

*Supplementary Material*

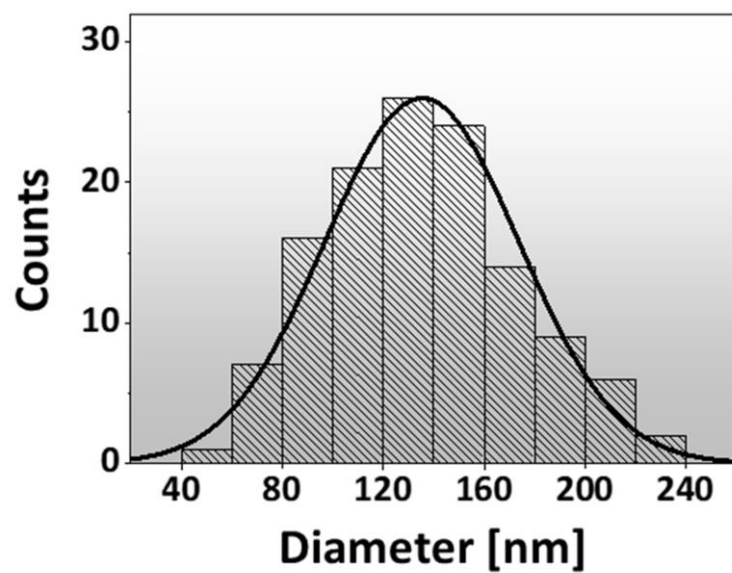
# **Hierarchically Assembled Plasmonic Metal-Dielectric-Metal Hybrid Nano-Architectures for High-Sensitivity SERS Detection**

**Puran Pandey<sup>1</sup>, Min-Kyu Seo<sup>1</sup>, Ki Hoon Shin<sup>1</sup>, Young-Woo Lee<sup>2,\*</sup> and Jung Inn Sohn<sup>1,\*</sup>**

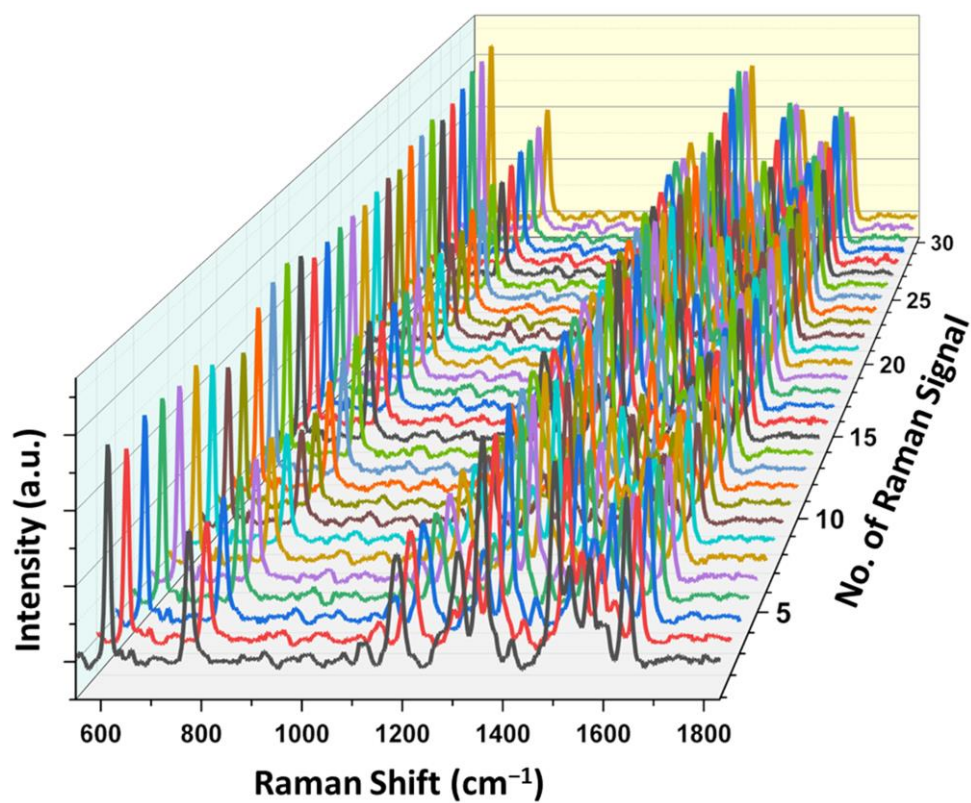
<sup>1</sup> Division of Physics and Semiconductor Science, Dongguk University-Seoul, Seoul 04620 Republic of Korea; ppcpurans@gmail.com (P.P.); seominkyuu@gmail.com (M.-K.S.); kihoonshin@dongguk.edu (K.H.S.)

<sup>2</sup> Department of Energy Systems, Soonchunhyang University, Asan-si, Chungcheongnam-do 31538, Republic of Korea

\* Correspondence: ywlee@sch.ac.kr (Y.-W.L.); junginn.sohn@dongguk.edu (J.I.S.)



**Figure S1.** Histogram of Au NPs arrays on Si/SiO<sub>2</sub> substrate by the annealing of 10 nm Au at 800 °C for 120 s.



**Figure S2.** Raman spectra of R6G molecules (10<sup>-6</sup> M) measured at 30 different locations on PMDM hybrid nanostructures to test the SERS reproducibility.

**Table S1.** Raman Band Assignments of R6G molecules.

Raman peak [cm <sup>-1</sup> ]		Assignment
This work	Literature Ref. [45]	
612	612	C–C ring in-plane bending in xanthene/phenyl rings
776	772	C–H out-of-plane bending
1128	1127	C–H in-plane bending in xanthene/phenyl rings
1185	1187	C–H in-plane bending in xanthene ring
1310	1312	hybrid mode (xanthene/phenyl rings and NHC <sub>2</sub> H <sub>5</sub> group)
1363	1363	C–C stretching in xanthene ring
1506	1509	C–C stretching in xanthene ring
1575	1575	C–C stretching in phenyl ring
1650	1651	C–C stretching in xanthene ring

**Table S2.** Comparison of SERS performance of current work and previously reported plasmonic NPs-based SERS substrates.

SERS material	Probe Molecule	LOD	EF	Reference
Au island over Nanosphere	R6G	-	$1.5 \times 10^6$	[46]
Nanoporous Au-Ag nanorod	4-MBA	$10^{-9}$ M	$1.5 \times 10^6$	[47]
Nanoporous Au thin films	R6G	$10^{-8}$ M	-	[48]
Au@AuAg multishell Nanostructures	4-ABT	$10^{-5}$ M	$9.18 \times 10^5$	[49]
Porous AuAg NPs	R6G	$2.37 \times 10^{-9}$ M	$7.8 \times 10^6$	[4]
Ag@ZrO <sub>2</sub>	R6G	$10^{-8}$ M	-	[50]
Sandwich-Like Sensor (AuA-pMIP)	R6G	$10^{-10}$ M	$1.24 \times 10^6$	[51]
Au/SiO <sub>2</sub> /Graphene/Au	CV	-	$0.23 \times 10^6$	[29]
Ag@SiO <sub>2</sub>	R6G	-	$1.07 \times 10^7$	[52]
Ag/Al <sub>2</sub> O <sub>3</sub> /Au Nanograting	p-thiocresol	$10^{-9}$ M	$5.2 \times 10^7$	[53]
Ag-Al <sub>2</sub> O <sub>3</sub> -Ag heterojunctions	MB	$10^{-10}$ M	$1 \times 10^8$	[30]
PMDM hybrid nanostructure	R6G	$10^{-11}$ M	$1.3 \times 10^8$	[This work]

LOD = Limit of Detection

EF = Enhancement Factor

CV = Crystal violet

MB = methylene blue

R6G = Rhodamine 6G

4-MBA = 4-Mercaptobenzoic acid

4-ABT = 4-Aminobenzenethiol