An overview of surface-enhanced Raman scattering substrates by pulsed laser deposition technique: fundamentals and applications

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Abstract

Metallic nanoparticles (NPs), as an efficient substrate for surface-enhanced Raman scattering (SERS), attract much interests because of their various shapes and sizes. The appropriate size and morphology of metallic NPs are critical to serve as the substrate for achieving an efficient SERS. Pulsed laser deposition (PLD) is one of the feasible physical methods employed to synthesize metallic NPs with controllable sizes and surface characteristics. It has been recognized to be a successful tool for the deposition of SERS substrates due to its good controllability and high reproducibility in the manufacture of metallic NPs. This review provides an overview about the recent advances for the preparation of SERS substrates by PLD technique. The influences of parameters on the sizes and morphologies of metallic NPs during the deposition processes in PLD technique including laser output parameters, gas medium, liquid medium, substrate temperature, and properties of 3D substrate are presented. The applications of SERS substrates produced by PLD in the environmental monitoring and biomedical analysis are summarized. This knowledge could serve as a guideline for the researchers in exploring further applications of PLD technique in the production of SERS substrate.

Keywords Pulsed laser deposition · Surface-enhanced Raman scattering · Metallic nanoparticles · Substrate morphology

1 Introduction

Raman spectroscopy, based on the inelastic scattering that has been firstly discovered by Raman [1], plays a major role in providing structure information, surface, and interfacial process at the molecular level. However, the signals of Raman scattering are too weak to be detected since 10^7 of incident phonons could excite only one Raman phonon. Later on, a phenomenon of an enhanced Raman signal in the pyridine adsorbed on the surface of a roughened silver electrode has been firstly observed by Fleischman et al. in 1974 [2], which was explained by the adsorbed more molecules on the surface of rough electrode arising from its larger specific surface area than that of smooth electrode. However, in 1977, Van Duyne [3] and Creighton [4] found that the

increased number of adsorbed molecules caused by the high specific surface area of rough electrode was not consistent with the enhancement of Raman scattering intensity. Subsequently, this new phenomenon was received tremendous interests and named as surface-enhanced Raman scattering (SERS). After development over more than 40 years, SERS is widely used in the fields of physics, chemistry, biology, medicine, etc. [5–8] resulting from its extremely high sensitivity, good selectivity, and rapid response. It is possible to characterize molecules adsorbed on the surface of SERS substrate, even to realize the detection of a single molecule [9].

Normally, it is well accepted that the electromagnetic mechanism and chemical mechanism are two major enhancement mechanisms in the SERS theory. Electromagnetic mechanism is regarded as the dominant model in the understanding of SERS theory because the intensity of SERS is proportional to the fourth power of the incident electromagnetic field amplitude [3, 4]. The origin of electromagnetic enhancement on the nanoscale is from the collective oscillation of electrons excited by the incident light on the metal surface. This collective oscillation is called surface plasmon resonance (SPR) that is influenced

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by the size and morphology of SERS substrate [10, 11]. By controlling the shape, size, and interparticle spacing of SERS substrate, the SPR frequency could be altered, further attaining an optimized SERS enhancement [12].

It is evident that the selection of proper methods for preparing substrates is significant because the efficiency of SERS technique distinctly depends on the characteristics of SERS substrates. Metallic nanoparticles (NPs) are recognized as the most studied substrates after the discovery of SERS effect. In general, the synthesis methods of metallic NPs could be divided into chemical methods and physical methods [13–17]. The reduction of metal salts in aqueous or nonaqueous solution [18–21] is the main chemical method for the fabrication of metallic NPs. Both experimental and theoretical results have indicated that a junction or close interaction of two or more metallic NPs (that has been called hot spot) would produce strong SERS enhancement owing to the coupling of electromagnetic field [9, 22, 23]. However, the SERS substrate produced by chemical reduction method has a poor reproducibility because of the randomness of interparticle distance and arrangement. Therefore, the enhancement effect is not easy to be predicted. Self-assembly method is a commonly used chemical method to improve the reproducibility of SERS signal. This is the reason that in the self-assembly process [24–26], the metallic NPs could be fixed on the glass slide surface via the presence of functional groups like amine or thiol groups by immersing the glass side surface into metallic NPs suspension, leading to a stable SERS effect. Then, the adjustable distance between two metallic NPs by changing the immersion time is contributed to the SERS enhancement [27]. Unfortunately, various reducing agents, surfactant, and adhesion agent have been added in the chemical process for fabricating the metallic NPs to prevent their agglomeration/oxidation and adjust their growth rate [28–30]. Consequently, a large number of reagents used in the chemical methods result in the contamination of fabricated metallic NPs.

The manufacture of metallic NPs is an easy task, but keeping the uniform dispersion of uncontaminated metallic NPs is a challenge. By contrast, the physical methods could be broadly exploited in the synthesis of metallic NPs thanks to the good controllability and high reproducibility. In addition, physical methods could also yield the uniformly monodisperse metallic NPs without solvent contamination. Thermal deposition [31, 32], sputtering deposition [33–36], electron beam evaporation [37–39], and pulsed laser deposition (PLD) [40–43] are some of the most commonly used physical methods to produce the metallic NPs. Mechanical pressure, high-energy radiation, thermal energy, and electrical energy have been applied to generate the metallic NPs by triggering the melting, evaporation, condensation, and ablation [28].

Among these physical methods, PLD is a relatively flexible and effective technique for the fabrication of metallic NPs with controllable sizes. Metallic NPs produced in ultrahigh vacuum feature are different from that in the conventional deposition techniques such as evaporation or sputtering [44]. In PLD technique, the pulsed laser beam like excimer laser or Nd:YAG laser is utilized for the ablation of any type of target material. It is easy to modify the morphology of fabricated metallic NPs by adjusting the laser wavelength, pulse duration, or laser fluence. Inert gas environment could also provide an additional condition to systematically vary the properties of metallic NPs [45]. For example, a good SERS performance of Ag NP film deposited by PLD was proved by Smyth et al. [46]. They investigated the SERS sensitivity of three different SERS substrates including pulsed laser-deposited Ag NP film (which was done in a high vacuum environment at room temperature), conventional silver colloids, and a commercial film (Klarite® substrate from Renishaw Diagnostics). The results showed that the SERS signals of both pulsed laser-deposited Ag NP film and conventional silver colloids were superior to that of commercial film. Moreover, pulsed laser-deposited Ag NP film offered much better reproducibility than conventional silver colloids.

Up to now, several review articles [23, 47–52] have displayed the various top-down, bottom-up, and combination technologies on the fabrication of SERS substrates. Nevertheless, PLD technique has been reported curtly in a few reviews. In this review, we have comprehensively summarized the detailed description of various SERS substrates fabricated by PLD technique. Firstly, we present the fundamental processes occurred in the ablation and deposition of metallic NPs in PLD technique. Secondly, many deposition process conditions of substrates using metallic NPs are considered. Lastly, the applications of SERS substrates fabricated by PLD in the environmental and biomedical analysis are described and the potential future trend in this area is outlined with the aim of stimulating the broader interest in developing PLD technique in the SERS substrates.

2 Fundamentals of pulsed laser deposition

Although the first experiment of PLD was carried out in the 1960s [53, 54], the extensive application in growing oxide film rapidly and efficiently was arisen in the late 1980s [55]. PLD technique has gradually developed over the past few decades. Today, it has been considered as one of the most popular techniques for depositing semiconductors [56, 57], metals [40–42], alloys [58, 59], and compounds [60, 61]. In addition, it could be applied to most of materials due to the high-power density near the focus of a pulsed laser ($> 10^6$ W/cm²).

Fig. 1 Schematic diagram of a PLD system

PLD technique presents many advantages in fabricating nanostructures. For example, benefiting from the intense energy of laser, almost all the materials could be deposited with the stoichiometry retained. The high instantaneous fluxes would lead to the high nucleation densities [62], which is beneficial for the formation of hot spots. Besides, since it could be operated at low temperature, the

temperature sensitive materials could also be manufactured with this technique. Remarkably, the composite films with complex composition could be deposited by using several targets as well, and their thickness and morphology could be easily controlled.

PLD is a physical vapor deposition process, which requires a vacuum system and a pulsed laser. As shown in Fig. 1, the construction of PLD experiment is relatively simple. A pulsed laser with high laser energy density concentrates onto a rotating target in a vacuum. The extreme energy of the focused beam vaporizes or ablates a small surface area of target material to create a plasma plume for film growth. The ejected plasma plume travels at an initial velocity $\geq 10^6$ cm/s that is formed with ions and atoms [63] through the vacuum chamber, causing a highly directional expansion perpendicular to the target. The ejected flux impinges on the surface of substrate, and the film deposition is fulfilled.

In contrast to its simplicity in construction, the mechanisms of PLD are rather convoluted. As shown in Fig. 2a, the PLD process is normally classified into four substages: (I) laser ablation and target vaporization, (II) plasma formation and interaction with the incident laser, (III) plasma expansion in vacuum, (IV) deposition at the substrate [64]. When

Fig. 2 **a** Schematic of the key elements of the PLD event. **b** Actual deposition process of PLD. **c** Schematic of film growth in a simple view of nucleation, island growth, coalescence

the laser is concentrated on the metal surface, part of the laser penetrates the metal target and gets absorbed. During the process of laser ablation, metal target starts to melt and evaporate since the vaporization temperature is acquired. Next, the plasma plume formed at the metal target surface further interacts with the incident laser. The effect of plasma plume on absorbing the incident laser is closely related to the distance between plasma and target surface. Strong absorption happens when the distance is close enough to allow high density of plasma plume to be built on the target. With the expansion of plasma plume, it retards the absorption of incoming radiation and the deposition rate of substrate turns to rely on the rate of plasma plume expansion. Therefore, it is usually deposited in vacuum or inert gas atmosphere to avoid the interaction between plasma plume and ambient molecules. Finally, the plasma plume reaches the substrate followed by the nucleation and growth of island. In the most simplified model, an atom meets the nearest neighbor atom to nucleate an island. The number of islands grows rapidly within the deposition of a few percent of a monolayer [65]. Then, the probability of atoms encountering another diffusion atom is reduced, whereas the probability of atoms bumps into the island is raised, causing the enlarged size of islands [66]. This leads to descending the number density of atoms while increasing the number density of step edge and islands [65]. For PLD, the number of nucleation sites hinges on the first laser pulse and the additional atoms coalesce the existing islands. The steps involved in the formation and growth of islands are shown in Fig. 2c. The first laser pulse results in the nucleation of islands at a characteristic distance that is greatly affected by the diffusion rates. Ideally, the size of island remains lower than this distance until the coalescence begins. New islands nucleate on the top of existing islands as soon as its size is comparable to or larger than the nucleation site spacing. Therefore, the ideal case of layer-by-layer growth is not accomplished [67].

Previous works [9, 68–71] have showed the excellent SERS effect in the gap between neighboring Au or Ag NPs, that is, hot spot. However, the SERS effect is weakened when the islands are cross-linked with each other resulting from the decreased numbers of hot spots. As a result, it is necessary to control the parameters in PLD technique and prepare the suitable SERS substrate in order to obtain a better SERS effect. The quality of SERS substrate deposited by PLD generally depends on the experimental conditions, including laser output parameters, ambient medium, substrate temperature, properties of PLD substrate, etc. With the appropriate laser wavelength and fluence, high-energy densities are absorbed by a small volume of material, leading to the stoichiometric transfer of ablated material from the target. In PLD process, some introduced background gas (e.g., oxygen) interacts with the ejected plume, causing the formation of complex compounds. The background gas (e.g.,

N_2 and Ar) could also be used to slow the plume expansion. Moreover, the substrate temperature influences the kinetic energy of metallic NPs and results in the formation of a highly ordered substrate structure [72]. Additionally, the morphology of PLD substrate could strongly affect the surface morphology of metallic NPs. Summarily, the convenient manipulation of sizes and morphologies of metallic NPs allow PLD technique to be an effective method for preparing SERS substrates.

3 Parameters effect in PLD technique for preparation of SERS substrates

It is well known that the Raman intensity is proportional to the square of the induced dipole moment that is a product of the incident electromagnetic field magnitude and the molecular polarizability, so it is reasonable to deduce two major enhancement mechanisms (i.e., electromagnetic mechanism and chemical mechanism). The latter involves in the charge transfer process [73]. The former is due to the interaction between incident or scattered light and SERS substrate and served as the most important mechanism in the understanding of SERS. When SPR is excited on the surface of metallic NPs, the electromagnetic fields would be greatly enhanced at the specific position around the metallic NPs.

Like the harmonic oscillator model, which is a classical physical model, the electrons in the metal could be coherently excited to oscillate by incident laser. Under the irradiation of incident light, electrons move toward the surface of metallic NPs, resulting in the separation of charges and generation of electric dipoles (Fig. 3). The electric field opposite to that of the light induced by the dipole makes the charges return to the equilibrium position and causes the resonance collectively [74–76]. The type that electrons oscillate around the surface of metallic NPs or nanoscale crevices is termed as localized SPR [10]. The oscillating

Fig. 3 Illustration of electric field and charge distribution at the surface of metallic NPs

amplitude could be determined by the extinction spectrum indirectly. According to Mie theory, the extinction spectrum, $E(\lambda)$, is given by Eq. (1) [74]:

$$E(\lambda) = \frac{24\pi^2 Na^3 \epsilon_{out}^{3/2}}{\lambda \ln(10)} \left[\frac{\epsilon_i(\lambda)}{(\epsilon_r(\lambda) + 2\epsilon_{out})^2 + \epsilon_i(\lambda)^2} \right] \quad (1)$$

where λ is the wavelength of incident light, N is the molecule surface density, a is the radius of a spherical nanoparticle, ϵ_{out} is the dielectric function of the surrounding medium, and ϵ_r and ϵ_i are the real and imaginary components of the metallic dielectric function, respectively.

Therefore, the wavelength and strength of the SPR are not only responsive to the dielectric function of both metallic NPs and surrounding medium, but also tunable by the surface morphology of substrate. Specific adjustment of the variables to change the surface morphology of metallic NPs assists SERS optimization because the strong amplification of local electromagnetic field is obtained by SPR, leading to the increase in SERS signal of analytes [77, 78]. PLD is a relatively simple and effective nanofabrication technique to immobilize metallic NPs upon a given substrate. Structure and optical property of the grown metallic NP film are strictly dependent on the deposition conditions. In this process, laser output parameters, gas medium, liquid medium, substrate temperature, and 3D substrate morphology significantly affect the sizes and morphologies of metallic NPs.

3.1 Effect of laser output parameters

Several laser output parameters such as laser pulse number, pulse duration, and mass per pulse must be taken into consideration to control the size and distribution of metallic NPs by PLD. A series of studies have been done to understand the effect of laser pulse number on the morphology of SERS substrates. Alonso et al. [79] initially realized the formation of Ag NPs by PLD without any ambient gas. They found that the condensation in the gas phase during the plume expansion was unnecessary for the formation of Ag NPs, and the surface morphology of Ag NPs arrays could be optimized by tuning the laser pulse number. The TEM images (Fig. 4a–d) showed that the size of Ag NPs was positively correlated to the laser pulse numbers. Because of the laser pulse number deposited was from 500 to 10,000, the mean Feret's diameters of isolated Ag NPs were in the range of 5 to 32 nm. Ag NPs turned to ellipsoidal and bean-like shapes. Then, the number of bean-like shapes was increased, and the islands started to be cross-linked with 15,000 laser shots. Additionally, AFM images suggested that the nanostructures had a columnar-like growth arising from the increasing height. The correlation SPR peak was red shifted from 425 to 737 nm, and the resonance band was widened as the pulse number was increased from 1000 to 15,000.

Furthermore, an important parameter of PLD process, namely the ablated mass per laser pulse (M_p), could obviously affect the structural properties of metallic NP films. Although the M_p depends on the fluence, the fluence is not

Fig. 4 TEM images of Ag NPs deposited in vacuum by: (a) 500, (b) 5000, (c) 10,000, and (d) 15,000 laser pulses from [79] with permission from Elsevier; SEM images of Ag deposited at 70 Pa in Ar by:

(e) 500, (f) 5000, (g) 10,000, and (h) 15,000 laser pulses from [83] with permission from Elsevier and [89] with permission from John Wiley and Sons, Inc

competent to determine the mass ablated by the laser pulse. At a fixed pulse energy density, the M_p could be affected by the irradiated area where the laser beam focuses on. Spadaro et al. [80] showed that the effect of M_p was controlled by the laser spot area on the structure of Ag NP films when the laser fluence was fixed. The morphologies were apparently distinguishable between two samples with different M_p . Isolated, nearly spherical Ag NPs were observed at a lower M_p , but the coalescent Ag islands were displayed at the higher M_p , resulting from the clustering of spherical Ag NPs. The SPR position and width of samples deposited at the same experimental conditions were determined by M_p . When the M_p was lower, the SPR peak showed an evident maximum at about 590 nm, while the broad maximum was detected around 820 nm at the higher M_p . The red shift of SPR and the increase of linewidths at the higher M_p pointed out the detuned optical transitions due to the dipole–dipole interactions.

The previous studies [81] suggested that a longer pulse laser ablation, like *ps* or *ns*, would suppress the formation of metallic NPs due to the effects of laser absorption, heating, and evaporation of the metal target. Unlike the *ps* or *ns* PLD, an *fs* laser pulse duration is shorter than the full ablation process and the ablated species do not interact with the laser pulse itself, thereby avoiding the complicated secondary laser interactions. Bonis et al. [82] investigated the characteristics of Ag film obtained by *ns* and *fs* ablation in PLD process. The atomic velocity distribution, excitation temperature, and electron density were evaluated to show the effect of plasma dynamics on the Ag film properties. Surprisingly, the features of Ag films like morphology, peak position, and widening of the SPR deposited by *fs* pulsed laser were similar to those of the films deposited by *ns* pulsed laser, which was different from the previous reports [79, 83]. Generally, the gas phase condensation occurred in the *ns* ablation could result in the formation of droplets. It is clearly different from the situation in the *fs* ablation, in which a new plume with a lower velocity than the primary plume is expanded [82]. They supposed that this similarity between *ns* and *fs* laser-induced plasmas was related to the Ag bulk properties.

Because of easily controllable tuning of SPR band, Ag–Au alloy films were considered by Verma et al. [84]. Unlike chemical methods, PLD technique is able to synthesize the uniformly mixed Ag–Au alloy film with various sizes within a single step without stabilizing agents or other extraneous chemical species [85]. The relationship between SPR response and Ag–Au alloy film grown under different process parameters (including the Ag–Au pulse ratio and total number of pulses) was studied further. The result depicted that the red shift of SPR band was accompanied with the decreasing Ag–Au pulse ratio and the increasing mass thickness which was under the control of pulse

number. Generally, the increase of mass thickness means the enlargement of metallic NP size [85]. In the present case, 20–50 nm size of Ag–Au alloy films boasted strong SERS response based on the size dependent near-fields [86, 87], in which the near-field enhancement was initially increased with increasing the size of metallic NPs and started to reduce after reaching a maximum, arising from the competition between nonradiative damping and radiative damping of the excited near-field. Moreover, SPR wavelength of the Ag rich alloy film and the pure Ag film closely corresponded to the laser excitation wavelength, resulting in the highest SERS response. Therefore, the fabrication of Ag–Au alloy film allowed the easy tuning of SPR band without changing the particles size, and the higher SERS intensity under different laser excitation wavelengths was obtained as a result of the alloy films with tunable SPR band.

3.2 Effect of gas medium

In the PLD process, a background gas is often introduced to provide a reactive species or reduce the kinetic energies of ablated species [44]. The expansion of laser ablation plume is strongly affected by the ambient gas pressure. To promote the formation of metallic NPs, an inert gas is adopted to confine the expansion of plume, which allows the sufficient time for nucleation.

For example, the performance of Ag NPs deposited in the Ar gas is not identical to that in the vacuum. Ossi et al. [83, 88, 89] reported a series of pulsed laser-deposited Ag NPs in the Ar. As shown in Fig. 4e–h, the mean size of Ag NPs is approximately the same under the different laser pulse numbers, while the number density of Ag NPs increases as a function of the number of laser pulses at a fixed Ar pressure of 70 Pa. This is supposed to be a typical model for the growth of metallic NPs at the high laser shot number and low buffer gas pressure.

Moreover, they [83, 88, 89] not only changed the pulse laser numbers, but also optimized the Ar pressure to figure out the highest SERS activity. Qualitative differences in the surface morphologies of Ag NPs deposited at the different Ar pressures were clearly visual. SEM images proved that the Ar pressure made a notable contribution to the morphology of Ag NPs. The Ag NPs grown at the high Ar pressure showed the isolated nearly spherical with no evidence of a coalescence phenomenon. At the low Ar pressure, the presence of Ag islands with smooth rounded edges indicated that they were caused by the coalescence of small nearly spherical particles. The SERS substrate formation mechanism was summarized into two steps: (I) the aggregation of Ag atoms induced by the Ar atmosphere caused the formation of Ag NPs; (II) the coalescence of Ag NPs led to the appearance of interconnected network of Ag islands [90]. In this process, the dimensions of Ag NPs were determined by the Ar

pressure, that is, the higher Ar pressure, the larger Ag NPs were formed [91]. In addition, at a constant Ar pressure, the number density of Ag NPs was increased with increasing the laser pulse number, resulting in the beginning of aggregation process. It might be the result of high resistance from the Ar environment to the plume motion [92].

The different morphologies of Ag films lead to the different optical properties. Bulk Ag showed an absorption peak in near-UV [93], while the SPR peak of isolated Ag NPs shifted to the visible region at about 390 nm [94]. It was clear that the plasmon position of deposited Ag films was red-shifted from 440 to 632 nm with decreasing Ar pressure and the full width at half maximum (FWHM) was broadened from 150 to 630 nm, indicating the non-visual aggregation of isolated spherical Ag NPs. The absorption spectra of Ag films deposited in the Ar gas at different pulse numbers were investigated as well. The SPR peaks were red-shifted and broadened when the number of laser shots was increased. This implied an effective aggregation process among Ag NPs with the collective optical behavior [89]. In this research, the highest SERS activity was attained at 70 Pa with 30,000 laser shots. The corresponding SEM and TEM images illustrated that the Ag NPs with appropriate sizes were closely arranged to form a dense network, arousing the red shift of SPR peak position and the largest FWHM. The enormous enhancement of SERS signal was attributed to the red shift of the coupled plasmon resonance and the increase of electromagnetic field at the junction between the two closely adjacent Ag NPs [95, 96].

In order to reduce the effect of gas on the plume, PLD is usually carried out in the vacuum or low-pressure gas. The formation of metallic NPs by PLD in the air is a complex and low efficient process as a result of re-deposition of the ablated material [97, 98]. It is reported that the growth of metallic NPs in the air was realized in the ablation plume due to the intensive interparticle collisions, while the metallic NPs deposited in the vacuum were created on the surface of PLD substrate [99]. Nedyalkov et al. [100] deposited Au NP film in the atmospheric pressure and air condition to understand this process. The red shift of SPR wavelength revealed that no significant oxidation or other chemicals generated in the air. Moreover, SERS spectra demonstrated that this Au NP film could be a cheap alternative for other SERS substrate deposited in the high vacuum conditions. Khan et al. [101] were also interested in using PLD at atmospheric pressure (which was named as APLD), generating strong collisional coupling of the ablated material with the ambient gas. Highly different morphological characteristics of Ag NP films made by a high repetition rate (HRR) vacuum PLD and HRR supersonic APLD were obtained. A substantial fraction of elongated Ag NPs was found in the film that was produced by vacuum PLD. By contrast, the Ag NP films made by HRR supersonic APLD were more

densely packed with more uniform size distribution. Then they found that the enhancement factor of HRR supersonic APLD film was about 30 times more sensitive than that of the PLD film prepared in the vacuum, which demonstrated the utility of APLD.

3.3 Influence of liquid medium

In principle, if the laser energy is not extremely attenuated and its density is still enough to perturb the material away from equilibrium state, the laser ablation could also occur in liquid [102]. The pulsed laser ablation in liquid (PLAL) attracts increasing attention since 1980s [103, 104]. Contemporarily, this technique is widely utilized for the nanostructure generation [105–108] because of its reliability and easy applicability without any contaminations. Desired metallic NPs could be obtained in one step due to the high energetic state of ablated species [109, 110]. Unlike conventional PLD technique, PLAL process does not need costly vacuum chambers and the products are easy to be collected from solutions.

Typical experimental setup for PLAL is laid out in Fig. 5a, which is composed of a pulsed laser, a set of focusing optics, and a vessel containing the metal target. The metal target might rotate to avoid a deep ablation trace, and the magnetic stirred liquid solution is adopted to achieve a better ablation and a higher reproducibility [111, 112]. Figure 5b–e schematically depict the evolution of the laser ablated plasma in liquid phase. Similar to the conventional PLD, the plasma plume is produced when the incident laser pulse irradiates the solid target through the liquid. Compared with ablation in vacuum or in gas, the expansion of plasma is strongly confined by liquid, and the thermodynamic and kinetic properties of plasma are able to be changed significantly. Different kinds of chemical reactions are yielded in the plasma and the interface between liquid and plasma provides the opportunities for the formation of new materials by the combination of target with liquid. Finally, the plasma cools down and condenses in the confining liquid and the metallic NPs start to nucleate, grow, and coalesce.

Up to now, a large number of literature [105–108, 113–115] about metallic NPs formation by PLAL and the effect of solvent types, solutes, stabilizers, and laser output parameters have been studied. Size and aggregation state of metallic NPs are controlled by the addition of charge-changing chemicals such as surfactants. Kadhim et al. [116] prepared Au NPs by PLAL in an aqueous solution of sodium dodecyl sulfate (SDS). They found that the size of Au NPs signified a negative correlation with the concentration of SDS. The concentration-dependent size relationship was explained by a dynamic formation mechanism [117, 118]. The rapid nucleation of Au NPs and further growth with supplying Au atoms were terminated when the surfaces were

Fig. 5 **a** Principal components of the experimental set-up for PLAL. The evolution of the laser ablated plasma in liquid. **b** Generation of the plasma due to the irradiation of the laser pulse on the metal target. **c** Expansion of the plasma plume in liquid. **d** Different kinds of chemical reactions taking place inside the plasma and liquid, and the interface between the plasma and liquid. The asterisk denotes the excited and/or ionized states. **e** Condensations and growth of the plasma plume in liquid

covered with the surfactant molecules. Alluhaybi et al. [119] demonstrated that the size of Au NPs could be altered by changing the laser energy and laser pulse repetition rate. The mean size of Ag NPs was decreased by increasing laser energy originated from the disintegration of large Au NPs at high effective transient temperature. Similarly, high repetition rates of the laser pulses with high effective transient temperature could also fracture large Au NPs into small ones.

Giorgetti et al. [105] investigated the Ag NPs acquired with different laser pulse durations by PLAL. They found that the Ag NPs attained with *ps* pulses in the pure water were more dispersible and spherical with a relatively small size than that achieved by *ns* pulses. Compared with theoretical spectra calculated with the Mie theory, both the SPR spectra of Ag NPs with *ps* and *ns* pulses were red shifted and the SPR band of *ps* pulses was wider than that of *ns* pulses. The formation of Ag_2O layer surrounding was proved to make a great contribution to the red shift of SPR peak position for Ag NPs [120]. Owing to the presence of Ag_2O , the ablation-prepared Ag NPs could keep stable for months but displayed a weakened SERS activity. However, the addition of chloride ions into Ag NPs was able to promote an efficient SERS signal [121].

Vinod et al. [106] synthesized Ag@Au core-shell NPs by ablating Au target in the Ag colloidal suspension using low energy laser pulses. Au target absorbed the energy of pulse laser and Au atoms jetted on the spherical surface of the fragmented core Ag NPs [122]. The thickness of Au shell was controlled by changing the ablating duration of Au target. Two distinct SPR bands appeared when the Au shell was thin or no dense, while peculiar SPR band was observed due to the dense and thick Au shell. The interparticle distance between Ag@Au core-shell structures was decreased with increasing the number of Ag@Au core-shell NPs per unit volume, leading to the formation of “hot spots.” Therefore,

SERS spectra strongly depended on the shape and interparticle distance of Ag@Au core-shells by controlling the ablation duration and colloid concentration. In addition, the pure metal colloidal solutions without stabilizer or polymer are preferred as an optimal SERS substrate for biological applications.

Besides, AgCl NPs were confirmed to be directly generated by pulsed laser ablation of Ag target in the sodium chloride (NaCl) solution [123]. The AgCl NPs were cubes with the sizes in several hundred nanometers. Cl^- ions could not only promote the generation of cubic morphology [124], but also prevent the aggregation of Ag nanoclusters [121]. Furthermore, the AgCl NPs possessed a high decontamination capability of gram-negative bacteria and obvious SERS activity as well.

3.4 Effect of substrate temperature

The ambient medium can control the morphology of grown substrates without changing their stoichiometry of the target material. The change of substrate temperature also affects the properties of formed metallic NPs; particularly, the size and number density of metallic NPs can be adjusted. Budner et al. [72] reported the effect of parameters in the PLD process on the morphology and SERS enhancement property of Ag NP films. The effective SERS enhancement was reached by altering the laser fluence, number of laser pulses, and substrate temperature. Especially, the substrate temperature played a key role in the formation of Ag NP films. The thermal energy increased the kinetic energy of Ag atoms, causing a highly ordered structure. The best SERS performance was noted at the coalesced Ag NPs with decrease in the dimensions of gaps.

Kamakshi et al. [125] showed the effect of substrate temperature on the morphology of Ag NPs, which were grown by means of the Volmer-Weber growth mode [126] at the

substrate temperature ranging from 100 to 500 °C and the maximum size of Ag NPs appeared at substrate temperature of 300 °C. They noticed that the Ag NPs size and substrate temperature followed a non-linear relationship, meaning that there was more than one competing growth mechanism. On the one hand, the surface mobility of Ag NPs was raised with the increase of substrate temperature, resulting in the increase in the Ag NPs size. On the other hand, the increase of substrate temperature aroused the decrease in the density of ambient gas and collisional rate inside the plasma, declining the size of Ag NPs [127]. Besides, at high substrate temperature of 600 °C, the average size and interparticle distance of Ag NPs were larger than that deposited in the substrate temperature range of 100–500 °C, which suggested that the growth mechanism of Ag NPs was changed. It was reported that appropriate size and interparticle spacing of Ag NPs played key roles on the SPR and SERS effect [128]. As expected, the highest SERS activity was reached at the deposition temperature of 300 °C.

3.5 Effect of 3D substrates

Recently, three-dimensional (3D) nanostructures with high specific surface area are regarded as one of the solutions to promote giant enhancement of SERS [68, 69, 129, 130]. Furthermore, the local electromagnetic field could be facilitated by additional charge transfer between the metallic NPs and the adjacent 3D nanostructures [68, 69]. These evidences have prompted the study of composite substrates formed by 3D nanostructures and metallic NPs.

D'Andrea et al. [131] fabricated the 3D Si nanowires with Ag NPs coating and investigated the effect of size and density for Ag NPs and length of 3D Si nanowires on the SERS performance. Their experimental results are depicted in Fig. 6A. It is observed that the Si nanowire array was still intact after PLD deposition, illustrating that the deposition process had no effect on the 3D Si nanowires. It was seen that the Ag NPs size was increased from the bottom to the top section of the 3D Si nanowires, whereas the distance between Ag NPs was decreased. At the bottom and central sections of the 3D Si nanowires with 1.7 µm long, the mean radii of the separated Ag NPs were estimated to be 5.5 ± 1.1 and 7.5 ± 2.7 nm, respectively. On the top of 3D Si nanowires, the mean radius was 8.3 ± 2.7 nm and the Ag NPs started to agglomerate. The coalesced Ag NPs with larger size and smaller interparticle spacing were formed on the top section relative to the bottom and central section due to the greater possibility for the top of 3D Si nanowires to catch the incident Ag NPs. In addition, the mean radius was decreased to 7.3 ± 4.7 nm on the top of 3D Si nanowires with 3.4 µm long. Accordingly, it was easy to change the size distribution and the interparticle distance by changing the

length of nanowires and the laser pulse number for increasing the SERS efficiency.

As the growing interest in the fabrication of flexible plasmonic substrates, Rebollar et al. [132] fabricated and characterized a range of Au-coated 3D nanostructured polymer films to explore the relationship between the Au-coated 3D nanostructured polymer films and the SERS signal (Fig. 6B). The polymer films were irradiated under different laser systems to generate the periodic surface of 3D structures with different periods, depths, and roughness. After coated with a thin layer of Au NPs by PLD, the 3D nanostructured polymer films exhibited no significant change in the period and height. Owing to the same morphology of Au layer on both flat and 3D nanostructured polymer films, they supposed that the SERS signal was more related to the period, depth, and roughness of polymer nanostructures. Particularly, the greater roughness, longer period, and deeper depth of 3D nanostructures, the higher SERS enhancement was obtained, attributing to the larger effective area of interaction between substrate and analyte.

Wang's group originally fabricated Ag NPs/Ni(OH)₂ nanosheet arrays to study the effect of 3D Ni(OH)₂ nanosheet arrays on the SERS intensity (Fig. 6C) [68]. These 3D Ni(OH)₂ nanosheets demonstrated a highly ordered and porous nanoarray structure, providing a large specific surface area for the deposition of Ag NPs. As the deposition time was varied from 10 to 60 min, the diameter of Ag NPs went up from 55 to 212 nm. When depositing for 50 min, SERS enhancement reached the maximum because of the optimal interparticle gap and particle size. Moreover, as shown in Fig. 6C, the red shift of SPR band for Ag NPs/Ni(OH)₂ nanosheet arrays verified the decrease in the electron density of Ag NPs. The charge transfer from Ag NPs to 3D Ni(OH)₂ was facilitated by the stacking of positively charged Ni(OH)₂ layers [133]. Finally, the positive charges were left on the Ag NPs, while 3D Ni(OH)₂ nanosheets were negatively charged and the charge density became higher at the adjacent surface of Ag NPs and Ni(OH)₂ nanosheets. The surface plasmon between 3D Ni(OH)₂ and Ag NPs would be strongly excited, devoting to the increase of SERS enhancement.

This group was further investigated the SERS effect of Ag-coated 3D nanostructure arrays (Fig. 6D) [69]. The naked 3D Ti(OH)₄ and TiO₂ nanobelt arrays with similar morphology feature were studied. The distribution of Ag NPs on the 3D Ti(OH)₄ nanobelt arrays was more uniform than that of Ag-decorated 3D TiO₂ nanobelt arrays. According to the XPS spectra, Ag₂O might be formed arising from the reaction between Ti(OH)₄ and Ag on the surface of 3D Ti(OH)₄ nanobelt arrays with the high kinetic energy supplied by PLD [63]. The Ag₂O could serve as a nucleation center to grow the ordered Ag NPs. The red shift of SPR band of both Ag decorated 3D Ti(OH)₄ and TiO₂

Fig. 6 **A** Top is a schematic of 3D Si nanowires fabrication. **(a–d)** SEM images of different views of Si nanowires and **e** TEM image of the bottom region of the Ag coated 3D Si nanowires. **(f–h)** Corresponding statistical analysis of the Ag NPs radii. From [131] with permission from IOP Publishing. **B** AFM height images of uncoated and Au-coated 3D nanostructured polymer films from [132] with permission from John Wiley and Sons, Inc. **C** **(a)** Schematic of the fabrication of Ag/3D Ni(OH)₂ nanosheet arrays. SEM images of **(b)** 3D Ni(OH)₂ nanosheet arrays and **(c)** Ag-coated 3D Ni(OH)₂ nanosheet

arrays. **(d)** UV–Vis spectra of Ag NPs, 3D Ni(OH)₂ nanosheet arrays and optimized Ag-coated 3D Ni(OH)₂ nanosheet arrays. **(e)** Schematic of charge transfer between 3D Ni(OH)₂ nanosheet and Ag NPs. From [68] with permission from Elsevier. **D** Schematic of fabrication processes of Ag-decorated 3D Ti(OH)₄ nanobelt arrays and 3D TiO₂ nanobelt arrays from [69] with permission from Elsevier. The insets are the SEM and TEM images of naked and Ag-coated 3D Ti(OH)₄ nanobelt arrays and 3D TiO₂ nanobelt arrays, respectively

nanobelt arrays in the UV–Vis spectra proved the charge transfer from Ag NPs to 3D nanobelt arrays. The more red shifts for Ag decorated 3D Ti(OH)₄ nanobelt arrays than that for Ag decorated 3D TiO₂ nanobelt arrays implied a higher charge density and a stronger local electromagnetic field, leading to an increased SERS effect. Additionally, the SPR of Ag decorated 3D Ti(OH)₄ nanobelt arrays was closer to the wavelength of the incident light than that of Ag-coated 3D TiO₂ nanobelt arrays, which also made contribution to the enhancement of SERS signals [12].

In summary, among these parameters, the laser output parameters are the simplest one to adjust the size and morphology of SERS substrates. Under the high-energy laser ablation, the metallic NPs jet with a very high initial velocity, which is easy to form the irregular nanoislands. The introduction of gas medium effectively alleviates the formation of nanoislands, especially in the deposition process of Au NPs. Moreover, the SERS substrate fabricated in the

liquid medium exists in the form of metallic NP colloidal solution. Hence, its SERS sensitivity is high since the molecules to be detected could easily combine with metallic NPs in the colloidal solution. The substrate temperature has a certain effect on the morphology of metallic NPs. In addition, the morphology of 3D substrate is changeable and its large specific surface area could increase the density of hot spots. More and more research reports on the 3D substrate suggest a great potential of 3D substrate in the SERS applications.

4 Applications

SERS has the wide applications in the field of food safety [18, 134, 135], environmental analysis [136–138], drug and explosive monitoring [139–142], biomedical analysis [143–146], etc. However, the application of SERS substrates

prepared by PLD technique mostly focuses on the environmental and biomedical analysis fields.

4.1 Environmental analysis

As a result of irrational human activities, the potential hazards in environments have been the center of gravity in the environmental detection field and analytical chemistry. Thus, it is very important to develop the corresponding analytical techniques for monitoring the trace pollutants. A number of analytical techniques, including spectrophotometry [147–149], fluorescence [150–152], chromatography [153, 154], and electrochemical methods [155, 156] have been reported. However, the complicated instruments, complex sampling processes, and insufficient recognition capability restrict their applications. SERS technique has attracted wide attention for its simple, and fast process, as well as simultaneously efficient detection of multiple contaminant classes.

There are several review papers that have already mentioned the application of SERS technique in the environmental analysis [157–161]. The analysis target of SERS mostly included organic pollutant such as dyes [68, 69], pesticides [136–138], and polycyclic aromatic hydrocarbon (PAHs) [162–166]. Some of the analytes with strong affinity toward a substrate could easily realize the SERS detection. Dye molecules, like rhodamine 6G (R6G) and crystal violet (CV), are representative SERS analytes that could be efficiently adsorbed on the Au and Ag surfaces. Unsurprisingly, most of literature referring to Sect. 3 used R6G as a probe molecule to verify the SERS effect of metallic NP substrates. Moreover, fungicide Dithane DG [167], neonicotinoid insecticide thiacloprid [168], and benzenethiol [132] with special functional groups containing sulfur or nitrogen that usually applied in the agriculture or industry could be easily detected.

However, the analytes with low affinity to the substrate are still challenges to be directly detected by SERS. For instance, PAHs are highly carcinogenic and bioaccumulative [169, 170], and they could accumulate to the harmful concentrations from very low levels. Unfortunately, the apolar nature of aromatic rings causes a low affinity to the metal surface, which limits the SERS application in the PAH detection. To overcome this limitation, SERS substrates were often modified by the thiol [163, 171], humic acid [162], calixarene [172, 173], cyclodextrin [164], and other molecules like carbon nanotubes (CNTs) [174]. The basic principles of above modification methods are similar, that is, they employed the interaction between PAH molecules and functional groups/molecules to adsorb PAHs on the surface of SERS substrate for SERS detection. However, thiol, calixarenes, and cyclodextrin have strong absorption peaks

in the range of characteristic peaks of PAHs, which is easy to interfere with the detection of PAHs.

Wang's group used a simple and rapid method to prepare Al_2O_3 nanosheet array structure [175] and utilized PLD technique to composite Ag NPs with Al_2O_3 , as shown in Fig. 7A. Figure 7B shows that the Ag NPs were uniformly distributed on the surface of Al_2O_3 nanosheet array with a mean diameter of 20 nm. Because of the porous structure of Al_2O_3 nanosheet array, the number density of hot spots was increased, which was beneficial for the enhancement of SERS effect. The SERS performance of Ag/ Al_2O_3 nanosheet array and Ag/Al foil are investigated in Fig. 7C by using the Al foil without Ag NPs as a comparison. Raman spectrum of R6G affirmed the excellent SERS performance of Ag/ Al_2O_3 nanosheet array. Pyrene was chosen as a molecular model because it was recognized as a key indicator of PAHs and occurred in all mixtures of PAHs. As shown in Fig. 7D, the limit for the detection concentration of pyrene whose absorption peaks were located at the blue region achieved based on the SERS results is evaluated to be ≤ 1 mM. Compared with other modification methods as mentioned above [164, 171–173], the Ag/ Al_2O_3 nanosheet array substrate depicted no obvious characteristic peak with no interference of PAH detection. The promising experimental results were acquired and demonstrated a relatively cheap and easy fabrication strategy of SERS active substrates by PLD for trace analysis of harmful pollutants.

CNTs were utilized to enrich and extract the pollutants because of the high specific surface area and π - π stack of non-covalent bonds [176–178]. Wang's group decorated CNTs that were prepared by the chemical vapor deposition (CVD) [179] with Ag NPs (Ag/CNTs) by the PLD technique. The fabrication process of Ag/CNTs is listed in Fig. 8A.

SEM images of CNTs and Ag/CNTs obtained from Ni-Ag catalyst are shown in Fig. 8B. The generated CNTs grew perpendicular to the substrate and formed an intersecting network structure, providing a larger specific surface area. Ag NPs with similar size were uniformly deposited on CNTs. All the characteristic peaks of pyrene were detected from the SERS spectrum (Fig. 8C). However, the differences of peak position between SERS spectra and Raman spectra for pyrene were speculated due to the different orientations of the PAH molecules relative to the substrate surface [163]. Although the SERS effect could be further optimized, the results suggested that the Ag/CNTs were a promising substrate in the optical sensor devices for detecting the PAHs.

4.2 Biomedical analysis

To comprehensively understand the biological process and early diagnose the complicated disease, the sensitive and specific methods are sought for the biomedical analysis.

Fig. 7 **A** Fabrication processes of Ag/Al₂O₃ nanosheet arrays. **B** SEM image of Ag/Al₂O₃ nanosheet arrays. **C** SERS spectra of 10^{−6} M R6G adsorbed on **(a)** Ag/Al₂O₃ nanosheet arrays and **(b)** silver-decorated Al foil. Raman spectrum of 10^{−2} M R6G adsorbed on **(c)** Al foil. **D**

SERS spectra of pyrene molecules **(a–d)** in different concentrations on Ag/Al₂O₃ nanosheet arrays, **(e)** Raman spectra of Ag/Al₂O₃ nanosheet arrays, and **(f)** solid pyrene

The changes of biomolecules like proteins, nucleic acids, and metabolites suggest the presence and course of disease [180–183]. Therefore, to monitor these changes is conducive to further understand the life process and disease mechanisms. Various techniques, including nuclear magnetic resonance, mass spectroscopy, electrochemistry, fluorescence microscopy, electron microscope, etc., are exploited for biomedical research [184–188]. With the discovery of SERS phenomena, SERS-based investigation in biomedical application is fascinated originating from its excellent molecular selectivity, high sensitivity, intense signal, and great precision [189]. Considerable investigations have been taken to extend the multi-functional SERS technology to realize the detection of tissues, cells (apoptosis, secretion, cellular stress, cellular cycle) and biomolecules (proteins, enzymes, nucleic acid), tumor makers, and metabolic products in vitro and in vivo, especially

for the detection of problems in the complex biological compositions [51, 190–192].

A feasible SERS application of in vivo analysis is the blood glucose quantification. The inconstant monitoring of blood glucose is potentially dangerous for patients. To overcome this obstacle, a direct sensing of glucose at physiologically relevant concentrations with SERS on the Au film-over-nanosphere (AuFON) substrates functionalized with bisboronic acid receptors was developed to resolve the glucose in the high background of fructose and accurately detect the glucose in the 1–10 mM range, which was promising in the SERS-based in vivo glucose monitoring sensors [193]. Recent progress by applying SERS substrate in lateral flow immunoassay (LFIA) strip instead of colloidal gold as signal reporters is valuable. SERS-based LFIA strips were introduced to detect trace amount of influenza H1N1 virus and human adenovirus (HAdV) with a value of 50 and 10

Fig. 8 **A** Fabrication processes of Ag/CNTs. **B** (a) FESEM cross-sectional image of CNTs grown with Ag–Ni catalyst. (b) SEM images of Ag/CNTs grown by PLD. **C** (a) Raman spectrum of solid pyrene and (b) SERS spec-

tra of 1 mM pyrene on Ag/CNTs. Absorption peaks of pyrene are located at the blue region

pfu mL⁻¹, respectively, which was 2000 times sensitive than the standard colloidal gold strip method [194]. Based on this, a pioneering study magnified that the trace spike protein (S protein) of SARS-CoV-2 in human saliva was detected by SERS biosensors with an extremely low concentrations of 10×10^{-9} g mL⁻¹, exhibiting the significant differences from Raman spectra. These results represented a facile, rapid, and generalizable strategy for the future investigation of SARS-CoV-2 by SERS-based detection [195].

The detection of complex molecular systems such as proteins is usually time-consuming and expensive. Common detection of proteins in clinical applications is not practical enough in medical emergency. Agarwal et al. [196] showed the fast response and low-cost SERS method obtained with Au NP substrates deposited by PLD technique (Fig. 9A).

Colloids systems were more popular for SERS detection, which was possible to probe the protein from all directions [197]. However, the interaction was easily uncontrollable between protein and metallic NPs, resulting in the poor molecular-structure sensitivity and scarcely reproducible. The SERS substrates obtained by PLD technique were more controllable, which made it possible to reveal the fingerprint of protein structure. The demonstration of binding for specific amino acid residues to the Au NPs indicated that the SERS enabled label-free detection of a protein.

Breast cancer has a high morbidity rates in female malignant tumor and is the major cause of cancer mortality in Chinese women. HER2, a kind of the human epidermal growth factor receptor, could be detected in the different biosensors for breast cancer diagnosis [199]. In order to simply

Fig. 9 **A** Left is a schematic of lysozyme lying flat on the Au substrate. Right is SERS spectra of lysozyme on Au substrate (a, red) and Ag colloid (b, green) from [196] with permission from Springer Verlag. **B** Left is a schematic of formation of a functionalized silver

nanostructured plate (FSNP) with a breast cancer cell attached on. Right is (a) a SERS spectrum of SKBR3 attached on a FSNP and a Raman spectrum of SKBR3 attached on a flat substrate of Au (111) from [198] with permission from MDPI

and directly detect the breast cancer, Téllez-Plancarte et al. [198] immobilized recombinant humanized anti-HER2 monoclonal antibody (trastuzumab) on an Ag nanoplate made by PLD technique and investigated its SERS activity (Fig. 9B). The number of laser pulse was adjusted to find the best experimental conditions that provided the highest SERS effect. A clean Au (111) substrate and the specially prepared Ag nanoplates with high affinity for HER2 were analyzed. Compared with the Au nanoplates, the SERS spectra of Ag nanoplates provided higher peak intensity, better peak definition, and higher S/N ratio. The remarkable peaks at 1002 and 1030 cm⁻¹ belonged to the phenylalanine symmetric ring breathing mode and in-plane C–H bend mode, respectively [200]. They proposed that the phenylalanine could be used as a label to identify the breast cancer cells with HER2 expression and monitored by SERS.

SERS has received extensive attention in the environmental and biomedical analysis thanks to its capability of high sensitivity and rapidness during the detection process. However, many of the experimental results remain at the

stage of laboratory research, and it still takes a long way to apply these results to the real life and production. For example, PLD technique is a small-scale production technique in the laboratory which may not fulfill the industrial demands for large amounts. In addition, even though it is possible to detect multiple pollutants simultaneously, the molecules with similar structure are not easy to be identified. Furthermore, the design of human-friendly SERS substrates is still the focus of future research to realize the in vivo detection.

5 Summary and outlook

The design and fabrication of SERS substrates with suitable morphology is the primary issue for SERS applications. In this review, we have outlined a variety of SERS-active substrates manufactured by PLD technique. Generally, PLD is utilized to synthesize conventional films, especially oxide-based compounds. However, recent

studies provide the strong support for broadening its application scope from traditional film to a new platform for SERS-active substrates. The PLD deposited SERS substrates exhibit the high SERS activity, high controllability, good adhesion, and great reproducibility resulting from the controllable growth conditions. However, there also exist some important challenges. For example, the morphology of SERS substrates prepared by PLD technique is relatively simple and limited to metallic NPs or films; the distribution of metallic NPs is random, which is easy to form the metallic islands, causing a weakened SERS effect. Thus, further research is necessary for solving or improving these problems. The manufacture of composites formed by 3D nanostructures and metallic NPs might be a feasible solution. In fact, metal oxides, carbon, and silicon materials with different morphologies are preliminarily used in the SERS substrates because of their intrinsic properties like magnetism, porosity, and photosensitivity and controllable preparation process. Furthermore, the electrochemical methods might be used to assist the growth of metallic NPs deposited by PLD. The growth process of NPs could be finely regulated to reduce the production of metallic islands. Moreover, the quantitative study on charge transfer between 3D nanostructures and noble NPs should be further carried out for the enhancement of SERS effect. In addition, it is still a challenge for PLD to satisfy the industrial demands with large amounts. The large-area PLD technique is a requisite prerequisite for SERS substrate to move from laboratorial fundamental research to practical application. However, conventional PLD grown sample with good uniformity is limited to a small area of 10×10 mm. Laser energy, repetition rate, deposition time, and gas pressure would be adjusted to realize the large-area deposition. High-temperature superconductor layer with length more than 600 m has been deposited by multi-beam pulsed laser deposition technique [201]. This method could be extended to the deposition of SERS substrates in the future. As a conclusion, this review paves the way to an extensive use of PLD technique as a reliable and efficient manufacture technology for SERS substrate. It is believed that the study and development of SERS substrates produced by PLD would make SERS technology a facile and indispensable tool in the future.

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Declarations

Conflict of interest The authors declare no competing interests.

References

1. Raman CV, Krishnan KS (1928) A new type of secondary radiation. *Nature* 121:501–502
2. Fleischmann M, Hendra P, McQuillan A (1974) Raman spectra of pyridine adsorbed at a silver electrode. *Chem Phys Lett* 26(2):163–166
3. Jeanmaire DL, Van Duyne RP (1977) Surface Raman spectro-electrochemistry: Part I Heterocyclic aromatic and aliphatic amines adsorbed on the anodized silver electrode. *J Electroanal Chem Interfacial Electrochem* 84(1):1–20
4. Albrecht MG, Creighton JA (1977) Anomalous intense Raman spectra of pyridine at a silver electrode. *J Am Chem Soc* 99(15):5215–5217
5. Cao Z, He P, Huang T, Yang S, Han S, Wang X, Ding G (2020) Plasmonic coupling of AgNPs near graphene edges: a cross-section strategy for high-performance SERS sensing. *Chem Mater* 32(9):3813–3822
6. Chen R, Jensen L (2020) Interpreting the chemical mechanism in SERS using a Raman bond model. *J Chem Phys* 152(2):024126
7. Jimenez de Aberasturi D, Henriksen-Lacey M, Litti L, Langer J, Liz-Marzán LM (2020) Using SERS tags to image the three-dimensional structure of complex cell models. *Adv Funct Mater* 30(14):1909655
8. Pannico M, Musto P (2021) SERS spectroscopy for the therapeutic drug monitoring of the anticancer drug 6-mercaptopurine: molecular and kinetic studies. *Appl Surf Sci* 539:148225
9. Nie S, Emory SR (1997) Probing single molecules and single nanoparticles by surface-enhanced Raman scattering. *Science* 275(5303):1102–1106
10. Willets KA, Van Duyne RP (2007) Localized surface plasmon resonance spectroscopy and sensing. *Annu Rev Phys Chem* 58:267–297
11. Lin X-M, Cui Y, Xu Y-H, Ren B, Tian Z-Q (2009) Surface-enhanced Raman spectroscopy: substrate-related issues. *Anal Bioanal Chem* 394(7):1729–1745
12. Nikoobakht B, El-Sayed MA (2003) Surface-enhanced Raman scattering studies on aggregated gold nanorods. *J Phys Chem A* 107(18):3372–3378
13. Jain B, Hashmi A, Sanwaria S, Singh AK, Susan MABH, Singh A (2020) Zinc oxide nanoparticle incorporated on graphene oxide: an efficient and stable photocatalyst for water treatment through the Fenton process. *Adv Compos Hybrid Mater* 3(2):231–242
14. Chowdhury P, Roy B, Mukherjee N, Mukherjee S, Joardar N, Mondal MK, Roy D, Sinha Babu SP (2018) Chitosan biopolymer functionalized gold nanoparticles with controlled cytotoxicity and improved antifilarial efficacy. *Adv Compos Hybrid Mater* 1(3):577–590
15. Khan SH, Pathak B, Fulekar MH (2020) A study on the influence of metal (Fe Bi and Ag) doping on structural optical and antimicrobial activity of ZnO nanostructures. *Adv Compos Hybrid Mater* 3(4):551–569
16. Kaur D, Bagga V, Behera N, Thakral B, Asija A, Kaur J, Kaur S (2019) SnSe/SnO₂ nanocomposites: novel material for photocatalytic degradation of industrial waste dyes. *Adv Compos Hybrid Mater* 2(4):763–776
17. Guo J, Chen Z, Abdul W, Kong J, Khan MA, Young DP, Zhu J, Guo Z (2021) Tunable positive magnetoresistance of magnetic polyaniline nanocomposites. *Adv Compos Hybrid Mater*
18. Pan T-T, Sun D-W, Paliwal J, Pu H, Wei Q (2018) New method for accurate determination of polyphenol oxidase activity based on reduction in SERS intensity of catechol. *J Agric Food Chem* 66(42):11180–11187

19. Zhou Y, Ding H, Liu J, LaChance AM, Xiao M, Meng Y, Sun L (2019) Gold nanoparticles immobilized on single-layer α -zirconium phosphate nanosheets as a highly effective heterogeneous catalyst. *Adv Compos Hybrid Mater* 2(3):520–529
20. Chen R, Bao J, Yan Z, Huang X, Yun J, Zeng X, Chen J (2019) Preparation of transparent dispersions with monodispersed Ag nanoparticles for TiO₂ photoelectrode materials with excellent photovoltaic performance. *Eng Sci* 8:54–65
21. Li N, Zhang F, Wang H, Hou S (2019) Catalytic degradation of 4-nitrophenol in polluted water by three-dimensional gold nanoparticles/reduced graphene oxide microspheres. *Eng Sci* 7:72–79
22. Kneipp K, Wang Y, Kneipp H, Perelman LT, Itzkan I, Dasari RR, Feld MS (1997) Single molecule detection using surface-enhanced Raman scattering (SERS). *Phys Rev Lett* 78(9):1667
23. Kleinman SL, Frontiera RR, Henry AI, Dieringer JA, Van Duyne RP (2013) Creating characterizing and controlling chemistry with SERS hot spots. *Phys Chem Chem Phys* 15(1):21–36
24. Zhong L-B, Yin J, Zheng Y-M, Liu Q, Cheng X-X, Luo F-H (2014) Self-assembly of Au nanoparticles on PMMA template as flexible transparent and highly active SERS substrates. *Anal Chem* 86(13):6262–6267
25. Zhang L, Dai L, Rong Y, Liu Z, Tong D, Huang Y, Chen T (2015) Light-triggered reversible self-assembly of gold nanoparticle oligomers for tunable SERS. *Langmuir* 31(3):1164–1171
26. Chen R, Shi J, Liu C, Li J, Cao S (2021) In situ self-assembly of gold nanorods with thermal-responsive microgel for multi-synergistic remote drug delivery. *Adv Compos Hybrid Mater*
27. Freeman RG, Grabar KC, Allison KJ, Bright RM, Davis JA, Guthrie AP, Hommer MB, Jackson MA, Smith PC, Walter DG (1995) Self-assembled metal colloid monolayers: an approach to SERS substrates. *Science* 267(5204):1629–1632
28. Dhand C, Dwivedi N, Loh XJ, Ying ANJ, Verma NK, Beuerman RW, Lakshminarayanan R, Ramakrishna S (2015) Methods and strategies for the synthesis of diverse nanoparticles and their applications: a comprehensive overview. *Rsc Adv* 5(127):105003–105037
29. Ul-Islam MU, Ali J, Khan W, Haider A, Shah N, Ahmad MW, Ullah MW, Yang G (2019) Fast 4-nitrophenol reduction using gelatin hydrogel containing silver nanoparticles. *Eng Sci* 8:19–24
30. Yu J, Sun L (2019) Facile one-pot synthesis of silver nanoparticles supported on α -zirconium phosphate single-layer nanosheets. *ES Mater Manuf* 5:24–28
31. Hao H, Li H, Wang S, Cheng Z, Fang Y (2020) Epitaxial growth of Ag-Cu bimetallic nanoparticles via thermal evaporation deposition. *Appl Surf Sci* 505:143871
32. Syu W-L, Lin Y-H, Paliwal A, Wang K-S, Liu T-Y (2018) Highly sensitive and reproducible SERS substrates of bilayer Au and Ag nano-island arrays by thermal evaporation deposition. *Surf Coat Technol* 350:823–830
33. Zhang H, Liu M, Zhou F, Liu D, Liu G, Duan G, Cai W, Li Y (2015) Physical deposition improved SERS stability of morphology controlled periodic micro/nanostructured arrays based on colloidal templates. *Small* 11(7):844–853
34. López-Lorente AI, Picca RA, Izquierdo J, Kranz C, Mizaikoff B, Di Franco C, Cárdenas S, Cioffi N, Palazzo G, Valentini A (2018) Ion beam sputtering deposition of silver nanoparticles and TiO_x/ZnO nanocomposites for use in surface enhanced vibrational spectroscopy (SERS and SEIRAS). *Microchim Acta* 185(2):153
35. Görgün N, Özer Ç, Polat K (2019) A new catalyst material from electrospun PVDF-HFP nanofibers by using magnetron-sputter coating for the treatment of dye-polluted waters. *Adv Compos Hybrid Mater* 2(3):423–430
36. Kala PV, Srinivasarao K (2019) Structural and luminescence studies on TiO₂-MoO₃ thin films. *Adv Compos Hybrid Mater* 2(4):735–742
37. Petti L, Capasso R, Rippa M, Pannico M, La Manna P, Peluso G, Calarco A, Bobeico E, Musto P (2016) A plasmonic nanostructure fabricated by electron beam lithography as a sensitive and highly homogeneous SERS substrate for bio-sensing applications. *Vib Spectrosc* 82:22–30
38. Durmanov NN, Guliev RR, Eremenko AV, Boginskaya IA, Ryzhikov IA, Trifonova EA, Putlyayev EV, Mukhin AN, Kalnov SL, Balandina MV, Tkachuk AP, Gushchin VA, Sarychev AK, Lagarkov AN, Rodionov IA, Gabidullin AR, Kurochkin IN (2018) Non-labeled selective virus detection with novel SERS-active porous silver nanofilms fabricated by electron beam physical vapor deposition. *Sensor Actuat B: Chem* 257:37–47
39. Li WQ, Wang G, Zhang XN, Geng HP, Shen JL, Wang LS, Zhao J, Xu LF, Zhang LJ, Wu YQ, Tai RZ, Chen G (2015) Geometrical and morphological optimizations of plasmonic nanoarrays for high-performance SERS detection. *Nanoscale* 7(37):15487–15494
40. Zakaria MA, Menazea AA, Mostafa AM, Al-Ashkar EA (2020) Ultra-thin silver nanoparticles film prepared via pulsed laser deposition: Synthesis characterization and its catalytic activity on reduction of 4-nitrophenol. *Surf Interfaces* 19:100438
41. Khan TM, Lunney JG, O'Rourke D, Meyer MC, Creel JR, Siewierska KE (2019) Various pulsed laser deposition methods for preparation of silver-sensitized glass and paper substrates for surface-enhanced Raman spectroscopy. *Appl Phys A* 125(9):659
42. Zanchi C, Lucotti A, Tommasini M, Pistaffa M, Giuliani L, Trusso S, Ossi PM (2019) Pulsed laser deposition of gold thin films with long-range spatial uniform SERS activity. *Appl Phys A* 125(5):311
43. Nie R, Wang Q, Sun P, Wang R, Yuan Q, Wang X (2018) Pulsed Laser Deposition of NiSe₂ film on carbon nanotubes for high-performance supercapacitor. *Eng Sci* 6:22–29
44. David PN (2006) Pulsed laser deposition of complex materials: progress toward applications. In: Eason R (ed) Pulsed laser deposition of thin films. Optoelectronics Research Centre, University of Southampton, UK, pp 1–31
45. Sturm K, Fähler S, Krebs H-U (2000) Pulsed laser deposition of metals in low pressure inert gas. *Appl Surf Sci* 154:462–466
46. Smyth CA, Mirza I, Lunney JG, McCabe EM (2013) Surface-enhanced Raman spectroscopy (SERS) using Ag nanoparticle films produced by pulsed laser deposition. *Appl Surf Sci* 264:31–35
47. Fan M, Andrade GFS, Brolo AG (2011) A review on the fabrication of substrates for surface enhanced Raman spectroscopy and their applications in analytical chemistry. *Anal Chim Acta* 693(1):7–25
48. Luo S-C, Sivashanmugan K, Liao J-D, Yao C-K, Peng H-C (2014) Nanofabricated SERS-active substrates for single-molecule to virus detection in vitro: a review. *Biosens Bioelectron* 61:232–240
49. Yin PT, Kim T-H, Choi J-W, Lee K-B (2013) Prospects for graphene-nanoparticle-based hybrid sensors. *Phys Chem Chem Phys* 15(31):12785–12799
50. Wang X, Shi W, She G, Mu L (2012) Surface-enhanced Raman scattering (SERS) on transition metal and semiconductor nanostructures. *Phys Chem Chem Phys* 14(17):5891–5901
51. Huang Z, Zhang A, Zhang Q, Cui D (2019) Nanomaterial-based SERS sensing technology for biomedical application. *J Mater Chem B* 7(24):3755–3774
52. Prasad SR, Teli SB, Ghosh J, Prasad NR, Shaikh VS, Nazeruddin GM, Al-Sehemi AGA, Patel I, Shaikh YI (2021) A review on bio-inspired synthesis of silver nanoparticles: their antimicrobial efficacy and toxicity. *Eng Sci*
53. Hass G, Ramsey JB (1969) Vacuum deposition of dielectric and semiconductor films by a CO₂ laser. *Appl Opt* 8(6):1115–1118
54. Smith HM, Turner AF (1965) Vacuum deposited thin films using a ruby laser. *Appl Opt* 4(1):147–148
55. Inam A, Hegde M, Wu X, Venkatesan T, England P, Miceli P, Chase E, Chang C, Tarascon J, Wachtman J (1988) As-deposited

- high T_c and J_c superconducting thin films made at low temperatures. *Appl Phys Lett* 53(10):908–910
56. Godel F, Zatkó V, Carrétéro C, Sander A, Galbiati M, Vecchiola A, Brus P, Bezencenet O, Servet B, Martin M-B, Dlubak B, Seneor P (2020) WS₂ 2D semiconductor down to monolayers by pulsed-laser deposition for large-scale integration in electronics and spintronics circuits. *ACS Appl Nano Mater* 3(8):7908–7916
 57. Zhang F, Teng X, Shi W, Song Y, Zhang J, Wang X, Li H, Li Q, Li S, Hu H (2020) SnO₂ nanoflower arrays on an amorphous buffer layer as binder-free electrodes for flexible lithium-ion batteries. *Appl Surf Sci* 527:146910
 58. Chen J, Ding Y, Li L, Zhang W, Li M, Chang G, Lu Y, Klar PJ, He Y (2019) Pulsed laser deposition and characteristics of epitaxial non-polar m-plane ZnO_{1-x}S_x alloy films. *J Alloys Compd* 773:443–448
 59. Lu T-W, Feng C-S, Wang Z, Liao K-W, Liu Z-Y, Xie Y-Z, Hu J-G, Liao W-B (2019) Microstructures and mechanical properties of CoCrFeNiAl_{0.3} high-entropy alloy thin films by pulsed laser deposition. *Appl Surf Sci* 494:72–79
 60. Gansukh M, López Mariño S, Espindola Rodriguez M, Engberg SLJ, Martinho FMA, Hajjafarassar A, Schjødt NC, Stamate E, Hansen O, Schou J, Canulescu S (2020) Oxide route for production of Cu₂ZnSnS₄ solar cells by pulsed laser deposition. *Sol Energy Mater Sol Cells* 215:110605
 61. Mostafa AM, Menazea AA (2020) Laser-assisted for preparation ZnO/CdO thin film prepared by pulsed laser deposition for catalytic degradation. *Radiat Phys Chem* 176:109020
 62. Geohegan DB, Puzosky AA, Boulesbaa A, Duscher G, Eres G, Li X, Liang L, Mahjour-Samani M, Rouleau C, Tennyson W, Tian M, Wang K, Xiao K, Yoon M (2018) Laser synthesis processing and spectroscopy of atomically-thin two dimensional materials. In: Ossi PM (ed) *Advances in the application of lasers in materials science*. Springer International Publishing, Cham, pp 1–37
 63. Lowndes DH, Geohegan DB, Puzosky AA, Norton DP, Rouleau CM (1996) Synthesis of novel thin-film materials by pulsed laser deposition. *Science* 273(5277):898–903
 64. Marla D, Bhandarkar UV, Joshi SS (2011) Critical assessment of the issues in the modeling of ablation and plasma expansion processes in the pulsed laser deposition of metals. *J Appl Phys* 109(2):2
 65. Amar JG, Family F, Lam P-M (1994) Dynamic scaling of the island-size distribution and percolation in a model of submonolayer molecular-beam epitaxy. *Phys Rev B* 50(12):8781–8797
 66. Christen HM, Eres G (2008) Recent advances in pulsed-laser deposition of complex oxides. *J Phys Condens Matter* 20(26):264005
 67. Tersoff J, Denier van der Gon AW, Tromp RM (1994) Critical island size for layer-by-layer growth. *Phys Rev Lett* 72(2):266–269
 68. Jing Y, Wang H, Chen X, Wang X, Wei H, Guo Z (2014) Pulsed laser deposited Ag nanoparticles on nickel hydroxide nanosheet arrays for highly sensitive surface-enhanced Raman scattering spectroscopy. *Appl Surf Sci* 316:66–71
 69. Jing Y, Wang H, Zhao J, Yi H, Wang X (2015) Pulsed laser deposition of Ag nanoparticles on titanium hydroxide/oxide nanobelt arrays for highly sensitive surface-enhanced Raman spectroscopy. *Appl Surf Sci* 347:499–504
 70. Dey P, Baumann V, Rodríguez-Fernández J (2020) Gold nanorod assemblies: the roles of hot-spot positioning and anisotropy in plasmon coupling and SERS. *Nanomaterials* 10(5):942
 71. Huang J-A, Mousavi MZ, Giovannini G, Zhao Y, Hubarevich A, Soler MA, Rocchia W, Garoli D, De Angelis F (2020) Multiplexed discrimination of single amino acid residues in polypeptides in a single SERS hot spot. *Angew Chem Int Ed* 59(28):11423–11431
 72. Budner B, Kuźma M, Nasiłowska B, Bartosiewicz B, Liszewska M, Jankiewicz BJ (2019) Fabrication of silver nanoisland films by pulsed laser deposition for surface-enhanced Raman spectroscopy. *Beilstein J Nanotech* 10:882–893
 73. Doering WE, Nie S (2002) Single-molecule and single-nanoparticle SERS: examining the roles of surface active sites and chemical enhancement. *J Phys Chem B* 106(2):311–317
 74. Stiles PL, Dieringer JA, Shah NC, Van Duyne RP (2008) Surface-enhanced Raman spectroscopy. *Annu Rev Anal Chem* 1:601–626
 75. Huber R, Tausser F, Brodschelm A, Bichler M, Abstreiter G, Leitenstorfer A (2001) How many-particle interactions develop after ultrafast excitation of an electron-hole plasma. *Nature* 414(6861):286–289
 76. Barnes WL, Dereux A, Ebbesen TW (2003) Surface plasmon subwavelength optics. *Nature* 424(6950):824–830
 77. Kerker M, Wang D-S, Chew H (1980) Surface enhanced Raman scattering (SERS) by molecules adsorbed at spherical particles: errata. *Appl Opt* 19(24):4159–4174
 78. Kreibig U, Genzel L (1985) Optical absorption of small metallic particles. *Surf Sci* 156:678–700
 79. Alonso JC, Diamant R, Castillo P, Acosta-García MC, Batina N, Haro-Poniatowski E (2009) Thin films of silver nanoparticles deposited in vacuum by pulsed laser ablation using a YAG: Nd laser. *Appl Surf Sci* 255(9):4933–4937
 80. Spadaro MC, Fazio E, Neri F, Ossi PM, Trusso S (2014) On the influence of the mass ablated by a laser pulse on thin film morphology and optical properties. *Appl Phys A* 117(1):137–142
 81. Amoroso S, Ausanio G, Bruzzese R, Vitiello M, Wang X (2005) Femtosecond laser pulse irradiation of solid targets as a general route to nanoparticle formation in a vacuum. *Phys Rev B* 71(3):033406
 82. De Bonis A, Galasso A, Ibris N, Sansone M, Santagata A, Teghil R (2012) Ultra-short pulsed laser deposition of thin silver films for surface enhanced Raman scattering. *Surf Coat Technol* 207:279–285
 83. Fazio E, Neri F, Ossi PM, Santo N, Trusso S (2009) Growth process of nanostructured silver films pulsed laser ablated in high-pressure inert gas. *Appl Surf Sci* 255(24):9676–9679
 84. Verma S, Rao BT, Sathe V, Bhartiya S, Patel H, Kaul R, Singh B (2018) Optical and surface enhanced Raman scattering responses of densely packed Ag-Au alloy nanoparticle films of varied composition and thickness. *J Alloys Compd* 753:395–406
 85. Verma S, Rao BT, Detty AP, Ganesan V, Phase DM, Rai SK, Bose A, Joshi SC, Kukreja LM (2015) Surface plasmon resonances of Ag-Au alloy nanoparticle films grown by sequential pulsed laser deposition at different compositions and temperatures. *J Appl Phys* 117(13):133105
 86. Stampelcoskie KG, Scaiano JC, Tiwari VS, Anis H (2011) Optimal size of silver nanoparticles for surface-enhanced Raman spectroscopy. *J Phys Chem C* 115(5):1403–1409
 87. Deeb C, Zhou X, Jrm P, Wiederricht GP, Bachelot R, Russell M, Jain PK (2013) Size dependence of the plasmonic near-field measured via single-nanoparticle photoimaging. *J Phys Chem C* 117(20):10669–10676
 88. D'Andrea C, Neri F, Ossi PM, Santo N, Trusso S (2009) The controlled pulsed laser deposition of Ag nanoparticle arrays for surface enhanced Raman scattering. *Nanotechnology* 20(24):245606
 89. Fazio E, Neri F, D'Andrea C, Ossi PM, Santo N, Trusso S (2011) SERS activity of pulsed laser ablated silver thin films with controlled nanostructure. *J Raman Spectrosc* 42(6):1298–1304
 90. D'Andrea C, Neri F, Ossi P, Santo N, Trusso S (2011) Synthesis by pulsed laser ablation in Ar and SERS activity of silver thin films with controlled nanostructure. *Laser Phys* 21(4):818–822
 91. Bailini A, Ossi P (2007) Expansion of an ablation plume in a buffer gas and cluster growth. *Europhys Lett* 79(3):35002
 92. Agarwal NR, Fazio E, Neri F, Trusso S, Castiglioni C, Lucotti A, Santo N, Ossi PM (2011) Ag and Au nanoparticles for SERS substrates produced by pulsed laser ablation. *Cryst Res Technol* 46(8):836–840

93. Taneja P, Ayyub P, Chandra R (2002) Size dependence of the optical spectrum in nanocrystalline silver. *Phys Rev B* 65(24):245412
94. Mulvaney P (1996) Surface plasmon spectroscopy of nanosized metal particles. *Langmuir* 12(3):788–800
95. Xu H, Aizpurua J, Käll M, Apell P (2000) Electromagnetic contributions to single-molecule sensitivity in surface-enhanced Raman scattering. *Phys Rev E* 62(3):4318
96. Michaels AM, Jiang J, Brus L (2000) Ag nanocrystal junctions as the site for surface-enhanced Raman scattering of single rhodamine 6G molecules. *J Phys Chem B* 104(50):11965–11971
97. Ganeev RA, Chakravarty U, Naik PA, Srivastava H, Mukherjee C, Tiwari MK, Nandedkar RV, Gupta PD (2007) Pulsed laser deposition of metal films and nanoparticles in vacuum using subnanosecond laser pulses. *Appl Opt* 46(8):1205–1210
98. Semaltianos NG, Perrie W, Vishnyakov V, Murray R, Williams CJ, Edwardson SP, Dearden G, French P, Sharp M, Logothetidis S, Watkins KG (2008) Nanoparticle formation by the debris produced by femtosecond laser ablation of silicon in ambient air. *Mater Lett* 62(14):2165–2170
99. Donnelly T, Krishnamurthy S, Carney K, McEvoy N, Lunney J (2007) Pulsed laser deposition of nanoparticle films of Au. *Appl Surf Sci* 254(4):1303–1306
100. Nedyalkov N, Nikolov A, Atanasov P, Alexandrov M, Terakawa M, Shimizu H (2014) Nanostructured Au film produced by pulsed laser deposition in air at atmospheric pressure. *Opt Laser Technol* 64:41–45
101. Khan TM, Lunney JG, O'Rourke D, Meyer MC, Creel JR, Siewierska KE (2019) Various pulsed laser deposition methods for preparation of silver-sensitized glass and paper substrates for surface-enhanced Raman spectroscopy. *Appl Phys A* 125(9):1–7
102. Yan Z, Chrisey DB (2012) Pulsed laser ablation in liquid for micro-/nanostructure generation. *J Photochem Photobiol C* 13(3):204–223
103. Patil P, Phase D, Kulkarni S, Ghaisas S, Kulkarni S, Kanetkar S, Ogale S, Bhide V (1987) Pulsed-laser-induced reactive quenching at liquid-solid interface: aqueous oxidation of iron. *Phys Rev Lett* 58(3):238
104. Neddersen J, Chumanov G, Cotton TM (1993) Laser ablation of metals: a new method for preparing SERS active colloids. *Appl Spectrosc* 47(12):1959–1964
105. Giorgetti E, Marsili P, Giammanco F, Trigari S, Gellini C, Muniz-Miranda M (2015) Ag nanoparticles obtained by pulsed laser ablation in water: surface properties and SERS activity. *J Raman Spectrosc* 46(5):462–469
106. Vinod M, Gopchandran KG (2015) Ag@Au core-shell nanoparticles synthesized by pulsed laser ablation in water: effect of plasmon coupling and their SERS performance. *Spectrochim Acta A* 149:913–919
107. Riedel R, Mahr N, Yao C, Wu A, Yang F, Hampp N (2020) Synthesis of gold-silica core-shell nanoparticles by pulsed laser ablation in liquid and their physico-chemical properties towards photothermal cancer therapy. *Nanoscale* 12(5):3007–3018
108. Mostafa AM, Mwafy EA, Awwad NS, Ibrahim HA (2021) Catalytic activity of Ag nanoparticles and Au/Ag nanocomposite prepared by pulsed laser ablation technique against 4-nitrophenol for environmental applications. *J Mater Sci: Mater El* 32(9):11978–11988
109. Saito K, Sakka T, Ogata YH (2003) Rotational spectra and temperature evaluation of C₂ molecules produced by pulsed laser irradiation to a graphite-water interface. *J Appl Phys* 94(9):5530–5536
110. Sakka T, Saito K, Ogata YH (2004) Confinement effect of laser ablation plume in liquids probed by self-absorption of C₂ Swan band emission. *J Appl Phys* 97(1):014902
111. Tsuji T, Iryo K, Watanabe N, Tsuji M (2002) Preparation of silver nanoparticles by laser ablation in solution: influence of laser wavelength on particle size. *Appl Surf Sci* 202(1):80–85
112. Barcikowski S, Menéndez-Manjón A, Chichkov B, Brikas M, Račiukaitis G (2007) Generation of nanoparticle colloids by picosecond and femtosecond laser ablations in liquid flow. *Appl Phys Lett* 91(8):083113
113. Wagener P, Ibrahimkutty S, Menzel A, Plech A, Barcikowski S (2013) Dynamics of silver nanoparticle formation and agglomeration inside the cavitation bubble after pulsed laser ablation in liquid. *Phys Chem Chem Phys* 15(9):3068–3074
114. Dell'Aglio M, Gaudioso R, De Pascale O, De Giacomo A (2015) Mechanisms and processes of pulsed laser ablation in liquids during nanoparticle production. *Appl Surf Sci* 348:4–9
115. Ibrahimkutty S, Wagener P, Menzel A, Plech A, Barcikowski S (2012) Nanoparticle formation in a cavitation bubble after pulsed laser ablation in liquid studied with high time resolution small angle x-ray scattering. *Appl Phys Lett* 101(10):103104
116. Raheem GK, Abdurrahman KA, Maryam FN (2012) Nanofabrication and characterization of gold metal by pulsed laser ablation in SDS solution. *Adv Phys The Appl* 11:23–27
117. Mafuné F, Kohno J-y, Takeda Y, Kondow T, Sawabe H (2000) Structure and stability of silver nanoparticles in aqueous solution produced by laser ablation. *J Phys Chem B* 104(35):8333–8337
118. Mafuné F, Kohno J-y, Takeda Y, Kondow T, Sawabe H (2000) Formation and size control of silver nanoparticles by laser ablation in aqueous solution. *J Phys Chem B* 104(39):9111–9117
119. Alluhaybi HA, Ghoshal SK, Shamsuri WNW, Alsobhi BO, Salim AA, Krishnan G (2019) Pulsed laser ablation in liquid assisted growth of gold nanoparticles: evaluation of structural and optical features. *Nano-Struct Nano-Objects* 19:100355
120. Amendola V, Meneghetti M (2013) What controls the composition and the structure of nanomaterials generated by laser ablation in liquid solution? *Phys Chem Chem Phys* 15(9):3027–3046
121. Šišková K, Vlckova B, Turpin P, Fayet C (2008) Ion-specific effects on laser ablation of silver in aqueous electrolyte solutions. *J Phys Chem C* 112(12):4435–4443
122. Han H, Fang Y, Li Z, Xu H (2008) Tunable surface plasma resonance frequency in Ag core/Au shell nanoparticles system prepared by laser ablation. *Appl Phys Lett* 92(2):023116
123. Dong C, Yan Z, Kokx J, Chrisey DB, Dinu CZ (2012) Antibacterial and surface-enhanced Raman scattering (SERS) activities of AgCl cubes synthesized by pulsed laser ablation in liquid. *Appl Surf Sci* 258(23):9218–9222
124. Zhang W, Wu X, Chen H, Gao Y, Zhu J, Huang G, Chu PK (2008) Self-organized formation of silver nanowires, nanocubes and bipyramids via a solvothermal method. *Acta Mater* 56(11):2508–2513
125. Kamakshi K, Silva JPB, Sekhar KC, Marslin G, Moreira JA, Conde O, Almeida A, Pereira M, Gomes MJM (2016) Influence of substrate temperature on the properties of pulsed laser deposited silver nanoparticle thin films and their application in SERS detection of bovine serum albumin. *Appl Phys B* 122(5):108
126. Floro JA, Chason E, Cammarata RC, Srolovitz DJ (2002) Physical origins of intrinsic stresses in Volmer-Weber thin films. *MRS Bull* 27(1):19–25
127. Sambri A, Radovic M, Wang X, Amoroso S, Granozio FM, Bruzzese R, (2007) Substrate heating effects on the propagation dynamics of laser produced plume during pulsed laser deposition of oxides. *Appl Surf Sci* 254(4):790–793
128. Xu H, Bjerneld EJ, Käll M, Börjesson L (1999) Spectroscopy of single hemoglobin molecules by surface enhanced Raman scattering. *Phys Rev Lett* 83(21):4357
129. Li F, Wu S, Zhang L, Li Z (2017) Ag nanoparticles decorated ZnO nanoarrays with enhanced surface-enhanced Raman scattering and field emission property. *J Mater Sci-Mater EL* 28(21):16233–16238
130. Szlachetko K, Piotrowski P, Sadecka K, Osewski P, Kasprzowicz D, Pawlak DA (2020) Selective surface-enhanced Raman scattering in

- a bulk nanoplasmonic Bi₂O₃-Ag eutectic composite. *Nanophotonics* 9(14):4307–4314
131. D'Andrea C, Faro MJ, Bertino G, Ossi PM, Neri F, Trusso S, Musumeci P, Galli M, Cioffi N, Irrera A, Priolo F, Fazio B (2016) Decoration of silicon nanowires with silver nanoparticles for ultrasensitive surface enhanced Raman scattering. *Nanotechnology* 27(37):375603
 132. Rebollar E, Hernández M, Sanz M, Pérez S, Ezquerro TA, Castillejo M (2015) Laser-induced surface structures on gold-coated polymers: influence of morphology on surface-enhanced Raman scattering enhancement. *J Appl Polym Sci* 132(45):42770
 133. Kamath PV, Annal Therese GH, Gopalakrishnan J (1997) On the Existence of hydrotalcite-like phases in the absence of trivalent cations. *J Solid State Chem* 128(1):38–41
 134. Shi R, Liu X, Ying Y (2018) Facing challenges in real-life application of surface-enhanced Raman scattering: design and nanofabrication of surface-enhanced Raman scattering substrates for rapid field test of food contaminants. *J Agric Food Chem* 66(26):6525–6543
 135. Wu L, Pu H, Huang L, Sun D-W (2020) Plasmonic nanoparticles on metal-organic framework: a versatile SERS platform for adsorptive detection of new coccine and orange II dyes in food. *Food Chem* 328:127105
 136. Hong J, Kawashima A, Hamada N (2017) A simple fabrication of plasmonic surface-enhanced Raman scattering (SERS) substrate for pesticide analysis via the immobilization of gold nanoparticles on UF membrane. *Appl Surf Sci* 407:440–446
 137. Xu Y, Hassan MM, Ali S, Li H, Ouyang Q, Chen Q (2021) Self-cleaning-mediated SERS chip coupled chemometric algorithms for detection and photocatalytic degradation of pesticides in food. *J Agric Food Chem* 69(5):1667–1674
 138. Hu B, Sun D-W, Pu H, Wei Q (2020) Rapid nondestructive detection of mixed pesticides residues on fruit surface using SERS combined with self-modeling mixture analysis method. *Talanta* 217:120998
 139. Burr DS, Fatigante WL, Lartey JA, Jang W, Stelmack AR, McClurg NW, Standard JM, Wieland JR, Kim J-H, Mulligan CC, Driskell JD (2020) Integrating SERS and PSI-MS with dual purpose plasmonic paper substrates for on-site illicit drug confirmation. *Anal Chem* 92(9):6676–6683
 140. Haddad A, Comanescu MA, Green O, Kubic TA, Lombardi JR (2018) Detection and quantitation of trace fentanyl in heroin by surface-enhanced Raman spectroscopy. *Anal Chem* 90(21):12678–12685
 141. Fan W, Yang S, Zhang Y, Huang B, Gong Z, Wang D, Fan M (2020) Multifunctional flexible SERS sensor on a fixate gel pad: capturing derivation and selective picogram indirect detection of explosive 2,2',4,4',6,6'-hexanitrostilbene. *ACS Sens* 5(11):3599–3606
 142. Gao R, Song X, Zhan C, Weng C, Cheng S, Guo K, Ma N, Chang H, Guo Z, Luo L-B, Yu L (2020) Light trapping induced flexible wrinkled nanocone SERS substrate for highly sensitive explosive detection. *Sens Actuat B Chem* 314:128081
 143. Choi N, Dang H, Das A, Sim MS, Chung IY, Choo J (2020) SERS biosensors for ultrasensitive detection of multiple biomarkers expressed in cancer cells. *Biosens Bioelectron* 164:112326
 144. Kapara A, Brunton V, Graham D, Faulds K (2020) Investigation of cellular uptake mechanism of functionalised gold nanoparticles into breast cancer using SERS. *Chem Sci* 11(22):5819–5829
 145. Liu J, Liu W, Huang Y, Zhao X, Feng Z, Wang D, Gong Z, Fan M (2021) Self-supporting liquid film as reproducible SERS platform for therapeutic drug monitoring of berberine hydrochloride in human urine. *Microchem J* 165:106122
 146. Panikar SS, Banu N, Escobar E-R, García G-R, Cervantes-Martínez J, Villegas T-C, Salas P, De la Rosa E (2020) Stealth modified bottom up SERS substrates for label-free therapeutic drug monitoring of doxorubicin in blood serum. *Talanta* 218:121138
 147. Liu X, Cai H, Zou J, Pang Z, Yuan B, Zhou Z, Cheng Q (2018) Spectrophotometric determination of trace permanganate in water with N, N-diethyl-p-phenylenediamine (DPD). *Chemosphere* 212:604–610
 148. Chen J, Wang X, Huang Y, Lv S, Cao X, Yun J, Cao D (2018) Adsorption removal of pollutant dyes in wastewater by nitrogen-doped porous carbons derived from natural leaves. *Eng Sci* 5:30–38
 149. Wu D, Zhang X, Zhu J, Cheng D (2018) Concerted catalysis on Tanghulu-like Cu@zeolitic imidazolate framework-8 (ZIF-8) nanowires with tuning catalytic performances for 4-nitrophenol reduction. *Eng Sci* 2:49–56
 150. Li M, Liu H, Ren X (2017) Ratiometric fluorescence and mesoporous structured imprinting nanoparticles for rapid and sensitive detection 2,4,6-trinitrophenol. *Biosens Bioelectron* 89:899–905
 151. Gu J, She J, Yue Y (2020) Micro/nanoscale thermal characterization based on spectroscopy techniques. *ES Energy Environ* 9:15–27
 152. Zhou R-S, Lin Z-Z, Xin L-D, Song J-F, Liu H, Guo Z (2018) Naked eye colorimetric multifunctional sensing of nitrobenzene, Cr(VI) and Fe(III) with a new green emission Ag₆S₆ multi-metal-cluster. *Adv Compos Hybrid Mater* 1(4):785–796
 153. Nedaei M, Zarei AR, Ghorbanian SA (2019) Miniaturized matrix solid-phase dispersion based on deep eutectic solvent and carbon nitride associated with high-performance liquid chromatography: a new feasibility for extraction and determination of trace nitro-toluene pollutants in soil samples. *J Chromatogr* 1601:35–44
 154. Danish M, Qamar M, Suliman MH, Muneer M (2020) Photoelectrochemical and photocatalytic properties of Fe@ZnS QDs/TiO₂ nanocomposites for degradation of different chromophoric organic pollutants in aqueous suspension. *Adv Compos Hybrid Mater* 3(4):570–582
 155. Dong S, Wang S, Gyimah E, Zhu N, Wang K, Wu X, Zhang Z (2019) A novel electrochemical immunosensor based on catalase functionalized AuNPs-loaded self-assembled polymer nanospheres for ultrasensitive detection of tetrabromobisphenol A bis(2-hydroxyethyl) ether. *Anal Chim Acta* 1048:50–57
 156. Panthi G, Ranjit R, Khadka S, Gyawali KR, Kim H-Y, Park M (2020) Characterization and antibacterial activity of rice grain-shaped ZnS nanoparticles immobilized inside the polymer electrospun nanofibers. *Adv Compos Hybrid Mater* 3(1):8–15
 157. Li D-W, Zhai W-L, Li Y-T, Long Y-T (2014) Recent progress in surface enhanced Raman spectroscopy for the detection of environmental pollutants. *Microchim Acta* 181(1):23–43
 158. Hao J, Han M-J, Han S, Meng X, Su T-L, Wang QK (2015) SERS detection of arsenic in water: a review. *J Environ Sci* 36:152–162
 159. Pang S, Yang T, He L (2016) Review of surface enhanced Raman spectroscopic (SERS) detection of synthetic chemical pesticides. *TrAC Trends Anal Chem* 85:73–82
 160. Wei H, Hossein Abtahi SM, Vikesland PJ (2015) Plasmonic colorimetric and SERS sensors for environmental analysis. *Environ Sci Nano* 2(2):120–135
 161. Song D, Yang R, Long F, Zhu A (2019) Applications of magnetic nanoparticles in surface-enhanced Raman scattering (SERS) detection of environmental pollutants. *J Environ Sci* 80:14–34
 162. Qu LL, Li YT, Li DW, Xue JQ, Fossey JS, Long YT (2013) Humic acids-based one-step fabrication of SERS substrates for detection of polycyclic aromatic hydrocarbons. *Analyst* 138(5):1523–1528
 163. Gu X, Tian S, Zhou Q, Adkins J, Gu Z, Li X, Zheng J (2013) SERS detection of polycyclic aromatic hydrocarbons on a bowl-shaped silver cavity substrate. *RSC Adv* 3(48):25989

164. Yu Z, Grasso MF, Sorensen HH, Zhang P (2019) Ratiometric SERS detection of polycyclic aromatic hydrocarbons assisted by β -cyclodextrin-modified gold nanoparticles. *Microchim Acta* 186(6):391
165. Li D, Cao X, Zhang Q, Ren X, Jiang L, Li D, Deng W, Liu H (2019) Facile in situ synthesis of core-shell MOF@Ag nanoparticle composites on screen-printed electrodes for ultrasensitive SERS detection of polycyclic aromatic hydrocarbons. *J Mater Chem A* 7(23):14108–14117
166. Zhou Z, Lu J, Wang J, Zou Y, Liu T, Zhang Y, Liu G, Tian Z (2020) Trace detection of polycyclic aromatic hydrocarbons in environmental waters by SERS. *Spectrochim Acta Pt A* 234:118250
167. Atanasov PA, Nedyalkov NN, Fukata N, Jevasuwan W, Subramani T, Hirsch D, Rauschenbach B (2019) Au and Ag films and nanostructures for detection of fungicide mancozeb: SERS analyses. *AIP Conf Proc* 2075(1):030001
168. Atanasov PA, Nedyalkov NN, Fukata N, Jevasuwan W, Subramani T (2019) Surface-enhanced Raman spectroscopy (SERS) of neonicotinoid insecticide thiacloprid assisted by silver and gold nanostructures. *Appl Spectrosc* 74(3):357–364
169. Harvey RG (1997) Polycyclic aromatic hydrocarbons. John Wiley & Sons, Chichester
170. Lee M, Novotny M, Bartle K (1981) Analytic chemistry of polycyclic aromatic hydrocarbons. Academic Press, New York
171. Cao J, Zhao D, Qin Y (2019) Novel strategy for fabrication of sensing layer on thiol-functionalized fiber-optic tapers and their application as SERS probes. *Talanta* 194:895–902
172. Kwon Y-H, Sowoidnich K, Schmidt H, Kronfeldt H-D (2012) Application of calixarene to high active surface-enhanced Raman scattering (SERS) substrates suitable for in situ detection of polycyclic aromatic hydrocarbons (PAHs) in seawater. *J Raman Spectrosc* 43(8):1003–1009
173. Shi X, Kwon Y-H, Ma J, Zheng R, Wang C, Kronfeldt HD (2013) Trace analysis of polycyclic aromatic hydrocarbons using calixarene layered gold colloid film as substrates for surface-enhanced Raman scattering. *J Raman Spectrosc* 44(1):41–46
174. Bao L, Sheng P, Li J, Wu S, Cai Q, Yao S (2012) Surface enhanced Raman spectroscopic detection of polycyclic aromatic hydrocarbons (PAHs) using a gold nanoparticles-modified alginate gel network. *Analyst* 137(17):4010–4015
175. Hongo H, Nihey F, Ichihashi T, Ochiai Y, Yudasaka M, Iijima S (2003) Support materials based on converted aluminum films for chemical vapor deposition growth of single-wall carbon nanotubes. *Chem Phys Lett* 380(1):158–164
176. Star A, Han T-R, Gabriel J-CP, Bradley K, Grüner G (2003) Interaction of aromatic compounds with carbon nanotubes: correlation to the Hammett parameter of the substituent and measured carbon nanotube FET response. *Nano Lett* 3(10):1421–1423
177. Leyton P, Gomez-Jeria JS, Sanchez-Cortes S, Domingo C, Campos-Vallette M (2006) Carbon nanotube bundles as molecular assemblies for the detection of polycyclic aromatic hydrocarbons: surface-enhanced resonance Raman spectroscopy and theoretical studies. *J Phys Chem B* 110(13):6470–6474
178. Nakashima N, Tomonari Y, Murakami H (2002) Water-soluble single-walled carbon nanotubes via noncovalent sidewall-functionalization with a pyrene-carrying ammonium ion. *Chem Lett* 31(6):638–639
179. Peng T, Wang H, Yi H, Jing Y, Sun P, Wang X (2015) Co(OH)₂ nanosheets coupled with CNT arrays grown on Ni mesh for high-rate asymmetric supercapacitors with excellent capacitive behavior. *Electrochim Acta* 176:77–85
180. Shaikh YI, Sameer A, Shaikh AVS, Ahmed K, Nazeruddin GM, Pathan AHM (2020) The revelation of various compounds found in *Nigella sativa* L. (black cumin) and their possibility to inhibit COVID-19 infection based on the molecular docking and physical properties. *Eng Sci* 11:31–35
181. Zhou P, Wang J, Du X, Huang T, Nallathamby PD, Yang L, Zou W, Zhou Y, Jault J-M, Chen S, Ding F (2018) Nanoparticles in biomedicine-focus on imaging applications. *Eng Sci* 5:1–20
182. Shen YQSZH, Shen J (2019) Nanotechnology advances in medicine: focus on cancer. *Eng Sci* 7:1–9
183. Jia W, Qi Y, Hu Z, Xiong Z, Luo Z, Xiang Z, Hu J, Lu W (2021) Facile fabrication of monodisperse CoFe₂O₄ nanocrystals@dopamine@DOX hybrids for magnetic-responsive on-demand cancer theranostic applications. *Adv Compos Hybrid Mater*
184. Shaikh VS, Nazeruddin GM, Bloukh YISSH, Edis Z, Pathan HM (2020) A recapitulation of virology modes of dissemination diagnosis treatment and preventive measures of COVID-19: a review. *Eng Sci* 10:11–23
185. Prasad RD, Charmode N, Shrivastav OP, Prasad SR, Moghe A, AnantSamant A, Sarvarkar PD, Prasad NR (2021) A review on concept of nanotechnology in veterinary medicine. *ES Food Agric* 4:28–60
186. Ahmed AAQ, Zheng R, Abdalla AME, Bakadia BM, Qi F, Xiao L, Atta OM, Mao L, Yang G (2021) Heterogeneous populations of outer membrane vesicles released from *Helicobacter pylori* SS1 with distinct biological properties. *Eng Sci* 15:148–165
187. Fu X, Su J, Hou L, Zhu P, Hou Y, Zhang K, Li H, Liu X, Jia C, Xu J (2021) Physicochemical and thermal characteristics of *Moringa oleifera* seed oil. *Adv Compos Hybrid Mater*
188. Ghosh S, Ghosh S, Pramanik N (2020) Bio-evaluation of doxorubicin (DOX)-incorporated hydroxyapatite (HAP)-chitosan (CS) nanocomposite triggered on osteosarcoma cells. *Adv Compos Hybrid Mater* 3(3):303–314
189. Huang Y, Luo Y, Liu H, Lu X, Zhao J, Lei Y (2020) A subcutaneously injected SERS nanosensor enabled long-term in vivo glucose tracking. *Eng Sci* 14:59–68
190. Vo-Dinh T, Liu Y, Fales AM, Ngo H, Wang H-N, Register JK, Yuan H, Norton SJ, Griffin GD (2015) SERS nanosensors and nanoreporters: golden opportunities in biomedical applications. *WIREs Nanomed Nanobi* 7(1):17–33
191. Picciolini S, Castagnetti N, Vanna R, Mehn D, Bedoni M, Gramatica F, Villani M, Calestani D, Pavesi M, Lazzarini L, Zappettini A, Morasso C (2015) Branched gold nanoparticles on ZnO 3D architecture as biomedical SERS sensors. *RSC Adv* 5(113):93644–93651
192. Baumberg J, Bell S, Bonifacio A, Chikkaraddy R, Chisanga M, Corsetti S, Delfino I, Eremina O, Fasolato C, Faulds K, Fleming H, Goodacre R, Graham D, Hardy M, Jamieson L, Keyes T, Królikowska A, Kuttner C, Langer J, Lightner C, Mahajan S, Masson J-F, Muhamadali H, Natan M, Nicolson F, Nikelspharg E, Plakas K, Popp J, Porter M, Prezgot D, Pytlik N, Schlücker S, Silvestri A, Stone N, Tian Z-Q, Tripathi A, Willner M, Wuytens P (2017) SERS in biology/biomedical SERS: general discussion. *Faraday Discuss* 205:429–456
193. Sharma B, Bugga P, Madison LR, Henry A-I, Blaber MG, Greeneltch NG, Chiang N, Mrksich M, Schatz GC, Van Duyne RP (2016) Bisboronic acids for selective, physiologically relevant direct glucose sensing with surface-enhanced Raman spectroscopy. *J Am Chem Soc* 138(42):13952–13959
194. Wang C, Wang C, Wang X, Wang K, Zhu Y, Rong Z, Wang W, Xiao R, Wang S (2019) Magnetic SERS strip for sensitive and simultaneous detection of respiratory viruses. *ACS Appl Mat Interfaces* 11(21):19495–19505
195. Huang J, Zhou M, Le W, Chen G, Ni S, Niu G, Li Z, Zhao Z, He Z, Li B, Zhao S, Zhang B, Du K (2020) Detection of spike protein of SARS-CoV-2 by surface enhanced Raman spectroscopy. *High Power Laser Part Beams* 32:069001

196. Agarwal NR, Tommasini M, Ciusani E, Lucotti A, Trusso S, Ossi PM (2018) Protein-metal interactions probed by SERS: lysozyme on nanostructured gold surface. *Plasmonics* 13(6):2117–2124
197. Kneipp J, Kneipp H, McLaughlin M, Brown D, Kneipp K (2006) In vivo molecular probing of cellular compartments with gold nanoparticles and nanoaggregates. *Nano Lett* 6(10):2225–2231
198. Tellez-Plancarte A, Haro-Poniatowski E, Picquart M, Morales-Mendez JG, Lara-Cruz C, Jimenez-Salazar JE, Damian-Matsumura P, Escobar-Alarcon L, Batina N (2018) Development of a nanostructured platform for identifying HER2-heterogeneity of breast cancer cells by surface-enhanced Raman scattering. *Nanomaterials* 8(7):549
199. Loo L, Capobianco JA, Wu W, Gao X, Shih WY, Shih W-H, Pourrezaei K, Robinson MK, Adams GP (2011) Highly sensitive detection of HER2 extracellular domain in the serum of breast cancer patients by piezoelectric microcantilevers. *Anal Chem* 83(9):3392–3397
200. Madzharova F, Heiner Z, Kneipp J (2017) Surface enhanced hyper-Raman scattering of the amino acids tryptophan, histidine, phenylalanine, and tyrosine. *J Phys Chem C* 121(2):1235–1242
201. Usoskin A, Betz U, Hofacker F, Dietrich R, Schlenga K (2017) Long HTS tapes with high in-field performance manufactured via multibeam PLD with “dynamic” drum concept. *IEEE Trans Appl Supercond* 27(4):1–5

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