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Review

Graphene-Based Surface-Enhanced Raman Scattering (SERS) Sensing: Bibliometrics Based Analysis and Review

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Abstract: Surface-enhanced Raman scattering (SERS) has received increasing attention from researchers since it was first discovered on rough silver electrode surfaces in 1974 and has promising applications in life sciences, food safety, and environmental monitoring. The discovery of graphene has stirred considerable waves in the scientific community, attracting widespread attention in theoretical research and applications. Graphene exhibits the properties of a semi-metallic material and has also been found to have Raman enhancement effects such as in metals. At the same time, it quenches the fluorescence background and improves the ratio of a Raman signal to a fluorescence signal. However, graphene single-component substrates exhibit only limited SERS effects and are difficult to use for trace detection applications. The common SERS substrates based on noble metals such as Au and Ag can produce strong electromagnetic enhancement, which results in strong SERS signals from molecules adsorbed on the surface. However, these substrates are less stable and face the challenge of long-term use. The combination of noble metals and graphene to obtain composite structures was an effective solution to the problem of poor stability and sensitivity of SERS substrates. Therefore, graphene-based SERS has been a popular topic within the last decade. This review presents a statistically based analysis of graphene-based SERS using bibliometrics. Journal and category analysis were used to understand the historical progress of the topic. Geographical distribution was used to understand the contribution of different countries and institutions to the topic. In addition, this review describes the different directions under this topic based on keyword analysis and keyword co-occurrence. The studies on this topic do not show a significant divergence. The researchers' attention has gradually shifted from investigating materials science and chemistry to practical sensing applications. At the end of the review, we summarize the main contents of this topic. In addition, several perspectives are presented based on bibliometric analysis.

Keywords: Surface-Enhanced Raman Scattering (SERS); graphene; substrate; bibliometrics; CiteSpace

1. Introduction

Since its discovery, Raman spectroscopy has had important applications in many fields, such as material structure analysis, biomolecular analysis, chemical and hazardous substance detection. However, the molecular scattering cross-section during conventional Raman detection is very small, making it challenging to detect molecular information at low concentrations and achieve quantitative detection of molecules.

Surface-enhanced Raman scattering (SERS) is a phenomenon in which the Raman signal intensity is significantly enhanced when the probe molecule is adsorbed on the surface of metallic nanoparticles (NPs) such as Au NPs, Ag NPs, or Cu NPs. This phenomenon was first discovered by Fleischmann et al. in 1974 [1]. They roughened the surface of the silver electrode using electrochemical methods and found a significant increase in the intensity of the Raman spectral signal of the pyridine molecules adsorbed on its surface. This phenomenon was then thought to increase the number of adsorbed molecules with the increase in surface area of the roughened silver electrode. It was not until 1977 that V. Duyne et al. [2] compared the Raman spectra of each pyridine molecule adsorbed on the rough Ag surface with the Raman spectra of pyridine in the solution phase. After a series of experiments and calculations, they found that the signal intensity of Raman spectra of pyridine molecules on Ag surfaces was enhanced by about 6 orders of magnitude and concluded that this surface enhancement effect was related to the roughness of the substrate surface.

After decades of continuous efforts and exploration by researchers, SERS has been developed as a new technique for non-destructive, rapid, and highly sensitive detection of structural information of chemical and biological molecules with enhancement factors up to 10^{14}~10^{17} [3–7]. Therefore, SERS can be applied in ultra-trace detection of hazardous substances, quantitative detection of molecular concentrations, and flow cytometry, which are beyond the reach of conventional Raman spectroscopy. The most commonly used metallic materials in SERS substrates are Au, Ag, and Cu, with Ag being the most effective enhancement. However, Ag substrates are susceptible to oxidation by oxygen in the air during preparation and storage. Therefore, most researchers currently use Au NPs to prepare various SERS substrates. Besides, many transition metals can be used to prepare SERS substrates, such as cobalt, iron, nickel, platinum, and lead [8,9], but the enhancement effect is weak and has not been widely investigated.

Although it has been more than 40 years since the discovery of SERS, its theoretical aspects have been relatively backward, mainly because the SERS effect has a very complex system. The physical structure of the surface of the SERS system and the electronic structure of the surface, the interaction of light with the rough surface and surface molecules, the orientation of the molecules on the surface, the bonding interaction and the surrounding environment of the molecules and the surface, the intensity, frequency, polarization and polarization direction of the incident laser all have a relatively complex effect on the SERS spectra, and therefore understanding of the SERS mechanism has remained divided until now [10,11]. Most researchers have recognized two mechanisms so far: electromagnetic enhancement (EM) and chemical enhancement (CM).

Graphene as a monolayer carbon film of sp^2 hybridized carbon atoms had been considered in scientific studies since the 1940s [12], but the available preparations were not stable. Novoselov et al. [13,14] have obtained the first stable graphene preparation on a film support using tape mechanical exfoliation of highly oriented pyrolytic graphite. Since then, graphene has been attracting the attention of researchers and industrial producers in fundamental physics research and advanced functional composites and devices for applications such as biological and chemical sensors, flexible displays, new energy batteries, and desalination of seawater. It was found that the Raman signal of molecules can be significantly enhanced when specific molecules are adsorbed on the graphene surface using graphene as a substrate [15]. By comparing the Raman signal intensity of phthalocyanine molecules on graphene and SiO_2/Si substrates, it was found that the Raman signal intensity of phthalocyanine molecules adsorbed on the surface of monolayer graphene was much stronger than that on SiO_2/Si substrates, indicating that monolayer graphene has a significant Raman enhancement effect. Despite the many advantages of graphene in SERS applications, its CM effect is weak, with enhancement factors (EFs) typically only in the range of 0.3–100 [16–19], far inferior to metallic SERS substrates.

In graphene-enhanced Raman substrates, observing the chemical enhancement mechanism in SERS is more convenient because there is no interference from electromagnetic field enhancement. However, at the same time, a considerable part of sensitivity is lost. For this purpose, researchers have prepared graphene-noble metal nanoparticle composite structures as SERS substrates. The strong electromagnetic fields distributed among the noble metal nanoparticles provide excellent Raman enhancement for the substrates, and the enhancement factor for the SERS substrates is well guaranteed. Graphene is also a new additive material in composite SERS substrates with many advantages such as fluorescence quenching, surface passivation, surface enrichment, and additional chemical enhancement. Until now, graphene-based SERS has been applied to detection in different fields, including environment, medicine and food. For example, Butmee et al. [20] reported a graphene-based SERS substrate for glyphosate direct detection in environmental water and soil. Xie et al. [21] reported the using of graphene/Ag SERS substrate for detecting the prohibited colorants in food. Ponlamuangdee et al. [22] reported a graphene/Au SERS substrate for detection of Mitoxantrone.

In recent years, several scholars have reviewed the work on graphene-based SERS. For example, Cao et al. [23] and Kang et al. [24] have summarized graphene-based SERS' sensing and catalytic applications. Analytical applications of graphene-based SERS are summarized by Zhang et al. [25]. The SERS properties of graphene/silver nanocomposites are summarized by Sharma et al. [26]. The series of reviews summarize important work on this topic in recent years, starting from the properties of graphene and conventional SERS materials. In this review, we attempt to analyze and summarize this topic using a bibliometric approach. A series of statistical indicators were used to analyze the different directions of investigation and important papers on this topic. We have tried objectively presenting the topic's historical development and current status. With a statistically based analysis and an interpretation of the highlighted literature, we tried to summarize this topic's challenges and future perspectives. Specifically, this review attempts to summarize and explain the following issues using bibliometrics:

(1) Can graphene effectively improve the sensitivity of conventional SERS substrates?
(2) Are the SERS properties of graphene itself promising for applications?
(3) What nanomaterials and morphologies are often used to compound with graphene to prepare SERS substrates?
(4) Do graphene-based SERS substrates already have a specific application at this stage?
(5) Has the enthusiasm for research on this topic waned, as attention has gradually shifted from graphene to other novel materials?

Two bibliometrics software have been used in this work. The first is CiteSpace, developed by Dr. Chaomei Chen, a professor at the Drexel University School of Information Science and Technology [27], which has become one of the commonly used software in bibliometrics analysis. CiteSpace 6.1R2 Advanced was used to calculate and analyze all documents. COOC is another emerging bibliometrics software [28]. COOC12.6 was used to calculate and analyze all documents.

We used the core collection on Web of Science as a database to assure the integrity and academic quality of the studied material. The following is the search criterion, where "Title" is used to retrieve the data.

"graphene SERS"

or "graphene 'surface-enhanced Raman scattering'"

or "graphene 'surface-enhanced Raman spectroscopy'"

The retrieval period was indefinite, and the date of retrieval was 30 December 2021. 516 research articles were retrieved, spanning the years 2010 to 2021.

2. Developments in the Research Field

2.1. Literature Development Trends

The number of published papers is an important indicator to evaluate whether a topic is attracting widespread attention. Figure 1 shows the annual and the cumulative number of publications on graphene-based SERS papers between 2010 and 2021. Although SERS is a topic with a long history, a graphene-based SERS paper undoubtedly needed to become possible after graphene was prepared. The publication of the graphene-based SERS paper was not first reported until the year graphene won the Nobel Prize in Physics. As shown in Figure 1, four papers were reported in 2010, including papers by Nobel laureates Andre Geim and Kostya Novoselov [29]. They investigated the advantages of graphene as a substrate for SERS. They proposed a graphene/SiO_2 (300 nm)/Si system and detected significant enhancements at 633 nm. The 2D nature of graphene allows for a closed-form description of the Raman enhancement. This is the starting point for graphene for SERS applications. Led by this paper and several others published in 2010, the topic quickly gained much attention. The superiority of graphene as a SERS substrate was also discussed by Wang et al. [30]. Specifically, they found that depositing a gold film of about 7 nm on the monolayer graphene surface could achieve the optimal SERS effect with the lowest photoluminescence background. Fu et al. [31] and Huang et al. [32] chose to investigate the potential of graphene derivatives, graphene oxide (GO) and reduced graphene oxide (rGO), for applications in SERS. This trend peaked in 2017 when the number of articles published reached 70.

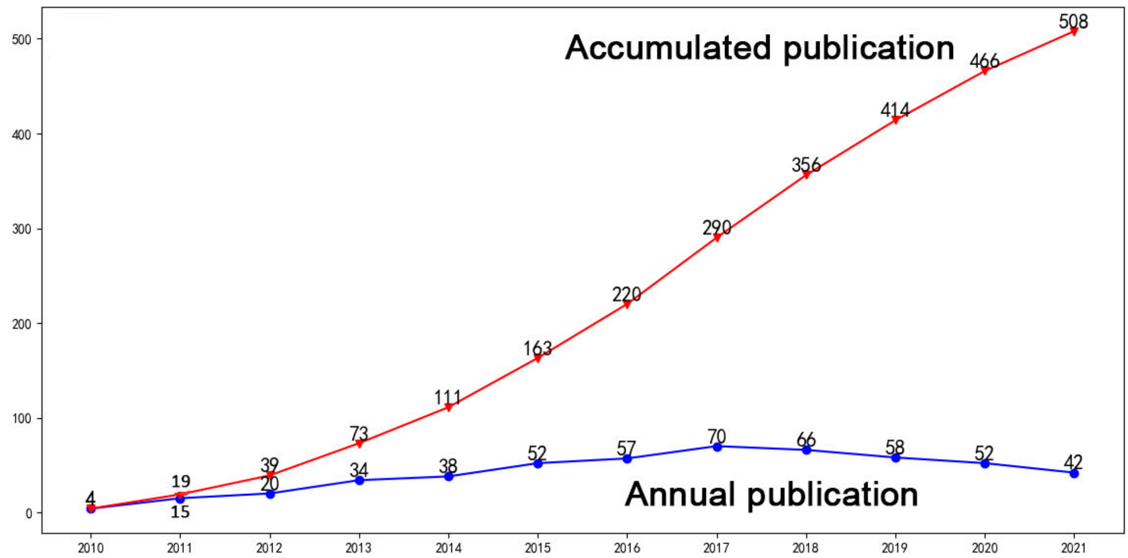


Figure 1. Annual and accumulated publications from 2000 to 2021 searched in the Web of Science about graphene-based SERS.

This topic immediately received widespread attention and great enthusiasm was devoted to the investigation, as shown by the rapid increase in the number of annual publications year after year. After 2017, the number of publications on this topic began to decrease yearly, with an annual of 42 articles in 2021. The publication history of this topic can be taken as a typical case in the bibliometric statistics. It has experienced a rise and a peak and is coming to the end of a cycle. It can be expected that the annual publication of this topic will continue to decrease after 2021 unless new important results are reported on this topic or new links are made with some popular fields, which will start a new life cycle of this topic. According to the overall publication process in the figure, an analysis of the papers around 2017 provides an understanding of the most important directions of investigation in this topic. In addition, analysis of papers published in recent years allows us to understand the most cutting-edge results on the topic and predict whether new directions of the investigation will emerge.

2.2. Journals, Cited Journals, and Research Subjects

Figure 2 shows the ten journals with the highest number of publications on graphene-based SERS. As can be seen, materials science-related journals dominate the tree diagram. Among them, the journal with the highest number of publications is Applied Surface Science. Although this journal focuses on the physical and chemical properties of interfaces, it publishes mostly on the properties and applications of interfaces of different materials. Similarly, RSC Advances is a comprehensive journal in chemistry, but it also publishes many papers related to materials science. In addition to journals related to materials science, journals in two other fields are included in Figure 2. The optics journal Optics Express and the sensing journal Sensors and Actuators B-Chemical published 11 and 14 papers related to graphene-based SERS, respectively. The appearance of these two journals is not unexpected, as SERS is an optical technology, and its most common application is the sensing and detection of specific analytes.

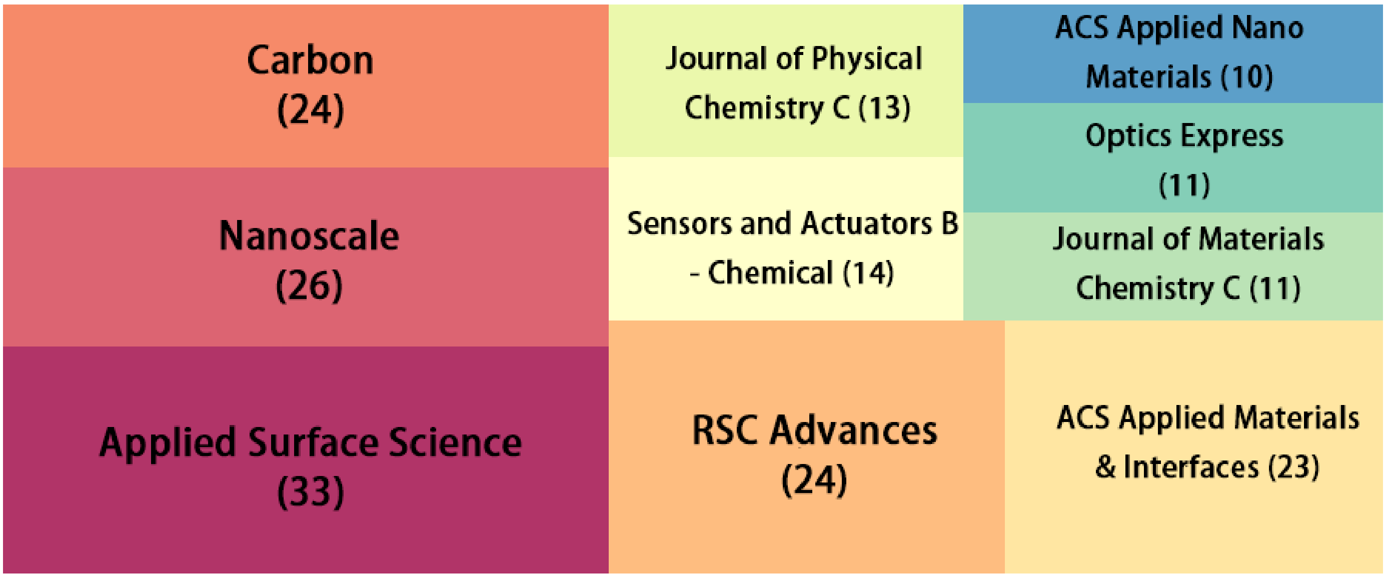


Figure 2. The top 10 journals that published articles on the graphene-based SERS.

In addition to the journals in which the papers are published, cited journals are also essential information to understand what areas the content of the topic is related to. Table 1 shows the top 20 cited journals on the graphene-based SERS. Comparing Figure 2 and Table 1, some interesting phenomena can be found. For example, although Applied Surface Science has published the most papers on this topic, it is ranked 20th in the cited journal. The fact that the ranking overlap between Figure 2 and the journals in Table 1 is not very high represents that this topic does not form very strict domain boundaries, which is different from the results of our previous bibliometric analysis of some topics [33–38]. The development and results of other themes will, effectively, have an impact on it. We found some physics-related journals in Table 1, such as Applied Physics Letters and Physical Review Letters, in addition to chemistry- and materials-related journals. This means that SERS' theoretical research on optics will also influence this topic. Two other journals of interest in Table 2 are Langmuir and Analytical Chemistry. Among them, Langmuir, a journal that publishes papers mainly in surface and colloidal chemistry, appears in Table 2, representing the adsorption properties of SERS substrates for target analytes as an essential content in this topic. On the other hand, Analytical Chemistry is a reputable journal in the field of analytical chemistry. Its emergence represents a move from theoretical analysis to practical detection in graphene-based SERS research.

Table 1. The top 20 cited journals on the graphene-based SERS.

No.	Citation	Cited Journal
1	375	ACS Nano
2	369	Journal of the American Chemical Society
3	359	Nano Letters
4	345	The Journal of Physical Chemistry C
5	311	Nanoscale
6	295	ACS Applied Materials & Interfaces
7	289	Small
8	275	Advanced Materials
9	260	Science
10	259	Carbon
11	234	Chemical Society Reviews
12	233	Analytical Chemistry
13	202	Chemical Communications
14	188	Langmuir
15	186	Nature
16	184	RSC Advances
17	183	Physical Chemistry Chemical Physics
18	182	Applied Physics Letters
19	180	Physical Review Letters
20	178	Applied Surface Science

Table 2. A list of journals has appeared in the co-occurrence network over the last two years.

Year	Journal Name
2021	ChemistrySelect, Journal of Materials Science & Technology, Science of The Total Environment, ACS Applied Bio Materials, The New England Journal of Medicine, Coatings, Frontiers in Chemistry, Nucleic Acids Research, Advanced Biology, Polycyclic Aromatic Compounds, Heliyon, Cellulose, Royal Society Open Science, Nature Reviews Chemistry, Fullerenes, Nanotubes and Carbon Nanostructures, Environmental Science: Nano, Dyes and Pigments, Journal of Biomolecular Structure and Dynamics
2020	Molecules, Biosensors, Journal of Pharmaceutical and Biomedical Analysis, NPG Asia Materials, Optical Materials Express, Nanoscale Horizons, Nanomaterials, JOSA B, Spectroscopy and Spectral Analysis, Bioinformatics, Composites Part B: Engineering, Nano Materials Science, Computational and Theoretical Chemistry, International Nano Letters, Synthetic Metals, Journal of Physics D, Trends in Food Science and Technology, American Journal of Psychiatry, American Mineralogist, Advanced Healthcare Materials, American Journal of Nuclear Medicine and Molecular Imaging, Advanced Synthesis & Catalysis, Advanced Device Materials, ACM Transactions on Sensor Networks

The relationship between different journals can be further understood using cited journals' co-occurrence network (Figure 3). In the network, the size of the node is proportional to the number of times the journal is cited. The links in the network represent the co-citation relationship between journals. The closer the co-citation relationship is proportional to the thickness of the line between them. Not surprisingly, the network is dominated by journals related to chemistry and materials science (large node radius), which are clustered in the lower right corner of the network. However, the darker color of the connecting lines between these journals represents that the connection between them occurred before the middle stage of this topic. The journals represented by these nodes do not correspond in the field to the journals in the lower right corner. Most of them belong to analytical chemistry, such as Talanta, Biosensors and Bioelectronics, Analytical and Bioanalytical Chemistry, Electrochimica Acta, Analytical Methods, etc. This phenomenon partially confirms the speculation in the previous paragraph that analytical detection using graphene-based SERS has become an important part of this topic in recent years.

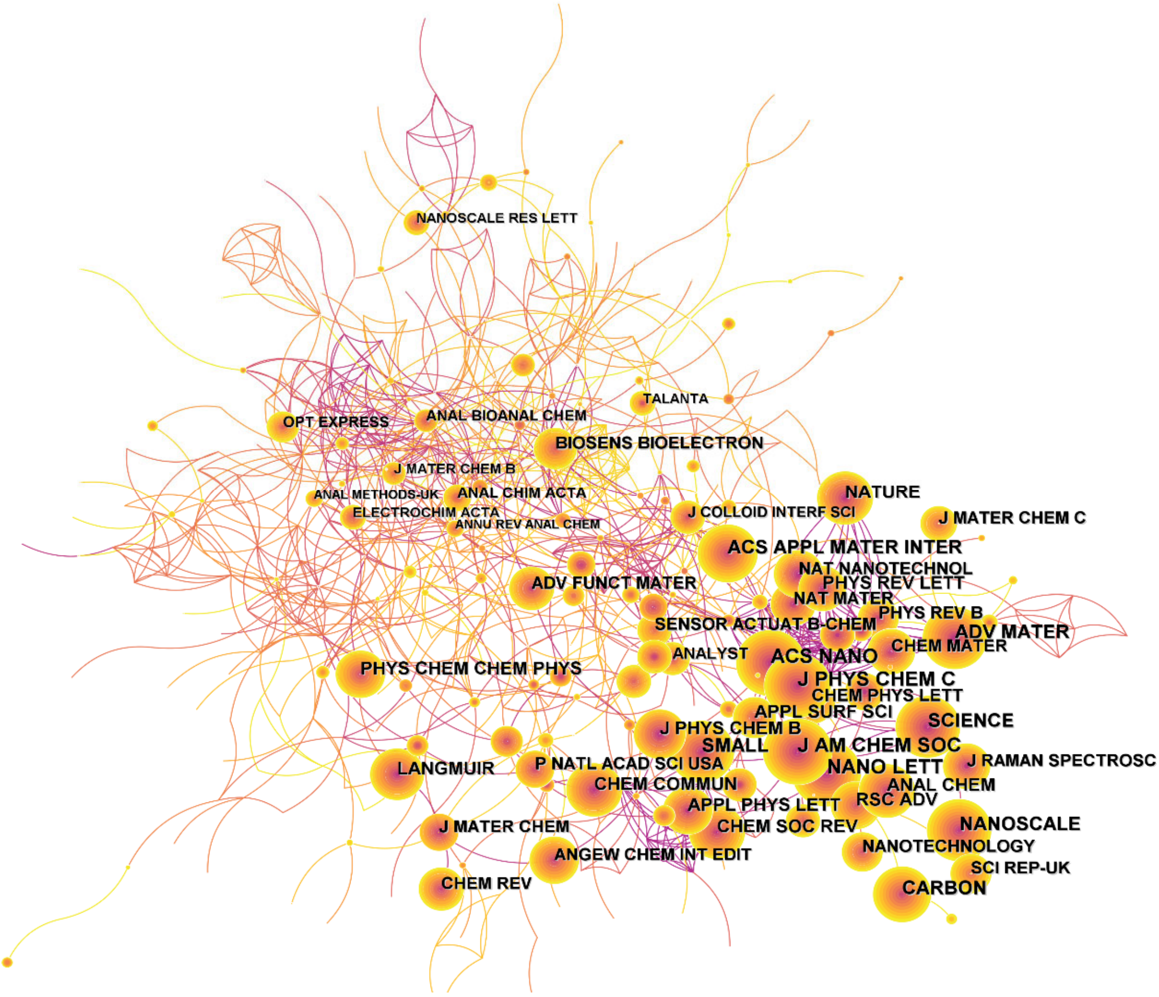


Figure 3. The co-occurrence network of cited journals for graphene-based SERS.

To learn more about the latest developments in graphene-based SERS, Table 2 shows journals that have begun interacting with graphene-based SERS for the first time over the last two years. As can be seen, the journals appearing in 2020 are mainly in the fields of materials science, analytical chemistry and sensing, with only a few journals belonging to fields not mentioned in the previous paragraph, including Trends in Food Science and Technology, American Journal of Psychiatry, Journal of Nuclear Medicine and Molecular Imaging. However, this emergence changed significantly in 2021, when a large number of journals in different fields started to interact with graphene-based SERS. These include journals in the field of environmental science (Science of The Total Environment, Environmental Science: Nano), medical journals (The New England Journal of Medicine) and biology journals (Nucleic Acids Research, Advanced Biology, Journal of Biomolecular Structure and Dynamics). This may be due to the application value of graphene-based SERS being able to meet the needs of certain specific detection fields. This speculation will be further explored in the keyword analysis.

Figure 4 illustrates the evolution of graphene-based SERS among different categories (The category to which a paper belongs is determined by the categories of the published journals indexed in WOS). This topic was widely investigated from the beginning in materials science, physics, optics and chemistry-related categories. In 2012, graphene-based SERS entered the field of analytical chemistry, especially in electrochemical and spectroscopy-based analytical assays. In 2013, graphene-based SERS was explored for specific applications and was first used in Biotechnology & Applied Microbiology, Food Science & Technology, Environmental Science and Pharmacology & Pharmacy in 2013, 2014, 2015 and 2016, respectively. Li et al. [39] used Ag-graphene nanocomposites in combination with electrophoretic preconcentration and SERS techniques to develop an in situ detection sensor that can be used for polar antibiotics in water. Nguyen et al. [40] prepared graphene-gold film-gold nanorod substrates, gold film-gold nanorod substrates, and graphene-gold nanorods for SERS detection of pesticides azinphos-methyl, carbaryl, and phosmet. Qiu et al. [41] developed a NIR SERS imaging technique using GO-coated gold nanorods, which is expected to be used for bioimaging. In 2020, this topic was used for the first time in Biochemistry & Molecular Biology and Computer Science, Interdisciplinary Applications, respectively. Muntean et al. [42] investigated the surface dynamics of graphene/AgNPs-based DNA functional groups at different acidic pH values using SERS. The evolutionary path of this category further illustrates that, currently, graphene-based SERS is a well-established technique that can be explored for different analytical assays.

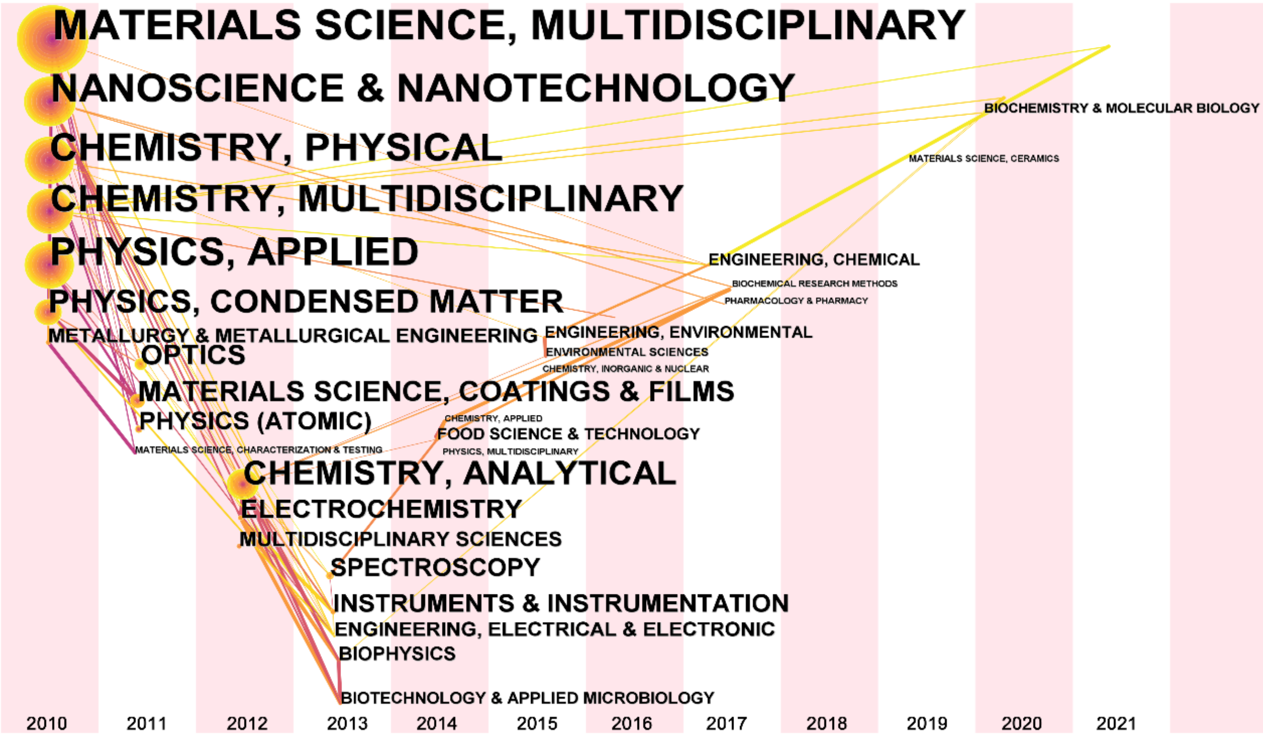


Figure 4. A time-zone view of research categories for graphene-based SERS.

2.3. Geographic Distribution

Figure 5 shows the 11 countries with the most publications. A total of 632 country labels were extracted from all publications. China contributed more than half of the papers. The country with the second-highest number of papers published is the USA, contributing 9.65% of the number. Both India and South Korea contributed an excess of 5% of the papers. The number of publications is not very intuitive data to evaluate the contribution of different countries on a topic, so the number of scientists varies considerably from country to country. However, it can be concluded from Figure 5 that graphene-based SERS attracts researchers from all regions of the world and does not show regionalization.

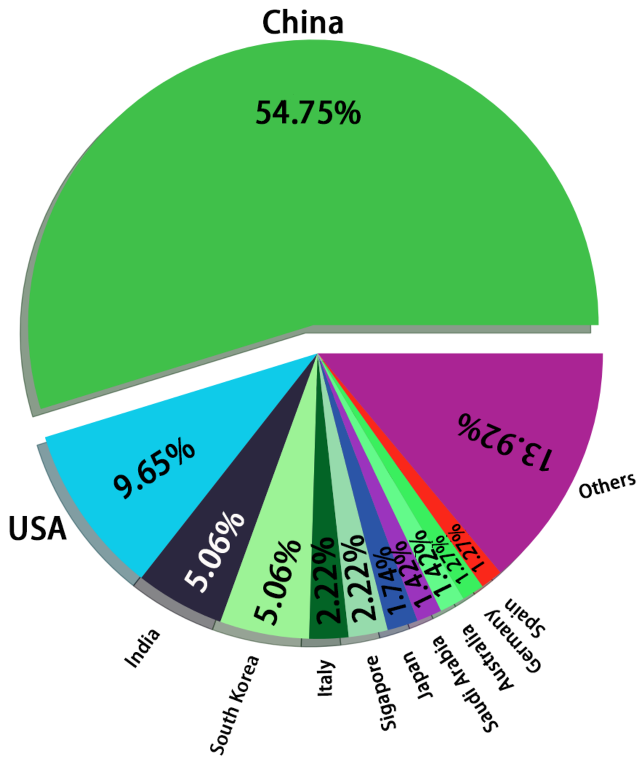


Figure 5. A pie chart of papers contributed by different countries.

Figure 6 shows the time-zone view of the geographic distribution for graphene-based SERS. China, USA, Singapore, England and Greece were involved in 2010 when this topic emerged. The following year, South Korea, Italy, and Australia joined in the investigation of this topic. The countries that appear in Figure 5 have joined the study on this topic by 2016. They are also the most densely connected countries on this topic, and the work on their behalf has generated extensive discussion and engaged other countries. Although the data in Figure 1 show that the number of graphene-based SERS publications has declined recently, the topic has attracted new countries to join it. Specifically, Malaysia, Vietnam, Pakistan and North Ireland joined them for the first time in 2019. Egypt joined it for the first time in 2020. In 2021, this topic again attracted scholars from Bangladesh and North Macedonia to participate in the survey for the first time.

Figure 7 illustrates the collaboration of the different institutes on this topic. It can be seen that graphene-based SERS presents a relatively wide range of collaborations. In addition to several small individual cooperation networks, this topic has a very close and large cooperation network. It can be seen that this complex collaborative network contains two institutions that contribute significantly in terms of the number of papers, namely the Chinese Academy of Sciences and Shandong Normal University. In addition to these two institutes, Chongqing University, Jiangsu University and University of Science and Technology of China have also contributed many papers on this topic. The number of publications is not the only indicator of an institution's contribution to the research on a topic. The centrality in CiteSpace can be used to measure the influence of a node in the network. Table 3 shows the 7 institutions with the highest centrality in this network. The Chinese Academy of Sciences contributed the most papers and received the highest centrality. However, Shandong Normal University does not appear in Table 4. On the contrary, Nanyang Technological University received second place for centrality, representing its influence on this topic. Similarly, the Massachusetts Institute of Technology presents an extraordinary impact although it has only published 3 papers in this field (it does not appear in Figure 7 due to the threshold we set for the node). Similarly, Jiangnan University and Academia Sinica are also in the same situation.

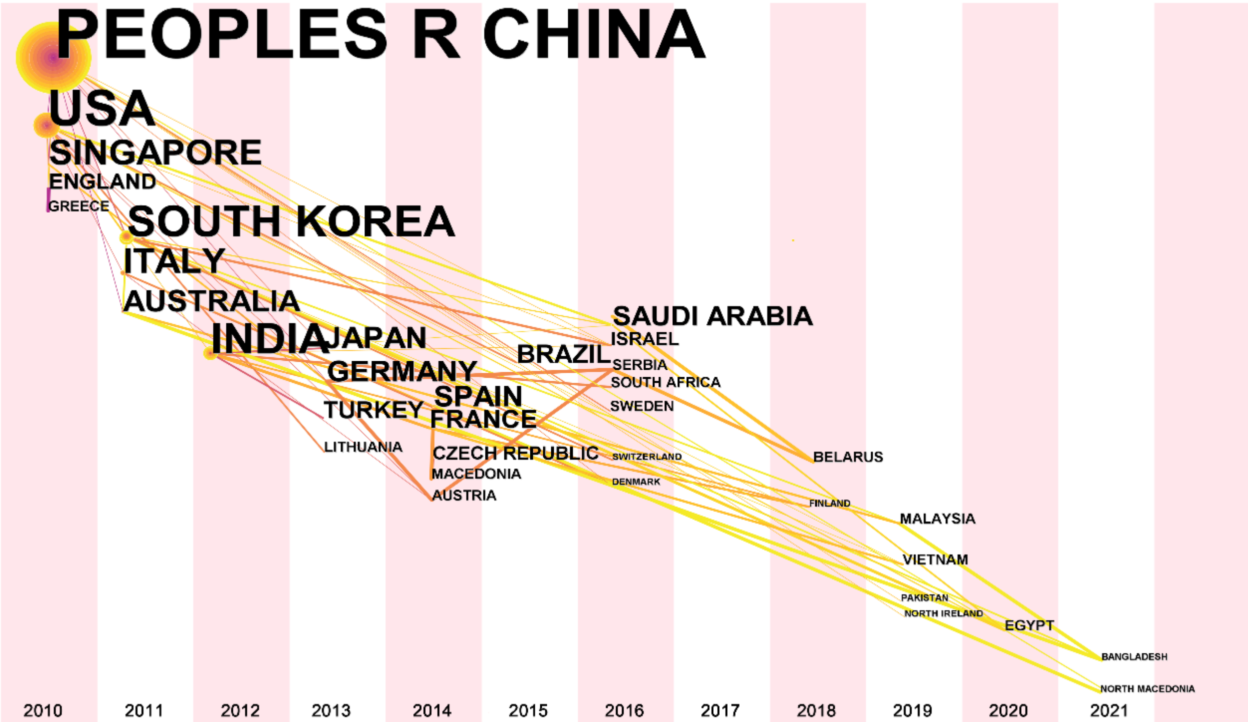


Figure 6. A time-zone view of geographic distribution for graphene-based SERS.

Figure 7. The institution cooperation network of published papers for graphene-based SERS.

Table 3. A list of the top 7 institutions using centrality order for graphene-based SERS.

No.	Count	Institution	Centrality
1	49	Chinese Academy of Sciences *	0.18
2	13	Nanyang Technological University	0.08
3	3	Massachusetts Institute of Technology	0.07
4	9	Jiangsu University	0.04
5	2	Jiangnan University	0.04
6	14	University of Science and Technology of China	0.03
7	3	Academia Sinica *	0.03

* Both Chinese Academy of Sciences and Academia Sinica contain a series of branch research institutions.

Table 4. A list of the top 15 keywords for graphene-based SERS.

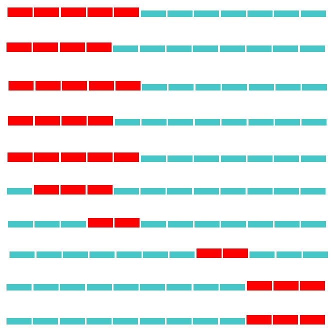
No.	Freq	Centrality	Keywords
1	179	0.26	Spectroscopy
2	129	0.21	Nanoparticle
3	125	0.15	Substrate
4	88	0.07	SERS
5	79	0.08	Film
6	76	0.07	Oxide
7	69	0.05	Fabrication
8	69	0.06	Platform
9	65	0.06	Nanostructure
10	62	0.07	Graphene Oxide
11	58	0.04	Molecule
12	50	0.05	Silver
13	49	0.04	Silver Nanoparticle
14	48	0.06	Gold Nanoparticle
15	45	0.04	Reduction

3. Keyword Analysis and Evolution of the Field

The analysis of keywords can be used to understand the different focuses of attention on a topic. Table 5 lists the top 15 keywords for graphene-based SERS. The content here does not give particularly obvious differentiating information. Some of these keywords are about optical techniques in the topic, such as Spectroscopy and SERS. Other keywords are the media used for SERS detection technology, such as Substrate, Film, and Platform. More interestingly, Graphene Oxide replaces graphene as a high-frequency keyword in Table 4, representing that GO will be more widely used than graphene for preparing SERS substrates. This may be because GO has more manipulable properties than graphene, especially its dispersibility in different solvents [43,44], making it easier to make it into substrates for SERS. On the other hand, this property also makes it possible to compound with other nanomaterials to further design advanced SERS substrates [45]. The high-frequency keywords in the table show that gold and silver nanoparticles are the most widely used nanomaterials. This is because they have the most significant SERS effect [46]. The eleventh most frequent keyword is Molecule, which represents the target used by SERS for detection. However, this table does not list specific molecules being used for detection, which may mean that SERS is not yet widely used for sensing a particular molecule or type of molecule.

Table 5. The 10 keywords with the strongest bursts during the research history of the graphene-based SERS.

Keywords	Strength	Begin	End	2010–2021
Sheet	5.53	2010	2014	
Delivery	3.39	2010	2013	
Graphene oxide	3.21	2010	2014	
Silver	3.07	2010	2013	
Spectra	2.95	2010	2014	
Gold nanostructure	2.78	2011	2013	
Pyridine	3.32	2013	2014	
SERS detection	4.07	2017	2018	
Graphene	3.92	2019	2021	
Surface-enhanced Raman spectroscopy	2.90	2019	2021	



Since the analysis of high-frequency keywords in Table 4 did not give much new information, we further filtered the keywords with Burst detection. Burst detection aims to identify an entity that is associated with a numeric function and the value of the function surges at least within a short period of time during the time frame we are observing. Burst detection can identify the direction of hot spots on which a topic is focused at different stages. As shown in Table 5, a total of ten keywords were retrieved. The first five of these keywords all start to come into focus with the appearance of this topic. The burst keyword Sheet with the highest strength value represented the SERS mechanism of the graphene sheet and was the first to receive attention and investigation. What is rather strange is the appearance of the keyword Delivery in Table 5. This burst keyword appeared from 2010 to the end of 2013 but linking it to SERS-related investigations is difficult. After carefully analyzing the literature, we found that Delivery stands for drug delivery. The potential drug delivery properties of graphene substrates were investigated at high frequencies during this period. In addition, a number of literatures have investigated the performance of graphene-based substrates in drug delivery and SERS [47–51]. Graphene oxide and Silver in Table 4 also appear at this stage in Table 5 at the same time, representing that they were also the object of attention from the beginning. Gold nanostructure also became a burst keyword in 2011. These results are well corroborated by the conclusions observed in Table 4. Pyridine was briefly a burst keyword in 2013 and 2014 due to its use as a probe molecule to investigate graphene-based SERS performance [52–54]. Starting from 2017, the research focus of this topic started to have shifted from the exploration of performance and mechanism to the evaluation of detection performance. Hence, SERS detection and Surface-enhanced Raman spectroscopy started to become burst keywords.

The cluster analysis of keywords can parse different documents by the similarity of shared keywords and can be used to understand different directions under a topic. Also, similar papers can be grouped together and used to sort out their contents quickly. Figure 8 shows the clustering results of keywords, with 16 clusters formed based on content similarity. From the figure, we can see that all the clusters except one (#15) overlap more or less with the neighboring clusters. Some of these clusters are entirely covered by surrounding clusters, meaning they overlap with each other in terms of content. Table 6 describes the clusters and their ID, size, silhouette, respective keywords and references. The following is a short explanation of each cluster:

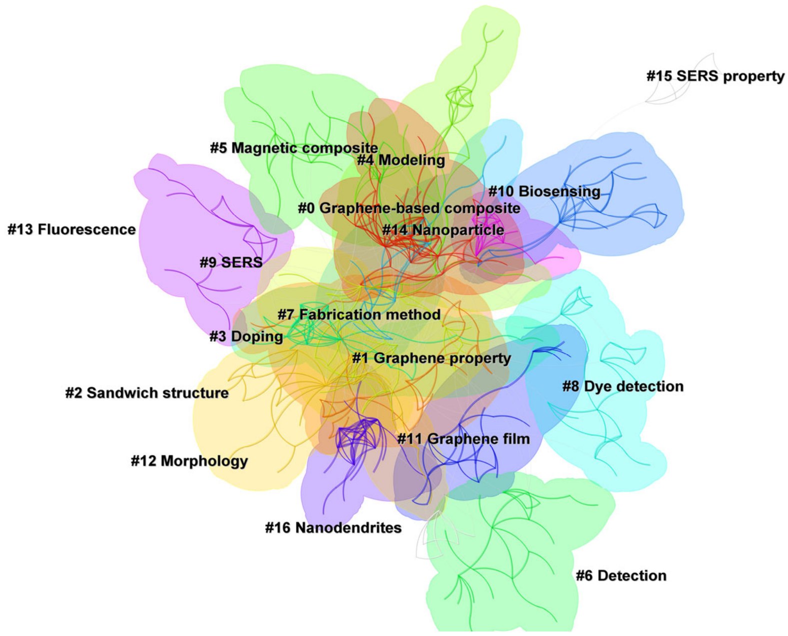


Figure 8. A grouping of keywords for graphene-based SERS.

Table 6. Knowledge clusters in the field of graphene-based SERS on keyword co-occurrences for each cluster.

Cluster ID	Size	Silhouette	Keywords	References
0	38	0.937	Nanoparticle, Substrate, Enhanced Raman scattering, Film, Fabrication, Platform, Reduction, Nanocomposite, Composite, Sheet	[32,54–142]
1	32	0.840	Nanostructure, Surface-enhanced Raman scattering, Array, Ag nanoparticle, Graphene, Water, Oxidation, Photoluminescence, Facile fabrication	[55,57,62,64,65,68,74,76,77,79,84,96,98,112,143–169]
2	30	0.935	Oxide, Silver nanoparticle, Reduced graphene oxide, Carbon, Light matter interaction, Green synthesis, Au nanoparticle,	[55,62,76,94,95,143,144,170–201]
3	29	0.824	Surface, Enhanced Raman spectroscopy, Rhodamine 6G, Graphite oxide, Electrode, Bacteria, Monolayer, Absorption	[56,60,70,73,77,104,146,147,171,177,185,202–220]
4	27	0.960	Growth, Raman, Plasmon resonance, Epitaxial graphene, Doped graphene, Facile synthesis	[72,97,221–225]
5	26	0.943	Sensitivity, Adsorption, Sensitive detection, SERS detection, Surface plasmon resonance	[21,57,75,89,171,217,226–233]

Table 6. *Cont.*

Cluster ID	Size	Silhouette	Keywords	References
6	24	0.941	Hybrid, Au, Deposition, Size, Acid	[47,50,86,91,100,108,234–244]
7	23	0.983	Spectroscopy, Molecule, Silver, Carbon nanotube, Few layer graphene, Pyridine	[16,31,55,56,61,66,67,69,82,88,101,106,109,170,172,173,175,178,182,186,218,220,226,234,235,237,245–290]
8	23	0.931	Surface enhanced Raman scattering, Surface-enhanced Raman spectroscopy, Monolayer graphene, Malachite green, Temperature	[42,58,74,80,85,92,247,291–295]
9	22	0.664	Chemical enhancement, Aromatic molecule, Chemistry, Cell, Optical property, Quantum dot	[54,246,249,253,264,292,296–300]
10	22	0.954	Performance, Single molecule, Immunoassay, Surface enhanced Raman, Ultrasensitive detection	[80,88,89,103,176,182,214,248,261,301–309]
11	21	0.926	Graphene oxide, Ag, Gold, Charge transfer, Gold nanostructure	[52,53,56,58,63,68,73,75,78,80,90,93,102,143,145,148,174,183,251,263,301,310–320]
12	19	0.908	Scattering, Spectra, Raman spectroscopy, Graphite	[29,40,60,66,120,124,210,214–216,219,234,236,246,255,256,258,259,291,310,321–335]
13	17	0.917	Fluorescence, Sensor, Gold nanorod, Shape	[41,59,143,181,245,260,336–340]
14	10	0.978	Gold nanoparticle, Label free detection, Hybrid film, Folic acid, Surface enhanced Raman scattering	[48,58,96,173,234,252,254,262,278,304,310,317,341–343]
15	6	0.988	Surface-enhanced Raman scattering (SERS), Anatase	[61,344]
16	6	0.968	Trace detection, Silver dendrite, Agent, Dendrite	[345]

#0 (Graphene-based composite) This one cluster contains the largest number of papers among all clusters. Most of them are concerned with preparing graphene and noble metal (Ag and Au) composites and investigating their SERS properties. The metal nanomaterials used for the composite have different morphologies such as nanocube [85,346], flower-like particle [57], nano-disc [61], nanorod [104], nanostar [136] and 3D butterfly wing structure [109]. In addition to Au and Ag, Cu [83], MoS_2 [347], Fe_3O_4 [58,282] and ZnO [98] have been used for the preparation of SERS substrates as well.

#1 (Graphene property) This cluster has a relatively low silhouette value, so its clustering effect is not particularly obvious. The graphene and Au/Ag composite continue to be a key content in this cluster. Unlike #0, this cluster contains a series of investigations on the SERS properties of graphene itself. For example, Ramanauskaite et al. [163] investigated the reduction process GO undergoes when used in SERS substrates and the changes in its properties. Li et al. [157] investigated that wrapping silicon nanowires with graphene allows silicon nanowires, which otherwise have no SERS properties, to become a novel SERS substrate. Han et al. [156] investigated the relationship between the chemistry and structure of graphene and its SERS properties.

#2 (Sandwich structure) The sandwich structure of the SERS substrate is the main content of interest in this cluster. Different investigations have found sandwich structures to be effective in enhancing the performance of SERS. For example, Wu et al. [172] synthesized an AgNP-graphene-AgNP sandwiched structure using a wet chemical method and an autonomous loading technique. Plasma coupling between AgNPs from both sides of the graphene can greatly enhance the performance of SERS. Zhao et al. [173] prepared AuNP-graphene-AgNP sandwiched substrates, which have a detection sensitivity of 10^{-13} M. Other sandwich structures include AgNPs-silica-GO [174], AuNPs-graphene-Au array [175], silicon nanowire-graphene-AuNPs [169,182], Ag-graphene-Au [183], AgNPs-TiO_2-graphene [144], Ag nanohole array-graphene-AuNPs [184] and AgNPs-silica-graphene [188].

#3 (Doping) The silhouette value of this cluster is low, so the direction of the papers contained in it is divergent. Two directions are worth noting. The first one is about the doping and modification of graphene. Some papers report that doping or modification of graphene can lead to more excellent SERS properties. For example, Kasztelan et al. [214] found that a simple treatment of GO with ammonia solution improved the SERS detection. This may be due to the partial reduction of GO by NH_3 and the introduction of nitrogen functionalization. Nair et al. [185] found that nitrogen sulfur co-doped RGO could be used to adsorb different forms of AgNPs and therefore exhibited more sensitive SERS performance. Another direction is the preparation of free-standing SERS substrates. Zhao et al. [220] synthesized a flexible film combining graphene and AgNPs for SERS applications. Fan et al. [346] also prepared a free-standing substrate containing GO and AgNPs for SERS. Lee and Kim [73] loaded AuNPs and GO on a hydrophobic paper, which can be used as a SERS substrate for analytical detection.

#4 (Modeling) This cluster focuses on the modeling of SERS. Al-Otaibi et al. [221] calculated the structural, nonlinear optical, electronic and biological properties of three anastrozole-based triazole analogues on graphene surfaces. The results demonstrated the enhancement of SERS for all three molecules. They also calculated three aminobenzoate derivatives and their SERS active graphene complexes [348]. Ullah et al. [349] performed theoretical calculations for adsorbed antimalarial-graphene dimers and predicted the SERS signal.

#5 (Magnetic composite) This cluster shares many papers with #0 and contains two directions. The first direction is the synthesis of graphene-Ag nanostructure-based composite for SERS. It is worth mentioning that this cluster does not contain any paper related to graphene-Au nanostructure-based composite. Another direction is the synthesis of graphene-based nanocomposites with magnetic properties. The fast magnetic response enables rapid separation of the composite material from the solution, and the practical application of SERS can be achieved by first using the material for adsorption on the analyte, followed by detection after rapid separation [217,231].

#6 (Detection) The papers in this cluster begin to focus further on the sensing applications of the prepared SERS substrates. Their titles will not only describe the preparation of a particular structure of the substrate but will also emphasize the detection of a particular analyte. For example, the work of Xu et al. [237] and Qiu et al. [242] both emphasized the detection of adenosine. Jinbin et al. [239] highlighted that their substrate could be used to detect circulating breast cancer cells. Naqvi et al. [240] highlighted that their SERS sensor is used for explosive detection. The SERS platform proposed by Dutta et al. [91] was used for uranyl ion sensing.

#7 (Fabrication method) This cluster mainly highlights the preparation techniques of different graphene-based SERS substrates and the way of optimization in the preparation process. Saha et al. [245] used stabilization of hot spots in GO liquid crystals to improve the reproducibility of SERS. Kovaricek et al. [246] investigated the covalent reaction during CVD to optimize the growth of graphene. Hu et al. [249] prepared SERS substrates by electrostatic self-assembly. Ouyang et al. [254] used a filtration-assisted fabrication technique to synthesize large-size SERS substrates.

#8 (Dye detection) Different dyes were used as analytes in this cluster. These dyes include malachite green [58,292,294], nile blue A [58], R6G [74,247,294,295] and methylene blue [294].

#9 (SERS) The silhouette value of this cluster is only 0.664. According to the CiteSpace manual, clusters with a silhouette value below 8.5 do not have a significant similarity. After analyzing the papers in this cluster one by one, we did not find any strong correlation between them.

#10 (Biosensing) This cluster mainly highlights the references of graphene-based SERS in biosensing. For example, the SERS substrate proposed by Fu et al. [302] to detect of cardiac troponin I. Chen et al. [303] focused on the detection of clenbuterol residues in animal-origin food samples by SERS. Lv et al. [306] tried the detection of adenine by SERS. Li et al. [308] attempted the detection of trace amounts of ferritin by SERS.

#11 (Graphene film) The content of this cluster is entirely covered by #1, #2, #6 and #8 as seen in Figure 8. The papers in this cluster mainly compare the SERS performance of noble metal nanomaterials enhanced with the assistance of graphene.

#12 (Morphology) The content of this cluster mainly emphasizes the effect of graphene morphology (number of layers) and location (center or edge) on SERS. For example, Xu et al. [322] investigated the SERS performance of highly ordered graphene-isolated silver nanodot arrays. Matz et al. [323] investigated the SERS fingerprint of monolayer graphene grown by CVD. D'Urso et al. [255] investigated the SERS properties of 1D-2D graphene-based structures.

#13 (Fluorescence) This cluster appears to utilize graphene quantum dots as a material for the SERS substrate. As a quantum dot, its fluorescence properties impact the Raman signal. Therefore, this series of work involves the investigation of the fluorescence properties. On the other hand, graphene has been observed to have a fluorescence quenching effect, which is one of the important reasons why it is widely used in SERS.

#14 (Nanoparticle) This cluster is also entirely covered by surrounding clusters, and its papers overlap with parts #0, #4, #5 and #10. It includes not only the composite of AuNPs or AgNPs with graphene but also the ternary composite of all three of them.

#15 (SERS property) This cluster includes only two papers. Guo et al. [344] used a photocatalytic method to grow Ag nanocrystals on the surface of TiO_2/RGO and examined their SERS properties. Liu and Luo [61] synthesized two gold nanostructures with different morphologies for compounding with graphene and evaluated their SERS properties.

#16 (Nanodendrites) This cluster contains only one paper. This paper describes the SERS properties after covering silver nanodendrites with graphene films [345].

We further summarize the highly cited papers. Table 7 listed the top 10 highly cited research papers and top 5 highly cited reviews in this topic. As can be seen from the table, the most concerned research papers are still the first series of papers in this field. This points to the fact that the topic has not gone through more than one cycle of research. The focus of the investigation at this stage is still on what concerns the topic when it was first proposed. Correspondingly, the most cited reviews did not show a large time span. There is a clear correspondence between the number of citations and the time of publication, which indicates that the focus of these reviews is not differentiated. More often, newly published reviews are updates to the original topic.

Table 7. The top 10 highly cited research papers and top 5 highly cited reviews in the field of graphene-based SERS.

No.	Title	Citation	Year	Reference
	Research Article			
1	Surface enhanced Raman spectroscopy on a flat graphene surface	455	2012	[350]
2	Surface-enhanced Raman spectroscopy of graphene	397	2010	[29]
3	Nanocomposites of size-controlled gold nanoparticles and graphene oxide: Formation and applications in SERS and catalysis	379	2010	[32]
4	A binary functional substrate for enrichment and ultrasensitive SERS spectroscopic detection of folic acid using graphene oxide/Ag nanoparticle hybrids	322	2011	[48]
5	UV/ozone-oxidized large-scale graphene platform with large chemical enhancement in surface-enhanced Raman scattering	297	2011	[16]
6	Tuning chemical enhancement of SERS by controlling the chemical reduction of graphene oxide nanosheets	274	2011	[17]
7	One-pot green synthesis of Ag nanoparticles-graphene nanocomposites and their applications in SERS, H_2O_2, and glucose sensing	261	2012	[257]
8	A facile one-pot method to high-quality Ag-graphene composite nanosheets for efficient surface-enhanced Raman scattering	238	2011	[351]
9	Silver nanoparticle decorated reduced graphene oxide (rGO) nanosheet: A platform for SERS based low-level detection of uranyl ion	226	2013	[91]
10	Surface enhanced Raman scattering of Ag or Au nanoparticle-decorated reduced graphene oxide for detection of aromatic molecules	225	2011	[71]
	Review Article			
1	Graphene: A platform for surface-enhanced Raman spectroscopy	393	2013	[352]
2	Graphene-gold nanoparticles hybrid-synthesis, functionalization, and application in an electrochemical and surface-enhanced Raman scattering biosensor	130	2016	[353]
3	Recent progress in the applications of graphene in surface-enhanced Raman scattering and plasmon-induced catalytic reactions	103	2015	[24]
4	Recent progress on graphene-based substrates for surface-enhanced Raman scattering applications	71	2018	[354]
5	Flexible and stretchable SERS substrate based on a pyramidal PMMA structure hybridized with graphene oxide assivated AgNPs	45	2018	[355]

Based on the above analysis, the investigation directions of the graphene-based SERS can be summarized as follows:

(1) The content of this topic does not show a considerable divergence. Most of the works have focused on investigating the performance of conventional SERS materials after graphene compounding.
(2) These SERS substrates prepared using graphene-based composites have much to investigate. For example, whether there is a difference in their SERS effect when different nanostructures and graphene are compounded. Whether the different oxidation states of graphene affect the SERS effect. Whether the number of layers of graphene affects the SERS effect of the composites.
(3) Investigation of the effect of graphene's own SERS. Mechanistic analysis of this phenomenon and whether it has practical value.
(4) The advantages of SERS in analytical assays. Which analytes are easier and more sensitive to detect using graphene-based SERS than other traditional detection methods.

Figure 9 shows the frequency of occurrence between keywords. Save for some similar keywords, the information in the figure can verify the conclusions drawn in the above keyword analysis. AgNPs and AuNPs are the most widely used compounds with graphene materials to prepare SERS substrates. On the other hand, nanostructures other than nanoparticles were often investigated. This is because different nanomorphologies can induce different SERS effects. The absence of any one molecule in terms of co-occurrence frequency represents that graphene-based SERS is not widely used for the actual detection of a specific class of analytes at this stage.

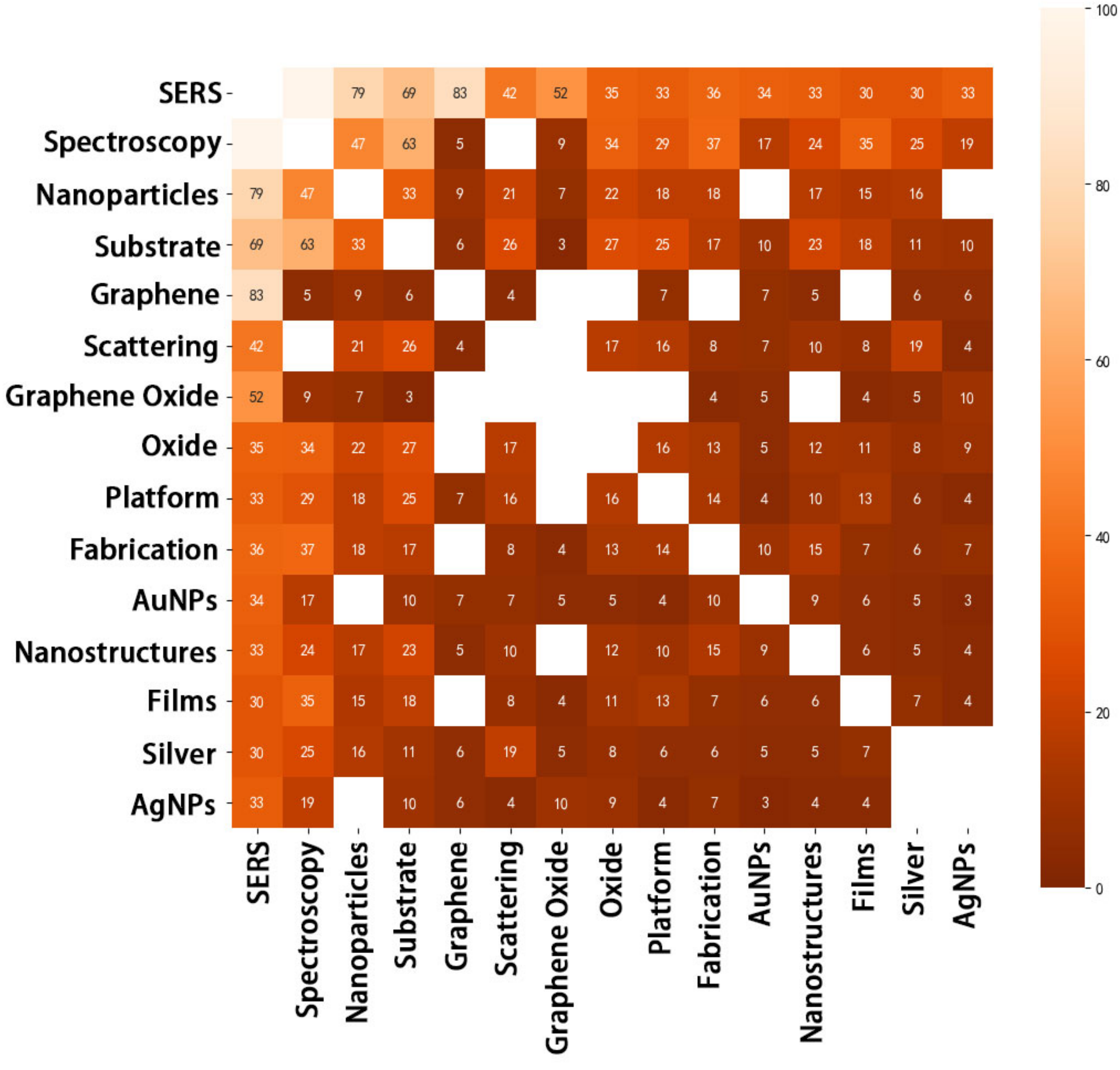


Figure 9. A keywords confusion matrix for graphene-based SERS.

4. Conclusions

The peculiar nature of SERS has led to its emergence as an analytical technique that has been successfully used as an alternative to other sensing techniques in several specific situations. The intrinsic SERS property of graphene and the advanced properties it exhibits when compounded with traditional SERS nanomaterials are breathing new life into the field. As a result, graphene-based SERS began to receive widespread attention starting in 2010. This bibliometrics-based review analyzes the history of the topic and its main content. Based on the above analysis, the following conclusions can be drawn:

(1) Graphene-based SERS has been widely discussed since it was proposed, and the publication of related papers gradually rose and peaked in 2017. This trend has not

continued until today. Starting in 2018, the annual number of publications on this topic began to decline, with only 42 in 2021. The annual number of publications shows that researchers are gradually shifting their attention from this topic to other areas.

(2) Although SERS is an optical-based analytical sensing technique, the investigations on this topic were initially focused on materials science and chemistry. This is because the SERS properties generated by graphene or by the composite of graphene and other conventional SERS nanomaterials need to be explained mechanistically. Therefore, most of the published papers on this topic simply choose a commonly used probe to evaluate the performance of the prepared SERS substrates rather than a custom development for specific detection needs. Starting in 2013, the topic gradually shifted from the investigation of materials science/optics/chemistry to different application areas, including food science, environmental science, pharmacology, molecular biology, etc.

(3) Chinese scientists contribute the most significant number of papers in this field, with the Chinese Academy of Sciences being the most influential institution. USA, India, and South Korea also play an important role in this topic. Nanyang Technological University in Singapore and Massachusetts Institute of Technology in USA have not published many papers on this topic. However, their work has had a significant impact. Based on the geographical analysis, this topic attracts the attention of scientists from global regions. Although the annual publication of this topic is decreasing yearly, countries continue to participate in this topic for the first time every year.

(4) The analysis of the keywords proves that the investigation of this topic focuses on the preparation of SERS substrates. Among them, GO in many cases replaces graphene for composite synthesis. The most commonly used SERS materials, AuNPs and AgNPs, continue to be the most widely used choices for composite with graphene. In addition to nanoparticles, other nanostructures have also been widely investigated. On the other hand, the nature of the graphene also affects the SERS performance, where a range of factors are included, such as the degree of oxidation, number of layers, size, folds, etc.

(5) Although graphene-based SERS has been studied for more than a decade, it has not yet presented a particular application dedicated to it in the sensing field. This may be due to the fact that while the assistance of graphene can provide enhancement of the SERS signal, it does not have the property of specific identification of the analyte. Therefore, it is indeed a very sensitive analytical tool when optimized, but it is more difficult to overcome the challenges posed by interferents in sensing.

As bibliometrics is an analytical technique based on statistics, there are still some deficiencies in the information analysis of scientific topics. In the process of writing this review, we found that bibliometrics had certain limitations in the discrimination of SERS materials. For example, SERS material contained a large number of composite materials or binary alloys. However, these materials are treated as separate words in the keyword analysis (because some authors refer to separate components of the composite when selecting keywords). Composites, on the other hand, tend not to have accepted acronyms. Therefore, the same composite material can be written differently in different papers. However, bibliometric software cannot merge these contents. This gives less weight to the information when it is analyzed.

5. Perspectives

Based on the review of this topic, we believe that the following issues need to be investigated regarding the graphene-based SERS:

(1) Graphene-based SERS substrates are an analytical platform that can be produced on a large scale with easily controlled stability. The development of practical applications based on this platform is a direction that needs to be focused on in the future. It is believed that with the participation of scientists from different fields, such as contaminant detection, food safety detection, drug detection, etc., it is possible to find suitable assay needs for this platform.

(2) Since graphene is a two-dimensional lamellar material, it changes its morphology when compounded with other nanomaterials. For example, it has the ability to combine into three-dimensional structures. These structural changes have been shown to affect the performance of SERS substrates. Some of these particular structures have also been shown to possess extraordinary properties. However, whether such structures can be controlled with high quality still needs to be verified. Therefore, how to tune graphene in SERS substrates is an important direction. Finding a balance between the reproducibility of the prepared substrates and SERS performance is challenging.

GO has been used in much of the work on this topic to prepare SERS substrates because it is easier to compound with other nanomaterials by chemical techniques than graphene. However, the properties of GO are very strongly linked to its degree of oxidation and GO in some of these works will also be partially reduced. The effect of this process on SERS properties needs to be investigated in depth. Similarly, the doping of graphene has been reported to lead to improved SERS performance. However, there is a lack of solid data on the relationship between the doped elements and the degree of doping with the SERS performance.

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