Supporting Information

Enhanced Light-Matter Interaction of Graphene-Gold Nanoparticles Hybrid Films for High-Performance SERS Detection

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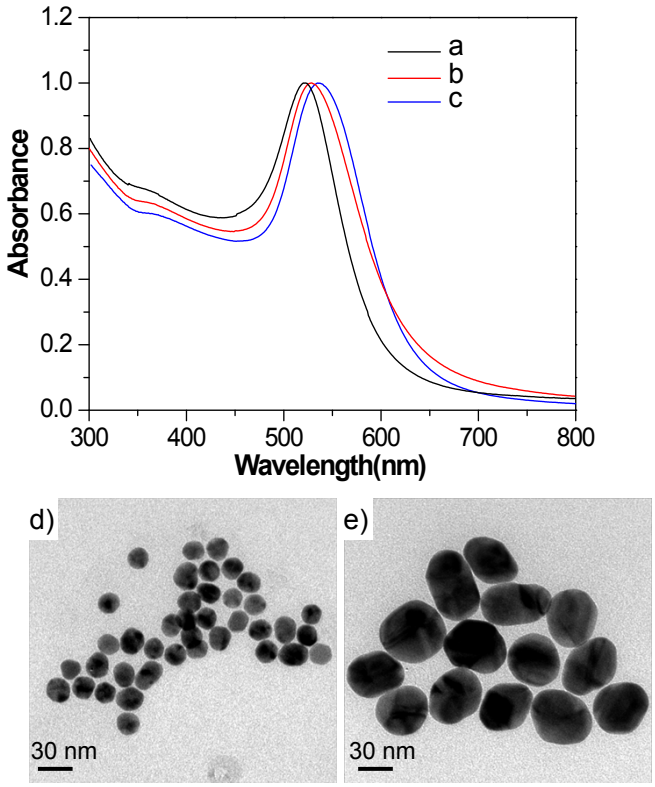


Fig. S1 The normalized absorbance spectra of Au NPs colloids with different sizes: a) ~15 nm, b) ~30 nm and c) ~45 nm. TEM images of Au NPs with different sizes: d) ~15 nm and e) ~45 nm.

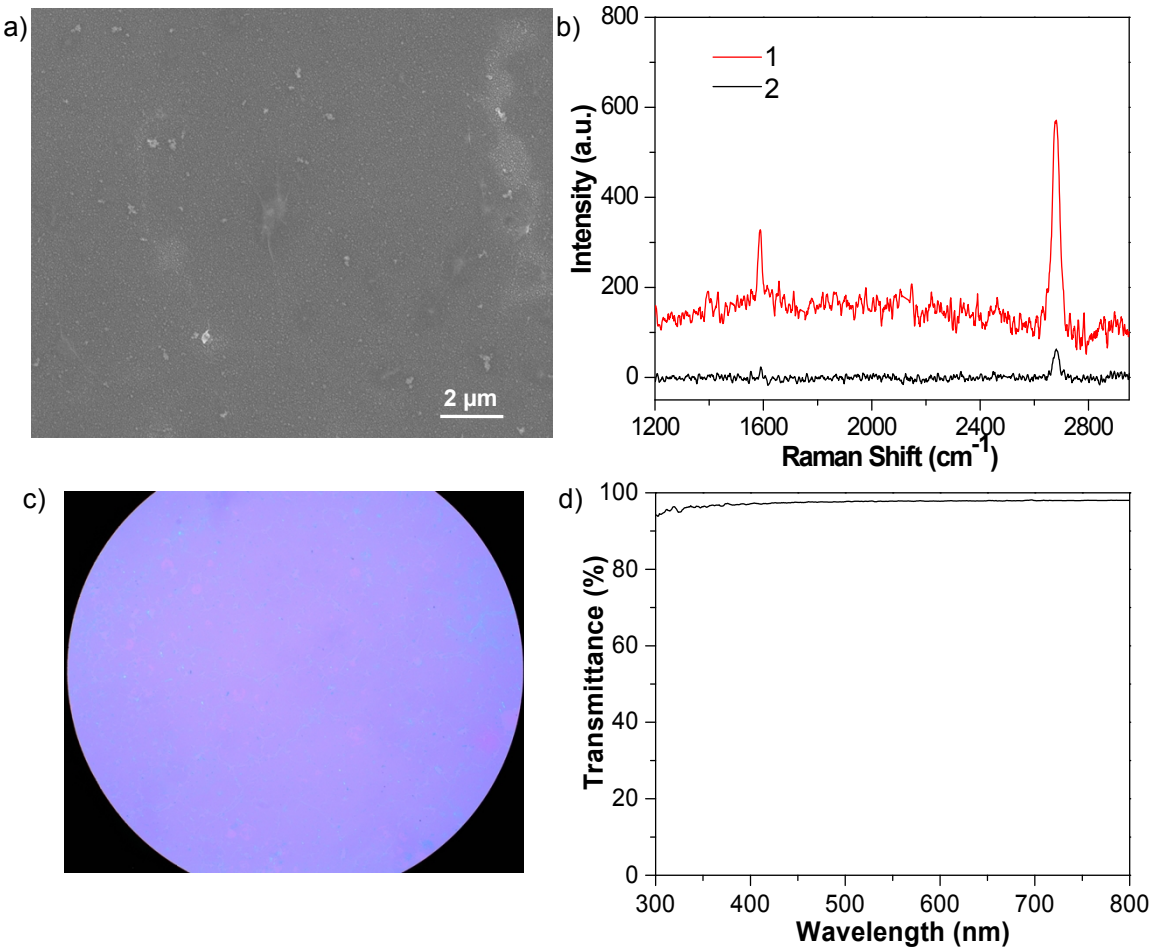
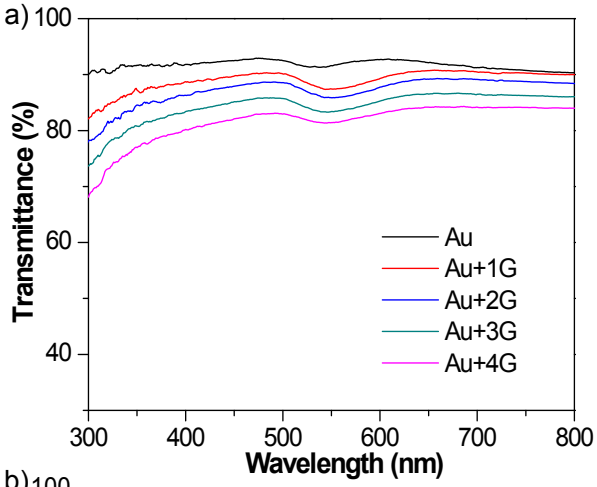
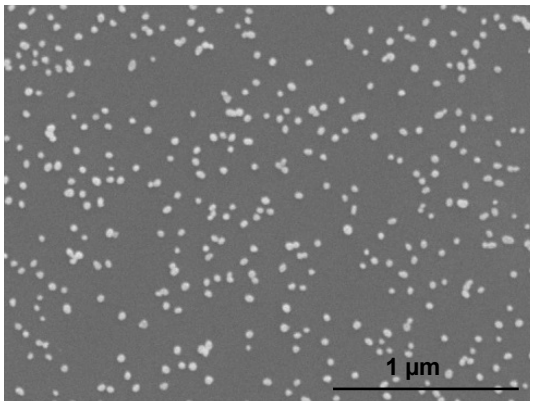
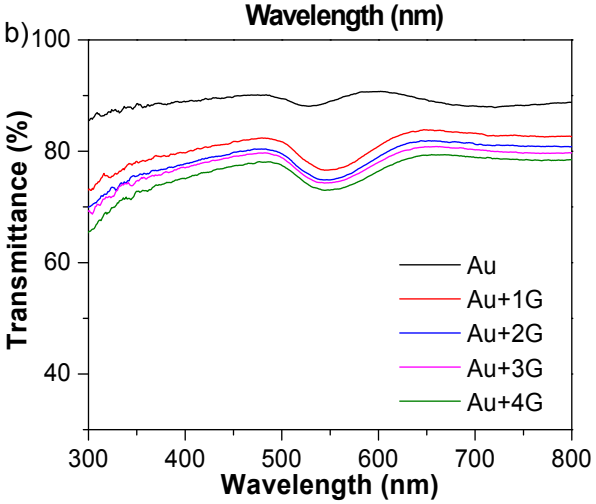
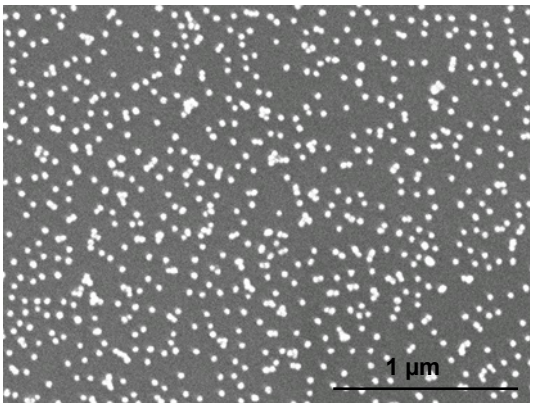


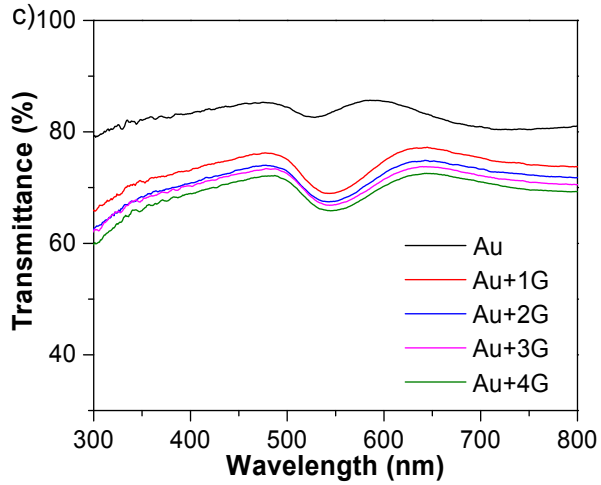
Fig. S2 a) SEM image of Au NPs covered with a large-area monolayer graphene. The dark region is covered by graphene and the coverage is estimated to be about 97%. b) Raman spectra of graphene on (1) Au NPs/quartz and (2) bare quartz substrate. c) Optical image of monolayer graphene transferred on a SiO_2/Si substrate. d) Transmission spectra of monolayer graphene on quartz.

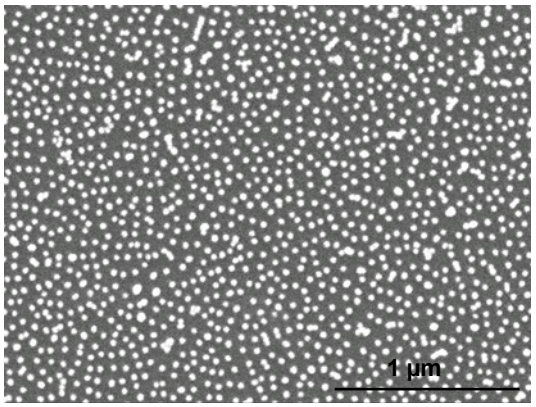

a)
Transmittance (%)
Wavelength (nm)
Au
Au+1G
Au+2G
Au+3G
Au+4G


1 μm


b)
Transmittance (%)
Wavelength (nm)
Au
Au+1G
Au+2G
Au+3G
Au+4G


1 μm


c)
Transmittance (%)
Wavelength (nm)
Au
Au+1G
Au+2G
Au+3G
Au+4G


1 μm

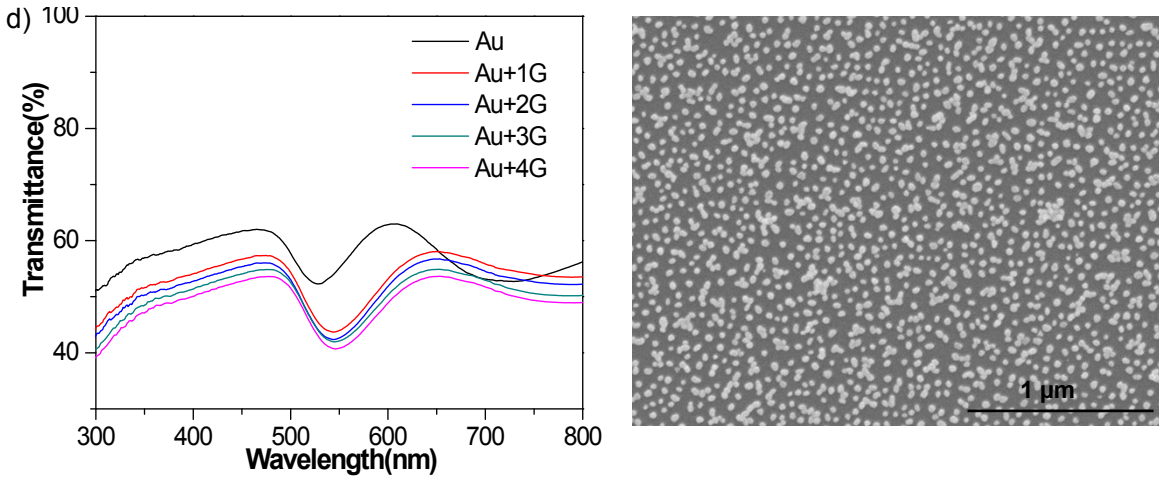


Fig. S3 The transmittance spectra of ~30 nm Au NPs immobilized on quartz substrate with different density a) ~69, b) ~122, c) ~221 and d) ~408 No./μm^2 before and after transferring different layers graphene (right: the corresponding SEM images of Au NPs on Si substrates before transferring graphene).

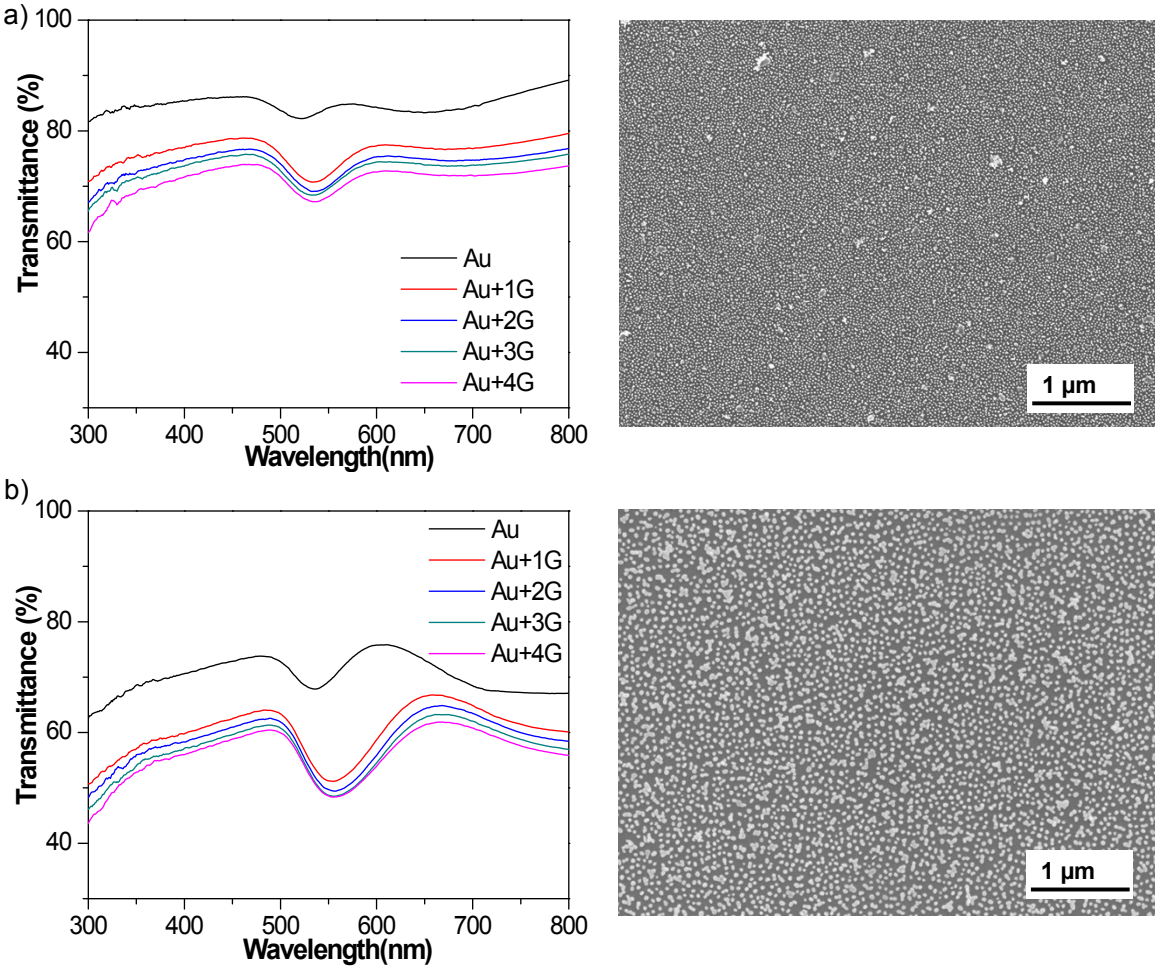


Fig. S4 Transmission spectra of a) ~15 nm Au NPs with density of ~1372 No./μm^2 (corresponding to a coverage of 24.23%) and b) ~45 nm Au NPs with density of ~309 No./μm^2 (corresponding to a coverage of 49.12%) immobilized on quartz substrates before and after different layers of graphene were transferred (right: the corresponding SEM images of Au NPs on Si substrates). Coverage is defined as the total area of particles above the projection per unit area. The decrease in absolute transmittance is about 11.4% and 15.6% for ~15 nm and ~45 nm Au nanoparticles coated by monolayer graphene, while the resonance peak red-shift is about 11 nm and 20 nm for ~15 nm and ~45 nm graphene-Au NPs hybrids, respectively.

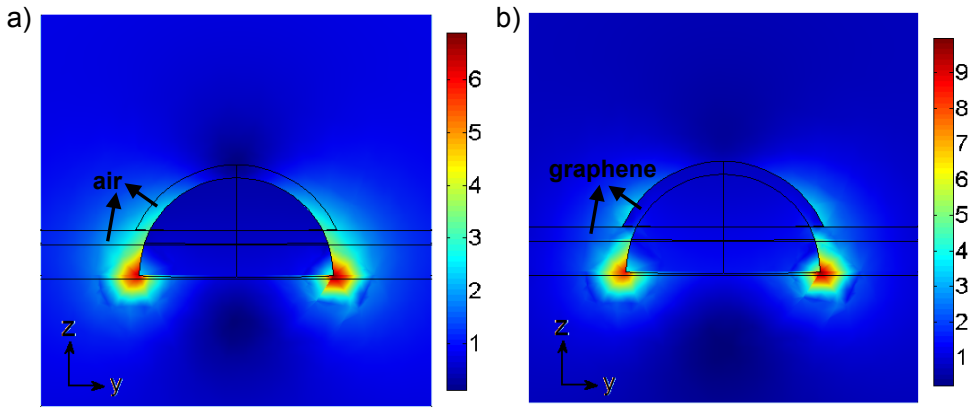


Fig. S5 Simulated electric field distributions in the y-z plane for AuNPs a) without graphene and b) with monolayer graphene, respectively.

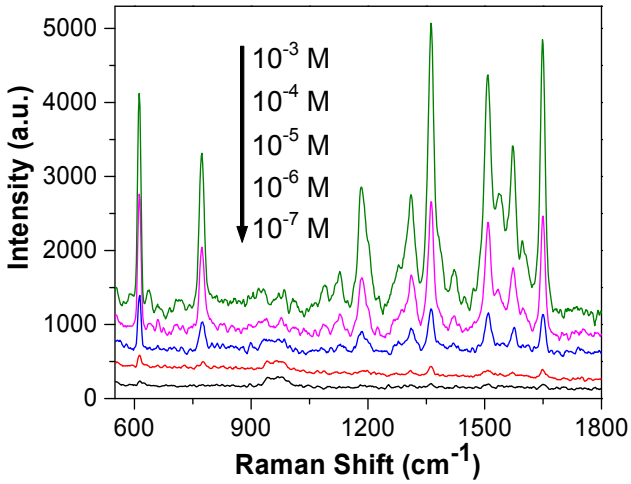


Fig. S6 Raman spectra of R6G on bare Au NPs on quartz substrates with different concentrations.

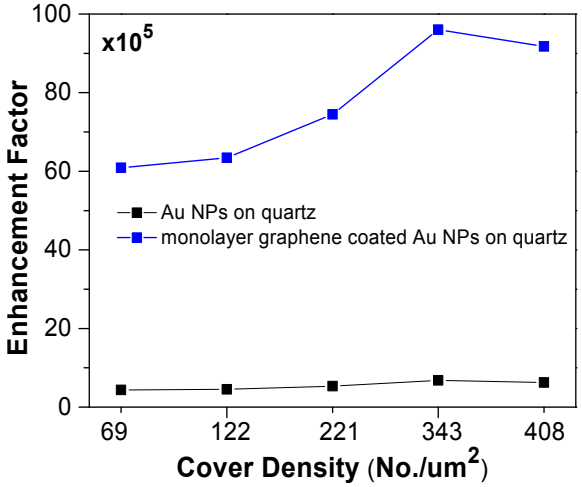


Fig. S7 SERS enhancement factors for monolayer graphene coated Au NPs hybrid films and bare Au NPs films, respectively, as a function of Au NPs cover density.

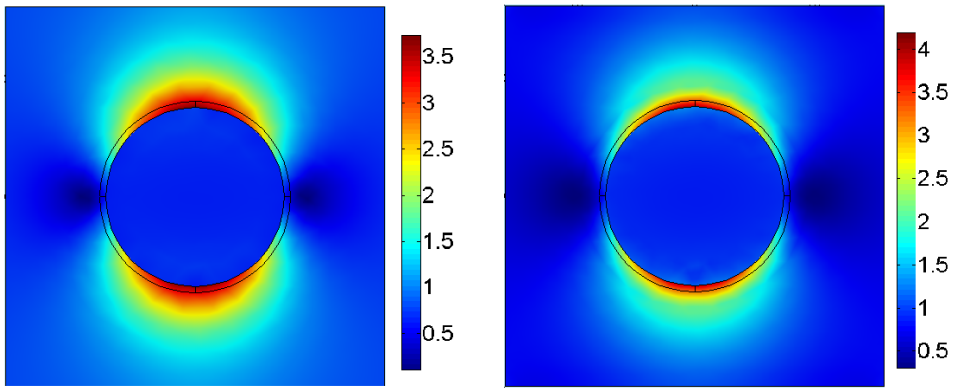


Fig. S8 The simulated electric field distributions at the laser wavelength of 532 nm for Au NPs without graphene (left) and with monolayer graphene covered (right).

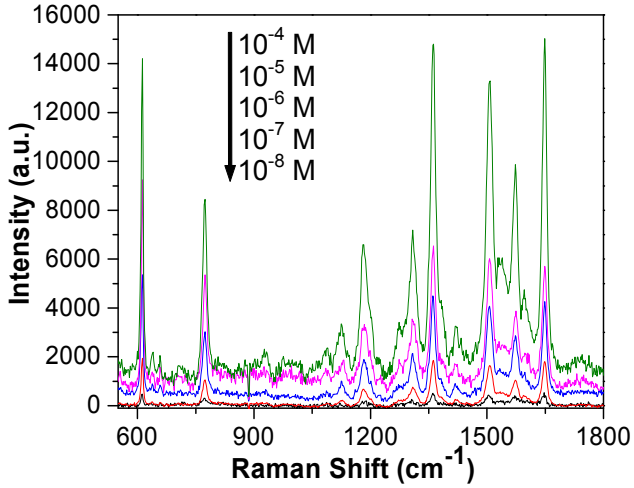


Fig. S9 SERS spectra of R6G with varied concentrations on the same graphene-Au NPs hybrid film done after half a year.

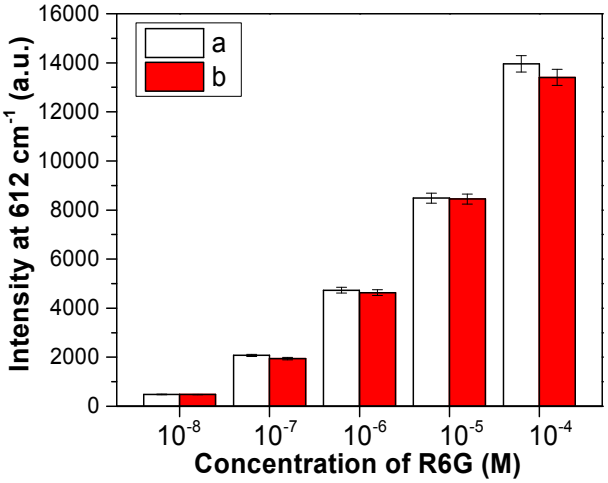


Fig. S10 Raman intensity of R6G at 612 cm^{-1} peak with varied concentrations on the same graphene-Au NPs hybrid film done a) half a year ago and b) a few days ago.

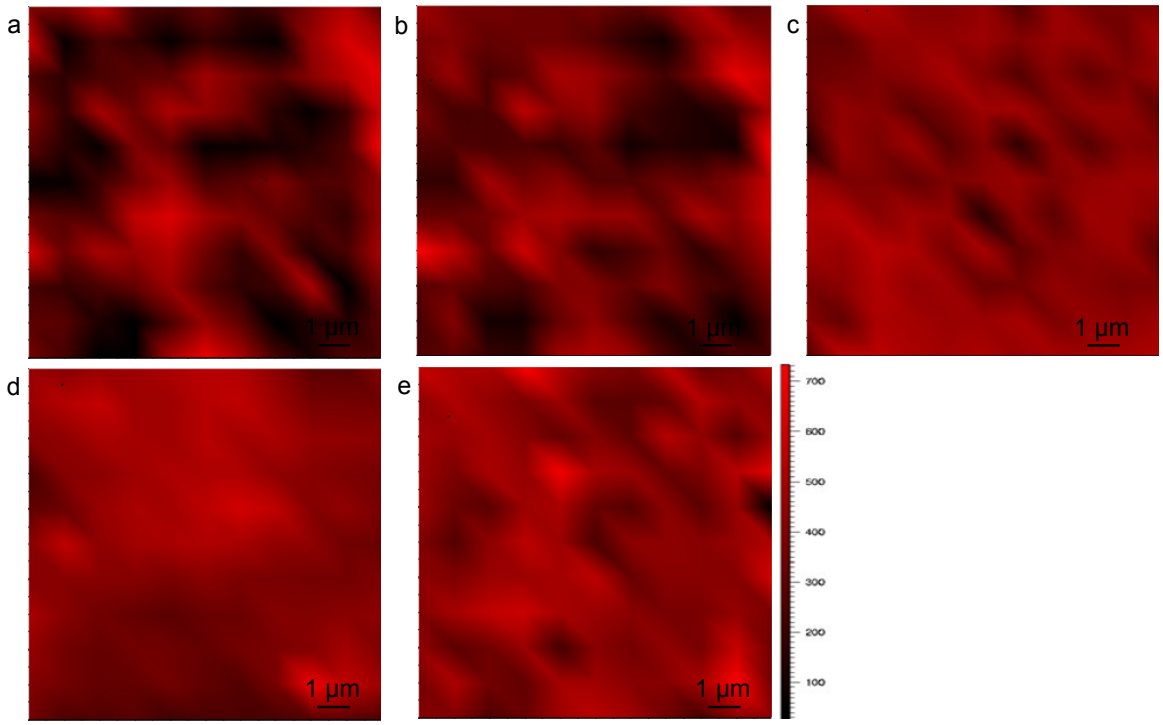


Fig. S11 Spatial resolved Raman intensity mappings of 612 cm^{-1} peak of 10^{-6} M R6G on Au NPs film with different cover density a) ~69, b) ~122, c) ~221, d) ~343 and e) ~408 No./μm^2. Clearly the Au NPs sample with the cover density of ~343 No./μm^2 has the best uniformity and sensitivity thus has been selected to be coated with graphene layers for further uniformity as discussed in the manuscript.

Cover density (No./μm^2)	Au NPs in aggregates (No.)	Au NPs total quantity (No.)	Aggregates ratio (%)
69	156	1180	13.22
122	299	2078	14.38
221	527	3748	14.06
343	800	5833	13.71
408	1365	6939	19.63

Table S1. Numbers of total Au NPs and of those in aggregates, and corresponding aggregation ratios for different Au NPs cover densities.