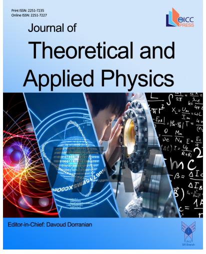

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Spatial self-phase modulation in plasmon excited graphene WS_2 heterostructure

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ORIGINAL RESEARCH

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Abstract:

In this work a spatial phase modulation and four-wave mixing in graphene tungsten disulfide (WS_2) heterostructure have been demonstrated, which is excited by plasmons of nano-plasmonic donate shape rings. For this purpose, a laser writing system was used to fabricate nanohole arrays and cover them with the plasmonic gold thin film via a sputtering machine and WS_2 and graphene heterostructure by chemical vapor deposition method. After evaluating the plasmon excitation in this system by the ellipsometric method, the probe laser excitation was set to 980 nm to record spatial phase modulation, and also, four-wave mixing was used. Our results show spatial self-phase modulation (SSPM) and temperature enhancement due to the nonlinear enhancement of two-dimensional structure by heterostructure substances suitable for new-generation optical devices.

Keywords: WS_2; Graphene; Spatial phase modulation; Four wave mixing

1. Introduction

Recent studies have shown that improving the motion of electrons in heterostructures like graphene over tungsten disulfide (WS_2) could make all-optical modulation and switching work much better. Even though graphene has very high carrier mobility, it can't be used in electric uses because it has no band gap [1]. the act of electrostatic gating applied to the graphene layer induces a modification in the exciton binding energies of WS_2 at different interlayer distances. This alteration has been measured and represented by a singular parameter in Ref. [2]. hole transfer is faster than electron transfer in the WS_2/graphene heterostructure, with hole transfer occurring in less than 200 fs and electron transfer taking about 1 ps. As a result, the structure of graphene/WS_2 allows for high electron mobility [3]. In all-optical modulation and switching uses, the higher movement of electrons can lead to faster response times and deeper modulation. Recent research has shown that this approach could make all-optical modulation and switching based on 2D materials work better.

Additionally, the growing area of thermo-plasmonics (TP) in photonics intends to use the kinetic energy of light to produce heat at the nanoscale. It entails the creation, detection, and manipulation of optical signals at nanometer-scale metal-dielectric contacts [4, 5]. The creation of plasmonic materials and structures that may improve light-matter interactions and produce heat at the nanoscale is one of the main applications of TP [6, 7]. Reduced losses and more design flexibility in thermo-plasmonic devices may be further aided by the creation of high-quality plasmonic substrates and better production methods [7]. Further study is required to get over this obstacle, though, as heat control in plasmonic circuits continues to be a problem [8, 9].

The investigation of spatial self-phase modulation (SSPM) is a topic of active research; however, the fundamental mechanism is yet unknown. It is unclear and misleading to say if SSPM is a result of the intrinsic nonlinear properties of 2D materials due to the proposal of several mechanisms. The earliest commonly accepted theory is the thermal lens effect. However, SSPM must not be connected to the material's nonlinear optical properties if it is produced from the thermal lens effect; specifically, it must be independent of $\chi(3)$, which led to a lot of uncertainty. Many scientists think that the molecule redirection and polymerization caused

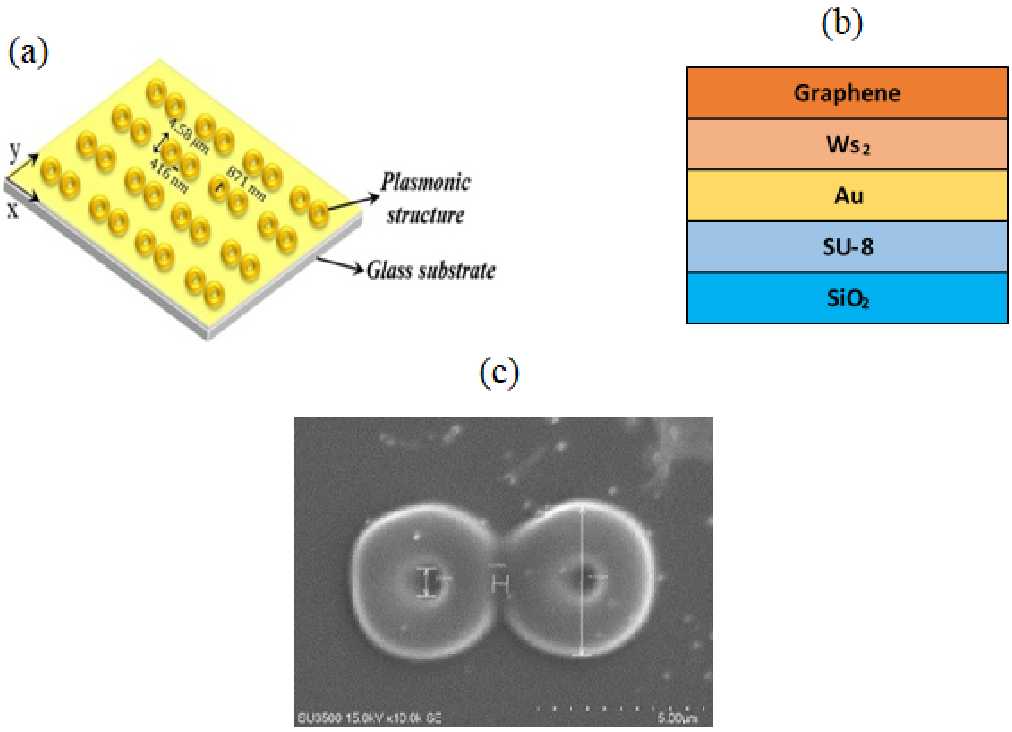

Figure 1. (a) Schematic diagram of sample with dimensions, (b) Schematic diagram of sample layers, and (c) real donut photo captured by SEM.

by lasers in SSPM results in a purely electronic coherent behavior. However, SSPM typically needs excitation with continuous light in order to produce sufficiently high average power that meets the excitation threshold. Thermally induced nonlinearity, which now becomes another competitive process, is inevitable. Some scientists have presented a way to distinguish between local and nonlocal nonlinearity because they think that thermally induced nonlinearity and pure electron coexist. Here, we have a summary of different approaches in an effort to serve as an inspiration for the overall SSPM mechanism [4].
Overall, there has to be a universal theory proposed to account for all of the laws and phenomena of SSPM because the current study on the physical genesis of SSPM is not mature enough. When hybrid graphene-TiO2 nanostructures were employed to stimulate SSPM, Jiang et al. discovered that there was no signal at 700 nm and an SSPM effect at 1100 nm [9]. Indicating that the nonlinear optical characteristics of graphene at 1100 nm play a significant role, TiO2 exhibits a large reverse saturation absorption (RSA) at 700 nm, making it unlikely that SSPM is merely a thermal phenomenon. Numerous investigations have also suggested that SSPM is only pure electrical coherence.
It has been demonstrated that silicon nitride (SiN) waveguides integrated with WS_2/graphene or graphene oxide may increase four wave mixing (FWM) efficiency by up to 7.3 dB (decibels). This implies that WS_2/graphene or comparable materials can improve the interaction between pump and signal waves and produce new frequencies via FWM processes [10]. with integrated photonics, electrically controllable FWM has been suggested using SiN waveguides wrapped with graphene. This suggests that control of FWM processes can be achieved by modifying the characteristics of graphene and its interactions with other materials using external electric fields [11]. In addition to WS_2 and graphene, the study also cites the utilization of additional stacked 2D materials as MoS2, MoSe2, and WSe2. This demonstrates the possibilities of several 2D material combinations for FWM and nonlinear optical applications [12].

2. Experimental part

A 2D micro cavity array was prepared via direct laser writing experimental setup using 405 nm laser beam. The laser beam was expanded and transferred to the silica substrate by objective lens with numerical aperture of 0.8. The substrate, covered with SU8 2002 polymer, was placed onto the motorized x-y stage and the array of microcavities was written onto the polymer. The laser power was set to 0.3 mW with the arrays consisting of 50 in 20 rings with width and radius set to 4.58 µm and 871nm respectively. The gap between two rings was selected as 416 nm and the periodicity of the structure was assigned to 20 µm. Figs.1 a and b depicts the sample structure and layers, while Fig.1 c, shows real photo captured by scanning electron microscopy (SEM).
The WS_2 nano sheets were prepared by CVD (chemical

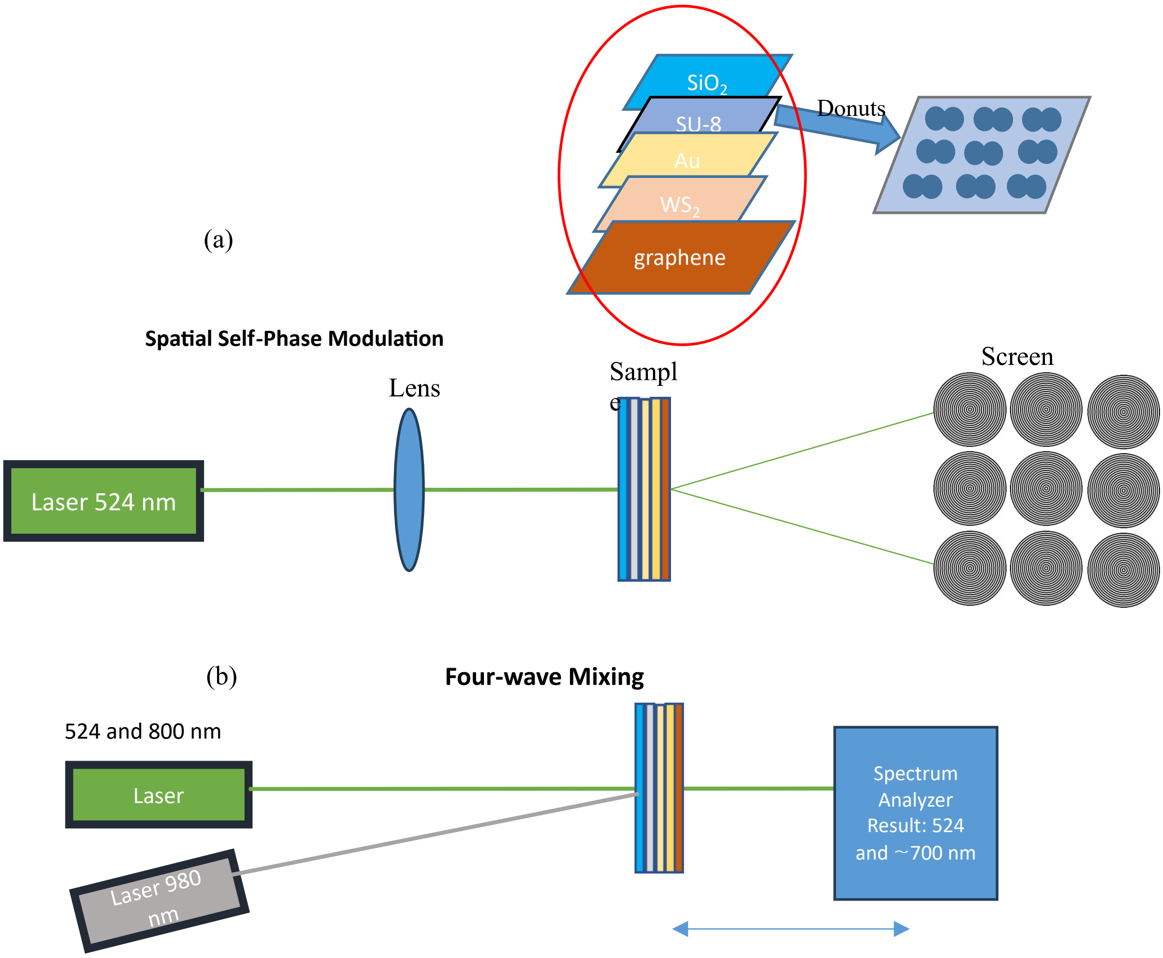

Figure 2. Schematic diagram of (a) SSPM experimental and (b) Four wave mixing setup.

vapor deposition) method. The Tungsten three oxide (WO3) and sulfur (S) powders were used as raw materials. Also, Potassium Chloride (KCl) was used as a catalyst and Ar/H_2 gas mixture was injected during the reaction. The furnace was first kept at a temperature of 250 °C for 10 minutes and then the temperature increased to 820 °C and kept for 30 minutes. After that, the graphene layer was fabricated by the same CVD machine, but under gas flow as described in our previous work [13].

To plasmon excitation and use of thermo-plasmonic phenomena, we used the near-field measurement system in transmission mode by using a high numerical aperture objective lens. The halogen lamp via the Glan-Taylor prim reaches the objective lens and thus the sample by two different polarizations. After plasmon excitation in this near field setup, the light is gathered by another normal objective lens and focused onto the spectrometer or charge coupled device (CCD) camera. To get efficient usage from the proposed heterostructure, we use the 980 nm as pumping light and the FLIR camera to record the temperature change in the sample. In addition, to record the SSPM of the sample, we use the continuous wave green laser as the main pump light and record by the screen as presented schematically in Fig. 2 a to record the SSPM. Finally, due to the nonlinear properties of the 2D materials, we record Four-wave mixing by experimental setup (Fig. 2 b).

3. Results and Discussion

The experimental setup has done the near field excitation of plasmons in the first gold layer based on a high numerical aperture objective lens with the visible light source and 980 nm laser light as the pump ones. The image of the CCD camera shows the structure without laser and on coupled structure, without laser and coupled and with pump laser and coupled systems, respectively (Figs. 3 a-c). Plasmonic coupling via near field excitation by high Numerical aperture objective lens confirmed in the comparison between b and a parts. In addition, when the sample exposed with pump laser, the intensity of coupled region enhances because the extra absorption of the laser light by two dimensional structures.

As explained in the experimental part, we measure the transmission spectra of the sample without (Figs. 4 a and b) and with pump laser (Figs. 4 c and d) and record the ellipsometric parameters of the sample as psi and delta parameters for both states of the heterostructure. One can see some new modes activation in the psi as the main change of the optical answer's amplitude in spite of this fact that in the phase diagram as the delta parameters, there are two important points (Figs. 4 b and d); at first, the delta amount of the heterostructure by the laser light enhances, and second, the main modes in the center of the visible region change hardly as it was supposed from the temperature enhancement in the

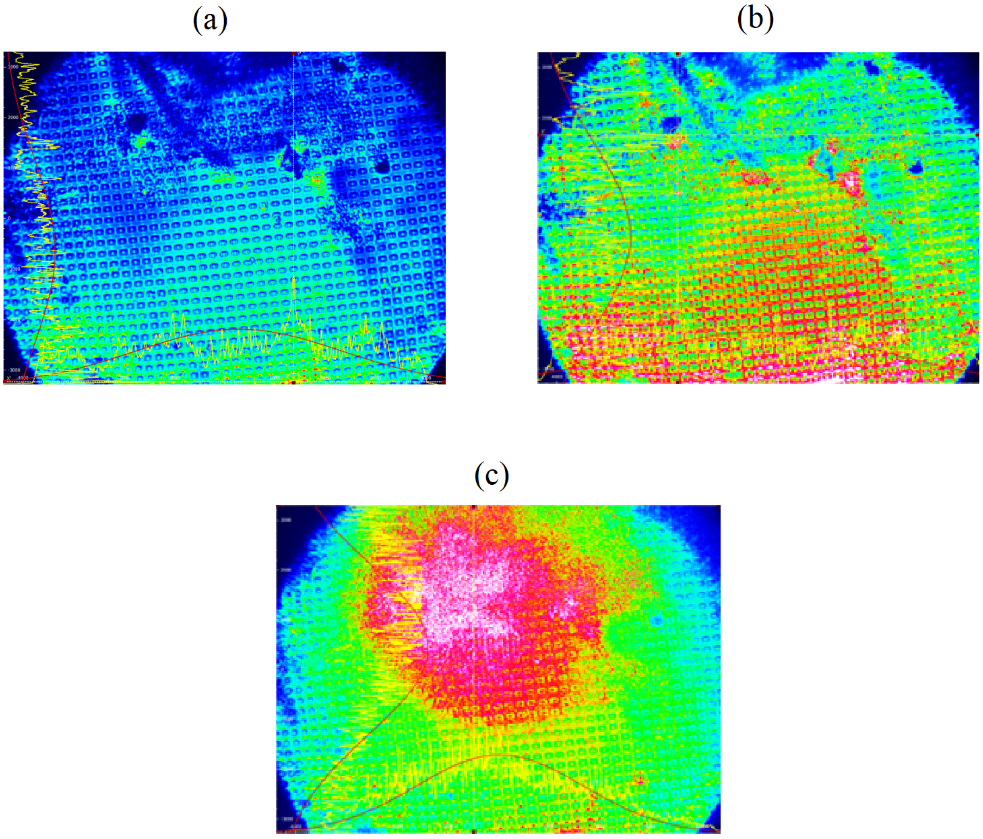

Figure 3. CCD camera's image from the sample for (a) without laser and un-coupled, (b) without laser and coupled and (c) with laser and coupled system.

heterostructure by absorption of the pump laser. According to the results of I. Paradisanos et al., the interaction between WS_2 and graphene considerably impacts how polarized WS_2 is in its valley region, with polarization levels reaching 24% at room temperature under near-resonant excitation. The research also revealed a strong relationship between the type of material utilized to create the heterostructure and the level of valley polarization of WS_2.

It was found that the interaction between WS_2 and graphene has a big effect on how the depolarization of WS_2 changes with temperature. The study shows that the model used needs to be changed to explain the actual data under near-resonant conditions. It also shows that the excitation energy has a big effect on how the temperature dependence changes [14].

Also, C. Lu et al. show that in mixed heterostructure films, the saturation absorption is higher. But nothing is said about how graphene is better than WS_2 at soaking heat or how it acts when it gets hot. [15]. On the other hand, Y. Zhang et al. look at how heat moves through a single layer of WS_2 that is touching graphene. They find that when graphene and WS_2 make hetero contacts, the heat conductivity goes down [16]. Due to this fact, a nonlinear optical phenomenon known as SSPM happens when a laser beam travels through a material having a nonlinear refractive index. Due to their distinct electrical structures, two-dimensional materials like graphene and WS_2 show remarkable nonlinear optical characteristics [17].

As a result of the previous statements, we supposed that the heterostructure has the larger nonlinear refractive index due to localized surface plasmon resonance of gold nano structure and also two dimensional structures and thus SSPM must be happens in this structure. The inherent nonlinear refraction coefficient of graphene/WS_2 and the radially dispersed intensity of a Gaussian beam are connected in the physical theory of spatial self-phase modulation. The material's refractive index varies with the strength of the laser beam due to the nonlinear refractive index of graphene/WS_2. The phase of the laser beam changes as a result of this change in refractive index, and this phase change is proportional to the laser's intensity. The Gaussian beam's spatially variable intensity distribution causes the phase shift to change spatially as the laser beam moves through the material, resulting in spatial self-phase modulation [18, 19]. It has been noted that rapid charge transfer occurs in WS_2/graphene heterostructures. It indicates a significant connection between the two materials, even if the precise microscopic process causing this charge transfer is

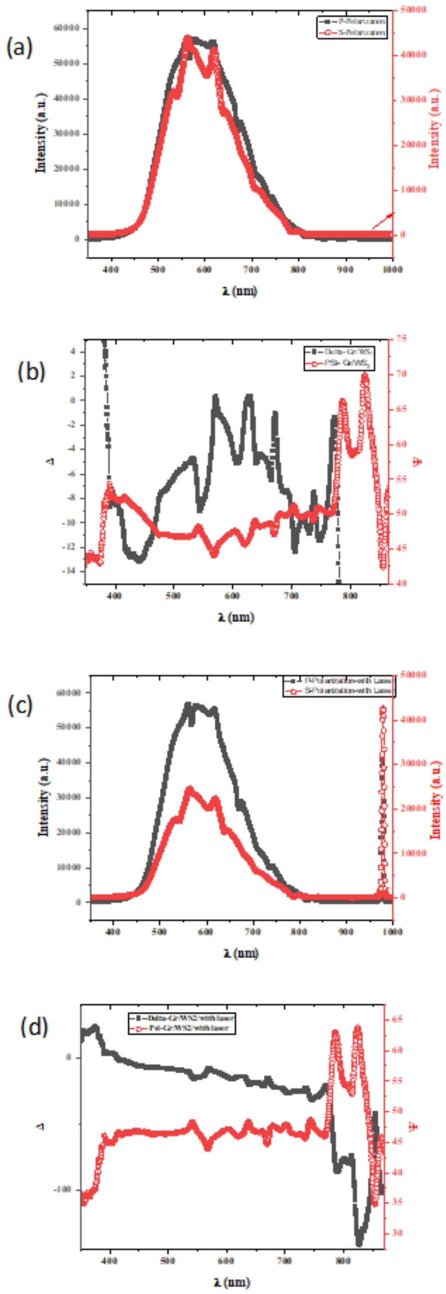

Figure 4. The optical spectra of the heterostructure for both polarizations (a) without laser and (c) with laser light, the ellipsometric parameters of the sample (b) without laser and (d) with pump laser.

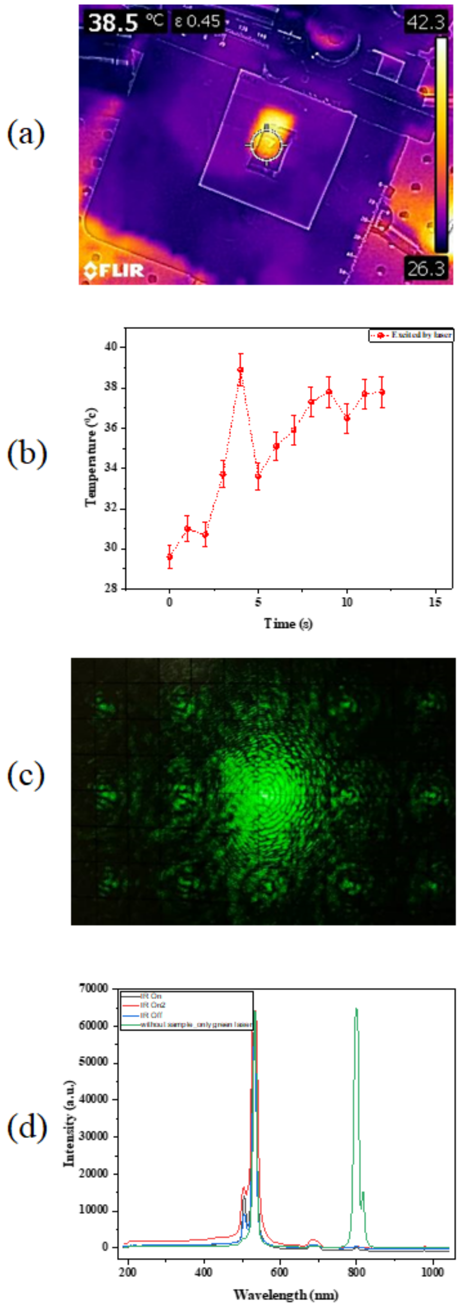

Figure 5. Temperature enhancement of the sample (a) with pump laser, 980 nm, after 5 minutes, (b) temperature enhancement in different elapsed times, (c) SSPM of the structure and (d) intensity recorded by spectrometer under CW green, 532nm, pump laser.

not yet completely known. By altering the carrier dynamics and exciton production, this interaction may have an impact on nonlinear optical characteristics, such as FWM [20]. Due to these facts, we record the temperature enhancement and SSPM of the heterostructure as shown in Figs. 5 a-d, respectively.

It has been demonstrated that the use of WS_2/graphene with waveguides or other structures improves nonlinear optical effects, including FWM. Strong light-matter interactions made possible by the special qualities of WS_2 and graphene, such as their different electronic band structures and carrier dynamics, may be the precise processes underlying this improvement [11]. In general, the interaction between excitons (bound states of electrons and holes) and phonons (lattice vibrations) can affect a material's nonlinear optical characteristics. It is possible that the existence of WS_2/graphene heterostructures may open up new pathways for exciton-phonon interactions, which might have an impact on the effectiveness of FWM processes. It is important to keep in mind that several mechanisms, including nonlinear intra-band conductivity in graphene, may interact to cause the nonlinear optical processes in WS_2/graphene heterostructures. The unique qualities of WS_2 and graphene combined with these processes may explain some of the reported FWM effects [21] and also shows SSPM in the proposed heterostructure.

Due to the high nonlinearity of the graphene/WS_2 heterostructure sample, FWM can take place. When lasers with 524, 800 nm wavelengths (as input wavelengths) and sample are pumped by a 980 nm laser, creating a new wavelength of almost 700 nm were recorded (Fig. 5 d). Figs. 5 b, and 5 c are present the FWM experimental setup and the recorded FWM results, respectively. Moreover, number of diffraction rings calculation formula can be changed to read [7].

$$N = \frac{2n_0 L_{eff} P}{\lambda}\left[\frac{n_2}{\pi\omega_0^2} + \frac{dn}{dT}\frac{\alpha}{4\pi K(1+P/P_s)}\right] \tag{1}$$

Based on this formula, the resulted clear rings number of the SSPM was 12.

4. Conclusion

In sum, plasmonic substrate was produced experimentally by laser writing system and covered with WS_2 and graphene layers to produce nonlinear heterostructure. After evaluation the main optical modes, due to the nonlinear properties of two-dimensional structures and thus the heterostructure, we assign the experimental setup of SSPM and FWM to record the nonlinear properties in the interface of both two-dimensional layers. Strong light-matter interactions were happening by the special qualities of WS_2 and graphene, such as their different electronic band structure. In addition, temperature enhancement in this heterostructure was measured by camera which can be used as good candidate in biocompatible thermos treatment systems.

Ethical approval

This manuscript does not report on or involve the use of any animal or human data or tissue. So the ethical approval is not applicable.

Authors Contributions

H. M. Hamodi design the main sample, measure all the processes and also write the main text, R. S. Fyath and S. M. Hamidi supervised the work and edit the main text.

Availability of data and materials

Data in this manuscript are available by request from the corresponding author.

Conflict of Interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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