Fabrication of Ag-modified nanocone frustum arrays with controlled shape as active substrates for surface-enhanced Raman scattering

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Abstract
Surface-enhanced Raman scattering (SERS) substrates utilizing the interparticle nanogaps as hot spots can provide ultrasensitive and geometry-dependent enhancement factors in analyte detection. Here, we report theoretical and experimental studies on the optical properties of Ag nanoparticles-decorated Si-nanocone frustum (NCF) arrays with various geometrical dimensions to enable engineering of the most effective SERS substrate. Our fabrication steps include electron beam lithography, deep reactive-ion etching, and metallization. In this regard, the effect of geometrical parameters including height, diameter, and slant angle, associated with Si-NCF arrays, on optical properties and SERS effect are investigated. Results show that the NCF array with larger height and slant angle exhibit higher antireflection property and excellent performance for low-concentration analyte molecules detection. Furthermore, our results show that the excitation of Mie resonances in NCF arrays leads to formation of near electric field distributions and enhancement of Si Raman lines. It is demonstrated that the strength of Si Raman line follows the trend of the integrated near electric fields in the excitation volume.

KEYWORDS
electron beam lithography, enhancement factor, nanocone frustum arrays, reflectance spectrum, surface-enhanced Raman spectroscopy

1 | INTRODUCTION

Surface-enhanced Raman scattering (SERS) is a powerful spectroscopy tool for chemical[1-3] and biomedical[4-6] detection and identification due to its large enhancement, high sensitivity, and rapid response.[7,8] This large enhancement arises from strong electromagnetic field localization in the vicinity of the metal nanoparticles (NPs),[9,10] called “hot spots,” which is established as the dominant mechanism of the SERS. According to the electromagnetic theory, the enhancement factor (EF) is proportional to the fourth power of the local electric field strength in hot spots, because both excitation and Raman scattering lights are enhanced.[11,12] Sharp tips[13] and closely packed NPs[14] improve the electromagnetic EF. Such nanometric structures can provide the capability of even a single molecule detection.[15,16]

Metal NPs coated on surface of the Si-based nanostructural substrates, such as nanocones[17,18] and nanowires,[19] form the high-performance SERS substrates. Several approaches have been utilized to fabricate these Si nanostructures including chemical reduction.
method,[20,21] wet chemical etching,[22,23] and inductively coupled plasma reaction ion etching.[24,25] Nevertheless, the random distribution of these Si nanostructures prevents any direct control of hot spots distribution and results in poor reproducibility and reliability. However, to enable SERS techniques for practical applications, SERS substrates need to be reproducible and highly sensitive.[26,27] Advanced lithographic techniques, such as ion beam or electron beam lithography[28–30] and nanosphere lithography,[31,32] in combination with reactive ion etching (RIE), are effective techniques that can produce well-controlled nanostructural arrays on Si substrate.[33] Deposition of metal NPs on the surface of these Si nanostructure arrays can provide high concentration of hot spots, which is essential for application of these substrates as SERS sensors.

In SERS measurement, the electric field strength reaches its maximum at the hot spots and drops sharply as the distance from the hot spots increases. A systematic study showed that only less than 1% of molecules resided on the hot sites (>10⁷) contribute almost 70% of SERS signal, and 61% of molecules resided on the cold sites (<10⁵) contribute only 4% of the SERS signal.[34] Therefore, substrates with higher concentration of hot spots facilitate residing more molecules on the hot sites and contribute to the enhancement of Raman signal. We have earlier shown that when Ag NPs are decorated on the Si nanostructure arrays, a high concentration of active sites is achieved and provides high-strength SERS substrates.[35] Moreover, the enhancement strength of these hybrid nanostructure arrays depends on the geometrical properties of the Si nanostructure array. However, there is no study concerning the relationship between geometrical dimensions and Raman EF for Ag NPs-modified Si nanostructure arrays.

Here, we report on the fabrication of great SERS substrates, Ag NPs-decorated Si-nanocone frustum (NCF) arrays, with various geometrical dimensions to engineer the most effective SERS substrate, which provide high electromagnetic field enhancement. These SERS substrates are prepared by electron beam lithography (EBL) together with RIE etching process and decorated with Ag NPs using electroless deposition technique. This fabrication process allows controlling the geometrical dimensions of these hybrid nanostructures and facilitates their study by using numerical simulation. The optical properties of the NCF arrays, with and without Ag decoration, are investigated both theoretically and experimentally. Results of SERS measurements for NCF arrays with different geometrical dimensions are discussed, and the optimum geometries for achieving highest EF are proposed. This information provides guidelines for the design and optimization of SERS substrates. Moreover, SERS capability of nanostructure arrays with highest EF is assessed using crystal violet (CV) solution with different concentrations.

2 | FABRICATION PROCEDURES

2.1 | Fabrication of silicon NCF array

Evolution of Si-NCF arrays is performed by means of a multistep process on p-type silicon (100) substrates. The process includes three important steps: (a) the EBL of a deposited resist, (b) deposition of Cr layer, and (c) plasma etching. The schematic of fabrication steps is illustrated in Figure 1 (all panels). First, hole arrays are patterned on the 300-nm thick layer of poly methyl methacrylate using EBL method. In the next step, a thin Cr film with an approximate thickness of 20 nm is deposited on the substrate as hard mask for the subsequent etching process. The fabrication process of Si-NCF arrays involves a deep RIE technique where a certain mixture of three gases of O₂, SF₆, and H₂ are used in an RIE unit with a 13.56-MHz radiofrequency plasma source, which operates at a low power density ranging from 30 to 300 W. The deep etching process involves two sequential steps, namely, etching and passivation. The etching step is achieved using merely SF₆ as a feed gas during a short period of time (9s) with a flow rate of 150 SCCM, whereas plasma power is set at 110 W. After every etching step, the reactor chamber is evacuated, and the mixture of hydrogen and oxygen gases with a trace of SF₆ is introduced into the reactor while plasma power is maintained at a value of 115–150 W. Indeed, this step provides an in situ protective mask over the sidewalls to protect them during the etching step. The etching/passivation subcycles are repeated several times to achieve the desired shape. Various shapes can be achieved by adjusting the parameters of these two steps such as plasma power, duration, and flow of gases. As detailed in Table S1, all the etching subcycle parameters are kept constant, whereas duration and plasma power of the passivation step are varied in order to achieve the desired shape. Figure 2 collects several scanning electron microscopy (SEM) images of Si-NCF arrays under various deep RIE process conditions. Figure 2c–e shows the morphological changes of nanostructures as a function of the total number of the sequences. Upon increasing cycle number, Si-NCFs’ height increase gradually. The base diameters of these nanostructures (Figure 2c–e) are maintained at about 145 ± 5 nm, whereas their height is increased. The shape of the Si nanostructures becomes tapered when the passivation step is intensified as shown in Figure 2f. This etching technique is a powerful method and can
accurately exert control on density, height, and thickness of Si-NCFs. This feature is of course very appealing because it provides the tuning required in optical properties. Furthermore, the diameter of each Si-NCF is accurately defined by initial lithographic mask as shown in Figure 2a–c.

2.2 | Chemical deposition of silicon NCF array

Decoration of Ag NPs on the side walls of Si-NCF arrays is performed using electroless deposition method. The NCF arrays are plunged into an aqueous solution AgNO$_3$/HF (0.5 mM AgNO$_3$, 9% HF) for 20-s time intervals at room temperature. According to our previous work, deposition time of 20 s provides more total number of hot spots and consequently higher enhancement power. Thereafter, samples are immediately rinsed with deionized water and dried. The SEM images of NCFs/Ag NPs arrays with different geometrical dimensions are shown in Figure 3. It should be noticed that this deposition method leads to formation of Ag NPs with larger size on the top of NCFs and smaller ones at the sidewalls.

2.3 | SERS measurements

To experimentally interrogate the Raman enhancement property of the fabricated silicon nanostructure arrays, CV molecules are used as the target analyte. For this aim, first, 15 μl of 1-μM CV solution is dropped on each substrate and let dry. Then, the Raman spectra are acquired using a Raman microscope spectrometer (Teksan_N1-541, Iran) with the 532-nm excitation wavelength.

2.4 | FDTD simulation method

Finite difference time domain (FDTD) simulation is used to investigate the optical properties of the fabricated nanostructures to understand their ability to enhance the local electric field. The reflection spectra and near-field distributions are calculated. Optical constants of all materials are taken from Palik handbook. Periodic boundary conditions are used in $x$ and $y$ directions to reduce the amount of computation. To eliminate the spurious reflections, perfectly matched layer boundary condition is used in the $z$ direction. Samples were illuminated using a linearly polarized plane wave light source.
FIGURE 2 70° tilt view scanning electron microscopy (SEM) images of nanocone frustum (NCF) arrays with periodicity of 600 nm and geometrical dimensions of top radius, bottom radius, and height, respectively: (a) 70, 120, and 440 nm; (b) 75, 130, and 440 nm; (c) 85, 145, and 440 nm; (d) 85, 145, and 560 nm; (e) 85, 145, and 1,000 nm; and (f) 25, 145, and 440 nm. Insets show the magnified images.

FIGURE 3 70° tilt view scanning electron microscopy (SEM) images captured from nanocone frustum (NCF) arrays in Figure 2 decorated with Ag nanoparticles (NPs). Deposition period was 20 s. Insets show the magnified images.
with normal incidence to the substrate, which has propagation direction opposite to the \( z \) direction. Two-dimensional monitors are used to calculate the electric field intensity distribution.

3 | RESULTS AND DISCUSSION

3.1 | Optical response characteristics of NCF arrays

In this section, characterization of the optical properties of NCF arrays with various geometrical dimensions is done, which lead to different optical responses over the desired wavelength range (400–800 nm). In fact, by employing various etching processes, one can fabricate Si-NCF arrays with different shapes, heights, and diameters. The schematic illustration of the fabrication process is shown in Figure 1. As seen in this figure, the NCF array has five geometrical parameters (including top radius \( r_1 \), base radius \( r_2 \), height \( H \), slant angle \( \theta \), and lattice constant), all of which can be controlled during the fabrication process. Following this, we discuss about optical response of the fabricated NCF arrays experimentally and theoretically.

3.1.1 | The effects of variation in height

To investigate influence of the height on the optical properties, we have fabricated three NCF arrays with almost the same base and top radius (average of 145 ± 5 nm and 85 ± 5 nm, respectively) at a constant pitch of 600 nm but with different heights including 440 nm, 560 nm, and 1,000 nm. The SEM images of these fabricated NCF arrays with different heights are shown in Figure 2 c–e. Their reflectance spectra are shown in Figure 4a. The results indicate that as the height of the Si-NCF arrays increases, the antireflection property of such nanostructures also increases. This fairly complies with results previously reported elsewhere.\(^{[39]}\) In other words, all NCF arrays with different heights show reduced reflectivity over the entire spectral range, with respect to the bare Si wafer. As seen, the reflectance spectrum of NCF array

![Figure 4](attachment:image_url)
with height of 1000 nm is less than 10% over a broad wavelength range, which has practical applications in photovoltaics, solar cells, optical emitters, and detectors. The optical reflection spectra of these dielectric nanostructures are also simulated, and results are shown in Figure 4b. It is worth mentioning that the observed disagreements between the simulated and experimental reflection spectra are due to some differences between NCFs’ topology and geometrical dimensions of the simulation unit cell and fabricated nanostructure.

Figure 4c depicts the Raman spectrum for each NCF array measured at 532 nm excitation wavelength. Clearly, Raman signal is enhanced when the height of the NCF array increases. Because the optical response is a complex combination of multiple resonances, the near electric and magnetic fields are simulated to facilitate understanding of the nature of excited resonant modes near excitation wavelength (532 nm), which leads to the Raman signal enhancement (shown in Figure S1). It can be seen that the Si-NCF arrays can support both magnetic and electric resonances, and the magnetic field is mostly confined inside the NCFs; however, the electric field is observed both inside and on the sidewalls of the NCFs. Accordingly, the Raman signal enhancement is attributable to the resonant increase of internal electric fields inside the NCFs due to the excitation of Mie resonant modes. The strong forward coupling of light from resonance modes of Si-NCFs into the high index substrate (as shown in Figure S1) causes the reduction in the reflected power. More interaction among pillars is evident from near distribution of electric field for NCF array with higher height. It is seen that the interpillar interactions have strong influence on the amount of absorption.

The Raman scattering intensity depends on the two parameters, incident laser excitation intensity and the amount of inelastically scattered light. For 532-nm excitation and Si material, wavelength of the Stokes scattered light is 547 nm. In the case of nanostructures, both incident and scattered light are subject to the near electric field enhancements, and this double enhancement leads to the Raman signal enhancement. The amount of signal amplification can be calculated by integrating the product $|E_{532}|^2 \cdot |E_{547}|^2$ within the excited volume:

$$I \propto \int E_{532}^2 dV \cdot \int E_{547}^2 dV.$$  \hspace{1cm} (1)

The enhancement power of each NCF array is calculated on the basis of the above equation, and results are shown in Figure 4d. As seen, there exists the same behavior between theoretical and experimental results. It is seen that the more the height increases, the more the amount of Si Raman increases.

3.1.2 | The effects of variation on slant angle

The slant angle is another geometrical parameter that affects the optical properties. To study the optical response as a function of the slant angle, we have fabricated two NCF arrays with different slant angles. The height and the base diameter of each NCF array are maintained at 440 ± 10 nm and 145 ± 5 nm, respectively, with a constant pitch of 600 nm. The first NCF array has a 90-nm top radius and 7° slant angle. The second one has a 25-nm top radius and 16° slant angle. The SEM images of the two NCF arrays are shown in Figure 2 a,c. The reflectance spectra of these two types NCF arrays are measured and shown in Figure 5a. The reflectance spectra are also simulated using FDTD method and are illustrated in Figure 5b. Again, good agreement is observed between theoretical and experimental results. In contrast to the NCF array with lower slant angle, the tapered NCF array with slant angle of 16° shows improved antireflection property in specific wavelength intervals. Actually, for any given wavelength, an optimum resonance condition can be established when the diameter of NCF cross section is decreased from base to the apex. Therefore, more optical mode coupling can occur for NCF array with higher slant angle, and a decrease in reflectance is observed. The Raman spectra are measured at 532-nm incident excitation wavelength to understand the enhancement strength of each NCF array, and results are shown in Figure 5c. From the acquired results, it can be concluded that the Raman signal is enhanced when the slant angle of NCF array increases. The enhancement strength of each type NCF array is also calculated according to the previous explanation, and results are shown in Figure 5d. As seen, the enhancement power is increased by an increase in slant angle, confirming the experimental results.

3.1.3 | The effects of variation on the base diameter

In order to assess the effect of base radius on the light trapping properties, three NCF arrays with the same height (with an average value of 440 ± 10 nm) but with base radii of 120, 130, and 145 nm have been fabricated. The SEM images of these NCF arrays are shown in Figure 2a–c. The calculated and measured reflectance spectra are shown in Figure 6a,b. A quite consistent trend is observed between simulated and experimental reflection spectra. According to this figure, the reflectance spectrum is red shifted when the diameter is increased. The measured Raman signals of these nanostructures show...
that the NCF array with base diameter of 130 nm provides the highest Raman signal enhancement. The calculated integration of near electric fields for these three substrates is shown in Figure 6d, which confirms the experimental results. These results indicate that the reflection dip of the NCF array having a diameter of 130 nm provides the best overlap with the excitation (532 nm) and Raman line (547 nm) wavelengths, hence the maximum enhancement is achieved on this substrate.

3.2 | SERS response characteristics of Ag-modified NCFs array

To experimentally investigate the SERS capability of the fabricated NCF/Ag NPs arrays with various geometries, CV is utilized as a target analyte. Geometrical parameters, including height, angle, and base diameter, influence the strength of SERS spectra. There is great interest to investigate how these parameters affect the SERS EF. In this section, we study the effect of geometrical dimensions on the enhancement capability of the substrates. As already explained, all structures are deposited under the same condition (20-s time deposition). Then, 15 μl of 1-μM CV solution is dropped onto all these substrates and left to dry. The SERS measurements are performed using incident excitation wavelength of 532 nm with power of 0.5 mW and integration time of 1s.

3.2.1 | The effects of variation on the height

The NCF arrays shown in Figure 2c–e are Ag deposited under the same condition to study the influence of the height on the amount of EF. The SEM images of these Ag NPs/NCF arrays are shown in Figure 3c–e. To characterize the optical properties of these nanostructures, their reflectance spectra are measured. Results are shown in Figure 7a. As reported in our previous work, larger size Ag NPs lye on the top of NCFs, whereas smaller size Ag NPs lye on the NCFs sidewalls. In order to envisage the
effect of changes in the NPs size, an Ag thin layer is assumed in a way that its thickness decreases from 40 nm (at the tip) to 1 nm (at the base). The size of NPs was measured using the SEM images. The simulated reflectance spectra are demonstrated in Figure 7b. A good agreement exists between the experimental and the numerically calculated data. These results indicate that the reflection intensity decreases as the height of the Ag-modified NCF arrays increases.

The SERS spectra of the substrates with different heights are measured for the same volume and concentration of CV solution (15 μl and 1 μM, respectively). Results are shown in Figure 8a. As can be seen, the SERS intensity has increased as the height of the NCFs increases from 440 to 1,000 nm. Actually, the SERS intensity is proportional to the number of Ag NPs, which increases with the height of NCFs. These Ag NPs play as active sites, which contribute to the enhancement of the Raman signal. The intensity of Raman line at wavenumber of 1,191 cm\(^{-1}\) and its EF as a function of height are plotted in Figure 8b. Results confirm that the increase of nanocone height corresponds to the generation of higher density of hot spots, which is responsible for higher electromagnetic enhancement. Thus, a higher density of NPs is essential to achieve higher SERS EF.

3.2.2 | The effects of variation on the slant angle

In this section, the effect of the slant angle on the amount of EF is evaluated. To do this, two NCF arrays with two different slant angles of 7° and 16° are deposited under 20-s time deposition. The SEM images of two Ag NPs-deposited NCF arrays are shown in Figure 3c.f. To characterize the optical properties, the reflectance spectra of these two types of NCF arrays are measured and demonstrated in Figure 7c. The reflectance spectra are also simulated and illustrated in Figure 7d. Again, a good agreement is observed between the theoretical and experimental results. As seen, the tapered NCF array with slant angle of 16° reduces the reflectivity over the entire spectral wavelength range.
To characterize the angle dependence of SERS EF, the SERS spectra are measured under the same condition as explained in the previous section (shown in Figure 8c). Results show that the NCF array with the larger slant angle creates more EF. The Raman intensities at Raman line of 1,191 cm\(^{-1}\) with their corresponding EFs are demonstrated in Figure 8d. It is seen that the Raman intensity is higher for the nanocone array. In fact, the NCF array with sharp tips creates more SERS active sites for scattering light, because after metal deposition, the sharpness of nanocones creates hot spots for SERS due to “lightning rod effect.” Furthermore, high density of silicon nanocones with sharp tips provide more active sites per unit surface area, which is the most important factor determining the amount of EF in this substrate work.
3.2.3 The effects of variation on the base diameter

In order to study the influence of the base diameter on the SERS signal, Ag NPs with three different base diameters are deposited under the same conditions already explained in Section 2.2. The SEM images of these NCF/Ag NPs arrays are shown in Figure 3c,e,f. As seen, the Ag NPs on the NCF arrays with higher diameter (130 and 145 nm) are relatively uniform, but for a small diameter (120 nm), the NCF size is short for Ag NPs to form the particle-like shape. Figure 7e,f illustrates the calculated and measured reflectance spectra of these NCF arrays with the same period and the same height but different diameters; geometrical parameters are given in Section 3.1.3, and a similar trend is observed between simulated and experimental reflection spectra. Afterwards, the SERS spectra are measured for the same volume and concentration of CV solution, and results are shown in Figure 8e. As can be seen, the SERS intensity is increased as the base diameter of the NCFs increases, which corresponds to an increase in the number of Ag NPs. The SERS intensity and EF as a function of base diameter are plotted in Figure 8f. It is seen that the Raman signal increases with increasing diameter.

The reported results provide practical guidelines for designing nanostructures with higher EF. Actually, the
EM field enhancement can be controlled by tuning the geometrical parameters of the Ag-modified NCF arrays.

3.3 | Sensitivity evaluation

As noted in the previous section, the NCF array with the height of 1,000 nm and NCF array with slant angle of 16° provides the highest SERS signal. Sensitivity of these two substrates is evaluated by employing an excitation power of 2 mW and integration time of 10 s. To this end, 15 μl of the CV solution with different concentrations including 10 pM, 10 nM, and 1 μM is dropped on each substrate. The SERS spectra obtained under the above conditions are shown in Figure 9a,b. We have precisely calculated the EFs of these substrates by comparing the intensity of Raman line at wavenumber of 1,191 cm\(^{-1}\) with the reference one. In this calculation, the fluorescence baseline is removed using an iterative multipolynomial algorithm to acquire the exact intensity of spectral peaks.\(^{[44]}\) The experimental EF is calculated by:\(^{[45]}\)

\[
\text{EF} = \frac{I_{\text{SERS}}}{I_{\text{ref}}} \frac{N_{\text{surf}}}{N_{\text{bulk}}},
\]

where \(I_{\text{SERS}}\) is the surface-enhanced Raman intensity of the selected peak, \(N_{\text{surf}}\) is the number of molecules contributing to the measured SERS signal, \(I_{\text{ref}}\) is the Raman intensity of the selected peak from the reference spectrum, and \(N_{\text{bulk}}\) is the number of molecules contributing to the measured reference signal. The number of molecules \(N\) is calculated as

\[
N = \frac{N_A C V A_{\text{laser}}}{A_{\text{spot}}},
\]

where \(N_A\) is the Avogadro's number, \(A_{\text{laser}}\) is the area of the excitation laser spot, \(A_{\text{spot}}\) is the total area of the CV

FIGURE 9  The surface-enhanced Raman scattering (SERS) spectra of crystal violet (CV) molecules for the concentration of 10\(^{-6}\), 10\(^{-8}\), and 10\(^{-11}\) M on the nanocone frustums (NCFs)/Ag nanoparticles (NPs) array with (a) height of 1,000 nm and (b) slant angle of 16°. The log-log plot of the intensity and enhancement factor (EF) at Raman line of 1,191 cm\(^{-1}\) versus CV concentration for NCFs/Ag NPs array with (c) height of 1,000 nm and (d) slant angle of 16° [Colour figure can be viewed at wileyonlinelibrary.com]
molecules on the substrate, $C$ is the molar concentration of the CV solution, and $V$ is the volume of the CV drop.

It is observed that the Raman lines of CV molecules can be identified from SERS spectra of both substrates down to $10^{-11}$ M. Therefore, these NCF/Ag NPs arrays are suitable SERS substrate with very high sensitivity. The intensity and the calculated EFs of Raman line at wavenumber of 1,191 cm$^{-1}$ are shown in Figure 9c,d (for different concentrations), and the results are summarized in Table 1. It is seen that the maximum SERS EF is $5.97 \times 10^9$ at $10^{-11}$ M CV solution for NCF array with a height of 1,000 nm. Our results indicate that the NCF/Ag NPs array with higher height and slant angle provides higher EF and consequently higher sensitivity for SERS measurements.

### 4 | CONCLUSION

In summary, we described the fabrication of NCF arrays with different geometrical dimensions and explained their SERS applicability when modified with Ag NPs. First, the optical response of each bare Si-NCF array was studied both theoretically and experimentally. Results showed that the excitation of Mie resonances leads to the Raman signal enhancement, and the amount of experimental Raman amplification of each NCF array was in good agreement with the theoretical ones. Our results indicated that the NCF array with larger height and slant angle will serve as a substrate with better anti-reflection property. Furthermore, the SERS applicability of the manufactured substrates was studied when they are decorated with Ag NPs. It was demonstrated that NCF/Ag NPs array with larger height and slant angles provide more density of active sites and consequently higher EF, which is very important for practical applications. Finally, it was demonstrated that EF in the order of $10^9$ and limit of detection of $10^{-11}$ M are possible using this technique.

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