



Full Length Article

## Platform for surface-enhanced Raman scattering in layered quantum materials

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## ABSTRACT

Raman spectroscopy has been used to study transition metal dichalcogenides (TMDs), quantum materials with promising properties. However, monolayer (1L) TMDs have limited observable Raman modes due to low light absorption. In this study, we propose a potential platform that can effectively enhance Raman scattering and increase the number of observable Raman modes in 1L TMDs. To enhance Raman scattering in 1L TMDs, a platform was fabricated by forming large-scale periodic arrays of gold micropillars (MPs) using conventional photolithography followed by gold film deposition. 1L WSe<sub>2</sub> was transferred onto the Au MPs and Raman scattering was observed. The Raman intensity of 1L WSe<sub>2</sub> on Au MPs was 19 times higher than that of 1L WSe<sub>2</sub> on SiO<sub>2</sub> plates, and Raman modes difficult to detect in typical 1L WSe<sub>2</sub> are observed. Confocal Raman spectroscopic mapping revealed that the strong local Raman enhancement at the edge of the Au MP resulted in the amplification of Raman scattering in 1L WSe<sub>2</sub> on Au MP. This local field enhancement was theoretically verified using finite difference time domain (FDTD). The platform has industrial advantages and wide applicability due to its low cost, simple process, large controllable area, and short process time.

### 1. Introduction

Transition metal dichalcogenides (TMDs) are a class of layered quantum materials that have attracted significant attention for various applications such as electronics, optoelectronics, spintronics, sensors, and quantum computing owing to their unique structural, electronic, and optical properties [1–10]. In particular, when the TMD is a monolayer (1L), it displays direct bandgap properties, strong light–matter interaction, enhanced quantum effects, flexibility, and scalability [11–20]. As the applicability of TMDs increases, significant attention is being paid to Raman scattering. It can accurately and comprehensively observe and analyze the physical properties of TMDs [21–28]. Because Raman scattering is the inelastic scattering of photons by materials, multiple Raman modes are observed in multilayer or bulk TMDs [29–33]. However, in the case of 1L TMDs, the number of observable Raman modes and Raman intensities are significantly smaller than those for multilayer TMDs because of the low light absorption due to ultra-thin two-dimensional (2D) crystal lattice in 1L TMDs [34–36]. To overcome this limitation, Raman scattering enhancement of 1L TMDs by coupling

with metal surface plasmons has recently been reported [37,38]. Yu *et al.* demonstrated Raman spectral enhancement in 1L MoS<sub>2</sub> by coupling plasmons with plasmonic nanogrooves [39]. Su *et al.* reported surface-enhanced Raman scattering of 1L TMDs on Ag nanorod arrays [40]. Using tip-enhanced Raman spectroscopy, Farhat *et al.* investigated the enhancement of Raman scattering in 1L MoS<sub>2</sub> by decorating it with gold nanoparticles [41]. However, the research results reported have been constrained by the requirement of expensive nanoscale processes, unavailability of controllable large-area fabrication, Raman scattering enhancement of less than 10 times, and non-observation of several Raman modes typically observed in multilayer TMDs.

In this paper, we propose an inexpensive, simple, and controllable large-scale substrate platform that can effectively enhance the Raman scattering of 1L TMDs. This platform enhanced the Raman scattering of 1L WSe<sub>2</sub>, which is one of the most widely used TMDs, by approximately 20 times. Furthermore, the platform allows for the observation of Raman modes that are difficult to observe in a typical 1L WSe<sub>2</sub>. The substrate platform was fabricated by creating periodic Au micropillars (MPs) by conventional photolithography, followed by deposition of an Au film. 1L

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WSe<sub>2</sub> was grown on an SiO<sub>2</sub> substrate using chemical vapor deposition (CVD). The surface morphology of periodic Au MP in a square array and 1L WSe<sub>2</sub> transferred to Au MP by wet transfer was verified by field emission scanning electron microscopy (FE-SEM). The thickness of the 1L WSe<sub>2</sub> and the diameter, height, and spacing of the Au MPs were verified by atomic force microscopy (AFM). Using Raman spectroscopy, the Raman spectrum of 1L WSe<sub>2</sub> was obtained for different substrate types. The number of observable Raman modes was compared and verified. Using confocal Raman spectroscopy mapping, we verified the specific region of 1L WSe<sub>2</sub> on the Au MPs that resulted in Raman scattering enhancement. Finally, the Raman scattering enhancement of 1L WSe<sub>2</sub> on Au MP was verified theoretically using finite-difference time-domain (FDTD).

## 2. Experimental

### 2.1. Synthesis of 1L WSe<sub>2</sub>

Using a two-zone furnace CVD technique, the 1L WSe<sub>2</sub> was deposited on an SiO<sub>2</sub>/Si substrate. In zone 1, the Se pellets were loaded to evaporate in the chamber. The cleaned SiO<sub>2</sub>/Si substrate was then spin-coated with a tungsten precursor. The substrate coated with the W precursor was placed in the center of zone 2. Zone 1 was heated to 380 °C for Se evaporation. Meanwhile, zone 2 was heated to 780 °C, where the W precursor was placed. After 1L WSe<sub>2</sub> synthesis, the furnace was cooled rapidly to room temperature. All the growth processes proceeded under N<sub>2</sub> and H<sub>2</sub> atmospheres at flow rates of 500 and 6 sccm, respectively. All the growth process were proceeded under N<sub>2</sub> and H<sub>2</sub> atmosphere at flow rates of 500 and 6 sccm, respectively.

### 2.2. Fabrications for 1L WSe<sub>2</sub> on Au MPs

A SiO<sub>2</sub> substrate with a clean surface was prepared using conventional chemical cleaning. Maskless photolithography (LithoMaskless, Standard Science Inc.) was used to form the photoresist (P/R) MPs on the cleaned SiO<sub>2</sub> substrates. Radio frequency sputtering was used to deposit a 100 nm-thick Au film over the entire surface of the P/R MPs on the SiO<sub>2</sub> substrate to obtain the Au MPs. CVD-grown 1L WSe<sub>2</sub> was transferred to Au MPs using the PMMA-assisted wet transfer method.

### 2.3. Characterization

FE-SEM (S-4800, Hitachi) was performed to observe the surface morphology of the 1L WSe<sub>2</sub> on the Au MPs. Atomic force microscopy (AFM (XE-100, Park system)) was used to measure the topography of the Au MPs and 1L WSe<sub>2</sub> in the non-contact mode. Raman spectroscopy was performed using a commercial confocal Raman system (LabRam HR Evolution, HORIBA) equipped with a 532 nm laser, a 100 × objective (N. A. = 0.90), and an 1800-groove grating. The spatial resolution of the 2D Raman mapping images is approximately 300 nm. Photoluminescence spectroscopy was performed using the commercial equipment used to perform Raman spectroscopy. The excitation source was a 532 nm laser, and a 150-groove grating was applied.

### 2.4. FDTD simulation

Lumerical (ver. 2023R1, ANSYS) FDTD solutions were used for the FDTD simulations. A perfectly matched layer method was used for the boundary conditions. The local electric field in the edge region of the Au MP was first calculated. Then, the enhancement factor was evaluated as  $EF = |E/E_0|^4$ . The excitation wavelength was 532 nm. The incidence normal to the Au substrate was also considered.

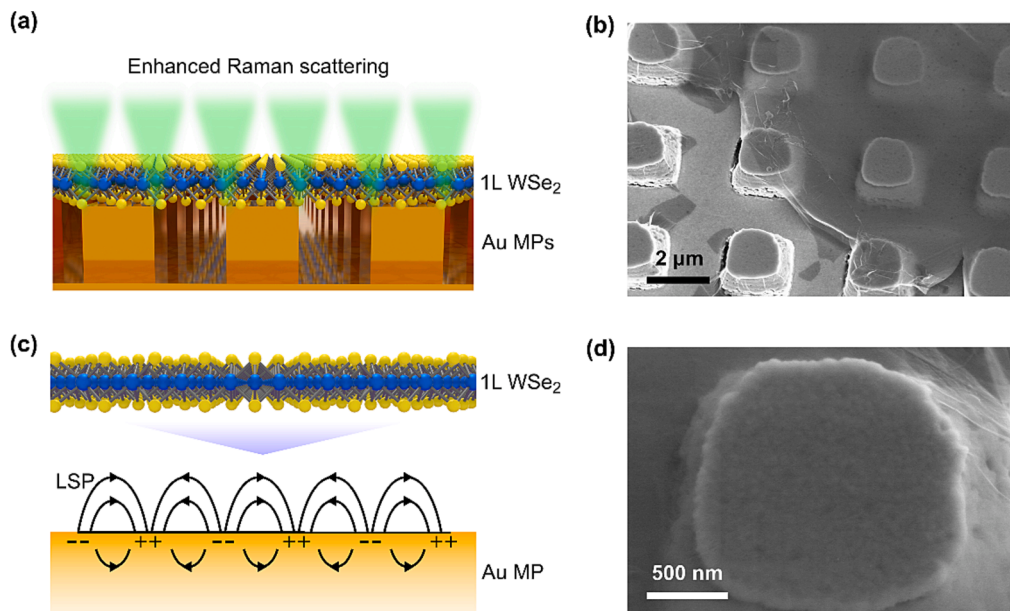
## 3. Results and discussion

### 3.1. Integrated structure of 1L WSe<sub>2</sub> and large-scale periodic Au MP

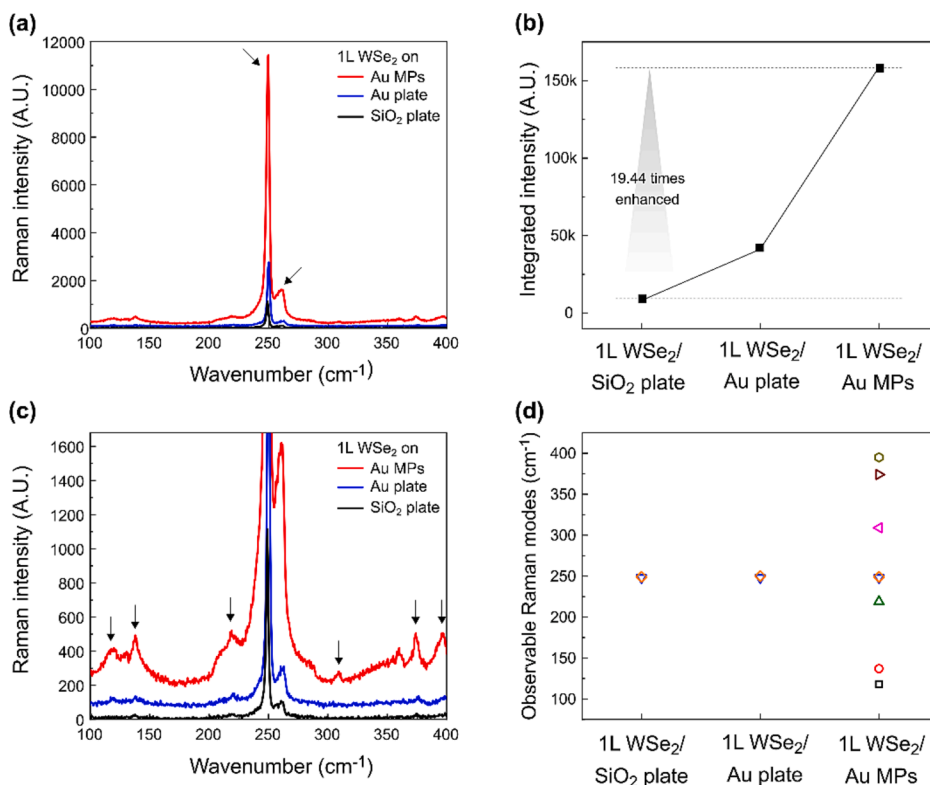
Large-scale periodic Au MP arrays were fabricated using conventional photolithography and gold film deposition on SiO<sub>2</sub> substrates. The Au MPs were arranged in a square array with uniform size and spacing. The surface edges of the Au MPs were sharp because the Au film was deposited after the MPs were formed. (Fig. S1) The diameter and height of the Au MPs were 2 μm and 1 μm, respectively, and the distance between individual Au MPs were 2 μm (Fig. S2). CVD-grown 1L WSe<sub>2</sub> has a triangular shape and an average lateral size of 35 μm. Therefore, there are approximately 50 Au MPs under one flake. (Fig. S3) The thickness of the 1L WSe<sub>2</sub> was verified to be 0.7 nm by AFM topography. This value is consistent with the thickness of a typical 1L WSe<sub>2</sub>, as reported previously (Fig. S4) [30,42–45]. CVD-grown 1L WSe<sub>2</sub> was transferred onto the Au MP array using the wet transfer method. During the transfer process, the 1L WSe<sub>2</sub> was subjected to a vertical downward force on the Au MPs. This resulted in a strong pulling force at the edge region of the Au MPs. This force caused the distance of 1L WSe<sub>2</sub> from the edge of the Au MP to be smaller than the van der Waals gap. Subsequently, this induced coupling of the surface plasmons localized at the edge of the Au MP. This, in turn, enhanced the Raman scattering of 1L WSe<sub>2</sub>. Fig. 1a shows a three-dimensional (3D) illustration of the selectively enhanced Raman scattering of 1L WSe<sub>2</sub> on Au MPs. The Raman scattering of 1L WSe<sub>2</sub> could be enhanced at specific locations on the edge of the Au MP. Moreover, this enhanced Raman scattering enabled the observation of Raman modes that are challenging to observe in monolayers. Fig. 1b displays an SEM image of 1L WSe<sub>2</sub> transferred onto an Au MP array. The 1L WSe<sub>2</sub> transferred by wet transfer cover several Au MPs and is strongly attached to the edge region of the Au MPs. Because of this strong attachment, the distance between the 1L WSe<sub>2</sub> and the Au surface at the edge region of the Au MP can be shorter than the distance between the 1L WSe<sub>2</sub> and a flat surface. Fig. 1c presents a schematic illustration of the localized surface plasmon (LSP) on the Au MP and an approaching 1L WSe<sub>2</sub>. The probability of surface plasmon coupling increases as the distance between the 1L WSe<sub>2</sub> and the surface of the Au MP decreases. This occurs because the intensity of the LSP electromagnetic field decreases exponentially with an increase in the distance from the Au surface. When the LSPs of the Au MPs interact with the 1L WSe<sub>2</sub> by reducing the distance between the 1L WSe<sub>2</sub> and Au MP, these induce a variation in the optical properties of the 1L WSe<sub>2</sub>, including Raman scattering. An SEM image of the surface of the 1L WSe<sub>2</sub> on an individual Au MP is shown in Fig. 1d. The wrinkles in the 1L WSe<sub>2</sub> at the upper right corner of the image indicate that the 1L WSe<sub>2</sub> is being subjected to an outward pulling force applied by the Au MP.

### 3.2. Enhanced Raman scattering of 1L WSe<sub>2</sub> on Au MPs

In order to investigate the phonon modes of 1L WSe<sub>2</sub> on Au MPs, Raman spectroscopy was performed. The excitation source for Raman scattering was a continuous-wave (CW) laser with a wavelength of 532 nm. Fig. 2a shows the Raman spectra of the 1L WSe<sub>2</sub> according to the substrate type. A SiO<sub>2</sub> plate is commonly used as a substrate for growing 1L TMD because SiO<sub>2</sub> is inexpensive material and electrical insulator. It was employed as the reference substrate. In addition, an Au plate was used as a substrate to compare the effects of the structural features of the Au MPs. The red, blue, and black solid lines represent the Raman spectra of 1L WSe<sub>2</sub> on the Au MPs, Au plates, and SiO<sub>2</sub> plates, respectively. The Raman mode showing the strongest intensity is the peak of the combination of E' and A'1 appearing at approximately 250 cm<sup>-1</sup>. The next strongest Raman mode is the 2LA peak at approximately 260 cm<sup>-1</sup> as indicated by the black arrows. As shown in Fig. 2a, the Raman intensity of 1L WSe<sub>2</sub> on the Au MPs is significantly higher than that of 1L WSe<sub>2</sub> on the SiO<sub>2</sub> plate as well as that on the Au plate. This indicates that the Raman scattering of 1L WSe<sub>2</sub> was enhanced by the interaction between



**Fig. 1. 3D Schematics and SEM Images of 1L WSe<sub>2</sub> on Au MPs.** (a) Three-dimensional (3D) illustration of the enhanced Raman scattering of 1L WSe<sub>2</sub> on Au MPs. The Raman scattering of 1L WSe<sub>2</sub> is enhanced at the edge region of Au MPs owing to LSP resonance. (b) SEM image of 1L WSe<sub>2</sub> transferred onto Au MPs by wet transfer method. The wet transfer method enables the transfer of 1L WSe<sub>2</sub> onto a large-area Au MP array. (c) 3D schematic of the interaction between 1L WSe<sub>2</sub> and an Au MP containing LSPs. As the 1L WSe<sub>2</sub> approaches the Au MP, the LSPs of the Au MP induce a transition in the optical properties of the 1L WSe<sub>2</sub>, including Raman scattering. (d) SEM image of 1L WSe<sub>2</sub> on an individual Au MP. The 1L WSe<sub>2</sub> is subjected to an omnidirectional pulling force on the individual Au MPs. This minimizes the distance between the 1L WSe<sub>2</sub> and the Au MPs in the edge region of the Au MPs.



**Fig. 2. Raman spectroscopy of 1L WSe<sub>2</sub> on different substrates.** (a) Raman spectra of 1L WSe<sub>2</sub> on Au MPs, Au plate, and SiO<sub>2</sub> plate. The Raman intensity of 1L WSe<sub>2</sub> was enhanced significantly on the Au MPs compared with that on the Au and SiO<sub>2</sub> plates. (b) Magnified Raman spectra of 1L WSe<sub>2</sub> on different substrates with the y-axis magnified. Significantly more Raman modes are observed in the Raman spectrum of 1L WSe<sub>2</sub> on the Au MPs compared with that of 1L WSe<sub>2</sub> on the Au and SiO<sub>2</sub> plates. (c) Integrated intensity plot of the spectral region from 100 cm<sup>-1</sup> to 400 cm<sup>-1</sup> in the Raman spectrum of 1L WSe<sub>2</sub> on Au MPs, Au plate, and SiO<sub>2</sub> plate. The integrated intensity of 1L WSe<sub>2</sub> on the Au MPs is 19 times higher than that of 1L WSe<sub>2</sub> on the SiO<sub>2</sub> plate. (d) Plot of Raman modes observed in 1L WSe<sub>2</sub> on different substrates. Three Raman modes are observed for 1L WSe<sub>2</sub> on the SiO<sub>2</sub> and Au plates, whereas nine Raman modes are observed for 1L WSe<sub>2</sub> on the Au MPs.

1L WSe<sub>2</sub> and the Au MPs. To quantify the Raman scattering enhancement, the intensity of each spectrum was integrated in the range from 100 cm<sup>-1</sup> to 400 cm<sup>-1</sup>. Fig. 2b is a plot of the integrated intensity of each Raman spectrum. It is evident that the integrated intensity of 1L WSe<sub>2</sub> on the Au plate increased marginally compared with that on the SiO<sub>2</sub> plate. However, the integrated intensity of 1L WSe<sub>2</sub> on Au MPs increased substantially compared with the other two samples. In particular, the integrated intensity of 1L WSe<sub>2</sub> on the Au MPs increased by 19 times compared with that of 1L WSe<sub>2</sub> on the SiO<sub>2</sub> plate. This enhancement was significantly higher than that reported previously [46,47]. It can result in the appearance of peaks concealed by background noise as well as the enhancement of the main peaks. To verify the appearance of the concealed Raman modes, Fig. 2c shows the magnified Raman spectra. Several low-intensity peaks are observed in the Raman spectrum of 1L WSe<sub>2</sub> on Au MPs. These are not present in the Raman spectra of 1L WSe<sub>2</sub> on the Au plate and 1L WSe<sub>2</sub> on SiO<sub>2</sub> plate. The Raman modes observed only in the Raman spectrum of 1L WSe<sub>2</sub> on Au MPs are E'(M)<sup>LO2</sup>-LA(M) at 118 cm<sup>-1</sup>, A'<sub>1</sub>(M)-LA(M) at 137 cm<sup>-1</sup>, LA(M) + TA(M) at 219 cm<sup>-1</sup>, B<sub>2g</sub> at 309 cm<sup>-1</sup>, E'(M)<sup>LO2</sup>-LA(M) at 374 cm<sup>-1</sup>, and 3LA(M) at 395 cm<sup>-1</sup> [23,30,48,49]. These are indicated with black arrows. These Raman modes have generally been observed in multilayer WSe<sub>2</sub> or only in resonant Raman spectra in the case of a monolayer. However, in this study, the corresponding Raman modes were observed using typical Raman spectroscopy for monolayer WSe<sub>2</sub>. In general, 1L TMDs have limited observable Raman modes because of their 2D features. However, the Au MPs template overcomes these limitations and enables the observation of concealed Raman modes in the 1L WSe<sub>2</sub>. Fig. 2d shows a plot of the observable Raman modes for each sample. Three Raman modes are observed in the Raman spectra of 1L WSe<sub>2</sub> on the Au plate and 1L WSe<sub>2</sub> on the SiO<sub>2</sub> plate, whereas nine are observed in that of 1L WSe<sub>2</sub> on Au MPs. Table 1 lists the Raman modes and peak positions for each sample.

### 3.3. 2D Raman mapping of 1L WSe<sub>2</sub> on Au MP

Raman spectroscopy verified that the Raman scattering of 1L WSe<sub>2</sub> on Au MPs was enhanced. However, it was necessary to verify which region on the Au MPs was responsible for this enhancement. Therefore, the 2D Raman mapping of 1L WSe<sub>2</sub> on an individual Au MP was performed using confocal Raman spectroscopy. The excitation source for confocal Raman spectroscopy was a 532 nm CW laser. Fig. 3a shows the 2D Raman intensity mapping image of 1L WSe<sub>2</sub> on an individual Au MP. The brighter the color in the mapping image, the higher is the Raman intensity. The scale bar at the bottom left of the image indicates 1 μm. In the 2D Raman mapping image, the edge of the Au MP is indicated by a white dotted line. The Raman intensity of 1L WSe<sub>2</sub> is significantly higher along the edge region of the Au MP than in the other regions. The Raman

**Table 1**

Observable Raman modes with peak positions in 1L WSe<sub>2</sub> depending on the substrate.

Raman mode	1L WSe <sub>2</sub>			Reference
	On SiO <sub>2</sub> plate (cm <sup>-1</sup> )	On Au plate (cm <sup>-1</sup> )	On Au MPs (cm <sup>-1</sup> )	
E'(M) <sup>LO2</sup> -LA(M)	-	-	118	[48]
A' <sub>1</sub> (M)-LA(M)	-	-	137	[23]
LA(M) + TA(M)	-	-	219	[30]
E'(Γ)	248	248	248	[48]
A' <sub>1</sub> (Γ)	249	250	249	[48]
2LA(M)	261	262	261	[48]
B <sub>2g</sub>	-	-	309	[49]
E'(M) <sup>LO2</sup> -LA(M)	-	-	374	[48]
3LA(M)	-	-	395	[48]

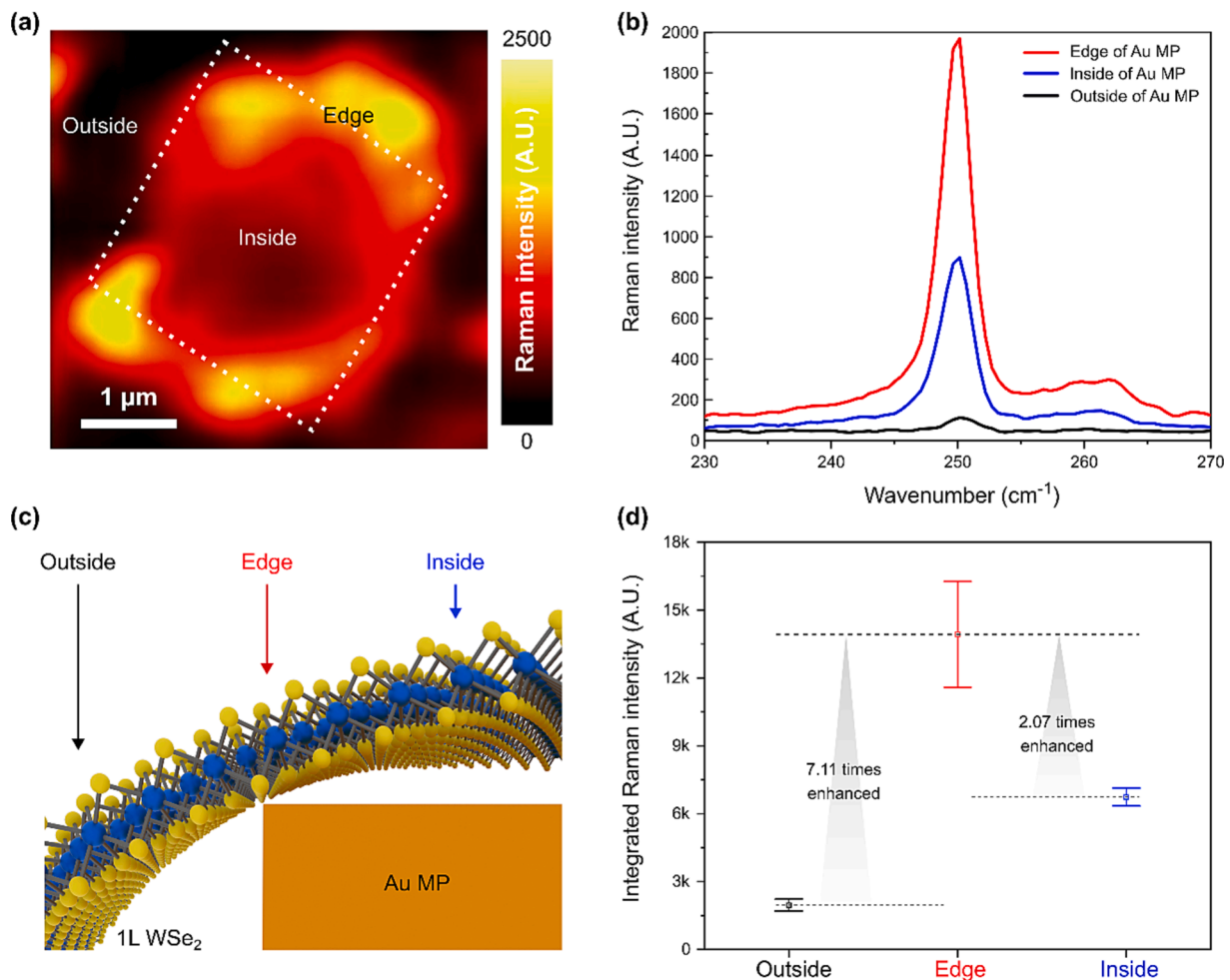
intensity of 1L WSe<sub>2</sub> in the inside region of the Au MP is higher than that outside the Au MP. However, it is significantly lower than that in the edge region of the Au MP. To compare and analyze the Raman spectra of each region, the local Raman spectra of 1L WSe<sub>2</sub> measured at the edge, inside, and outside of the Au MP were extracted, as shown in Fig. 3b. The red, blue, and black solid lines represent the local Raman spectra of 1L WSe<sub>2</sub> at the edge, inside, and outside of the Au MP, respectively. The wavelength range of the Raman spectrum was set from 230 cm<sup>-1</sup> to 270 cm<sup>-1</sup>. These included two main peaks to compare the Raman intensity. Similar to the results of the 2D Raman mapping image, the intensity of the Raman spectrum of 1L WSe<sub>2</sub> is significantly higher in the edge region of the Au MP. These results imply that the enhanced Raman scattering in 1L WSe<sub>2</sub> on the Au MPs was impelled by the Raman enhancement of the 1L WSe<sub>2</sub> at the edge region of the Au MPs. To understand the relationship between the strongly enhanced Raman scattering of 1L WSe<sub>2</sub> in the edge region of the Au MP and the structural features of the sample, a 3D schematic of the cross-sectional structure of 1L WSe<sub>2</sub> on the Au MP (including the edge region of the Au MP) is shown in Fig. 3c. Based on the location of the Au MP, the region of 1L WSe<sub>2</sub> was divided into edges, inside, and outside. These are indicated with arrows. During the wet-transfer process, 1L WSe<sub>2</sub> was subjected to a pulling force from all the directions in the edge region of the Au MP. (Fig. S5) This force caused the distance between 1L WSe<sub>2</sub> and the Au surface to be shorter in the edge region than in the other areas. This is depicted in Fig. 3c. The reduced distance between 1L WSe<sub>2</sub> and the Au surface caused the coupling of the 1L WSe<sub>2</sub> to the surface plasmon of Au. This resulted in enhanced Raman scattering. The SEM image and 2D Raman mapping results also indicate that the distance between the 1L WSe<sub>2</sub> and the Au surface is the shortest in the edge region of the Au MP. To quantify and compare the Raman scattering enhancement, the integrated Raman intensity according to the measurement position of 1L WSe<sub>2</sub> on the Au MP is shown in Fig. 3d. The integrated intensities extracted from each representative area (edge, inside, and outside the Au MP) are displayed in red, blue, and black, respectively. Five Raman spectra were extracted from each representative region, and the minimum, maximum, and average values were plotted. Based on the average value, the integrated intensity of 1L WSe<sub>2</sub> in the edge area was 7.11 and 2.07 times higher than those outside and inside, respectively. The relatively higher-intensity fluctuation of 1L WSe<sub>2</sub> at the edge region of the Au MP is attributed to the non-uniform edge of the Au MP. It resulted in different Raman scattering enhancements. Nevertheless, the average value for 1L WSe<sub>2</sub> in the edge region of the Au MP was remarkably high compared with that in the other regions. This supports the assertion that the Raman scattering of 1L WSe<sub>2</sub> was enhanced by surface plasmon coupling in the edge region of the Au MP. In addition, the significantly enhanced photoluminescence (PL) intensity of 1L WSe<sub>2</sub> on Au MPs compared with that for Au plate and SiO<sub>2</sub> plate supports the coupling between the PL of 1L WSe<sub>2</sub> and the LSP of the Au MP at the edge of the Au MP [50,51]. (Fig. S6).

### 3.4. Finite-difference time-domain (FDTD) for local field enhancement

To verify that the Raman scattering of 1L WSe<sub>2</sub> was enhanced in the edge region of the Au MP, the field enhancement was calculated using FDTD. The FDTD was performed using Lumerical, which is a commercial photonic simulation software package. A 1L WSe<sub>2</sub> was modeled as a 1 nm-thick flat layer on top of a step-edge Au-MP. The enhancement factor at a wavelength of 532 nm was calculated as

$$EF = \left| \frac{E}{E_0} \right|^4$$

E<sub>0</sub> is the initial amplitude of the input field, and E is the enhanced field at the 1L WSe<sub>2</sub> of the Au MP [52]. Fig. 4a-d shows the distribution of the local field enhancement when the distance between the 1L WSe<sub>2</sub> and Au MP is 0.5 nm, 1.0 nm, 3.0 nm, and 5.0 nm, respectively. The direction of

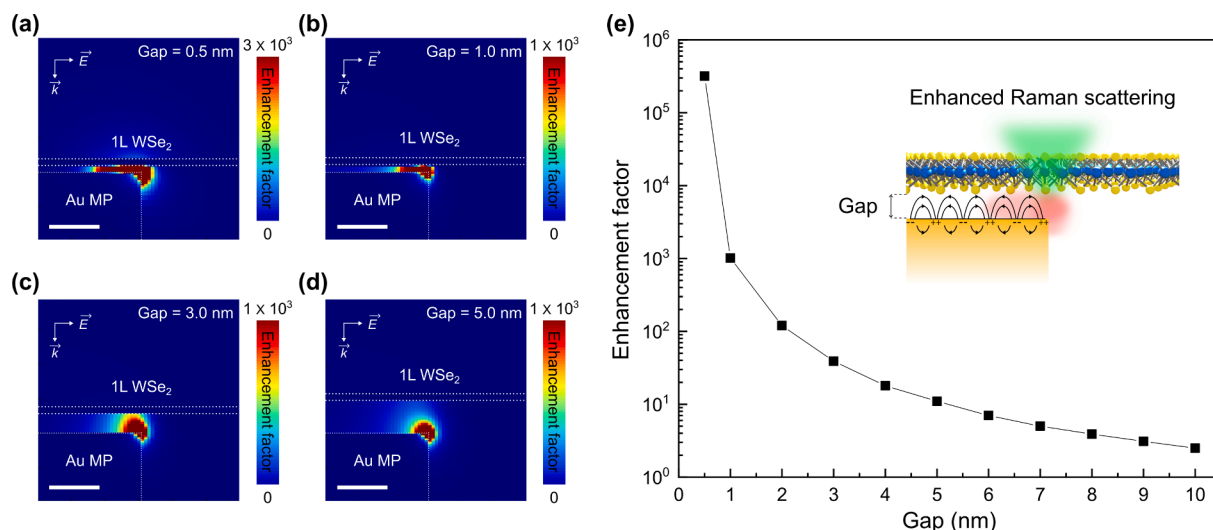


**Fig. 3. Raman mapping of 1L WSe<sub>2</sub> on an individual Au MP.** (a) Two-dimensional (2D) mapping image of the Raman intensity of 1L WSe<sub>2</sub> on an individual Au MP. The Raman intensity is highest at the edge region of the Au MP and lowest in the other regions. (b) Local Raman spectra extracted from 1L WSe<sub>2</sub> at the edge, interior, and exterior of the Au MP. The Raman intensity of 1L WSe<sub>2</sub> is highest at the edge of the Au MP and lowest outside the Au MP. (c) 3D schematic of 1L WSe<sub>2</sub> at the edge of the Au MP, showing the distance between 1L WSe<sub>2</sub> and the Au MP. The 1L WSe<sub>2</sub> is subjected to tensile forces in all the directions on the Au MP. This causes it to be stretched. The distance between the 1L WSe<sub>2</sub> and the Au surface is shortest at the edge of the Au MP, where the tensile forces are strongest. (d) Comparison of the integrated Raman intensities of 1L WSe<sub>2</sub> at different regions of the Au MP. The average integrated intensity of 1L WSe<sub>2</sub> at the edge of the Au MP is 7.11 times higher than that outside the Au MP.

propagation of the incident plane wave,  $E_0$ , was selected along the z-axis. Additionally, it was assumed that the electrical field was polarized along the  $x$  axis. As the 1L WSe<sub>2</sub> approached the Au MP, a strong field enhancement occurred because of the interaction between 1L WSe<sub>2</sub> and the Au surface. The variation in the enhancement factor with that in the gap between the 1L WSe<sub>2</sub> and the Au surface is plotted in Fig. 4e. The enhancement factor increased exponentially as the gap distance between 1L WSe<sub>2</sub> and the Au surface decreased. In particular, EF increased rapidly below 1 nm, where the gap distance was smaller than the van der Waals gap. This demonstrated the effect of the gap distance on the Raman enhancement of 1L WSe<sub>2</sub>. The gap between the 1L WSe<sub>2</sub> and Au surface was reduced significantly in the edge region of the Au MP. It enabled the LSPs at the edge of the Au MP to be coupled to the 1L WSe<sub>2</sub>. This significantly increased the field enhancement. As a result, the Raman scattering of 1L WSe<sub>2</sub> was enhanced substantially in the edge region of the Au MP, as shown in the inset of Fig. 4e. Therefore, FDTD theoretically supports the experimental results including Raman spectroscopy, 2D Raman mapping, SEM images, and Raman scattering enhancement of 1L WSe<sub>2</sub> by the surface plasmon coupling of Au MP.

#### 4. Conclusions

In this study, an Au MP array with a low-cost, simple process and large-scale fabrication was introduced as a platform to effectively enhance the Raman scattering of 1L WSe<sub>2</sub>. The SEM images show that multiple Au MPs were covered by 1L WSe<sub>2</sub> transferred using the wet transfer method. Raman spectroscopy was performed to verify the enhanced Raman scattering in 1L WSe<sub>2</sub> on the Au MPs. The Raman intensity of 1L WSe<sub>2</sub> on the Au MPs was enhanced by 19 times compared with that of 1L WSe<sub>2</sub> on the SiO<sub>2</sub> substrate. Moreover, nine Raman modes were observed in 1L WSe<sub>2</sub> on Au MPs, whereas only three Raman modes are observed in typical 1L WSe<sub>2</sub> owing to the 2D crystal lattice. Confocal Raman spectroscopy revealed that the Raman-scattering enhancement of 1L WSe<sub>2</sub> on the Au MPs occurred because of the locally enhanced Raman intensity in the edge region of the Au MPs. This was owing to the local field enhancement caused by the coupling between the LSPs of Au and 1L WSe<sub>2</sub> as the gap between 1L WSe<sub>2</sub> and the Au surface became exceptionally small in the edge region of the Au MP. According to the theoretical analysis of the FDTD results, a significant field enhancement occurred at the edge of the Au MP. The observations of this study would enable a more accurate and informative characterization of the optical, structural, and physical properties of layered



**Fig. 4. Finite-difference time-domain (FDTD) of local field enhancement.** FDTD simulations for local field enhancement between 1L WSe<sub>2</sub> and Au MPs with gap distance of (a) 0.5 nm, (b) 1.0 nm, (c) 3.0 nm, and (d) 5.0 nm. The simulations were performed in the edge region of the Au MP. The field enhancement factor was observed to be stronger when the distance between the 1L WSe<sub>2</sub> and Au MP was smaller. (e) Calculated enhancement factor as a function of the distance between 1L WSe<sub>2</sub> and the Au MP. The inset is a 3D schematic of the Raman enhancement mechanism. As the gap between 1L WSe<sub>2</sub> and the Au MP decreases, the Raman intensity is enhanced owing to the resonance of the phonons of 1L WSe<sub>2</sub> and LSPs of the Au MP.

quantum materials using Raman spectroscopy for applications in quantum technology.

#### Author contributions

The manuscript was written with contributions from all the authors. All the authors have approved the final version of the manuscript.

#### CRediT authorship contribution statement

**Hyun Jeong:** Conceptualization, Methodology, Investigation, Formal analysis, Data curation, Writing – original draft, Writing – review & editing. **Hyung Chan Suh:** Methodology, Data curation. **Ga Hyun Cho:** Methodology, Data curation. **Rafael Salas-Montiel:** Software, Validation. **Hayoung Ko:** Methodology. **Ki Kang Kim:** Resources. **Mun Seok Jeong:** Conceptualization, Funding acquisition, Resources, Project administration, Writing – review & editing, Supervision.

#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Data availability

Data will be made available on request.

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#### Appendix A. Supplementary material

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