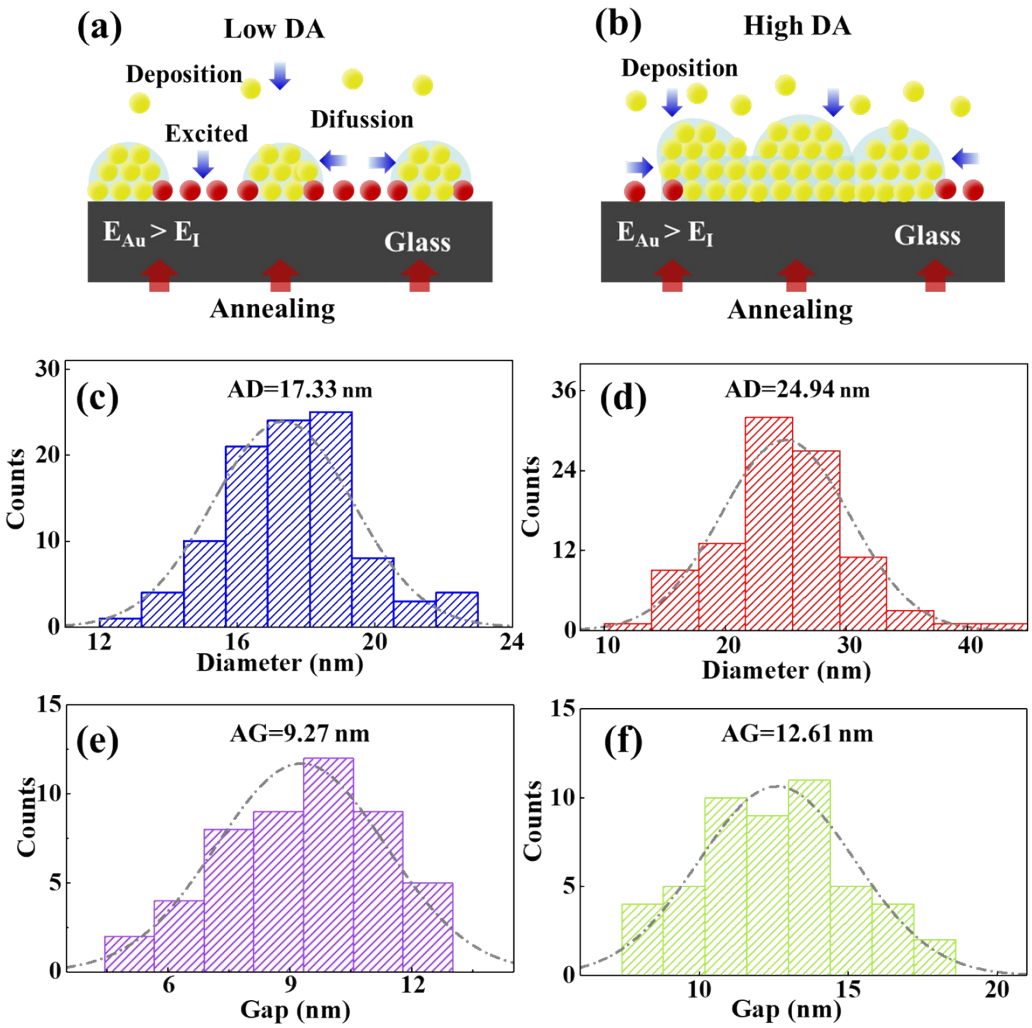

Figure S1 (a) – (b) Scheme of the growth mechanism of the Au NSs as a function of deposition amount (DA).The average diameter (AD) of the self-assembled (c) Au nano-spheres and (e) Au nano-ellipsoids. The average gap (AG) between nanostructures of the (d) Au nano-spheres and (f) Au nano-ellipsoids.

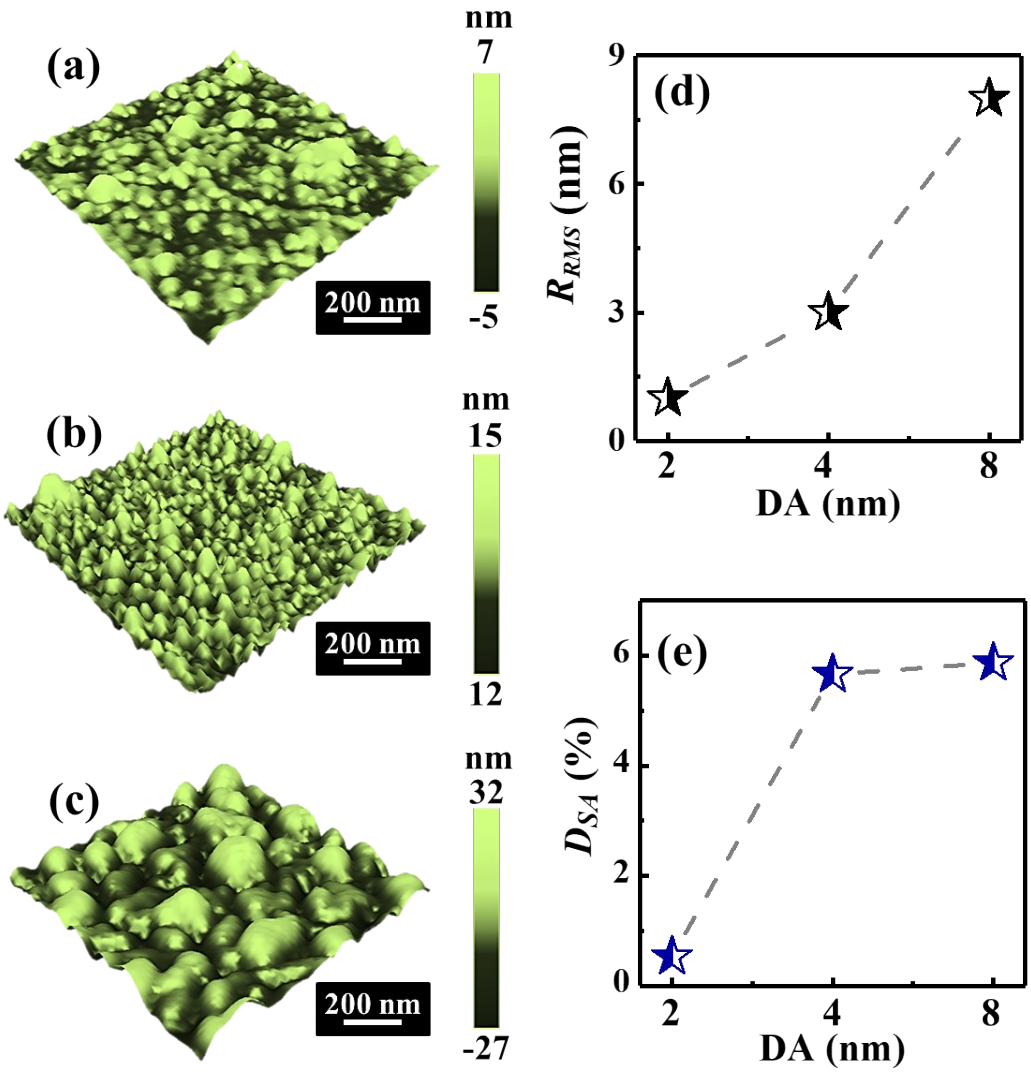

Figure S2 Atomic force microscopy (AFM) images of the Au NSs fabricated with various deposition amounts: (a) 2 nm, (b) 4 nm, (c) 8 nm. (d) The root-mean-squared roughness (R_{RMS}) and (e) surface area difference (D_{SA}) of the corresponding samples.

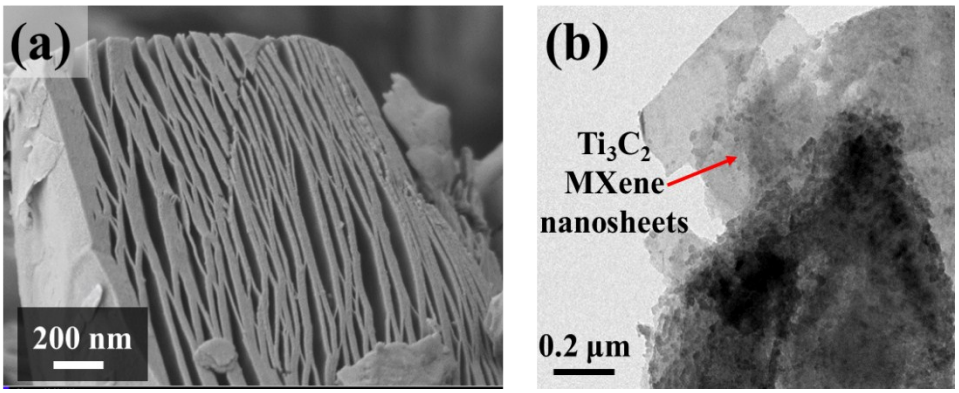

Figure S3 (a) SEM image of accordion-like $Ti_3C_2T_x$ after HF acid etching. (b) TEM image of MXene nano-sheets.

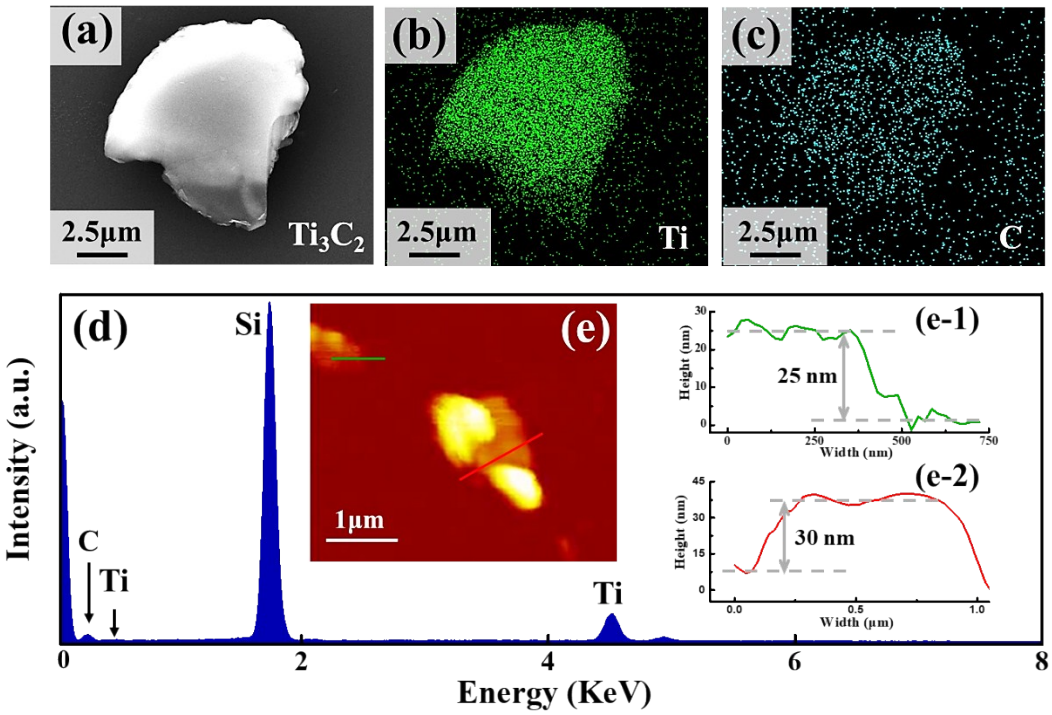

Figure S4 (a) SEM image of MXene nano-sheets. Elemental maps of (b) Ti and (c) C. (d) EDS spectrum of the MXene nano-sheets between 0 and 8 KeV. (e) AFM side-view of MXene nano-sheets. (e-1) and (e-2) The corresponding cross-sectional line-profiles acquired from the white line drawn area in the AFM side-view.

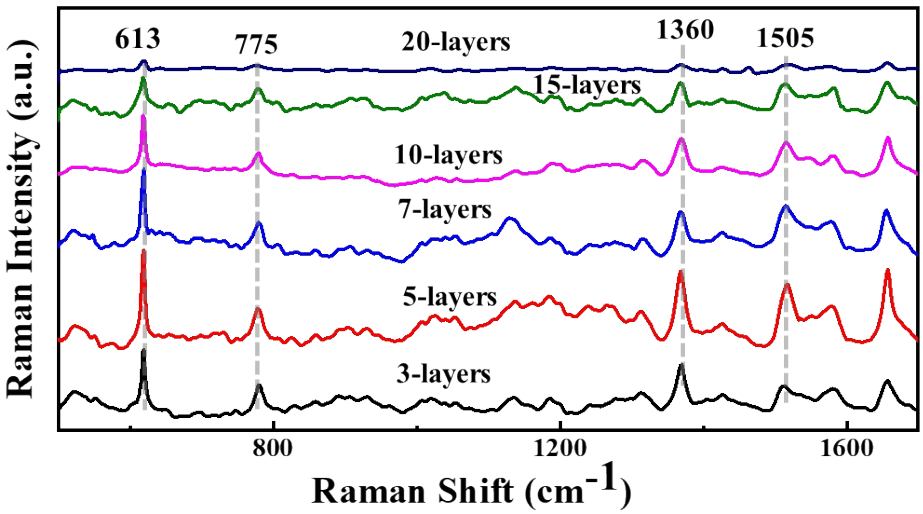

Figure S5 Raman spectra on the substrate MXene nano-sheet/Au nano-ellipsoids of R6G molecules at a concentration of 10^{-6} M at various layers.

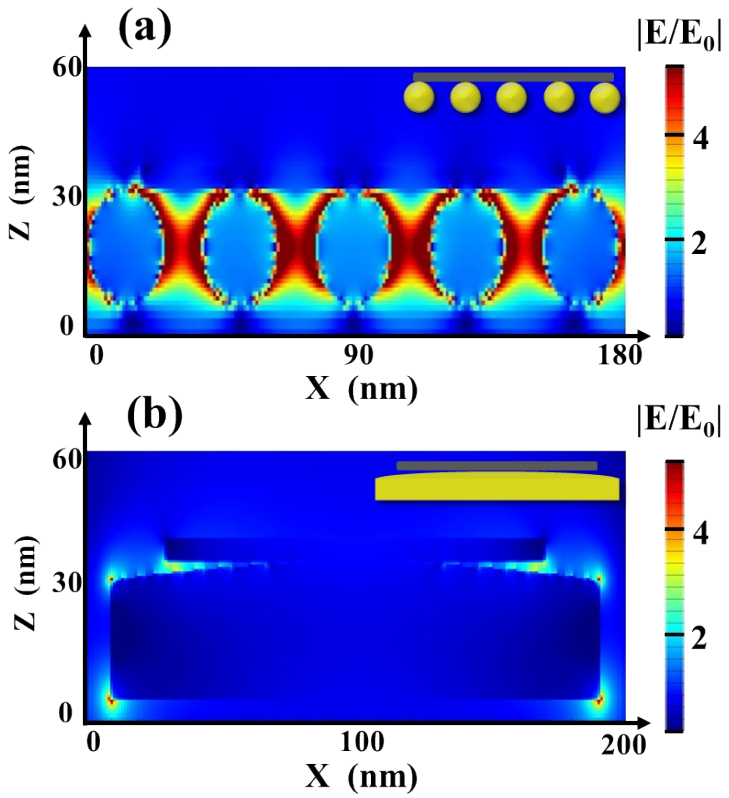

Figure S6 The X-Z plane electromagnetic (EM) field distribution simulation of the (a) Au nano-ellipsoids and (b) Au coralline nano-islands covered with 1-layer MXene nano-sheets excited by a 532 nm plane light illumination.

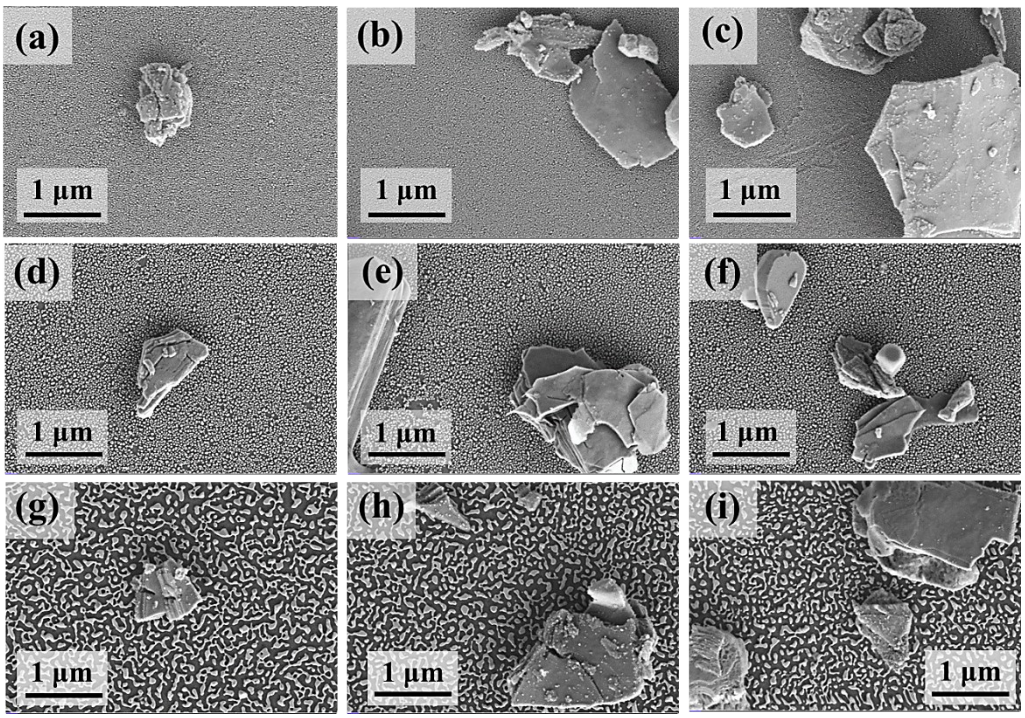

Figure S7 SEM images of the (a) MXene/Au-1, (b) MXene/Au-2, (c) MXene/Au-3, (d) MXene/Au-4, (e) MXene/Au-5, (f) MXene/Au-6, (g) MXene/Au-7, (h) MXene/Au-8, (i) MXene/Au-9.

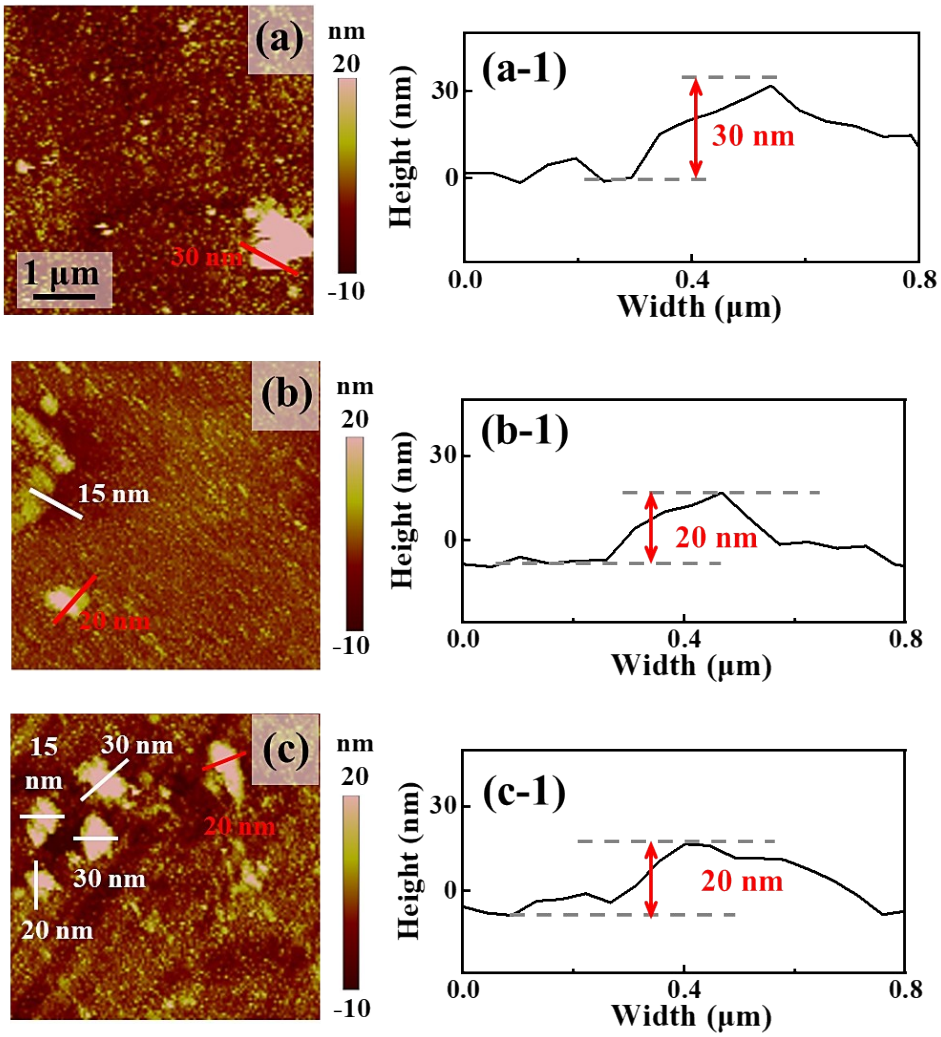

Figure S8 AFM top-views of (a) MXene/Au-1, (b) MXene/Au-2, (c) MXene/Au-3. The corresponding height of the area drawn with line was provided in each AFM top-views. (a-1) - (c-1) Cross-sectional line-profiles obtained from the red lines drawn area.

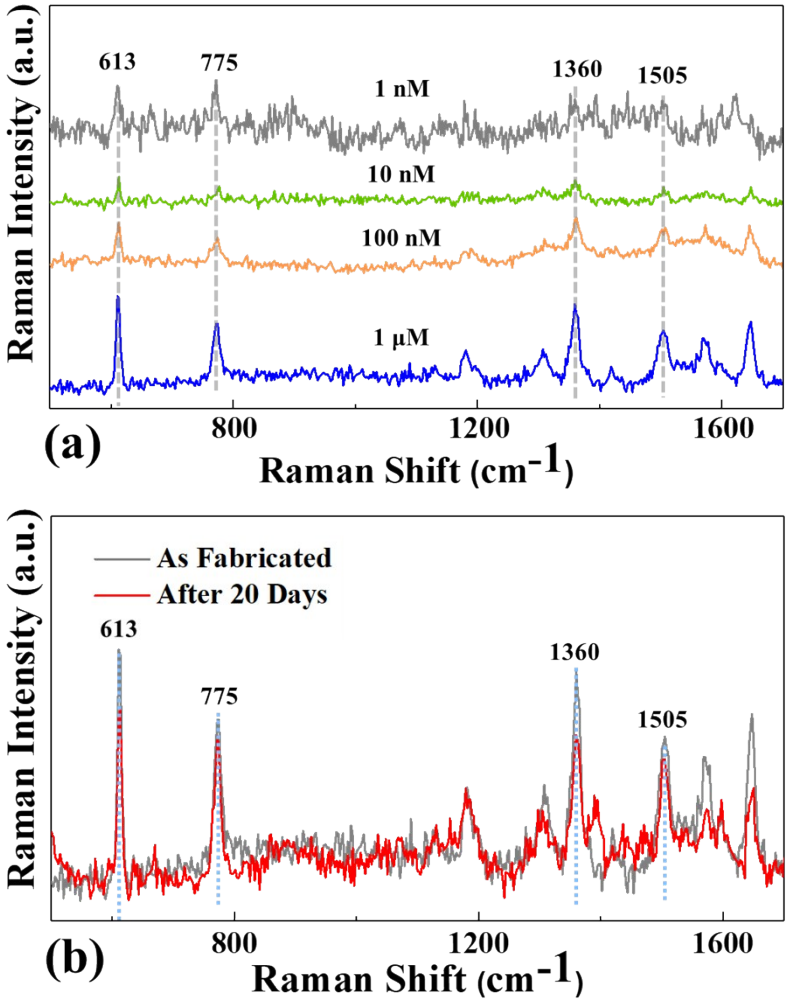

Figure S9 (a) Raman spectra on the of R6G molecules on the substrate MXene/Au-3 at different concertrations. (b) The Raman spectra of 1μM R6G molecules on the as-fabricated sample (MXene/Au-6) and after 20 days.

Table 1 Average absorption and transmittance for the Au nanostructures coated with (W) and without (W/O) MXene nano-sheets. The Au deposition thicknesses (DA) varied between 2 and 8 nm.

	DA (nm)	2	4	8
Absorption (%)	W/O MXene	4.3	14.9	39.8
	W MXene	5.9	15.9	46
Transmittance (%)	W/O MXene	91.2	72.5	39.2
	W MXene	87.4	66.9	36.2

Table 2 Raman intensity of various MXene nanosheet/Au nanosturcture architectures at 613, 775, 1360 and 1505 cm^{-1} Raman peaks.

Peak Position (cm^{-1}) / Sample	613	775	1360	1505
MXene/Au - 1	21.8	17.44	17.18	17.59
MXene/Au - 2	38.15	24.08	33.78	27.12
MXene/Au - 3	101.34	52.32	75.64	54.98
MXene/Au - 4	23.8	15.3	23.4	13.1
MXene/Au - 5	111	60.9	107.1	87.2
MXene/Au - 6	208.4	106.9	173.3	110.33
MXene/Au - 7	20.5	17.2	15.17	18.3
MXene/Au - 8	30.2	32.9	28.5	23.4
MXene/Au - 9	69.5	49.8	65.8	38.9

Table 3 Comparison of the calculated enhancement factors (EF) between different SERS substrates reported in the literatures.

Substrate	Probe molecule	Sensitivity	EF
Au nanocubes[1]	4-ATP	10^{-2} M	7×10^{5}
Au nanoboness[2]	4-MBA	10^{-5} M	4.76×10^{4}
non-tapered Au nanopillars[3]	benzenethiol	2×10^{-3} M	5.7×10^{6}
Ag@MXene[4]	methylene blue	10^{-6} M	1.5×10^{5}
Au@MXene[4]	methylene blue	10^{-6} M	1.17×10^{5}
Pd@MXene[4]	methylene blue	10^{-6} M	9.61×10^{4}
MXene[5]	R6G	10^{-6} M	1.2×10^{6}
This work	R6G	10^{-10} M	2.9×10^{7}

Note S1

To calculate the EF of the MXene/Au architectures for the R6G molecules detection, the ratio of surface enhanced Raman spectra (SERS) to normal Raman spectra (NRS) is determined by using the following calculating formula:

$$EF = (I_{SERS}/N_{SERS})/ (I_{Raman}/N_{Raman}) \quad (1)$$

, where I_{SERS} and I_{Raman} refer to the peak intensities of the SERS and NRS, respectively. N_{SERS} and N_{Raman} denote the number of probe molecules excited by laser beam on the SERS substrates and glass. N_{Raman}/N_{SERS} can be expressed with:

$$N_{Raman}/N_{SERS} = C_{Raman}\times V_{Raman}/C_{SERS}\times V_{SERS} \quad (2)$$

, where C_{SERS} and V_{SERS} represent the volume and concentration of R6G aqueous solution dip-coated on the SERS substrates, respectively. Meanwhile, C_{Raman} and V_{Raman} represent the volume and concentration of R6G aqueous solution dip-coated on the bare glass. In this work, the Raman vibrational peak at 1505 cm^{-1} was selected for the EF calculation. As a comparison, 1×10^{-3} M R6G aqueous solution was chosen for NRS. For each sample, the Raman spectra was averaged out from Raman signals acquired at 3 random locations with an acquisition time of 0.5 s.

Note S2

The local electric field properties of the samples are calculated and analyzed using Finite-Difference Time-Domain (FDTD) solutions software. The theoretical models are built based on the SEM results shown in Figure 1e - f and the coressponding parameters of Au NPs displayed in Figure S1c - f. According to the report[6],refractive index of MXene nano-sheets is set as 2.64 + i. A periodic boundary condition is applied for x-axis and y-axis, and the perfectly matched layer condition is used for z-axis. The excited source along z-axis is 532 nm wave-plane. To get relatively high resolution, the mesh grid size is set as 1 nm in x-y-z direction, and mesh accuracy is 8. Furthermore, shutoff level is 10^{-5}.

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