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Research paper

Graphene-Enhanced Raman Spectroscopy: Mechanisms, Optimization, and Applications

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Abstract

Raman spectroscopy has long been recognized as a powerful optical technique for materials analysis, garnering sustained scientific interest due to its ability to provide detailed vibrational fingerprints of molecular structures. Among the various Raman spectroscopy enhancement methods, Graphene-Enhanced Raman Spectroscopy (GERS) is an advanced variant that leverages the unique properties of graphene-based substrates to significantly amplify Raman signals. It has emerged as a pivotal technique for boosting the sensitivity of Raman spectroscopy, primarily through the exploitation of chemical enhancement pathways inherent to graphene. This article offers a thorough examination of the underlying mechanisms of GERS. It then presents various advanced graphene-based substrates including pristine graphene, graphene oxide (GO), nitrogen-doped graphene, and other functionalized derivatives engineered for superior Raman signal amplification. Finally, to highlight the importance of GERS and its potential in diverse and wide-ranging fields, this article comprehensively outlines the significant applications that GERS facilitates across various research disciplines and technological frontiers from biomedical diagnostics and environmental monitoring to food safety and materials characterization thereby establishing its importance as a powerful analytical tool.

1. Introduction

Raman spectroscopy, as a non-destructive analytical method that does not require complex sample preparation, plays a vital role in identifying molecular structure, determining crystal phases, and investigating chemical changes in various materials. This technique provides a unique fingerprint of molecular vibrations, allowing the study of a wide range of samples [1, 2]. The principle of Raman spectroscopy hinges on the inelastic scattering of light by molecules; this spontaneous process is exceedingly weak only about one in ten million photons are inelastically scattered, primarily through Rayleigh scattering. This low signal yield has been a persistent limitation [3, 4]. To overcome this, the field has seen the continuous innovation of Raman enhancement strategies, culminating in more than 25 distinct techniques. These include prominent methods such as Resonance Raman Spectroscopy (RRS), Surface-Enhanced Raman Spectroscopy (SERS), Tip-Enhanced Raman Spectroscopy (TERS) [5], Coherent Anti-Stokes Raman Spectroscopy (CARS), Stimulated Raman Spectroscopy (SRS) [6], and Grephene-Enhanced Raman Spectroscopy (GERS) [7]. GERS has emerged as a significant advancement in molecular detection, garnering substantial attention [8]. Since its isolation in 2004 [9], graphene -a singular layer of sp^2- hybridized carbon atoms structured in a unique two-dimensional hexagonal honeycomb lattice [10, 11], has emerged as a groundbreaking material. Its remarkable suite of properties high surface area, exceptional mechanical strength, transparency, superb electrical conductivity, and favorable biocompatibility [12] has driven intense research and development. These attributes are particularly valuable in fields like chemical and biological sensing. A key application is its role as an advanced substrate in Raman spectroscopy, where it significantly enhances spectral quality and analytical capabilities within diverse scientific domains [13]. In this review, the principles of GERS is first explained. Subsequently, it delves into the fabrication and characterization of various graphene-based substrates designed for signal amplification. The article concludes by outlining the method's applications across a broad spectrum of disciplines.

2. Results and Discussions

2.1. GERS Mechanisms

The Raman enhancement processes are predominantly governed by the synergistic interplay between an electromagnetic mechanism (EM) and a chemical mechanism (CM). Within the framework of the EM contribution, the electromagnetic field associated with the incident light is significantly amplified through plasmonic resonance effects, especially in regions referred to as "hot spots" that typically form on the rough or nanostructured surface of a metal. On the other hand, the CM originates from direct interactions between the substrate and the adsorbed molecules, which involve both photo-induced charge transfer processes and the mixing of molecular orbitals with the electronic states of the metallic substrate. As a result, the enhancement factors (EFs) derived from each mechanism exhibit vibrational mode dependence, albeit for fundamentally different reasons. In the case of the EM, variations in EF are primarily dictated by the frequency match between the incident light and the localized surface plasmon resonance. In contrast, for the CM, the EF is governed by the specific nature of the molecule substrate interaction, thereby displaying a pronounced molecular selectivity [14]. In Surface-Enhanced Raman Scattering (SERS), the dominant contribution to the overall enhancement is widely attributed to the electromagnetic mechanism (EM). Conventionally, SERS substrates are fabricated from noble metals such as silver (Ag), gold (Au), or copper (Cu), which possess rough or nanostructured surfaces capable of supporting localized surface plasmons [15]. GERS wherein a graphene monolayer serves as the enhancing platform instead of a roughened metal surface is primarily governed by the chemical mechanism (CM) [16]. It is noteworthy that the surface plasmon resonance inherent to graphene lies in the terahertz frequency regime, rather than in the visible range of the electromagnetic spectrum. Consequently, based on these physical considerations, graphene is not capable of supporting the EM mechanism under visible light excitation [17]. The EM mechanism imposes stringent requirements on the substrate architecture, including a sufficiently rough surface morphology, the presence of metallic particles with high local curvature, and the ability to absorb incident radiation efficiently so as to excite surface plasmons. In contrast, the CM mechanism necessitates an ultra-short separation distance typically below 0.2 nm between the molecule and the substrate. Moreover, the Fermi level of the metal or conductive substrate must align symmetrically with both the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) of the analyte molecule, thereby facilitating bidirectional charge transfer (i.e., from the substrate to the molecule or vice versa). When considering graphene as a Raman-enhancing substrate, several distinctive features emerge. Its surface is atomically smooth, and its optical transmittance in the visible spectral range exceeds 95%, implying minimal light absorption and thus negligible plasmonic excitation [18, 19]. Figure 1 shows a schematic image of the sensing molecules on the graphene substrate on the SiO2/Si layer and the Raman experiments [19]. Within a GERS system, the strength of interaction between graphene and the adsorbed molecule is predominantly determined by two critical factors: (i) the degree of energetic alignment between the molecular orbitals and graphene's electronic structure, and (ii) the compatibility of molecular symmetry with that of the graphene lattice. Furthermore, the observed Raman scattering intensity is strongly influenced by the band structure of the constituent materials. Accumulating evidence indicates that the molecular energy levels play a decisive role in governing the enhancement effect in GERS. Specifically, for a given set of laser excitation energies, the HOMO and LUMO levels of the analyte molecule must lie within an appropriate energy window relative to the Fermi level of graphene.

This principle has been consistently corroborated by experimental observations as well as theoretical analyses based on third-order perturbation theory within the quantum theory of Raman scattering.

As mentioned before, a second key selection rule pertains to molecular symmetry: molecules possessing certain symmetry characteristics exhibit substantially larger enhancement effects.

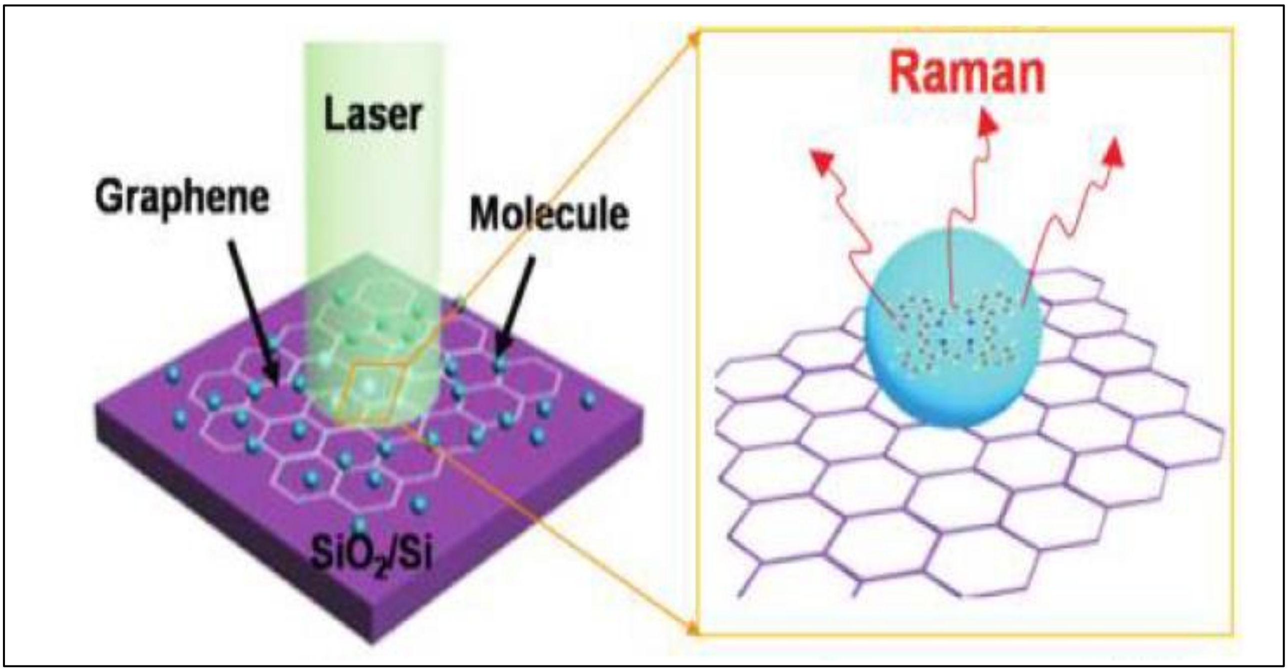


Figure 1. Schematic illustration of molecules on graphene and SiO2/Si substrate and Raman experiments [19]

The coupling between the molecule and the graphene substrate can be exceptionally strong, contributing significantly to the overall GERS enhancement. Of particular importance is the presence of conjugated aromatic systems, such as interconnected benzene rings, which tend to strengthen the molecule–graphene coupling and thereby amplify the GERS response. Accordingly, when the fundamental structural framework of a molecule resembles that of graphene, and such molecules are deposited as a sub-monolayer onto a graphene surface, they tend to adopt a planar orientation parallel to the graphene plane. This parallel arrangement is mediated by π–π stacking interactions, which not only ensure a minimal separation distance between the molecule and the graphene surface but also promote efficient electronic coupling [14, 20].

2.2. Various types of Substrates used in GERS

The recent observation of Raman signal enhancement for various molecules adsorbed onto graphene has sparked a significant debate regarding the underlying mechanism of this phenomenon, commonly known as the graphene-enhanced Raman spectroscopy (GERS) effect. Following the demonstration that graphene offers several advantages over metallic substrates for Raman enhancement such as easier fabrication, lower cost, and superior biocompatibility research in this area expanded considerably [21]. Consequently, researchers have pursued the development of graphene-based substrates through various fabrication methods to further investigate their effect on the enhancement of Raman signals.

In this context, Xie et al. reported that the quenching of luminescence from R6G molecules on the graphene surface enables clearer observation of the molecular Raman spectrum [22]. However, the observation of an enhanced Raman scattering effect on graphene was first reported at the end of 2010 by Ling et al., where it was demonstrated that probe molecules deposited on graphene gave significantly higher Raman signals than those deposited on bare substrates. The researchers evaporated sub-monolayer amounts of phthalocyanine (Pc), rhodamine 6G (R6G), protoporphyrin IX (PPP), and crystal violet (CV) onto mechanically exfoliated graphene, and compared the Raman intensities with those observed on SiO2/Si substrates. It was found that the intensity of Pc vibrational modes was 2–17 times (depending on the vibrational mode) greater when deposited on monolayer graphene. Furthermore, this paper presented a comparative analysis of Raman enhancement using substrates of monolayer, few-layer graphene, multilayer, graphite, and highly ordered pyrolytic graphite (HOPG). It was demonstrated that as the number of graphene layers increases, the intensity of the Raman signals diminishes, ultimately resulting in no observable signal for graphite and HOPG [19].

In a 2019 study by Alexander Silver and colleagues, a range of substrates were systematically investigated, including pristine graphene, graphene doped with elements such as nitrogen, boron, and silicon, as well as graphene-based nanocomposites incorporating gold or silver nanoparticles. In addition, the influence of the layer count of graphene-based substrates and other analogous two-dimensional materials on the Raman spectroscopy enhancement was further examined. It was demonstrated that an increase in the number of layers leads to a reduction in the intensity of the Raman signals. The paper provides a comprehensive review of graphene-enhanced Raman scattering (GERS) as a sensing methodology, emphasizing that the synthesis of graphene and related two-dimensional materials, along with the precise modulation of doping levels and Fermi energy, constitutes the fundamental prerequisite for developing highly sensitive and controllable GERS-based sensors. For instance, it was demonstrated that incorporating nitrogen into graphene lattices can modulate the electronic properties by shifting the Fermi level and, in certain cases, opening a band gap. Additionally, such doping can enhance catalytic and sensing performance by altering the local charge distribution in the vicinity of the dopants [7].

In another study published in 2022, graphene oxide, amino- and guanidine-functionalized graphene oxide, exfoliated graphene, and commercial graphene nanosheets were employed to investigate the GERS response through the modification of graphene's properties. Various graphene nanostructures were embedded within organic-inorganic microporous films to create a substrate platform enabling rapid and sensitive detection. Subsequently, the GERS performance of the graphene nanostructures deposited on silicon substrates was comparatively assessed against that of those embedded in the nanocomposite films. In this work, it was further demonstrated that graphene nanostructures differ in terms of the number of layers, lateral size, degree of oxidation, and surface functionalization. In this context, it has been shown that functionalization of graphene with specific functional groups is not an effective method to enhance the Raman signal of analyte molecules. Consequently, single-layer graphene nanostructures represent promising candidates for the fabrication of sensing platforms with rapid detection capabilities and the comparison among different types of graphene shows that monolayers are more efficient than the few-layer nanostructures in enhancing the Raman signal [23].

In a 2020 study by Václav Valeš et al. the graphene-enhanced Raman scattering (GERS) effect was investigated using four distinct graphene-based substrate models. They have been studied on isotopically labeled bilayer and a single layer of pristine and partially hydrogenated graphene. The hydrogenated graphene sample showed a shift in the relative intensities of the Raman bands of the analytes deposited on the monolayer and bilayer graphene. This shift is qualitatively consistent with the different graphene doping in both regions. The pristine graphene sample showed no difference in doping or relative intensities of the Raman peaks samples in the monolayer and bilayer regions. This approach was tested on copper phthalocyanine and R6G molecules [24].

2.3. Applications of GERS

GERS as a molecular vibrational spectroscopy has emerged as a highly effective and innovative technique for the identification of nanoscale materials, drawing significant research interest in recent years. In this approach, the distinctive physicochemical properties of graphene including its high electrical conductivity, superior charge carrier mobility, and strong light matter interaction lead to a substantial enhancement of the Raman scattering signal [12]. The significance of GERS arises from its capacity to detect substances at extremely low concentrations, enable the investigation of molecular interactions at the nanoscale, and facilitate the rapid identification of sensitive biological and chemical compounds [14]. Therefore, a thorough examination and understanding of the applications of this technique are of great importance. GERS holds broad promise in diverse fields such as biosensing, nanomaterials, pharmaceuticals, and environmental science [25].

Globally, around 2 million tons of pesticides are applied each year to boost agricultural productivity. Over time, these substances can build up in the environment and infiltrate the food chain, ultimately endangering human health. Even minute, trace levels of pesticides are enough to cause considerable health damage. therefore, it is crucial to establish systems that not only curb the overuse of pesticides but also detect them at very low concentrations. While traditional analytical methods are commonly used for both qualitative and quantitative analysis of pollutants, and despite their high sensitivity and accuracy, they suffer from drawbacks such as high costs, lengthy procedures, and a lack of portability to testing sites. As a result, there is a growing need for cost-effective, dependable tools that can streamline the detection process by integrating multiple steps into a single, efficient assay for pollutant monitoring. Notably, Swapneel Thakkar and colleagues developed a platform utilizing various graphene-based materials to detect pesticides in water, further illustrating the versatility and potential applications of GERS in environmental monitoring and beyond. They succeeded in achieving detection limits of 10^{-7} M, 10^{-5} M, and 10^{-4} M for the pesticides paraoxon, parathion, and glyphosate, respectively. The results of the experiments in this paper are reported in Figure 2. This Figure shows the Raman spectra measured from a 10^{-7} M paraoxon ethyl water solution deposited over a flat silicon substrate, hybrid porous silica, and nanocomposite substrates containing five different types of graphenes. It is evident that the presence of graphene leads to a substantial enhancement of the Raman peaks [23].

Another application of GERS lies in the detection of dye molecules. Fluorescent dyes are extensively utilized in the manufacturing of various commercial products, including paper, plastics, cosmetics, food items, and textile dyes. The expansion of these industries has led to the discharge of dye-contaminated effluents into water bodies, resulting in direct and indirect serious environmental pollution and posing risks to human health. Most dyes possess complex structural characteristics, such as aromatic frameworks and high chemical stability, rendering them resistant to degradation by conventional physical, chemical, or biological treatment methods. Among the various classes of dyes, cationic dyes exhibit the highest degree of toxicity. Rhodamine 6G (R6G) is a representative cationic dye known for its strong fluorescence and highwater solubility [26]. Although R6G is not classified as highly hazardous, prolonged exposure through inhalation or dermal contact can lead to severe health issues, including tachycardia, as well as an increased risk of cardiac or pulmonary cancer. Consequently, the removal of R6G from wastewater streams prior to environmental discharge is critical for mitigating water pollution and reducing associated health risks. Given these concerns, the detection of R6G, even at trace concentrations, is of paramount importance [27]. Another dye molecule of significant importance as an analyte across various applications is crystal violet (CV). This triphenylmethane dye, which functions as an antifungal agent, is widely employed in aquaculture. The residual presence of CV in aquatic environments exerts detrimental effects on ecosystems, and its accumulation in food products poses potential risks to human health. Owing to its low cost and high efficacy against specific fish diseases, CV continues to be utilized in aquaculture on a relatively large scale—a practice that raises concerns among consumers and regulatory bodies alike. Consequently, safety assessments and analytical investigations are routinely conducted to ensure compliance with health and environmental standards. However, most conventional analytical methods for CV detection are time-consuming and require highly skilled operators. In this context, Raman spectroscopy presents itself as a suitable alternative, leveraging its inherent advantages such as rapid analysis, minimal sample preparation, and high molecular specificity [28].

A significant portion of the research has employed pigments as analytical probes to investigate the effects of graphene-based substrates fabricated through various methods, owing to the pigments' high fluorescence and the typically weak or undetectable nature of their Raman spectra [25].

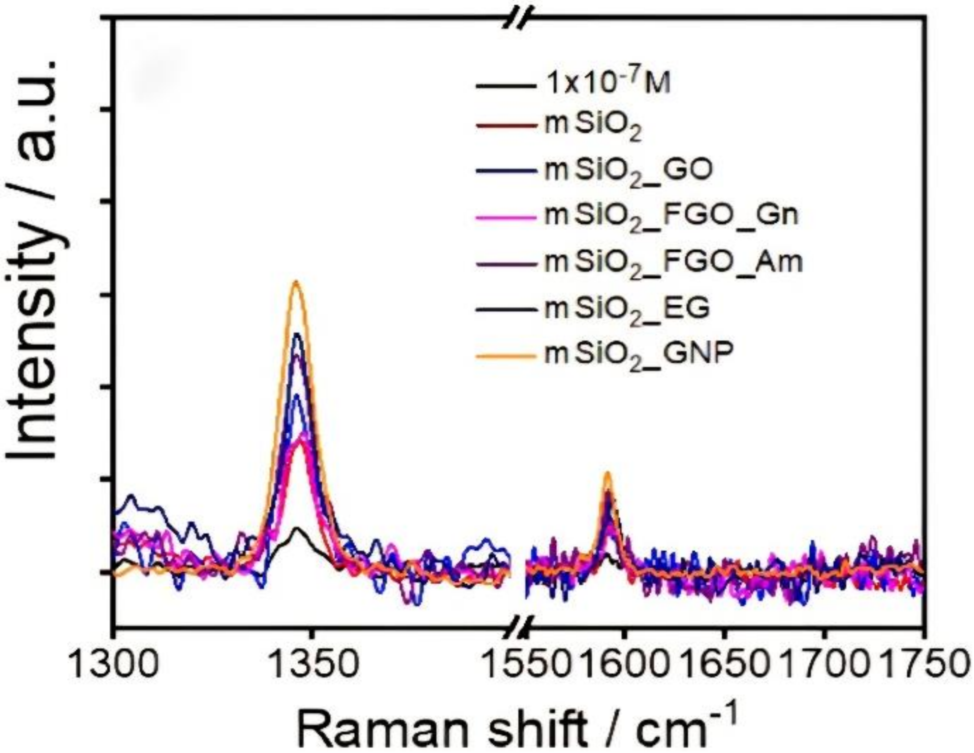
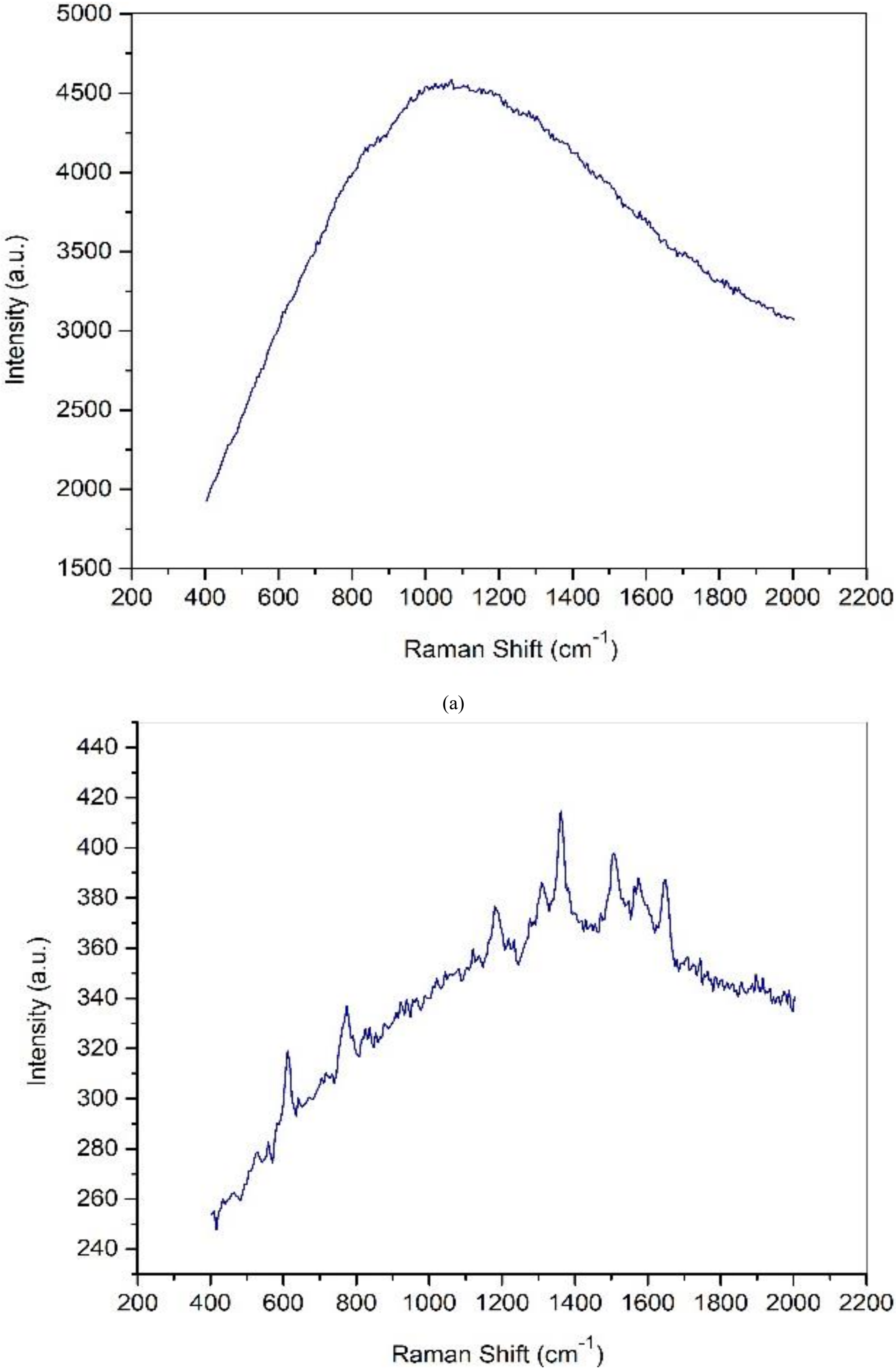


Figure 2. Raman spectra of paraoxon ethyl solutions cast on the nanocomposite films (the black line refers to the solution cast on bare silicon) [23]

The fluorescence quenching effect, notably observed in dyes such as R6G, and CV is facilitated by the π–π interactions of sp2 carbon atoms in 2D sheets [29, 30]. For instance, one of the pioneering articles in this field investigated the potential of utilizing graphene as a substrate to enhance the Raman signal of target molecules. The study demonstrated that the intensity of Raman signals on monolayer graphene is substantially higher than that on conventional SiO_2/Si substrates. Notably, this enhancement is observable even at extremely low concentrations a finding of considerable significance [19].

In a study, to investigate the influence of various graphene-based substrates on GERS, the Raman spectra of R6G solutions deposited onto different substrates were compared. Raman spectroscopic measurements were conducted following the deposition of a specific concentration of R6G solution onto substrates composed of graphene oxide and a glass slide. The resulting Raman spectra obtained from the R6G molecules are presented in Figure 3. The Raman spectra analysis indicates that the peaks associated with R6G on the glass are absent. Furthermore, as is clearly evident from the acquired graphical data, the presence of the graphene-based substrate has, as anticipated, resulted in a reduction of fluorescence intensity and a concomitant enhancement of the Raman peaks [25].

In general, an ideal sensor should be sensitive, specific, and minimally perturbative to the target analyte. That is, it must not respond to anything other than the intended analyte, and it should measure the analyte without altering the property being measured. Conventional surface-enhanced Raman spectroscopy (SERS) systems have struggled with the latter requirement, as the metal surfaces needed for plasmonic enhancement are chemically active, which undermines reproducibility. In contrast, graphene is relatively inert, making graphene-enhanced Raman spectroscopy (GERS) a promising candidate for sensing applications.



(a)

(b)

Figure 3. The Raman spectra of R6G deposited on the a) glass slide, and on the b) Substrate fabricated with graphene oxide [25]

Nevertheless, several challenges remain. The selectivity and specificity of GERS require further improvement, particularly because the Raman modes of graphene overlap with those of many organic molecules in the same spectral regions. Recent developments in GERS-based sensing have begun to address these limitations [7]. GERS is valuable for the detection of biomarkers, which may include cells, molecules, proteins, or enzymes, and is employed to identify or monitor disease states, drug responses, or normal biological processes. As an illustration, Huang et al. utilized GERS to detect blood constituent proteins such as hemoglobin and albumin key indicators of health disorders including leukemia and pulmonary diseases [31].

The investigation of biological and biochemical species frequently necessitates the use of substrates with enhanced chemical inertness, for which graphene-enhanced Raman spectroscopy (GERS) presents a suitable approach. Suitable substrates for GERS can be derived from carbon allotropes, including graphene and carbon nanotubes.

Notably, a study by Olena Fesenko and colleagues demonstrated the enhancement of Raman scattering and anti-Stokes coherent Raman signals from thymine adsorbed onto graphene oxide. As shown in Figure 4, their findings indicate that graphene can serve as an effective substrate for amplifying the Raman signals of biomolecules such as thymine [32].

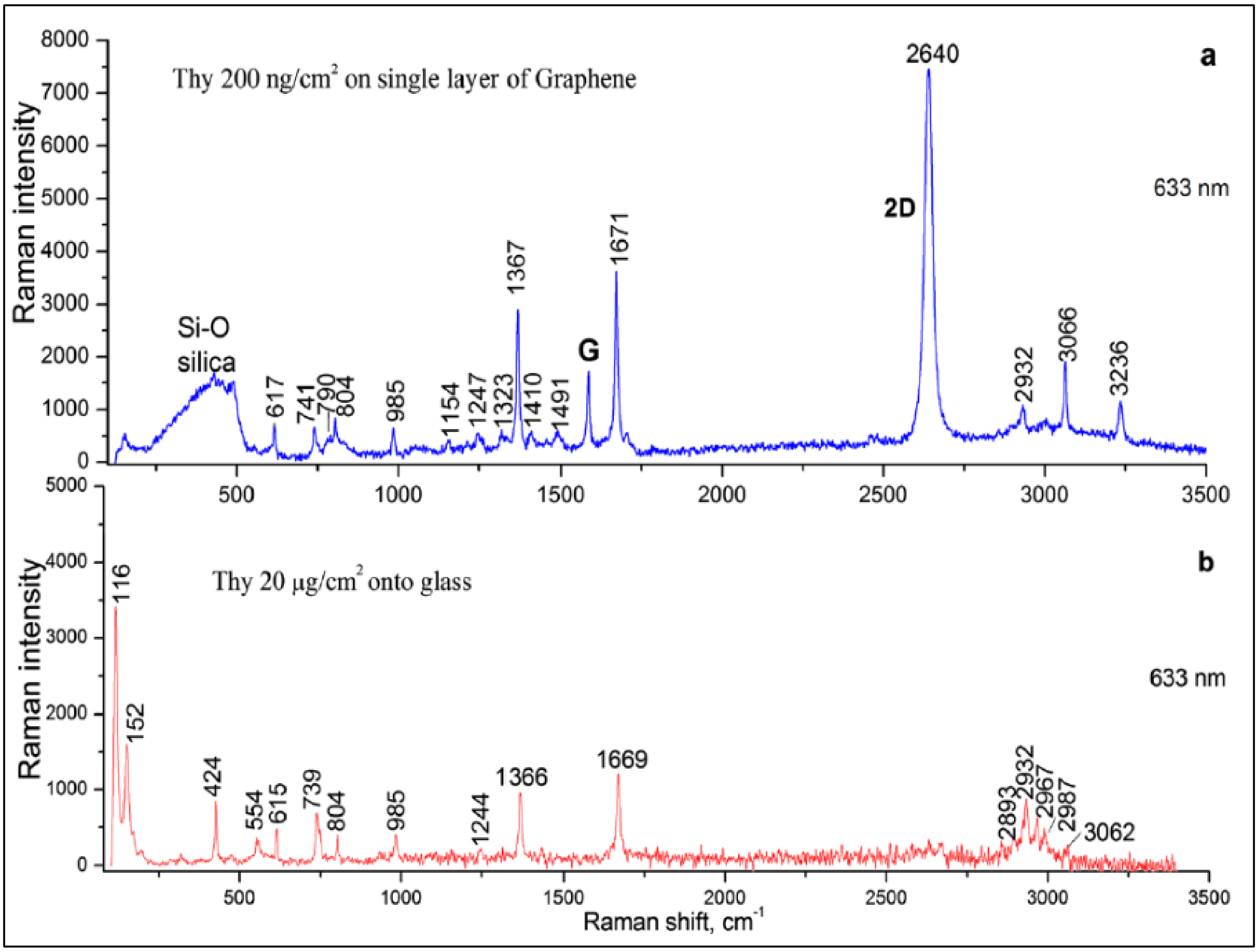
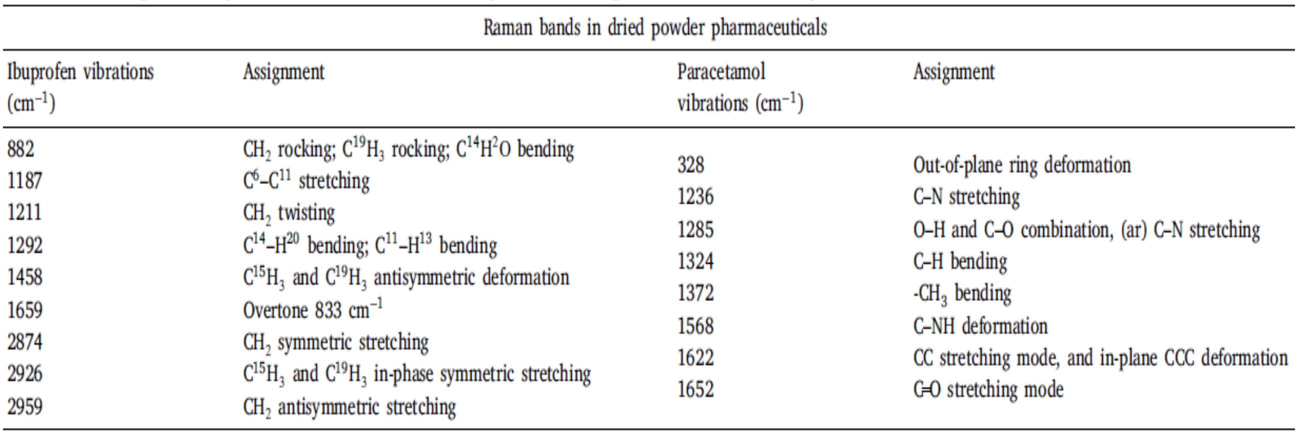


Figure 4. Raman spectra for Thy adsorbed (a) on single-layer graphene, and Raman spectra of Thy adsorbed (b) on the glass surface [32]

In 2025, Nayeli Y. and colleagues introduced a novel detection methodology based on GERS for identifying trace concentrations of ibuprofen and paracetamol dissolved in water. Their findings revealed that low drug concentrations induce an enhanced Raman spectroscopic response on the graphene substrate, enabling the detection of pharmaceuticals at concentrations as low as 100 µg/mL. The graphene monolayer was observed to both amplify the characteristic vibrational bands of the drugs and effectively quench their fluorescence, thereby facilitating the acquisition of a distinct spectral fingerprint. Based on the findings of this investigation, the researchers successfully identified the Raman bands of ibuprofen and paracetamol using the GERS method. The obtained results are presented in Table 1 [33].



Raman bands in dried powder pharmaceuticals			
Ibuprofen vibrations (cm^{-1})	Assignment	Paracetamol vibrations (cm^{-1})	Assignment
882	CH_2 rocking; $C^{19}H_3$ rocking; $C^{14}H^2O$ bending	328	Out-of-plane ring deformation
1187	C^6–C^{11} stretching	1236	C–N stretching
1211	CH_2 twisting	1285	O–H and C–O combination, (ar) C–N stretching
1292	C^{14}–H^{20} bending; C^{11}–H^{13} bending	1324	C–H bending
1458	$C^{15}H_3$ and $C^{19}H_3$ antisymmetric deformation	1372	-CH_3 bending
1659	Overtone 833 cm^{-1}	1568	C–NH deformation
2874	CH_2 symmetric stretching	1622	CC stretching mode, and in-plane CCC deformation
2926	$C^{15}H_3$ and $C^{19}H_3$ in-phase symmetric stretching	1652	C=O stretching mode
2959	CH_2 antisymmetric stretching		

Table 1. Identification of ibuprofen and paracetamol Raman bands in dried powder [33]

This unique spectrum can serve as a reliable biomarker for the presence of drug molecules. The proposed approach offers an innovative tool for the ultrasensitive detection of pharmaceutical compounds deposited onto graphene substrates. The study underscores the considerable potential of GERS in applications such as pharmaceutical contamination monitoring, water quality assessment, and public health protection [33].

3. Conclusion

Graphene-enhanced Raman spectroscopy (GERS) has emerged as a promising analytical technique offering significant advantages for materials characterization. The unique capability of graphene-based substrates to enhance Raman scattering signals while effectively suppressing interfering spectral contributions has positioned GERS as a valuable tool across a broad range of research domains. Consequently, a thorough and systematic examination of this technique is of considerable importance.

This review begins by elucidating the underlying mechanisms of signal amplification in GERS. Subsequently, in light of the distinctive properties and structural versatility of graphene, we explore various classes of graphene-based substrates and evaluate their influence on the overall Raman enhancement efficiency.

Finally, selected applications of GERS in the detection and analysis of diverse materials are discussed. Given its demonstrated potential, GERS represents a viable and increasingly attractive alternative to conventional and less sensitive methods for materials analysis. Accordingly, future research efforts should be directed toward addressing existing challenges and advancing the practical deployment of GERS in real-world analytical settings

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