Supplementary Information (SI) for Nanoscale.

Supporting Information

Lead-free Halide Double Perovskites Nanoflakes as High-Performance SERS Substrates for Detection of Trace Organic Pollutants: Chemical Enhancement versus Electromagnetic Enhancement

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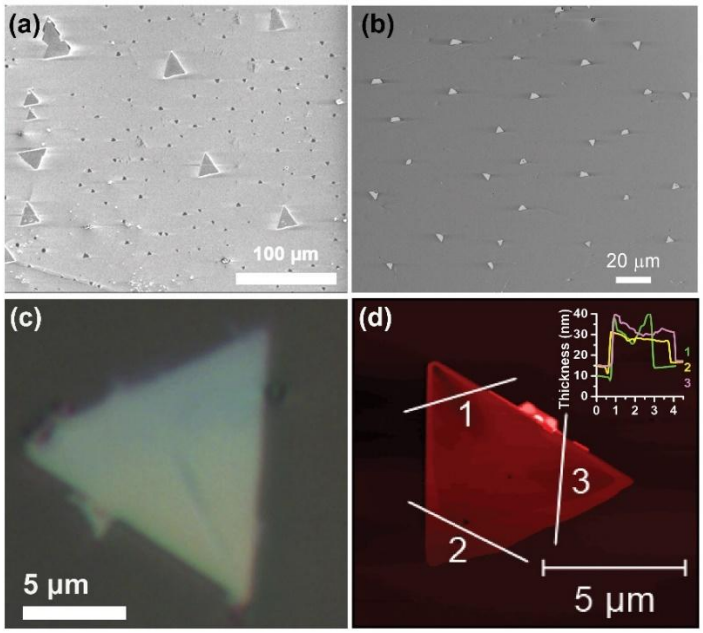

Figure S1. (a) FESEM image of the large area uniform growth of $Cs_2AgBiBr_6$ DP flakes prepared by the space-confinement method. **(b)** shows the magnified FESEM image. **(c)** Optical image of the smaller triangles of $CS_2AgBiBr_6$ DP flakes. **(d)** AFM image of 2D $Cs_2AgBiBr_6$ DP flake. The inset shows the height profile of the corresponding flake.

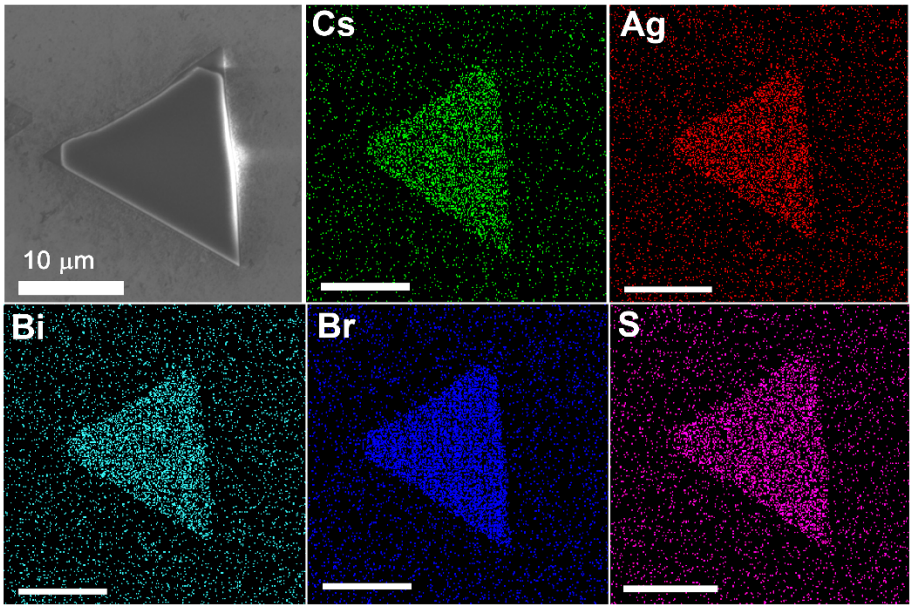

Figure S2. FESEM-EDX elemental mapping of each constituent element of the $Cs_2AgBiBr_6$/MB complex.

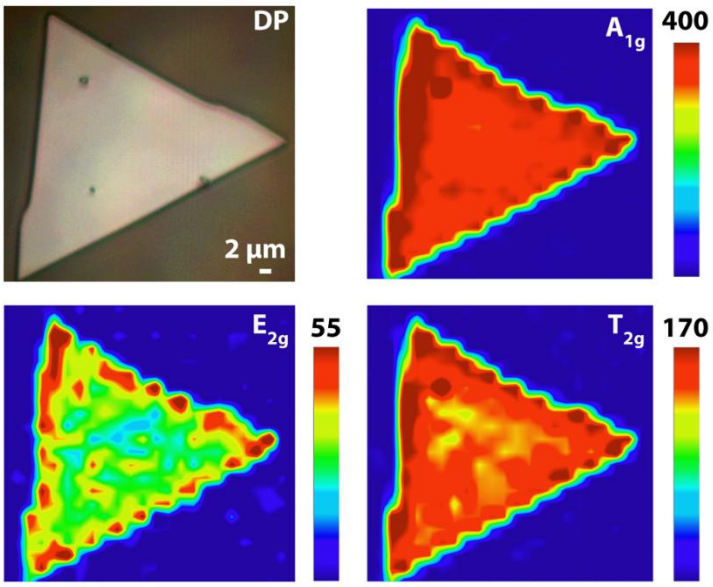

Figure S3. Raman mapping of all three characteristic peaks of $Cs_2AgBiBr_6$ DP flake.

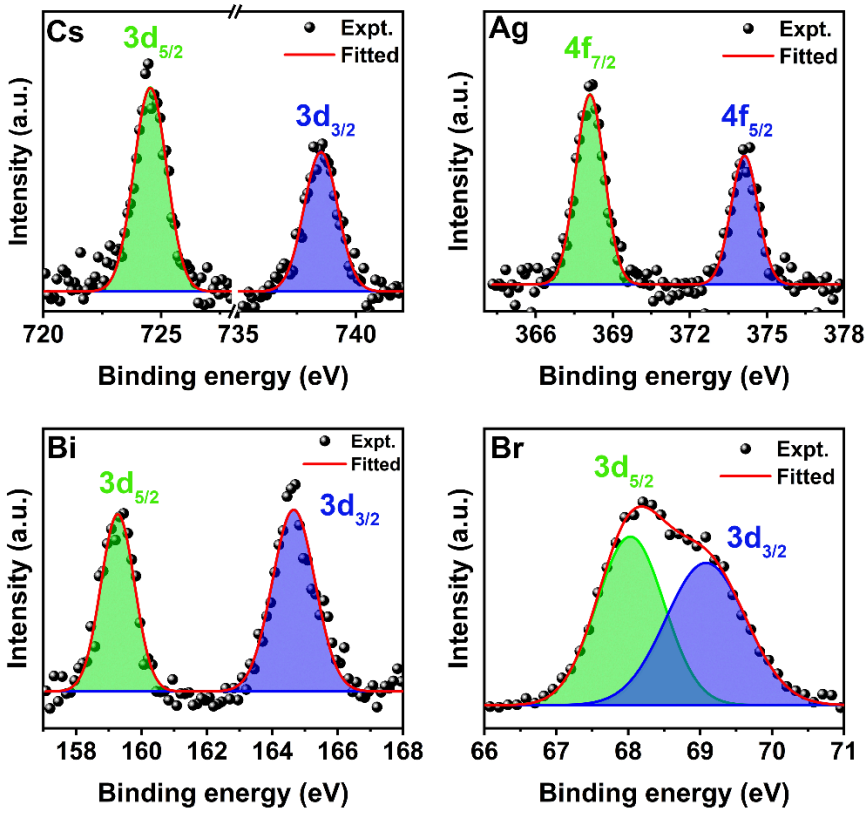

Figure S4. High-resolution XPS spectra of each element of pristine $Cs_2AgBiBr_6$ DP.

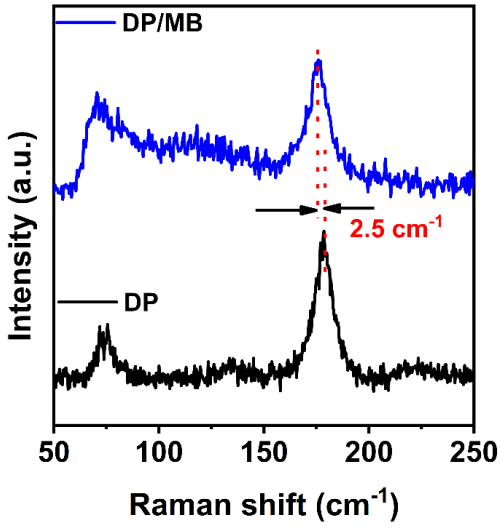

Figure S5. Comparison of Raman spectra of $Cs_2AgBiBr_6$ DP before and after MB adsorption.

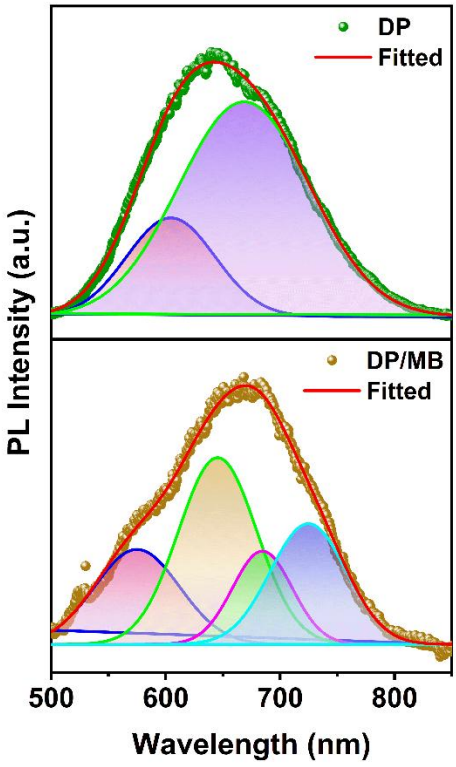

Figure S6. Depicts the deconvoluted PL spectra of $Cs_2AgBiBr_6$ DP before and after MB adsorption.

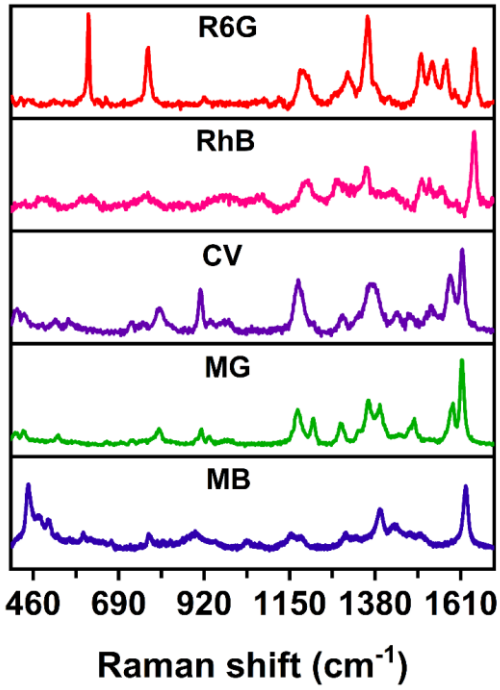

Figure S7. Raman spectra of different analytes adsorbed on the $Cs_2AgBiBr_6$ DP flakes.

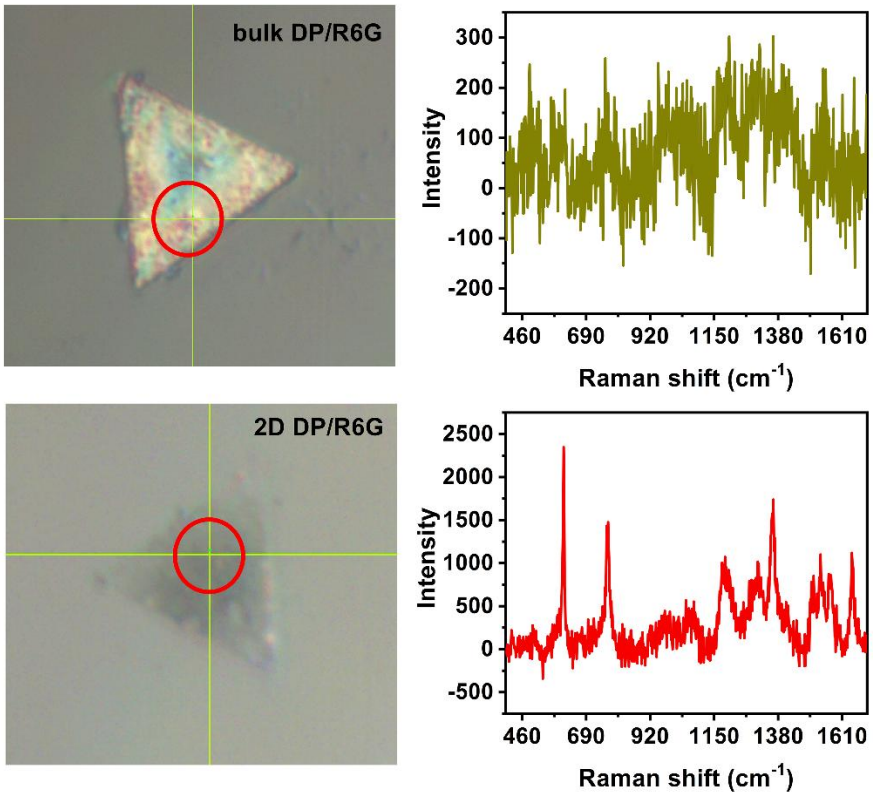

Figure S8. Comparison of the SERS performance of $Cs_2AgBiBr_6$ DP flakes of R6G analyte with varying thicknesses of the DP.

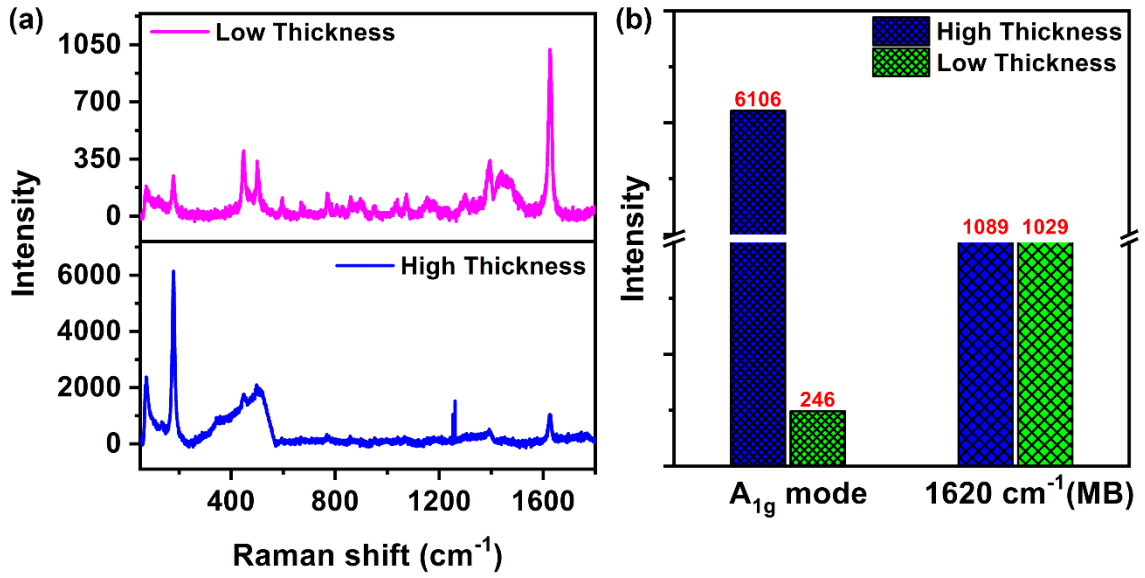

Figure S9. (a) Raman spectra of MB on $Cs_2AgBiBr_6$ DP with varying thicknesses. **(b)** depicts the comparison of the intensities of the A_{1g} mode and the 1620 cm^{-1} mode of MB adsorbed on different thicknesses.

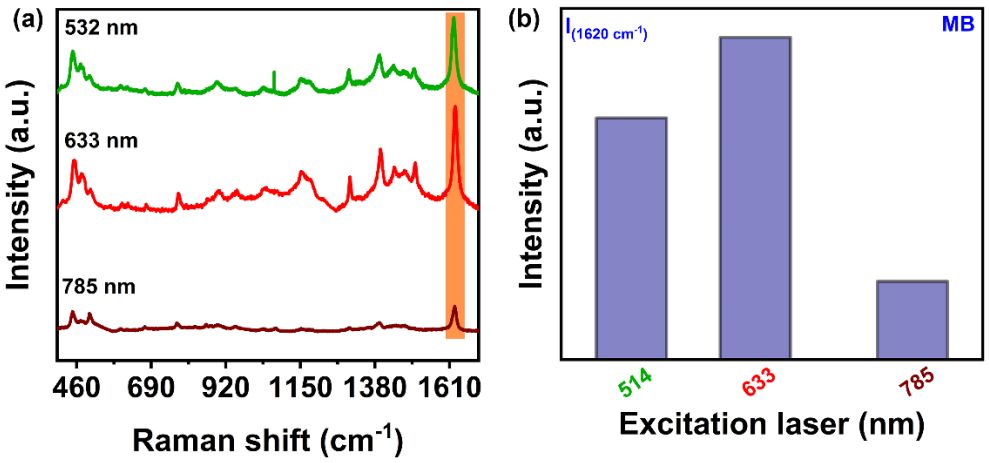

Figure S10. (a) SERS spectra of MB analyte on $Cs_2AgBiBr_6$ DP substrate using different laser excitation sources. **(b)** indicates the intensity of the characteristic mode of MB analyte with different laser excitation.

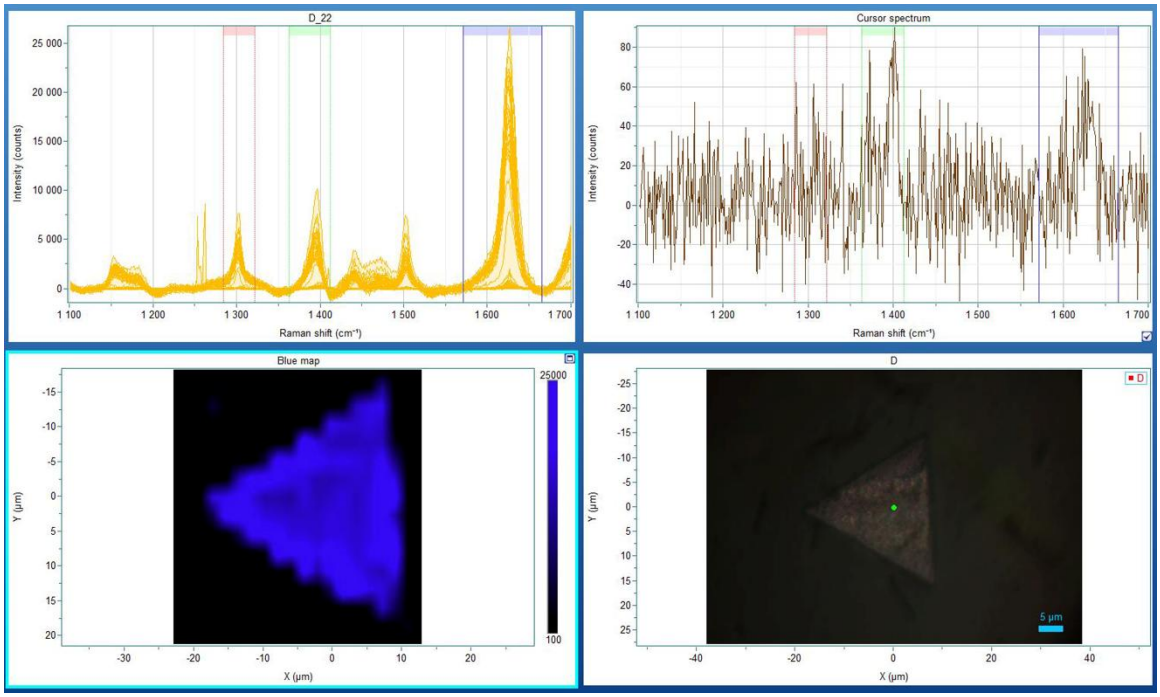

Figure S11. Raw image of the SERS measurement of MB on $Cs_2AgBiBr_6$ DP flake.

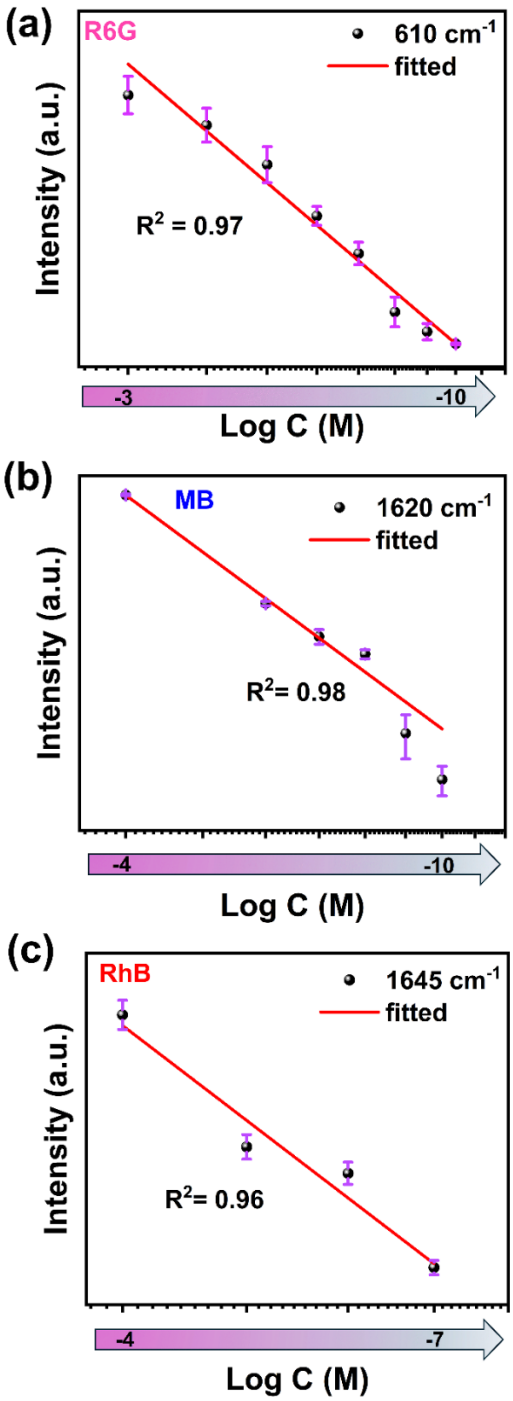

Figure S12. Variation of the SERS intensities of the characteristic modes of different molecules with the molar concentrations.

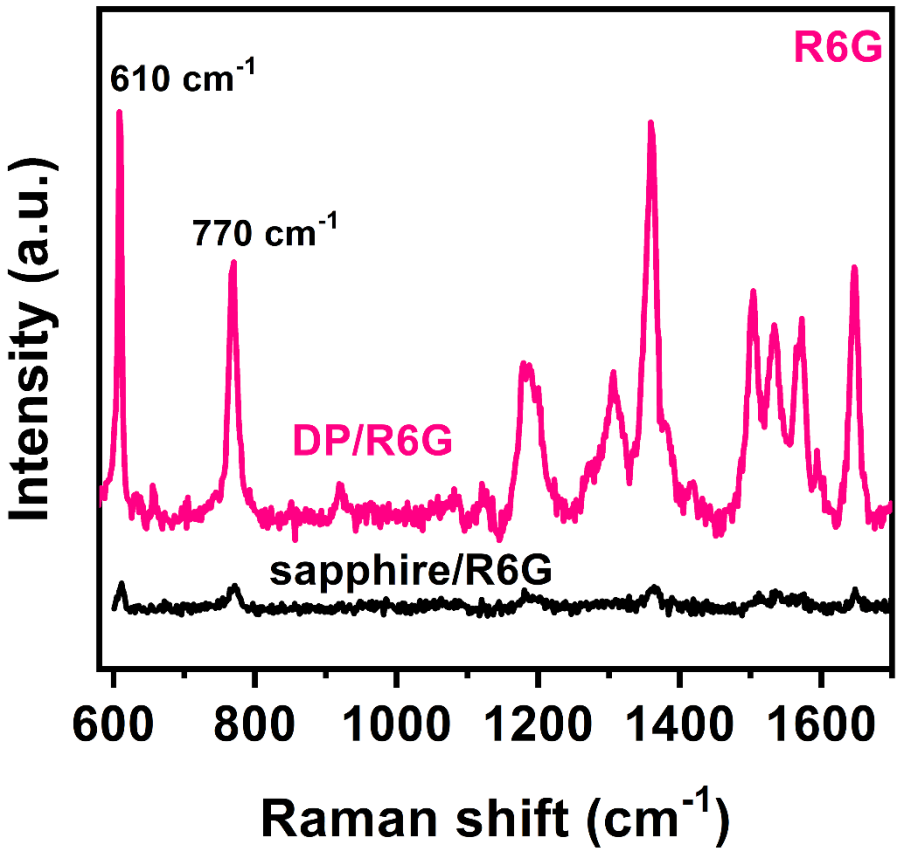

Figure S13. Comparison of the SERS performance of R6G analytes on $Cs_2AgBiBr_6$ DP (10^{-6} M) and sapphire substrates (10^{-3} M).

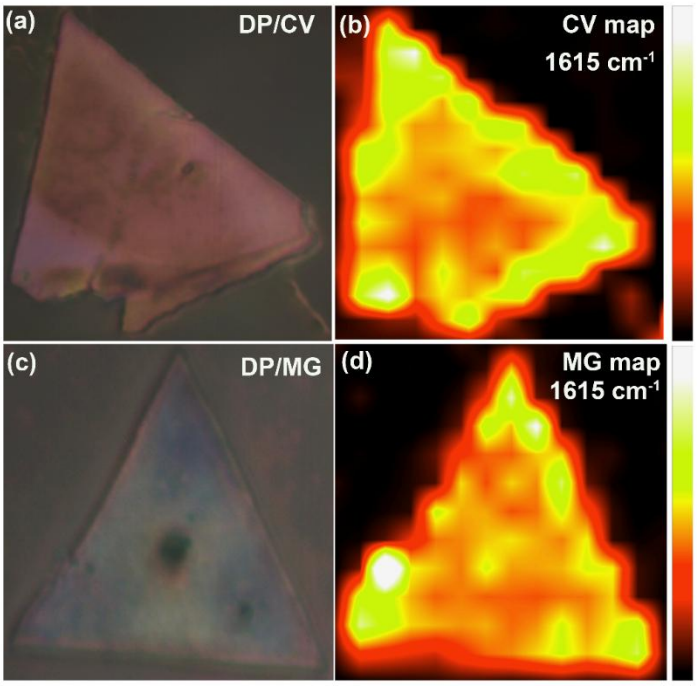

Figure S14. (a, c) shows the $Cs_2AgBiBr_6$ DP flakes with CV and MG adsorbed on the surface, respectively. **(b, d)** shows the Raman mapping of the characteristic modes of CV and MG on the Cs_2AgBiB_6 DP SERS substrate, respectively.

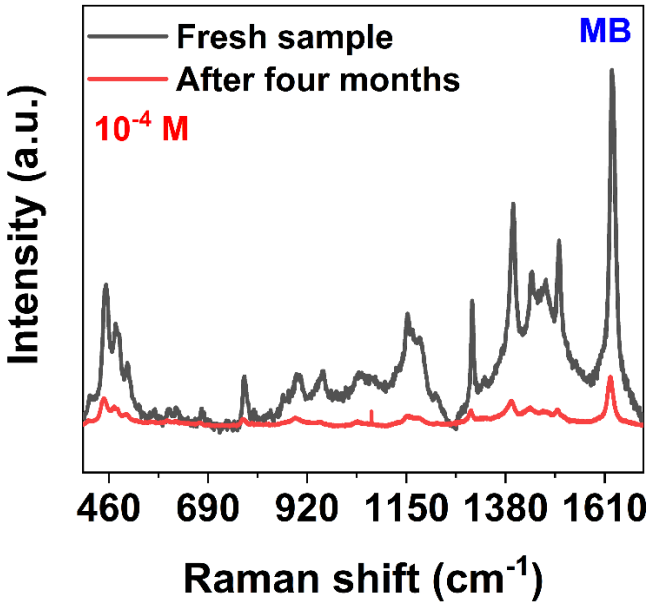

Figure S15. Comparison of SERS performance of the freshly prepared $Cs_2AgBiBr_6$ DP sample and after storing it for four months in ambient conditions.

Table S1. Binding energies of $Cs_2AgBiBr_6$ DP sample before and after adsorption of methylene blue analyte obtained from XPS analysis.

Sample / Elements		DP (eV)	FWHM (eV)	DP/MB (eV)	FWHM (eV)	Shift (eV)
Cesium	3d3/2	724.87	1.7	724.53	1.6	0.34
	3d5/2	738.84	1.9	738.51	1.7	0.33
Silver	3d3/2	368.01	1.3	367.59	1.3	0.42
	3d5/2	374.12	1.2	373.64	1.1	0.48
Bismuth	4f5/2	159.27	1.2	159.14	1.1	0.13
	4f7/2	164.65	1.5	164.45	1.1	0.25
Bromine	3d3/2	68.09	1.2	68.03	1.1	0.06
	3d5/3	69.07	1.3	69.02	1.2	0.05

Table S2. Comparison of the reported band positions of MB and our SERS spectrum.

Sr. No.	**Wavenumber (cm^{-1}) (Reported)**	**Peak assignment**	**Obtained SERS spectrum (cm^{-1})**
1.	949	C-H in-plane bending	945.6
2.	1067	C-H in-plane bending	1069
3.	1181	C-N stretching	-
4.	1301	β(CH); υ (C-N) $_{Ring}$	1296
5.	1392	C-H in-plane ring deformation	1389
6.	1444	C-N asymmetric stretching	-
7.	1618	C-C ring stretching	1620

Table S3. Calculation of the EF of different modes of the MB, R6G, and RhB molecules.

Molecules	**Peak position (cm^{-1})**	**I_S/I_R**	**C_R/C_S**	**Enhancement factor (EF)**
MB ($C_S=10^{-10}$ M) ($C_R= 10^{-10}$ M)	441	1.41	10^7	1.41×10^7
	1296	5.04	10^7	5.04×10^7
	1386	2.73	10^7	2.73×10^7
	1620	1.15	10^7	1.15×10^7
R6G ($C_S=10^{-10}$ M) ($C_R=10^{-3}$ M)	610	7.8	10^6	7.8×10^6
	766	9.8	10^6	9.8×10^6
	1356	1.37	10^7	1.37×10^7
	1646	7.8	10^6	7.8×10^6
RhB ($C_S=10^{-7}$ M) ($C_R=10^{-2}$ M)	1193	2.31	10^5	2.31×10^5
	1356	1.3	10^5	1.3×10^5
	1645	0.74	10^5	0.74×10^5