

Supporting Information for

Metallo-dielectric Photonic Crystals for Surface-enhanced Raman Scattering

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S1. Normalized I_p distribution.

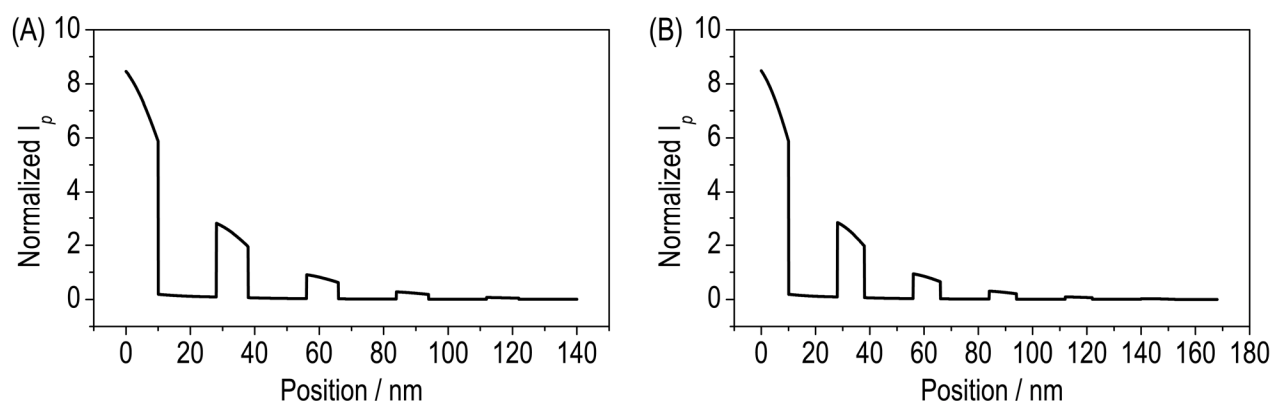


Figure S1. Normalized I_p distribution across MD structures composed of five (A) and six (B) MD units.

S2. Large-scale fabrication of PS SAM as template for MDPCs.

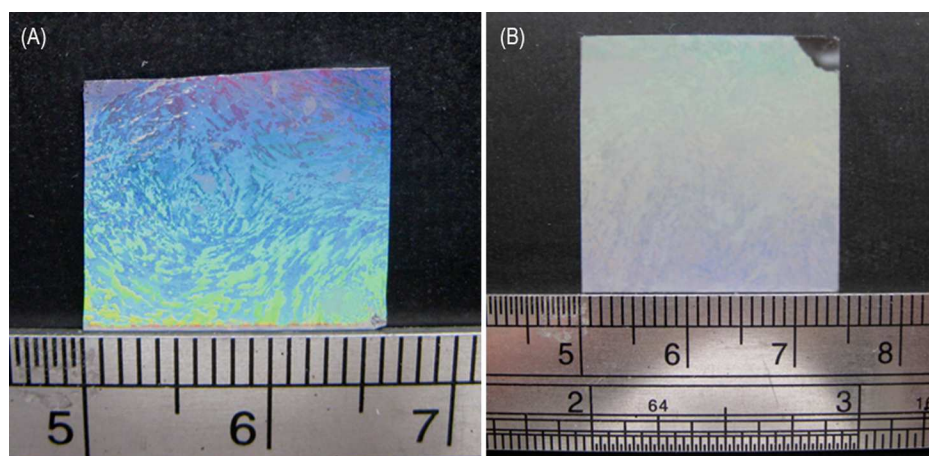


Figure S2. Digital image showing the obtained PS SAM on (A) silicon wafer and (B) traditional cover glass.

S3. Detailed dimension and structure of the as-prepared MDPCs.

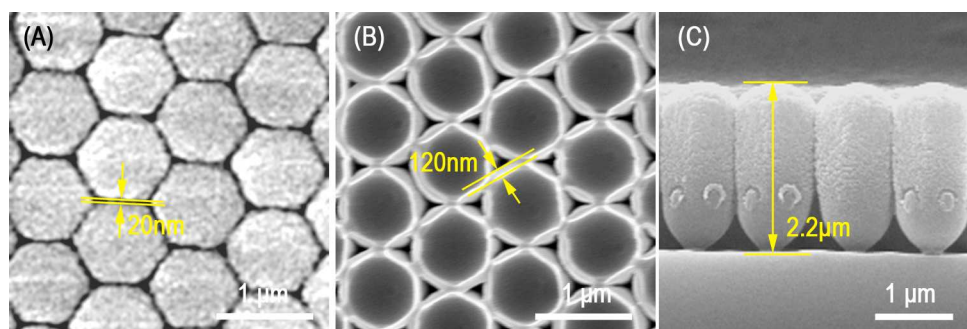


Figure S3. Detailed dimension and structure of the as-prepared MDPCs. (A) Top-view of MDPCs showing the distance between adjacent particles is about 20 nm; (B) reverse side of MDPCs showing an average wall thickness of about 120 nm; (C) cross-sectional view of MDPCs showing a height of 2.2 μm . By compare the diameter of shrank PS spheres of ~ 800 nm, the thickness of supporting a-Si (Figure 1A in the manuscript) is about 1.3 μm .

S4. Determination of enhancement factors (EFs).

The approximate calculation of N_{normal} is determined based on 20 μL of 10^{-3} M R6G solution spread on a-Si PCs with a spreading diameter of 2 mm. The number of probed molecules can be calculated to be $\sim 1.2 \times 10^{10}$ per spot-size. In determining N_{SERS} in the illuminated volume of our laser, we assume that all R6G molecules in the solution are absorbed as a monolayer onto the surface of MDPCs. Such assumption represents a theoretical maximum number of R6G molecules adsorbed on the surface of MDPCs and is surely an overestimate, thus the EF reported here is likely an underestimate rather than an overestimate of the actual EF value.

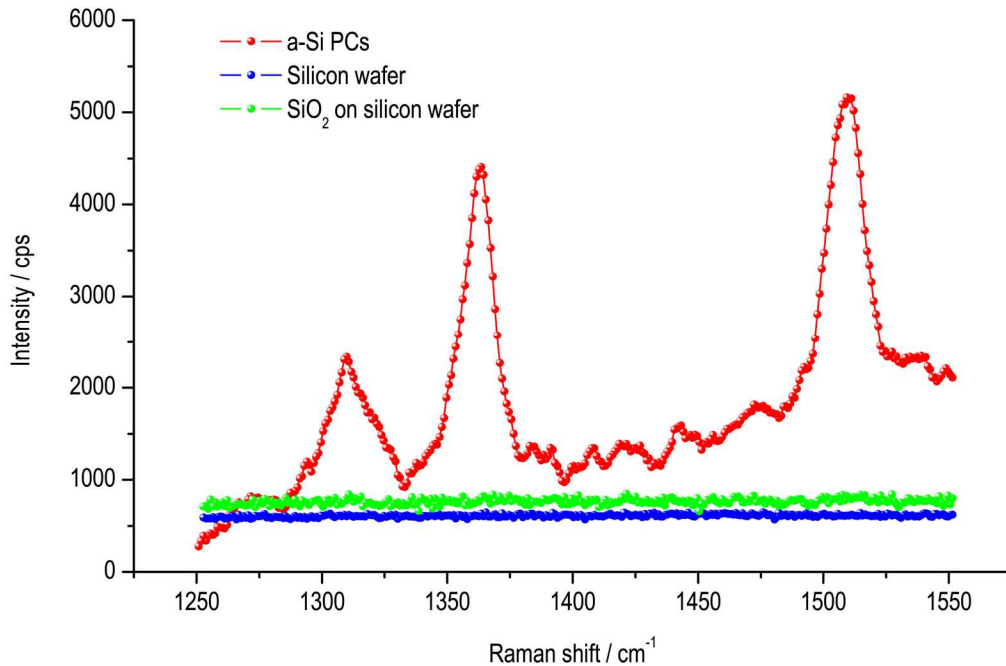


Figure S4. Raman spectra recorded from a-Si PCs (red), silicon wafer (blue) and SiO₂ on silicon wafer (green) with C_{R6G} of 10^{-3} M. For spectra recording, 20 μ L of 10^{-3} M R6G solution was spread on the three substrates with a spreading area of 2 mm in diameter, and the solution was allowed to evaporate completely before test. The laser power was fixed to 25 mW, 1%, and integration time was 20 s.

S5. Raman spectra of 4-MBA molecules on MDPCs.

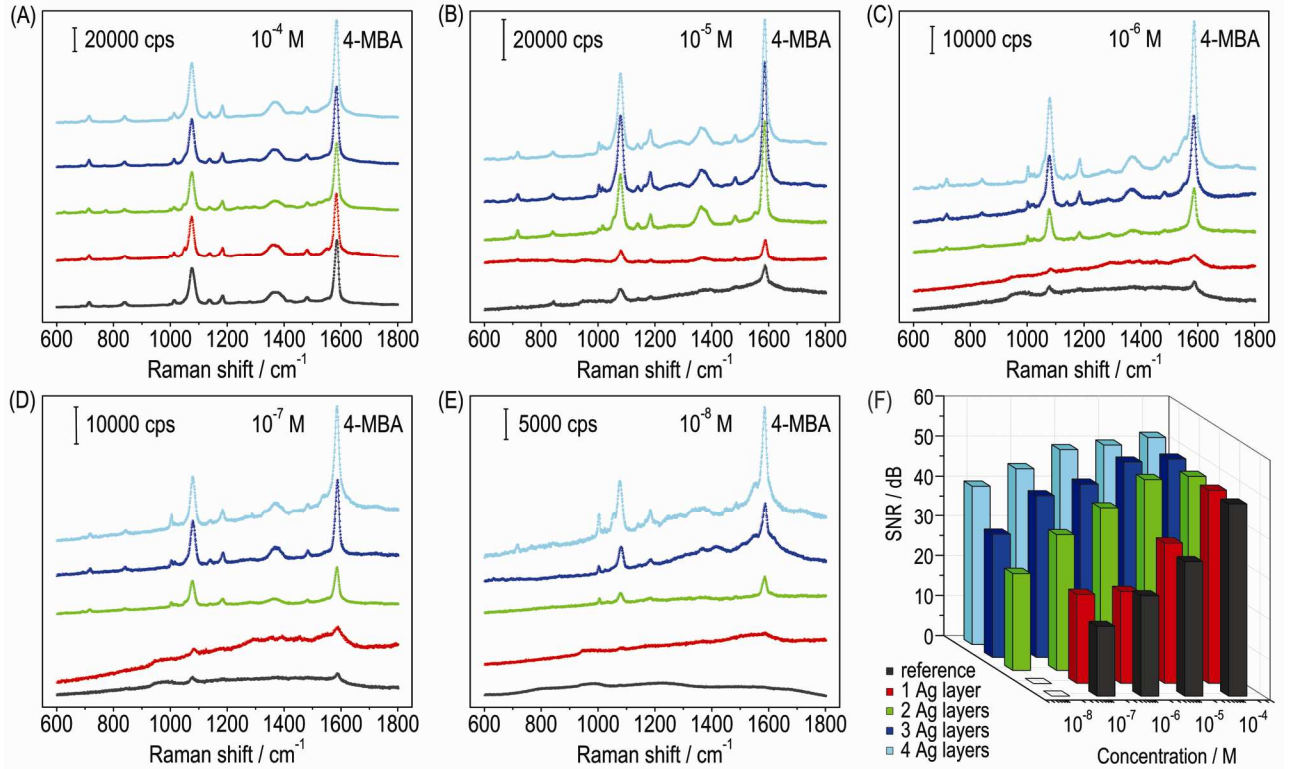


Figure S5. (A)–(E) Raman spectra recorded from 4-MBA on MDPCs with concentration decreasing from 10^{-4} to 10^{-8} M. The spectra in each diagram correspond to the MDPCs with one (red), two (green), three (blue), and four (cyan) MD units, respectively. The dark gray curves are

obtained from the reference substrates. (F) SNR analysis of a characteristic 1584 cm^{-1} peak for all 25 substrates. Colors are assigned in accordance with the spectra.

S6. Raman spectra of R6G molecules on other physical evaporated, patterned silver films

Two kinds of islands films were fabricated: silver network-like islands film (Figure S6A) and prism-like islands film (Figure S6B). The islands films were fabricated by sputtering silver film onto the PS SAM with different time of 300 s and 60 s (ion current density: 5 mA/cm^2), followed by PS SAM removal. The surface of silicon wafer thus remained with network-like and prism-like silver particles. Subsequently, the same Raman measurement as described in the manuscript was carried out to evaluate the Raman enhancement on such islands films. Figure S6C shows the Raman spectra of the two kinds of islands films at C_{R6G} 1 nM. The two islands film didn't show comparable Raman enhancement to MDPCs.

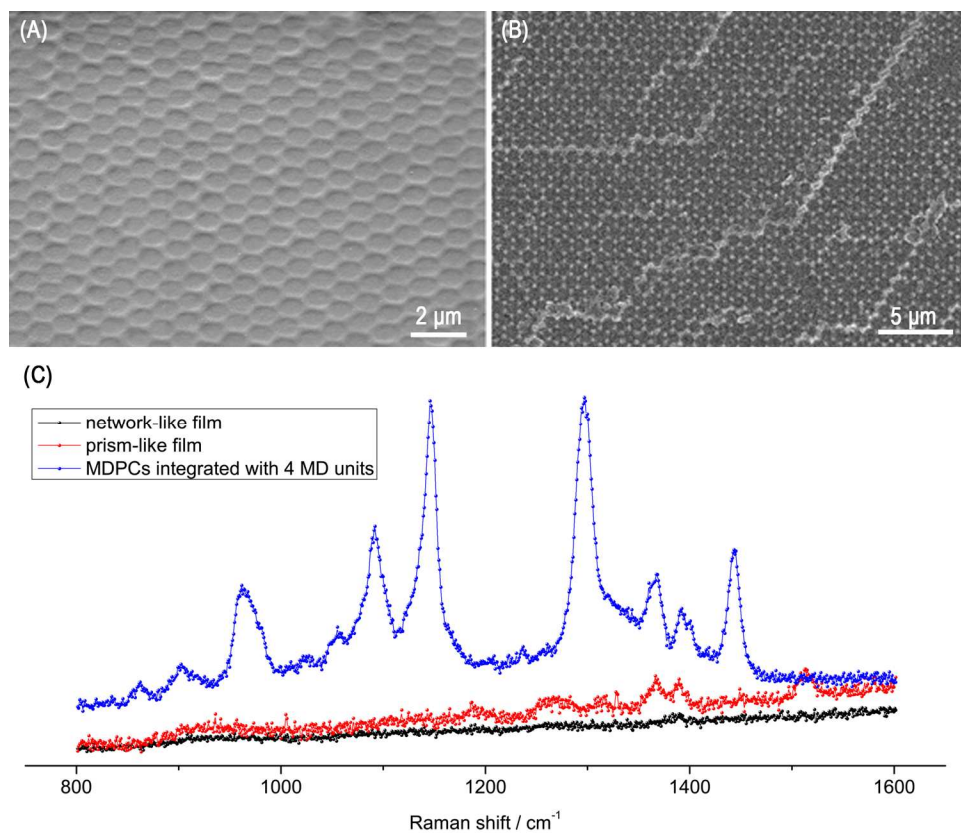


Figure S6. SEM images showing the physical evaporated silver island film: (A) network-like; (B) prism-like; (C) Raman enhancement of network-like (black) and prism-like (red) silver islands films. The reference blue curve stands for the Raman spectrum of MDPCs with 4 MD units.

S7. SERS intensity mapping of MDPCs substrates.

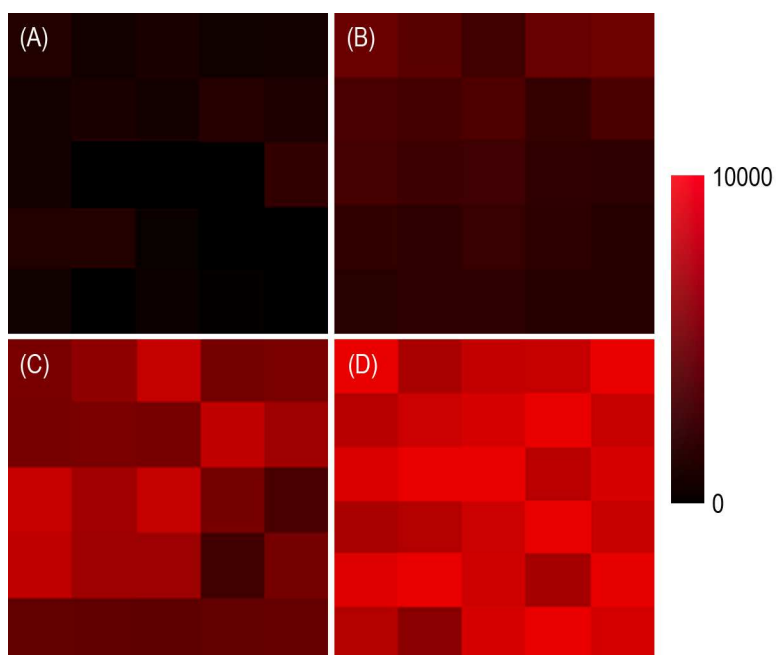


Figure S7. 5×5 SERS intensity mapping of 1509 cm^{-1} peak on MDPCs integrated with (A) one, (B) two, (C) three, and (D) four MD units. The concentration of R6G solution is 1 nM.

S8. Stability test.

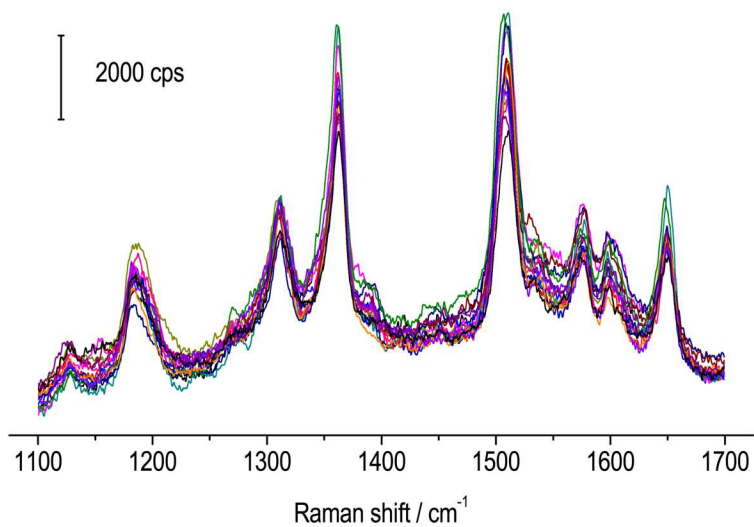


Figure S8. Stability of Raman spectra collected from the MDPCs with four MD units at C_{R6G} of 10^{-9} M. The 15 spectra were measured every two days on the same MDPCs stored in ambient air conditions.