Surface-Enhanced Raman Scattering on Silver Sinusoidal Nanograting: Impact of Interactions of Grating-Coupled Surface Plasmon Polaritons

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Abstract
In this study, we reported a silver sinusoidal nanograting used in microchannels, forming H2O/Ag/NOA/SiO2 heterostructure and studied the impact of interactions of grating-coupled surface plasmon polaritons (SPPs) on surface-enhanced Raman scattering (SERS). There are two modes of $E_x(z)$ in the Ag grating, namely, the anti-symmetric mode (AM) at the high SPP resonance frequency and the symmetric mode (SM) at the low-SPP resonance frequency. When the refractive indices of the upper and lower dielectrics of the thin Ag grating are close, there will be a strong coupling between the SPPs of the two interfaces, which will increase the transmittance but reduce the maximum electromagnetic (EM) of the SERS substrate. Additionally, the thinner the Ag grating, the stronger the coupling, accompanied by the frequency shift of the two coupling modes. Our experimental results are supported by FDTD calculations, which have corroborated the importance of the interactions of grating-coupled SPPs in the design of SERS substrate.

Keywords Surface plasmon polariton (SPP) · Surface-enhanced Raman scattering (SERS) · Sinusoidal nanograting · SPP coupling · Anti-symmetric mode (AM) · Symmetric mode (SM)

Introduction
Nanostructures that support surface plasmon resonance (SPR) have been widely used for sensing applications through different techniques including SPR spectroscopy [1, 2], SPR imaging [3], and surface-enhanced Raman scattering (SERS) spectroscopy [4–7]. SERS, given its ability in highly sensitive detection and specific identification of analytes, has been extensively employed in multiple applications including trace molecules detection [8, 9], biosensors [10, 11], food safety [12, 13], and environmental safety [14, 15]. Metallic nanogratings [16, 17], in particular, experience large-area uniform electromagnetic enhancement [18, 19], which increases the chances for analyte detection via SERS spectroscopy [19–21]. Their properties are connected to surface plasmon polariton (SPP) whose resonance wavelength depends on the geometrical parameters of the nanograting, the incident light, and the surrounding medium [7]. 1D metallic nanograting may enhance the electromagnetic field intensity at a metal-dielectric boundary near-surface region by 10 times [22]. However, Raman spectroscopy is greatly affected by the surrounding medium when metallic nanograting is used to detect in microchannels [23]. When the refractive index of the superstrate and substrate is close, the excitation of SPP takes place at the upper interface or the lower interface leading to the interaction of different modes [24].

In this work, we studied the coupling mode of SPPs supported by silver sinusoidal nanograting deposited on Norland Optical Adhesive (NOA)-coated glass substrate used in microchannels, forming a H2O/Ag/NOA/SiO2 heterostructure. Then we estimated the impact of interactions of grating-coupled SPPs on the SERS, by varying the thickness of Ag grating, the refractive index of the surrounding medium of Ag grating. The FDTD simulation results have corroborated the importance of the grating-coupled SPP interactions in the design of the SERS substrate.
Model and Theoretical Analysis

Model

As shown in Fig. 1, the SERS structure of sinusoidal nanograting with period $\Lambda$ consists of 3 layers (Ag, NOA, SiO$_2$). The medium above the Ag layer is water, which functions as a concentrator effectively increasing the volumetric density of trace molecules. The sinusoidal nanograting can be easily fabricated over a large area using the rapid and high-throughput interferometric technique described elsewhere [1, 25], avoiding the expensive and time-consuming electron beam-based nano-fabrication method. The Ag sinusoidal nanograting with pattern periodicity has the advantage over the random configuration of surface structures due to its reproducibility and pattern scalability. It also has a large-area approach for target analyte detection, with the metallic interface for the SPP excitation.

The optical responses of the heterostructure are computed using FDTD simulations by assuming that the grating volume extends uniformly in the Y-direction. Therefore, periodic boundary conditions (PBCs) are used in the Y-direction, while perfectly matched layers (PMLs) are used in the Z-direction. The normal incidence of light (plane waves) with transverse magnetic (TM) polarizations are considered excitations. The dispersive dielectric function of Ag and H$_2$O is extracted from the experimental data from Refs [26] and [27], respectively. The non-dispersive refractive index of glass (SiO$_2$) is considered as 1.45 and the refractive index of NOA varies with different dielectrics. For example, NOA61, with a refractive index of 1.56, is the preferred adhesive for military optics and is widely used due to low shrinkage and excellent glass and metal adhesion, and NOA133 has a low refractive index of 1.33, which is very close to that of H$_2$O in the wavelength range [28]. Relatively fine grid spacings set to 1 nm and simulation time to 1000 fs are applied for well-converged results.

Theoretical Analysis

The dispersion relation of SPP, propagating at a metal/dielectric interface, is defined as Formula (1).

$$\beta = k_0 \sqrt{\frac{\varepsilon_m(\omega) \times \varepsilon_d}{\varepsilon_m(\omega) + \varepsilon_d}}$$

(1)

where $\beta$ is the SPP wave vector, $k_0 = \frac{\omega}{c}$ is the incident wave vector, $\omega$ and $c$ are the frequency and speed of the incident light, $\varepsilon_m(\omega)$ and $\varepsilon_d$ are metal and dielectric permittivity.

When an incident beam of light is focused on a metallic grating, the diffraction wave will be generated [29] and directly coupled to SPP, if the momentum of the diffracted wave is matched with the grating surface [30] via the grating equation [31]:

$$\beta = k_0 n_d \sin \theta_i \pm m \frac{2\pi}{\Lambda}$$

(2)

where $n_d$ is the refractive index of the medium, $\theta_i$ is the angle of the incident light with respect to the normal on the grating, $m$ is an integer and represents the grating diffractive order, $\frac{2\pi}{\Lambda} = k_g$ is the reciprocal vector of the grating, and $\Lambda$ is the period of the grating.

The periodicity of the Ag sinusoidal grating is set to 570 nm calculated by Eq. (2) in order to generate SPP resonance at the TM wave of 785 nm. The amplitude of Ag sinusoidal grating is set to 20 nm with high electromagnetic enhancement [22]. The SPP fields fall off exponentially along the direction perpendicular to the grating surface [32] with the decay length of the fields calculated by Formula (3):

$$\delta = \frac{1}{\sqrt{\beta^2 - \epsilon k_0^2}}$$

(3)
In the H$_2$O/Ag/NOA/SiO$_2$ heterostructure, as seen in Fig. 2, when the thickness of Ag grating is comparable to or smaller than the decay length of the interface mode, where $\delta=23.484$ nm in the Ag at $\lambda_0=785$ nm, each single interface can sustain bound SPP [33], and interactions between SPPs give rise to coupled modes [34], even if the refractive index of NOA/SiO$_2$ is close to H$_2$O.

For TM light, according to Maxwell equations, the EM field components in H$_2$O are

$$H_y = Ae^{i\beta z} e^{-k_z z}$$
$$E_x = iA \frac{1}{\omega_0 \varepsilon_3} k_3 e^{i\beta z} e^{-k_z z}$$
$$E_z = -A \frac{\beta}{\omega_0 \varepsilon_3} e^{i\beta z} e^{-k_z z}$$

(4)

While in NOA, we get

$$H_y = Be^{i\beta z} e^{k_z z}$$
$$E_x = -iB \frac{1}{\omega_0 \varepsilon_2} k_2 e^{i\beta z} e^{k_z z}$$
$$E_z = -B \frac{\beta}{\omega_0 \varepsilon_2} e^{i\beta z} e^{k_z z}$$

(5)

where $k_i \equiv k_{z,i}, i = 1, 2, 3$ is the component of the wave vector perpendicular to the interface. In the Ag grating, the modes localized at the H$_2$O/Ag and Ag/NOA interface couple, yielding

$$H_y = Ce^{i\beta z} e^{k_z z} + De^{i\beta z} e^{-k_z z}$$
$$E_x = -iC \frac{1}{\omega_0 \varepsilon_1} k_1 e^{i\beta z} e^{k_z z} + iD \frac{1}{\omega_0 \varepsilon_1} k_1 e^{i\beta z} e^{-k_z z}$$
$$E_z = C \frac{\beta}{\omega_0 \varepsilon_1} e^{i\beta z} e^{k_z z} + D \frac{\beta}{\omega_0 \varepsilon_1} e^{i\beta z} e^{-k_z z}$$

(6)

The requirement of continuity of $H_y$ and $E_z$ leads to Eq. (7) at the H$_2$O/Ag interface (suppose $z = h$) and Eq. (8) at the Ag/NOA interface (suppose $z = -h$).

$$A e^{-k_z h} = C e^{k_z h} + D e^{-k_z h}$$
$$A \frac{1}{\varepsilon_3} k_3 e^{-k_z h} = -C \frac{1}{\varepsilon_1} k_1 e^{k_z h} + D \frac{1}{\varepsilon_1} k_1 e^{-k_z h}$$

(7)

$$Be^{-k_z h} = Ce^{k_z h} + De^{k_z h}$$
$$-B \frac{1}{\varepsilon_3} k_3 e^{-k_z h} = -C \frac{1}{\varepsilon_1} k_1 e^{k_z h} + D \frac{1}{\varepsilon_1} k_1 e^{k_z h}$$

(8)

$H_y$ further has to fulfill the wave equation $\frac{\partial^2 H_y}{\partial z^2} + (k_z^2 \varepsilon_1 \beta^2) H_y = 0$ in three distinct regions, via

$$k_i^2 = \beta^2 - k_0^2 \varepsilon_i, i = 1, 2, 3$$

(9)

Solving the system of linear equations results in the relation of $k_i$ via

$$e^{-k_i h} = \frac{k_1/\varepsilon_1 + k_2/\varepsilon_2}{k_3/\varepsilon_3} \times \frac{k_1/\varepsilon_1 + k_3/\varepsilon_3}{k_2/\varepsilon_2}$$

(10)

Results and Discussion

Coupling Mode

Since NOA133 has a refractive index of 1.33, which matches well with the H$_2$O index, it forms a symmetrical environment. We first analyze the coupling between SPPs of the H$_2$O/Ag/NOA/SiO$_2$ heterostructure with 60-nm Ag and NOA133. As the decay length of SPP in Ag is about 23.5 nm at $\lambda_0=785$ nm according to Eq. (3), very few plasmons penetrate this 60-nm thick Ag if there is no SPP coupling. It is convenient to study the influence of SPP coupling on SERS. The variation of reflectance, absorbance, and transmittance of SERS with incident wavelength are shown in Fig. 3.

It can be seen that strong coupling of SPPs occurs at 779 and 792 nm, where the reflectance curve shows two sharp troughs, while two sharp peaks appear in the corresponding transmittance and absorbance curves. These two wavelengths are not the SPP resonance wavelengths of the upper or lower interface, according to Eq. (1), but the resonance wavelengths of the heterostructure after interactions between SPPs. The electric field distributions of the SERS substrate at resonance wavelengths are shown in Fig. 4.

When $\lambda=779$ nm, $E_z(z)$, the component of $E_z$ perpendicular to the interface, is the anti-symmetric mode (AM, $E_z(z)$ at the two interfaces in phase) in the Ag grating. In this mode,
the SPPs of the upper and lower interfaces propagate back along the Ag grating surface and appear to repulse each other in the middle of the Ag layer. As a result, there is extremely little energy in the middle of Ag grating, but almost in the surrounding medium. The decay length of the evanescent field is large in the dielectric, which forms a wide range of EM enhancement both in the upper and lower dielectrics.

When $\lambda = 792$ nm, $E_z(z)$ is the symmetrical mode (SM, $E_z(z)$ at the two interfaces out of phase) in the Ag grating, where the SPPs of two interfaces propagate in the same direction along the Ag grating surface. In this mode, Ag grating has a strong bond on the EM field, and the decay length of the evanescent field is small in the dielectric. The SPP couplings of both modes are strong in such a structure where the EM of the lower interface is almost equal to that of the upper interface, with the maximum electric intensity ($E_{\text{max}} / E_{\text{0}}$) of 10 times.

**Influence of the Upper and Lower Dielectrics of the Ag Grating**

When the SERS substrate is used for trace detection, in order to absorb and concentrate the target molecules, specific substances can be added to the solution such as NaOH or NaCl when detecting explosive DNT. At the same time, in order to prevent the oxidation of the Ag grating, a thin layer of $\text{Al}_2\text{O}_3$ can be deposited on the surface of the Ag grating. These measures could improve the detection performance of the SERS substrate but would change the refractive index of the surrounding medium of the Ag grating.

We further studied the influence of the change of the refractive index of the medium surrounding the Ag grating on the electromagnetic field of the SERS substrate. The $\text{H}_2\text{O}/\text{Ag}/\text{NOA61}/\text{SiO}_2$ heterostructure is demonstrated as the SERS substrate, as NOA61 is commonly used with excellent glass and metal adhesion. The reflectance of SERS varies with the refractive index of the upper dielectric of Ag grating, as shown in Fig. 5.

It can be seen that the SERS substrate has two resonance frequencies due to the SPP excitation at the two interfaces. The SPP resonance wavelength of the lower interface ($\text{Ag/NOA61}$), about 918 nm, hardly changes, while the SPP resonance frequency of the upper interface ($\text{H}_2\text{O}/\text{Ag}$) red-shifts with the increase of the refractive index of the upper dielectric, as shown by the blue curve in the figure, which is consistent with the SPP dispersion Eq. (1). The higher the refractive index of the dielectric, the longer the SPP resonance wavelength.

When the refractive indices of the upper and lower dielectrics are similar or the same, the SPP coupling will increase, as shown by the lower reflectance at the SPP resonance wavelengths. On the contrary, when their refractive indices differ greatly, the SPP of the upper interface is strong at the resonance wavelength, while that of the lower interface is quite weak. Because the SPP coupling is weak or almost non-existent, the frequency and phase of the SPPs do not match.

The $E_z(z)$ in Ag grating at high resonance frequency is AM, while it is SM at the low resonance frequency, although $E_z(z)$ is quite weak at the lower interface when SPP coupling is weak. The frequencies of AM and SM are shown by the white dashed lines in the figure. When the upper and lower dielectrics have the same refractive index, the SPP resonance frequencies of the two interfaces are the same, but the AM and SM curves have an anti-cross at this resonance frequency. The anti-crossing of frequencies due to SPP coupling may be explained by classical [35], semi-classical, and fully quantum mechanical calculations [36].

We selected several special points (such as ①②③④) on the curves, and analyzed their electric field distributions, as shown in Fig. 6.
The $E_z(z)$ in Ag grating at the high resonance frequency (such as ①③) is AM, while it is SM at the low resonance frequency (such as ②④), although the SPP of the lower interface is much smaller than the SPP of the upper interface when the SPP coupling is weak.

When the SPP coupling increases at $\text{Ag} = 60 \text{ nm}$, the EM of the upper interface becomes smaller, while the EM of the lower interface becomes larger, as shown in Fig. 6 (③) and (④). When the refractive indices are the same, $E_z(z)$ even becomes odd or even vector in the Ag grating at SPP resonance wavelength. On the contrary, the electric field on the upper interface is much stronger when the SPP coupling is weak, which can be understood that the charge in the Ag grating is concentrated towards the upper interface, as shown in Fig. 6 (①) and (②).

We also investigated the influence of the lower dielectric on the interaction of SPPs, by using different NOA, as seen in Fig. 7.

Compared with NOA61 and NOA142, the refractive index of NOA133 is closer to that of H$_2$O, and the coupling of SPP is stronger at the SPP resonance wavelength, which results in a smaller EM of the upper interface. The EM of the SERS substrate with NOA61 is similar to that of the substrate with NOA142 at the SPP resonance wavelength.

**Influence of the Thickness of Ag Sinusoidal Grating**

In order to study the influence of the thickness of Ag sinusoidal grating on SPP coupling and EM, we further used H$_2$O/Ag/NOA133/SiO$_2$ and H$_2$O/Ag/NOA61/SiO$_2$ for analysis. The reflectance and absorbance of the substrate are shown in Fig. 8.

Since the skin depth of incident light in the Ag grating is small in the visible wavelength range, the thicker the Ag grating, the less the light penetration and the weaker the SPP excited at the lower interface, which leads to the lower coupling of SPPs. If the Ag grating is quite thick, even infinite ($h \to \infty$), there is no EM at the lower interface, let alone the coupling of SPPs, where Eq. (10) can be approximated as two uncoupled SPP wave equations at their respective interfaces, as shown in Formula (11).

\[
\frac{k_1}{\varepsilon_1} = -\frac{k_2}{\varepsilon_2} \\
\frac{k_1}{\varepsilon_1} = -\frac{k_3}{\varepsilon_3} 
\]

As shown in Fig. 8, when the Ag grating is thick (such as greater than 100 nm), the SM and AM wavelengths are the theoretical SPP resonance wavelengths of the interfaces.
which are the result of Formula (11). In this case, SPPs almost do not interact with each other.

In Fig. 8A, B, there is SPP coupling when the Ag is thin. It can be seen from Fig. 8A that the SPP coupling increases as the silver thickness decreases and the resonance frequency difference between the two modes becomes larger. The frequency of SM is red-shifted, while that of AM is blue-shifted, which is consistent with Eq. (10). From Fig. 8B, we can see that the absorption of AM is lower than that of SM. Because in AM, the plasmon is weakly localized, which results in the main part of the energy contained in the dielectric but not in the Ag grating. The transmittance increases in both modes, as shown in Fig. 9.

In Fig. 8C, as the refractive indices of upper and lower dielectrics are so different that the interaction of SPPs is quite weak, even when the Ag grating is thin. But $E_z(z)$ is also AM at high SPP resonance frequency, which is the same as SPP$_U$ and SM at SPP$_L$, even though SPPs are quite weak on the lower interface.

For a certain thickness of Ag grating, the transmittance peaks when SPPs are strongly coupled, such as in AM and SM. We also noted that the transmittance is larger in the AM than that in SM, mainly because the SM has a stronger ability to bind the electromagnetic field.

For an incident wavelength $\lambda$, the transmittance increases as Ag grating thickness decreases, as electromagnetic waves can penetrate the thin silver layer.

For SERS substrates of these two structures, the resonance frequency of the upper interface SPP is higher than that of the lower interface, according to Eq. (2). $E_z(z)$ is AM at this higher resonance frequency when SPP coupling is strong. The electric field distributions of the SERS substrate at the resonance frequency are shown in Fig. 10.

As shown in Fig. 10, the maximum EM of the SERS substrate increases in both modes with the Ag thickness, it may be that more plasmons gather in the upper interface in the thick Ag grating. When Ag is thick enough, such as greater than 100 nm, the maximum EM of the SERS substrate with different NOA will be similar or even the same.

Comparing Fig. 10A with B, AM has a larger electric field enhancement range in the surrounding medium than SM, and the main energy is in the medium rather than in the Ag grating. Comparing Fig. 10A with C, the SPP coupling between two interfaces of Ag grating increases the EM of the lower interface, but reduces the maximum EM of the SERS, when the Ag thickness is thin, such as 40–70 nm.

**Conclusion**

It has been shown that the interaction of grating-coupled SPPs significantly affects the SERS for Ag sinusoidal nanograting. The thickness of Ag grating and the surrounding medium has an important influence on SPP coupling and EM of the SERS. There are two modes of $E_z(z)$ in Ag grating, namely, AM at the high SPP resonance frequency and SM at the low SPP resonance frequency when there is SPP coupling between interfaces.

This interaction peaks at SPP resonance wavelength when the refractive indices of the upper and lower dielectrics of the thin silver grating are close. With the decrease of Ag thickness, the interaction of SPPs is gradually enhanced and the resonance frequency of AM is blue-shifted, while that of SM is red-shifted. Generally speaking, the transmittance of SERS substrate is largely enhanced in AM, while the
absorbance increases in SM. It can be said that the coupling of SPPs enhances the transmittance and the electric fields of the lower interface, but at the same time reduces the electric intensity on the upper interface.

As a consequence, the SERS is strongly dependent on the interaction of SPPs for Ag sinusoidal nanograting if the Ag grating is thin. The research may serve as a guideline for the design of multilayer metal/medium SERS substrate, as well as the substrate for the trace sensor used in microchannels.

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**Declarations**

**Consent to Participate** Not applicable.

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