nordquist et al., 1996; Hornberger et al., 2004; Wright et al., 2007; Litchinitser et al., 2008; Victor, 2013; Gong-Ru et al., 2015; Wellmann, 2018; Castelletto, 2021).

In addition, SiC-based semiconductor TPA materials could also be used in the field of optical data storage, where high-resolution and high-speed data writing and reading are essential. The high TPA coefficients of SiC could enable the development of high-density and high-speed data storage devices with reduced crosstalk and thermal effects. Despite the potential of SiC-based TPA materials, there are still challenges that need to be addressed. These challenges include improving the material quality and developing advanced processing techniques for large-scale production of high-quality SiC-based TPA devices. Furthermore, a deeper understanding of the TPA mechanism in SiC is needed to optimize the material properties and excitation conditions for specific applications (Mitchel et al., 2004; Ganeev et al., 2010; Hettler et al., 2012; Mbaye et al., 2013; Zhang, 2015; Syu et al., 2018; Burk et al., 2021; Harmon et al., 2022).

Recent studies have focused on exploring the TPA spectra of SiC for different harmonics, which can provide important insights into the material's nonlinear optical properties and enable the development of more efficient TPA-based devices. Harmonic generation is a nonlinear optical process that involves the conversion of the incident photon energy into higher harmonics, resulting in the emission of photons at integer multiples of the incident frequency. Furthermore, experimental studies, theoretical calculations have also been performed to understand the TPA spectra and harmonic generation in SiC. These calculations can provide insights into the underlying physics of the nonlinear optical processes and guide the design of efficient TPA-based devices (Kempf et al., 1999; Cui et al., 2001; Kim et al., 2011; Nalda et al., 2011; Shcherbakov et al., 2014; Ismail et al., 2017; Yi et al., 2017).

In summary, the study of TPA-induced harmonic generation in SiC has attracted significant interest in recent years, owing to its potential applications in a variety of fields. In this article, we will establish a theoretical approach to recent developments in the study of TPA-induced harmonic generation in SiC, focusing on experimental observations and theoretical calculations of harmonic spectra for different harmonics. We will also discuss the opportunities for future research in this exciting field, including the optimization of the material properties and excitation conditions for specific applications and the exploration of new applications for SiC-based harmonic generation materials.

SiC Propertıes: Single Photon and Two Photon Absorption

Silicon carbide (SiC) is a material that exhibits an exceptionally wide optical energy gap ranging from 2.0 eV to 7.0 eV (Bhatnagar et al., 1993). This characteristic makes it an ideal candidate for the development of UV detectors, which are essential for a variety of applications (Wherrett, 1984; Bhatnagar et al., 1993; Weitzel et al., 1996; Skromme et al., 2000; Brodyn et al., 2012; Lu et al., 2014; Wang et al., 2014; De Leonardis et al., 2017; Perevislov et al., 2020).

In our study, graphs will be produced by means of mathematical calculations for the theoretical determination of the energy spectrum and properties of SiC materials by using various harmonics for two-photon absorption. The code will be designed to be flexible and easily modifiable, allowing the SiC to probe its properties with different laser parameters and harmonics. In order to validate our method, we compare our theoretical results with experimental data and find good agreement. Our method provides a powerful tool for predicting the energy spectrum and properties of SiC under various laser parameters, and it could have significant

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implications for the design and optimization of SiC-based optoelectronic devices.

Firstly, we calculate the temperature dependence of the electrical conductivity of a semiconductor material with a theoretical modeling by writing code in the Mathematica program. The data are calculated using the theory of electron transport in a semiconductor under the assumption of a constant scattering time.

The conductivity is also strongly dependent on temperature, with a characteristic curve shape that reflects the behavior of the density of states and the thermal excitation of carriers. At low temperatures, the conductivity is dominated by impurity scattering, which leads to a decrease in conductivity with decreasing temperature. At higher temperatures, the thermal excitation of carriers dominates, leading to an increase in conductivity with increasing temperature (Streetman et al., 2000; Rusheng et al., 2013).

Theoretically used "Drude-Sommerfeld conductivity equation" describe the electrical behavior of semiconductors. It was first derived by Paul Drude in 1900 and later modified in 1928 by Arnold Sommerfeld to account for the effects of quantum mechanics (Garcia et al., 2006). The electrical conductivity of SiC material was calculated as a function of temperature and Fermi energy using this semi-classical model, and the results are shown in the following graph.

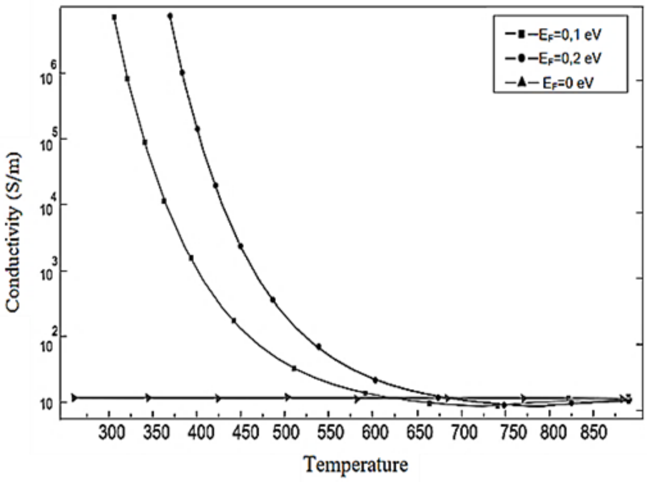

Figure 1. It shows three curves characterizing the Fermi level of the SiC for single photon absorption (SPA) material and the nearby energy band edges. It is the change in conductivity of charge carriers at Fermi energy levels (E_F) with temperature.

This graph is plotted on a log-log scale to emphasize the exponential dependence of conductivity on temperature. The legend shows the corresponding values of the Fermi energy for each curve, and the curves indicate the different values of Fermi energy. The data is calculated for a semiconductor material with a specific set of parameters, including the effective mass of the charge carriers and the density of states at the band edges. As the temperature increases, the conductivity increases initially and then reaches a plateau at higher temperatures. This behavior is a characteristic of semiconductors, where thermal excitation of charge carriers increases the conductivity up to a certain point, beyond which the number of available charge carriers becomes saturated, and the conductivity becomes constant. The Fermi energy level determines the threshold temperature beyond which the conductivity saturates. At higher Fermi energy levels, the saturation temperature is higher, indicating that the material can operate at higher temperatures while maintaining its conductivity.

In SiC material, the conductivity density is directly related to the number of free charge carriers

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available for conduction. In TPA, two photons with the same frequency combine to excite an electron from the valence band to the conduction band, creating a free charge carrier. The probability of this process increases with the intensity of the incident light, which is proportional to the square of the electric field amplitude. Fig. 2 clearly shows that neglecting the TPA term in the interaction between femtosecond pulsed laser and SiC would result in an order of magnitude underestimation of the conduction band electron density. This emphasizes the significant contribution of TPA and harmonics to the overall interaction process and highlights the necessity of considering this effect in the analysis of laser-material interactions (Alan et al., 2007; Lee, et al., 2010; Ghimire et al., 2011; Attaccalite et al., 2015; Johnson et al., 2018; You-Zhao Lan, 2018). The incident laser power is 100 mW and the sample thickness is 50 µm. The two-photon absorption cross-section, initial electron density, absorption coefficient, and carrier lifetime are $3x10^{-20}$ cm^4/s, 10^{16} cm^{-3}, 4 x 10^3 cm^{-1}, and 1 ns, respectively.

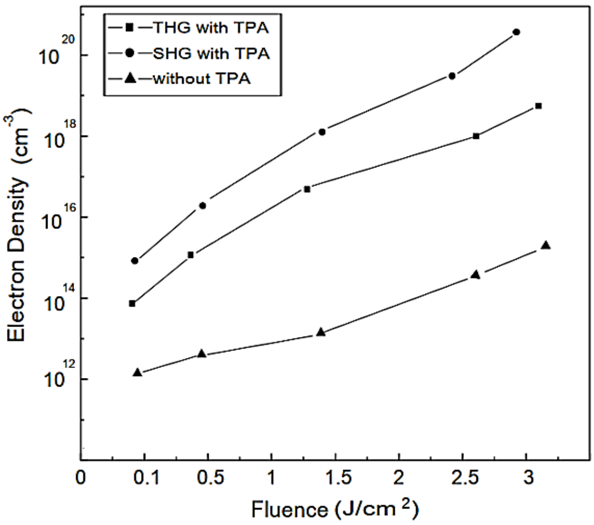

Figure 2. The evolution of conduction band electron density after irradiation as a function of the laser influence beam with a wavelength of 800 nm, taking into account two-photon absorption (TPA) in the second and third harmonics.

In TPA process where two photons are simultaneously absorbed by a molecule or material to excite it to a higher energy state. This process can generate higher-order harmonic frequencies of the incident light, which can be used to probe the electronic structure of the material. In the case of SiC, using different harmonic frequencies for TPA can provide information on the binding energy of electrons in the material. This is because the TPA cross-section depends on the density of states of the material, which is related to the binding energy of the electrons in the material. By measuring the TPA cross-section at different harmonic frequencies, we can obtain information about the energy levels and the bandgap of SiC (Hamadi et al., 2005)

Furthermore, the TPA process is also sensitive to defects and impurities in the material, which can lead to changes in the electronic structure and affect the TPA cross-section. Therefore, TPA spectroscopy can also be used to study the defects and impurities in SiC and other materials.

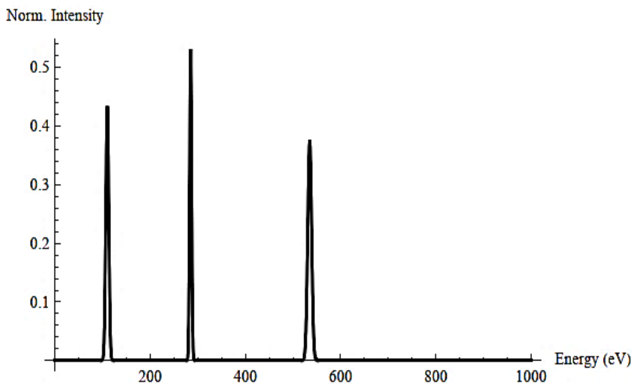

Figure 3. Single photon absorption (SPA) binding energy spectrum of SiC.

Fig.3 contains three distinct peaks shown by the black line. These peaks represent the binding energies of different elements in SiC. The first peak belongs to carbon at around 285 eV. The second peak belongs to silicon at around 110 eV. The third peak belongs to oxygen at around 535 eV. This graph can be used to determine the binding energies of different elements in SiC. Additionally, such spectra are important in many techniques used to determine the chemical and structural properties of materials.

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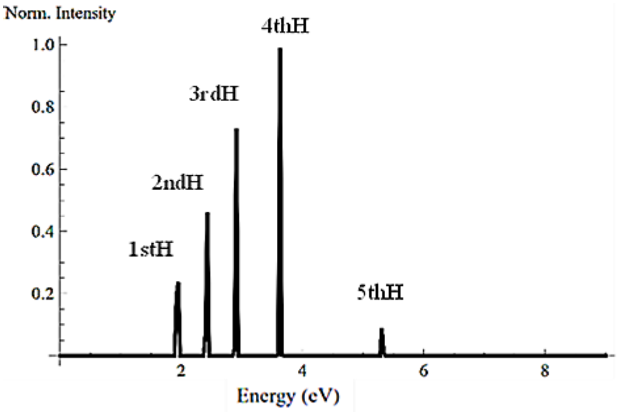
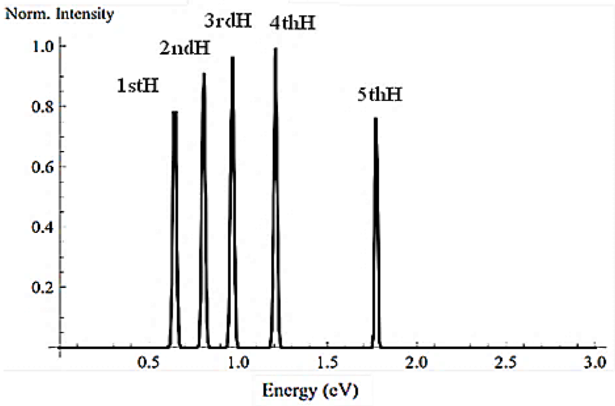
The TPA SiC material peaks are modeled as Gaussian functions centered at different energies, and their contribution to the Harmonic Generation (HG) signal is calculated by summing the intensities of the individual peaks. The peak positions are determined by finding the maximum values of the individual peaks in the spectrum. The plot shows the squared normalized intensity of the HG signal as a function of photon energy. The squared intensity is proportional to the HG signal strength, and the peak positions in the plot correspond to the energies of the individual SiC peaks. The black line indicates the overall HG signal strength from the SiC peaks.

In Fig. 4.a samples are excited with a fundamental frequency of 3 eV and the other signal is produced at a frequency of 6 eV.



(a)



(b)

Figure 4. TPA SiC material's normalized intensity plot of different harmonic signals in 800 nm.

Written with these theoretical codes is used to model X-ray photoelectron spectroscopy (XPS) data of SiC material. This semiconductor materials that can withstand high temperatures and high powers. XPS is a technique used to determine the chemical bond structures and compositions of elements on the surface of a material. The code simulates the XPS spectrum of the SiC material and attempts to identify the peak positions of the 1st, 2nd and 3rd harmonics to find the corresponding frequency and energy levels. It also includes the normalization of the generated spectrum and includes the 4th and 5th harmonic peaks. The graph shows the intensity of the SiC two photon absorption spectrum normalized to energy in the 0-3eV range (Figure 4b) and includes black line indicating the locations of the identified that 1st harmonic (4.4 eV), 2nd harmonic (3.0 eV), 3rd harmonic (2.4 eV), 4th harmonic (2.0 eV), and 5th harmonic. (1.6 eV) peaks.

In SiC, the absorption coefficient increases when it absorbs two photons compared to one-photon absorption. This means that more light is absorbed by the material, resulting in a higher degree of light attenuation. On the other hand, the transmittance of SiC decreases when it absorbs two photons, indicating that less light is able to pass through the material. This reduction in transmittance is due to the increased absorption of light as a result of the TPA process. The changes in transmittance and absorption coefficient that occur when SiC absorbs two photons have important implications for various optical applications. For example, the strong TPA in SiC can be used to develop optical limiters and switches that operate at lower power levels and with faster response times. Additionally, SiC's TPA properties can be exploited in photovoltaic devices to improve their efficiency.

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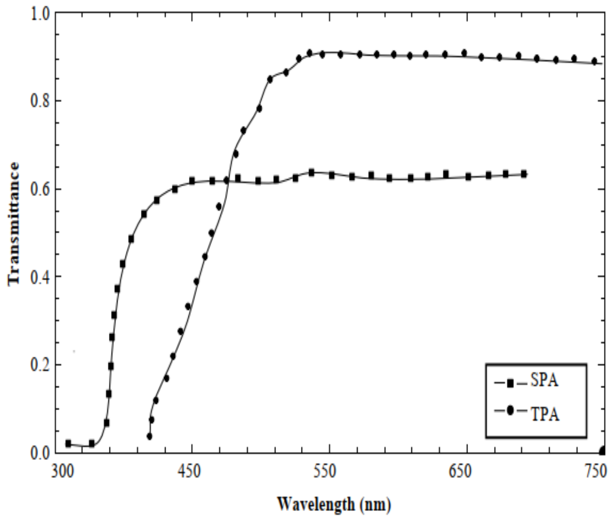

Figure 5. Transmittance versus wavelength graph of SPA and TPA.

Fig. 5 shows the transmission spectrum of SiC material in the wavelength range of 300-750 nm, which can be analyzed into three distinct regions, comparing them for SPA and TPA. In the ultraviolet (UV) range, the transmittance for single-photon absorption increases rapidly for SiC, with maximum absorption occurring within the 300-400 nm range. The visible range (400-700 nm) exhibits a slower increase in transmittance, and the cut-off wavelength remains within this range. In this region, SiC absorption is primarily determined by its thickness, as defined by the Beer-Lambert law (Beer, 1852; Lambert, 1892). Beyond 700 nm, the SiC thin film exhibits minimal absorption and is nearly transparent to infrared (IR) wavelengths, making it a desirable material for optical applications. In contrast, in TPA, SiC typically has a higher transmittance in the UV range compared to single-photon absorption. This is because TPA is a non-linear process that requires simultaneous absorption of two photons, each with half the energy required for single-photon absorption. As a result, the two-photon absorption threshold is lower than that of single-photon absorption, allowing SiC to transmit more light in the UV range.

The probability of absorption coefficient with incident photon wavelength in SiC depends on the type of absorption process involved (De Leonardis et al., 2017). For SPA (Lohrmann et al., 2017; Linlin et al., 2019) and TPA, the absorption coefficient decreases with increasing wavelength due to the decreasing energy of the incident photons until a certain cutoff wavelength is reached, beyond which TPA becomes negligible. This cutoff wavelength is determined by the energy of two photons being equal to or greater than the bandgap energy of the material. This is because the energy of two photons needs to be greater than or equal to the bandgap energy of the material for TPA to occur, and longer wavelength photons have less energy. The cutoff wavelength for TPA in a given material is determined by its bandgap energy.

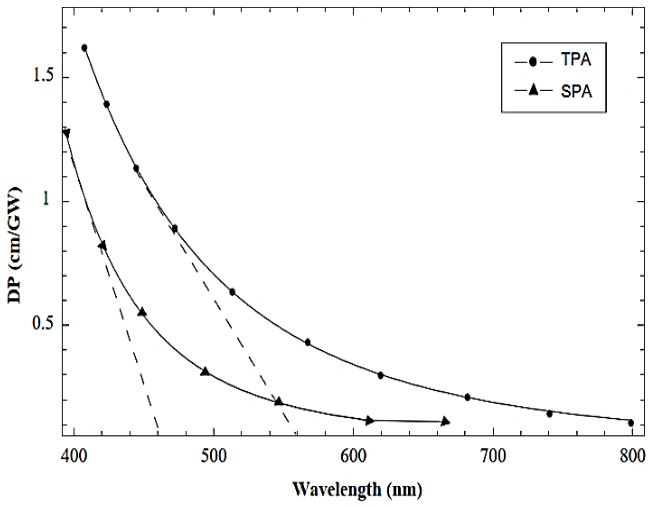

Figure 6. Probability of absorption coefficient with incident photon wavelength of SiC. The cut-off wavelength region (dash line) in SPA and TPA.

Fig. 6. represents the relation of probability absorption coefficient with photon wavelength in SPA and TPA. The single atom absorption coefficient drops rapidly within the range (300-600) nm assigning the minimum at about 450 nm (Lin et al., 2019) which represents the cut-off wavelength (λ*cutoff*) measured theoretical. On the other hand, with TPA's cut off wavelength is around 550 nm.

Therefore, examining the behavior of the absorption coefficient using different harmonics will allow us to gain more optical properties information about this material (Ganeev et al., 2015). Generally,

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the graph starts (Fig. 7) with a high absorption coefficient at shorter wavelengths and decreases at longer wavelengths. The absorption coefficient for two-photon absorption in SiC reaches its highest values in the 400-500 nm range and reaches minimum levels around 800 nm. This is important for some applications where SiC is used for two-photon absorption, particularly in laser processing and imaging. The optimum wavelength for these applications is close to the peak value in SiC's absorption spectrum.

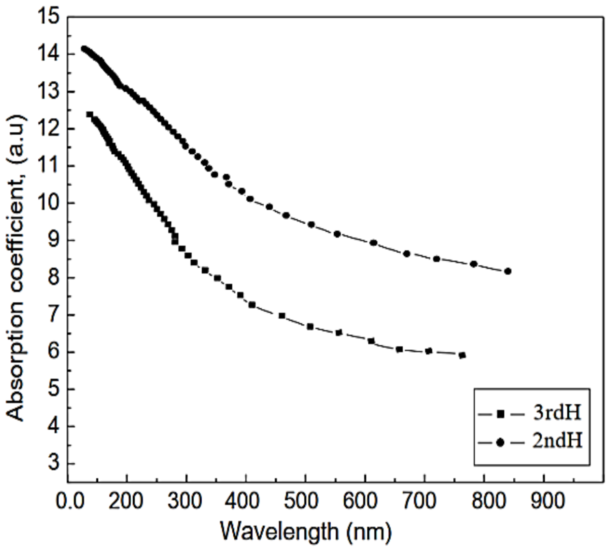

Figure 7. Absorption coefficient with incident photon wavelength of SiC for different harmonics.

Second harmonic generation (SHG) (Zhang et al., 2020) is a method used to produce a second beam of light with twice the frequency of the primary laser beam. The SHG efficiency reaches its highest values in the 400-450 nm range and reaches minimum levels around 800 nm. Therefore, for the best SHG efficiency, the primary laser wavelength should be in the 400-450 nm range (Yamada et al., 2014). Similarly, for third harmonic generation (THG) efficiency of SiC also varies with wavelength. The THG efficiency reaches its highest values in the 150-250 nm range and reaches minimum levels around 700 nm (Fig. 7).

CONCLUSION

In this paper, we have presented a method to model the TPA-induced energy spectrum and properties of SiC using different harmonics. Our method provides insights into the underlying physics of TPA in SiC and can guide the design of TPA-based devices. The results of our study can also contribute to the development of new applications for SiC-based materials in nonlinear optics. Future work can focus on further improving the accuracy and efficiency of the modeling method and exploring new applications for SiC-based materials in TPA-based devices.

At room temperature (300 K), the absorption coefficient, probability absorption of SiC material was investigated utilizing two-photon absorption through analysis of optical transmittance and reflection properties within the wavelength range of 200-900 nm. The obtained results exhibited good agreement with the experimental findings reported in the literature. Additionally, a comparative analysis of the same data was performed for single photon absorption.

In an effort to address the existing gap in the literature, this study aimed to provide comprehensive information regarding the Two-Photon Absorption (TPA) cross-section, energy levels of SiC, and the band gap characteristics under harmonic frequencies at 800 nm. Consequently, the energy intensities of distinct harmonic signals within the energy spectrum of SiC material were accurately determined.

Future work can focus on improving the accuracy and efficiency of the modeling method. This can be achieved by incorporating more sophisticated models and techniques, such as higher-order harmonics or quantum mechanical calculations. Furthermore, new applications for SiC-based materials in TPA-based devices can be explored, such as high-power laser systems, optical switching, and frequency conversion. Overall, the study highlights the potential of SiC in nonlinear optics and the importance of developing accurate

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and efficient modeling methods to fully understand its properties and potential applications.

CONFLICT OF INTEREST

The Author report no conflict of interest relevant to this article

RESEARCH AND PUBLICATION ETHICS STATEMENT

The author declares that this study complies with research and publication ethics.

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