

# Plasma amplifiers: multiscale light-enhanced uniform SERS composite substrates for breaking through resonance limitations

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### Preparation process of AuNPs/grating SERS substrate

The preparation process of AuNPs/gratings SERS substrate mainly includes photolithography, PDMS mold turning, and nanoparticle self-assembly. In this paper, a 500 um 4-inch silicon wafer is used, and the steps of lithography and PDMS mold turning are as follows:

(1) Stepper mask plate production: purchase mask plate material, get the mask plate by laser direct writing and developing, 5 times magnification mask.

(2) Homogenizer: AZ5214 homogenizer 4000 rpm 30 seconds, before baking 95 ° 90 seconds.

(3) Stepper exposure machine exposure: select the coordinate point lower left and upper right. Lower left: -25000um, -25000um. upper right 25000um, 25000um. actual step 10\*10mm.

(4) Developing: 2.38% developer for 45 seconds, deionized water for 30 seconds.

(5) Post-bake/firm film: 120° for 1 min.

(6) Deep Silicon Etching Slow: DRIE Process Steps (Bosch Process): Passivation ---- Etching - Passivation ---- Etching - Passivation: C4F8 gas is

passed into the reaction chamber to form a polymer film through a chemical reaction; Etching: SF<sub>6</sub> gas is passed into the reaction chamber to perform physical and chemical etching. Equipment Configuration: He, SF<sub>6</sub>, C<sub>4</sub>F<sub>8</sub>, O<sub>2</sub>, Ar. Center RF: 3000W Edge RF: 2000W Lower Electrode LF: 600W Chiller: -20-40°C. Fast deep silicon etching: 30mT/1800W(300W)/0W/180C4F8(50)/8T/20deg/0.8S 40mT/2500W(500W)/60W\*30%/200SF6(50)/8T/20deg/1.7S 240cycles

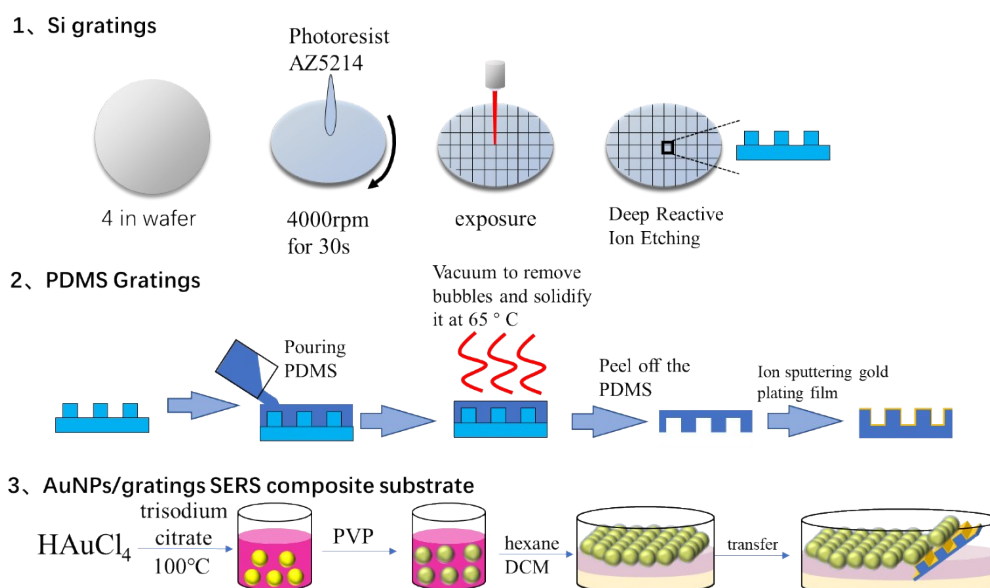
500nm etching rate approx. 400nm/min. Selection ratio 1:75-1:94.

(7) PDMS pouring: poly(dimethyl-methylvinylsiloxane) and poly(dimethyl-methylhydrogenosiloxane) were mixed 10:1, defoamed in vacuum and poured on the prepared silicon grating wafers to defoam naturally. Then put into the drying box at 85°C curing 15 min.

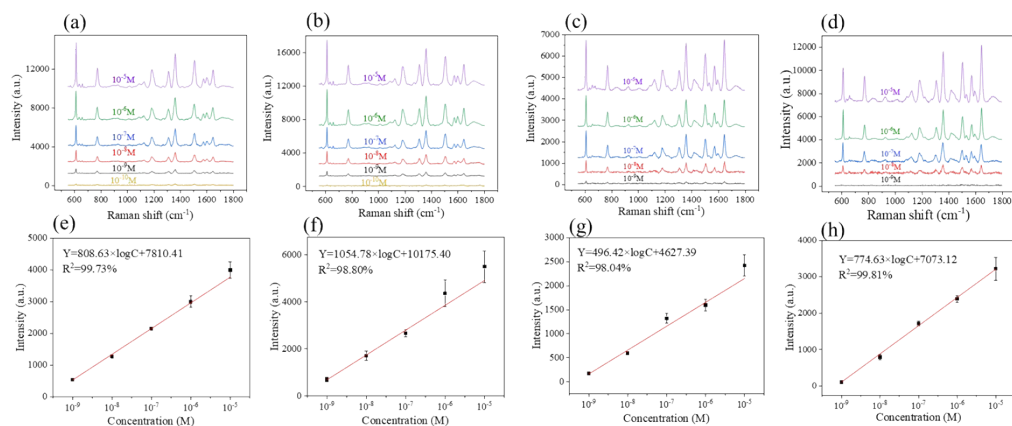
(8) After curing inverted mold: PDMS curing, the use of tweezers to carefully peel the PDMS layer of Si wafers and cut into 1\*1cm PDMS grating samples with a razor blade.

(9) Magnetron sputtering gold plating: in a relatively stable vacuum state, the cathode and anode generated between the glow discharge, the gas molecules between the poles are ionized and produce a charged charge, in which the positive ions by the cathode of the negative potential accelerated by the movement of the cathode and impact on the target material on the cathode, the atoms and other particles will be sputtered, sputtered particles will be deposited on the substrate and form a thin film at the anode. Magnetron sputtering is in the cathode target surface above the formation of an orthogonal electromagnetic field, due to the presence of an external magnetic field, the complex movement of electrons increases the ionization rate, to achieve a high-speed Au sputtering process, sputtering rate of 1 kw, 4.6 nm/s.

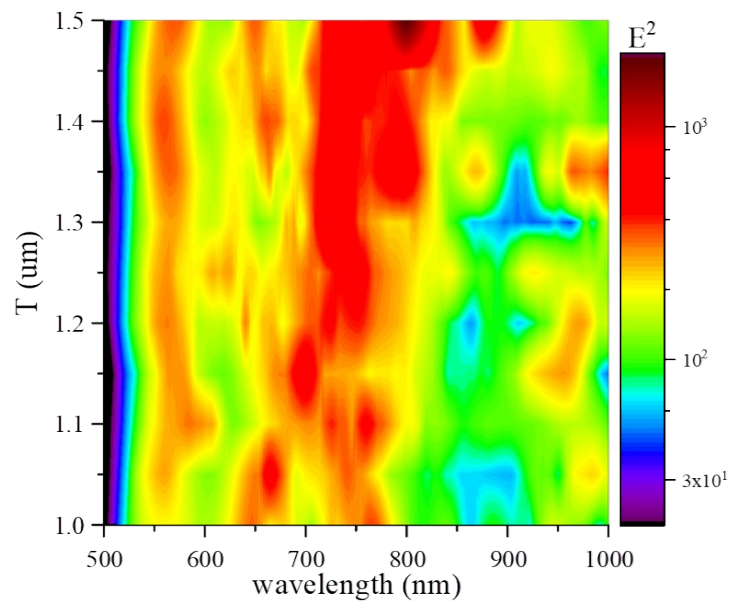
(10) Nanoparticle self-assembly technology reference main manuscript.



**Figure S1.** Schematic drawings of AuNPs/gratings SERS substrate preparation.



**Figure S2.** SERS spectrum of R6G. (a, b) Raman spectroscopy of R6G solutions at concentrations of 10<sup>-5</sup> to 10<sup>-10</sup> M for 633 nm laser using 40 and 30 nm AuNPs/gratings SERS substrates, respectively. (c, d) Raman spectroscopy of R6G solutions at concentrations of 10<sup>-5</sup> to 10<sup>-9</sup> M for 532 nm laser using 40 nm and 30 nm AuNPs/gratings SERS substrates, respectively. (e, f) Linear fitting diagrams of R6G at concentrations of 10<sup>-5</sup> M to 10<sup>-9</sup> M on 40 and 30 nm AuNPs/gratings SERS substrates, respectively. (g, h) Linear fitting diagrams of R6G at concentrations of 10<sup>-5</sup> to 10<sup>-9</sup> M on 40 and 30 nm AuNPs/gratings SERS substrates, respectively.



**Figure S3.** Spectral response of the grating period to  $E^2$