Optimization of Nanoparticle-Based SERS Substrates through Large-Scale Realistic Simulations
-- Supporting Information --

Diego M. Solís,[†] **José M. Taboada,**[‡] **Fernando Obelleiro,**[*,†]
Luis M. Liz-Marzán, [¶,∥,⊥] **and F. Javier García de Abajo**[*,§,#]

[†]Departamento de Teoría de la Señal y Comunicaciones, University of Vigo, 36301 Vigo, Spain
[‡]Departamento de Tecnología de Computadores y Comunicaciones, University of Extremadura, 10003 Cáceres, Spain,
[¶]Bionanoplasmonics Laboratory, CIC biomaGUNE, Paseo de Miramón 182, 20014 Donostia-San Sebastian, Spain
[∥] Ikerbasque, Basque Foundation for Science, 48013 Bilbao, Spain
[⊥] CIBER de Bioingeniería, Biomateriales y Nanomedicina, CIBER-BBN, 20014 Donostia-San Sebastian, Spain
[§]ICFO-Institut de Ciencies Fotoniques, The Barcelona Institute of Sci. and Techn., 08860 Castelldefels (Barcelona), Spain
[#]ICREA-Institució Catalana de Recerca i Estudis Avançats, Passeig Lluís Companys 23, 08010 Barcelona, Spain

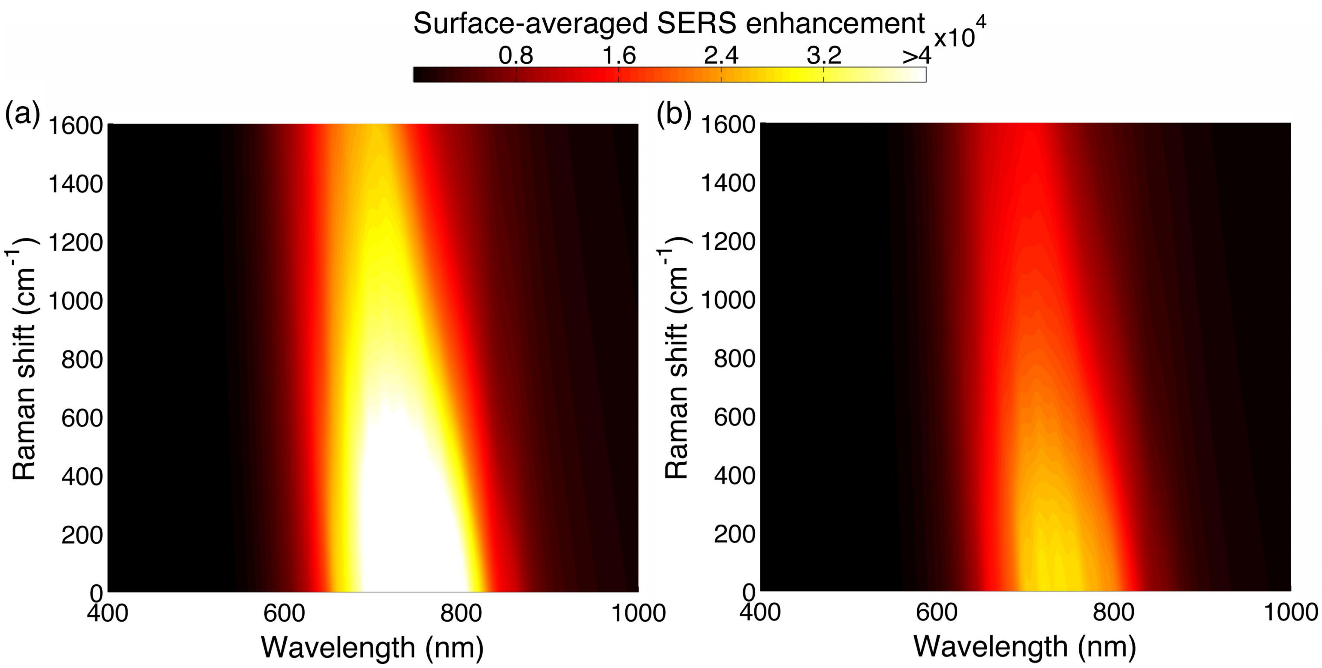

Figure S1. Same as Figure 1 of the main paper for a monolayer of gold nanorods (GNRs). (a) and (b) correspond to skin-type and sheet type molecular coverage, respectively. The layer consists of 740 GNRs (65 nm × 21 nm) compacted in a 1.2×1.2 μm^2 film, with a minimum surface-to-surface separation of 1 nm. The molecule-surface separation is also 1 nm. The surrounding medium is assumed to be homogeneous air for simplicity.

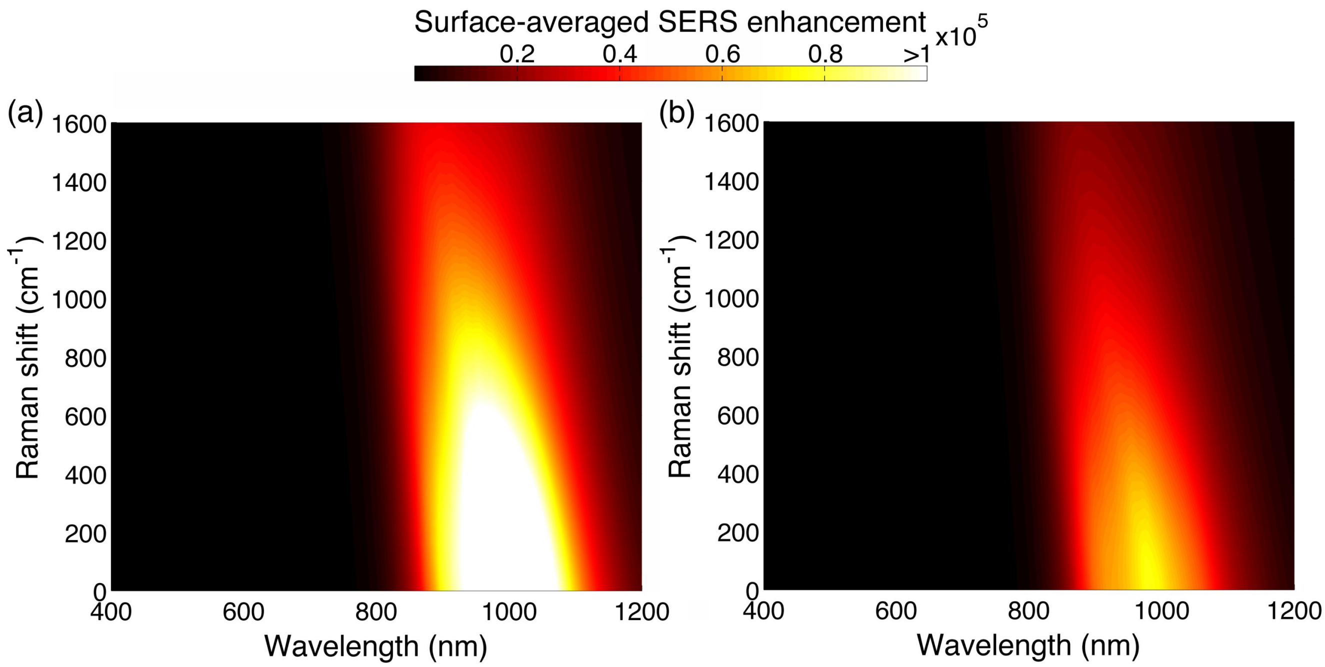

Figure S2. Same as Figure 1 of the main paper for a monolayer of gold nanostars (GNSs). (a) and (b) correspond to skin-type and sheet type molecular coverage, respectively. The layer consists of 504 GNSs (20 nm spherical core diameter and 10 tips with 1 nm apex rounding radius, 15.5 nm length, and 16.6^{o} angle) compacted in a 1.2×1.2 μm^2 film, with a minimum surface-to-surface separation of 1 nm. The molecule-surface separation is also 1 nm. The surrounding medium is assumed to be homogeneous air for simplicity.

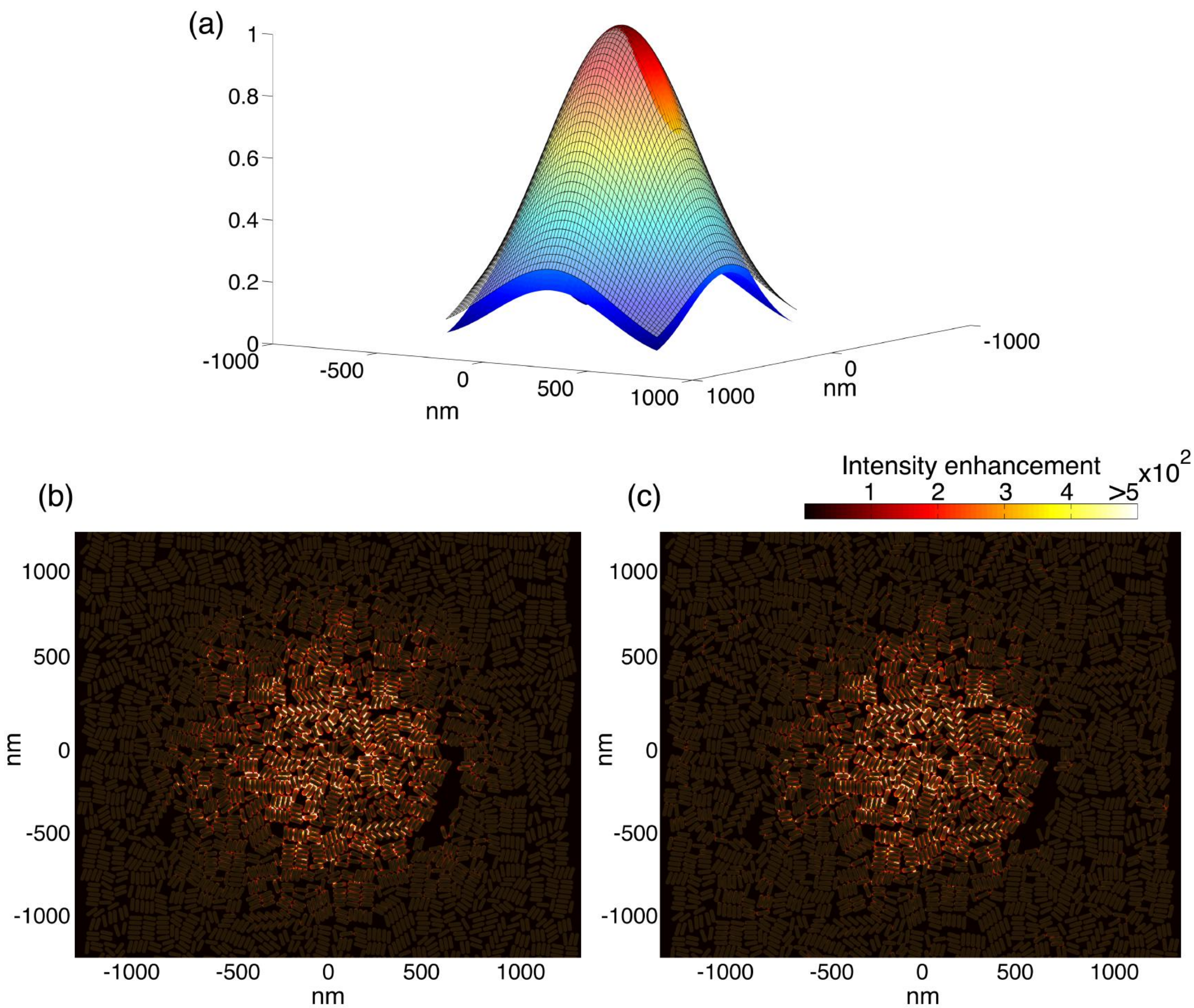

Figure S3. Gaussian light-beam (NA=0.58) versus Gaussian-weighted (0.39 λ standard deviation) plane-wave illumination. (a) Beam profiles, with the upper gridded surface corresponding to the Gaussian beam. (b,c) Near-electric-field distribution on a random GNR monolayer as produced by the Gaussian-weighted plane wave (b) and the Gaussian light beam (c), respectively.

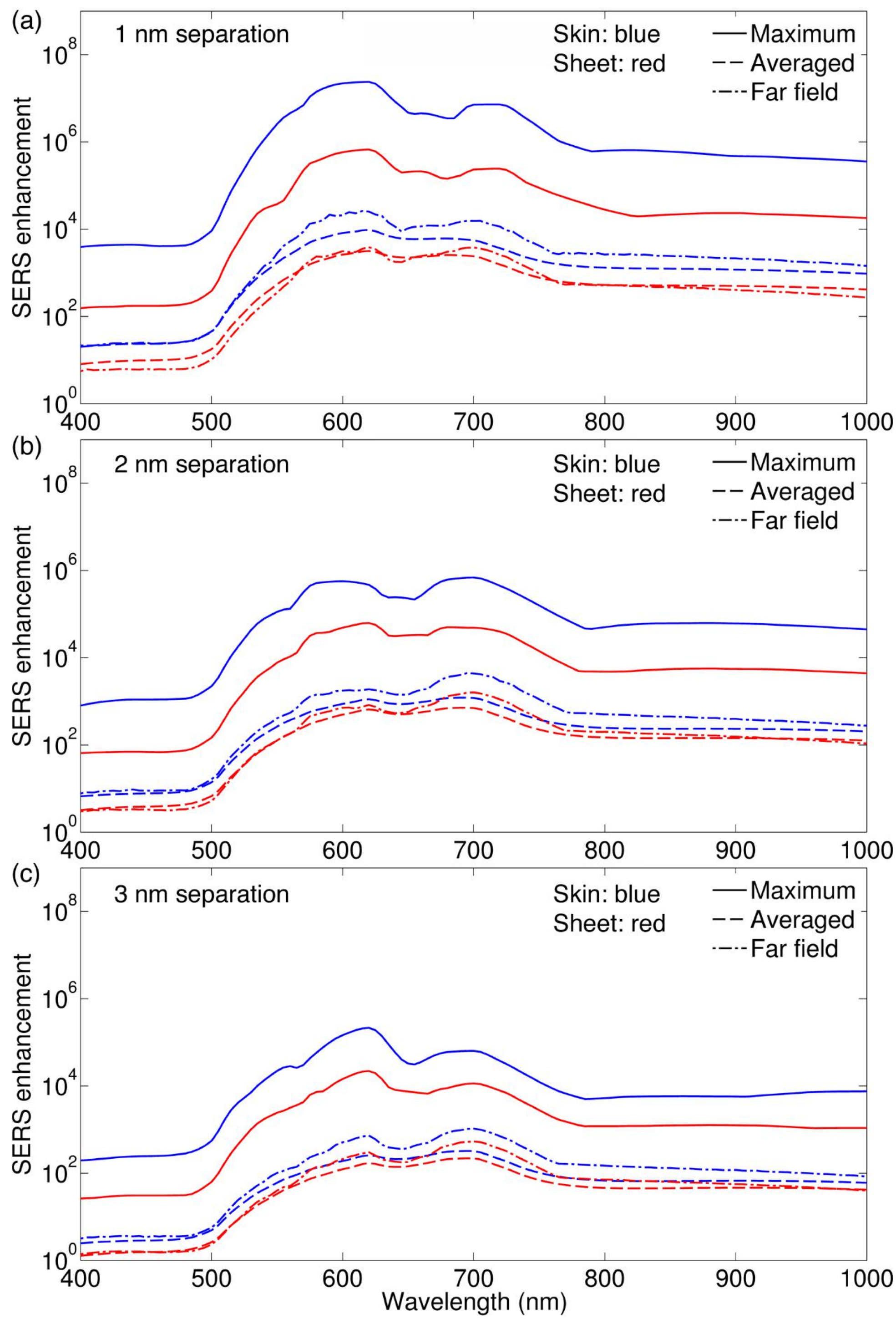

Figure S4. SERS performance of arrays of gold nanospheres (GNPs) for different molecule-surface distances and coverage models. We show the incidence-wavelength dependence of the maximum value of the SERS enhancement on the entire sample (solid curves), the surface-averaged SERS enhancement (dashed curves), and the far-field enhancement observed with a NA=1.4 objective (dash-dotted curves) for three different separations of the molecules relative to the gold surface and either skin-type (blue) or sheet-type (red) coverage. The array is the same as in the GNP sample in Figure 3 of the main paper. We consider normal light incidence averaged over polarizations.

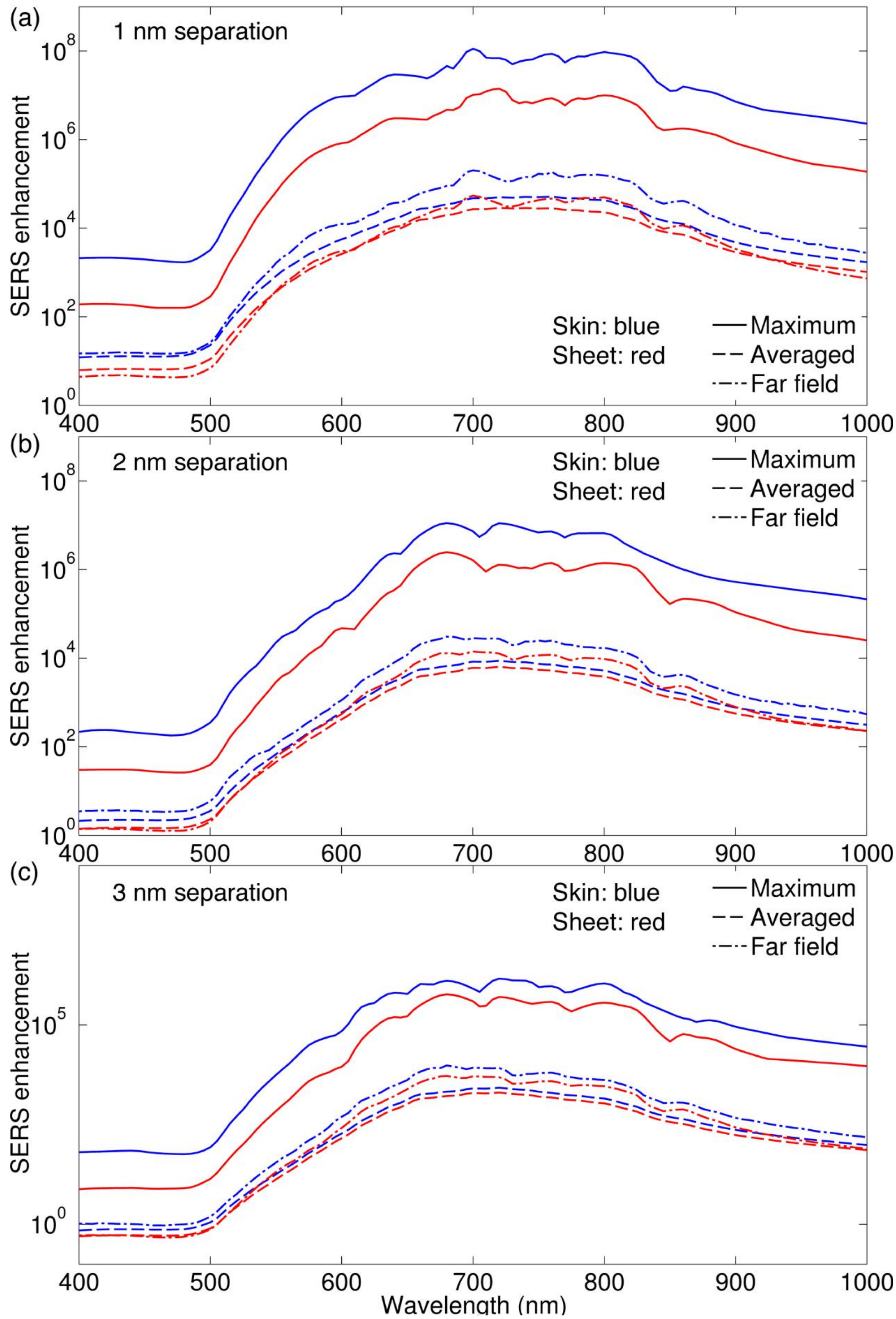

Figure S5. SERS performance of arrays of GNRs for different molecule-surface distances and coverage models. We show the incidence-wavelength dependence of the maximum value of the SERS enhancement on the entire sample (solid curves), the surface-averaged SERS enhancement (dashed curves), and the far-field enhancement observed with a NA=1.4 objective (dash-dotted curves) for three different separations of the molecules relative to the gold surface and either skin-type (blue) or sheet-type (red) coverage. The array is the same as in the GNR sample in Figure 3 of the main paper. We consider normal light incidence averaged over polarizations.

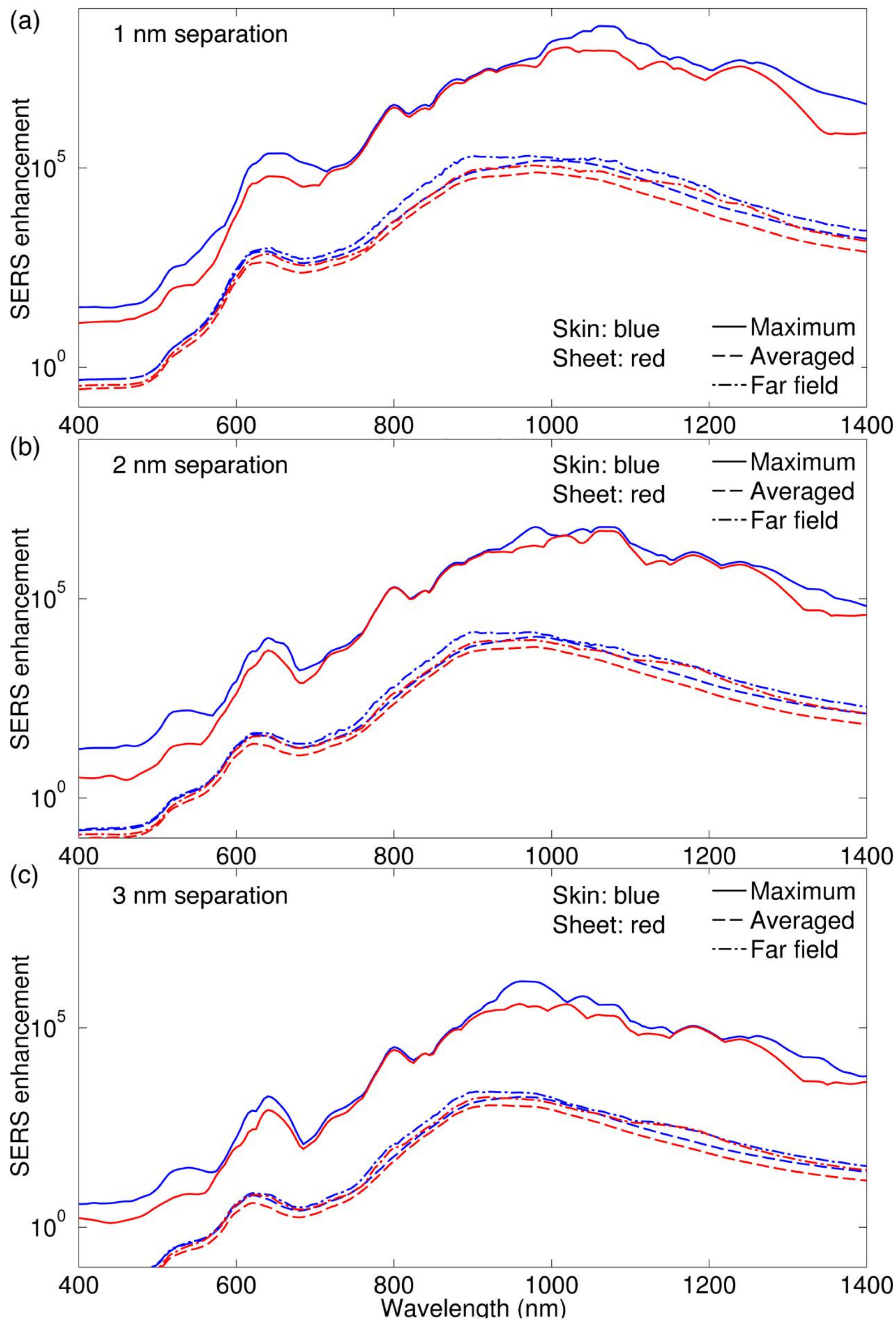

Figure S6. SERS performance of arrays of GNSs for different molecule-surface distances and coverage models. We show the incidence-wavelength dependence of the maximum value of the SERS enhancement on the entire sample (solid curves), the surface-averaged SERS enhancement (dashed curves), and the far-field enhancement observed with a NA=1.4 objective (dash-dotted curves) for three different separations of the molecules relative to the gold surface and either skin-type (blue) or sheet-type (red) coverage. The array is the same as in the GNS sample in Figure 3 of the main paper. We consider normal light incidence averaged over polarizations.

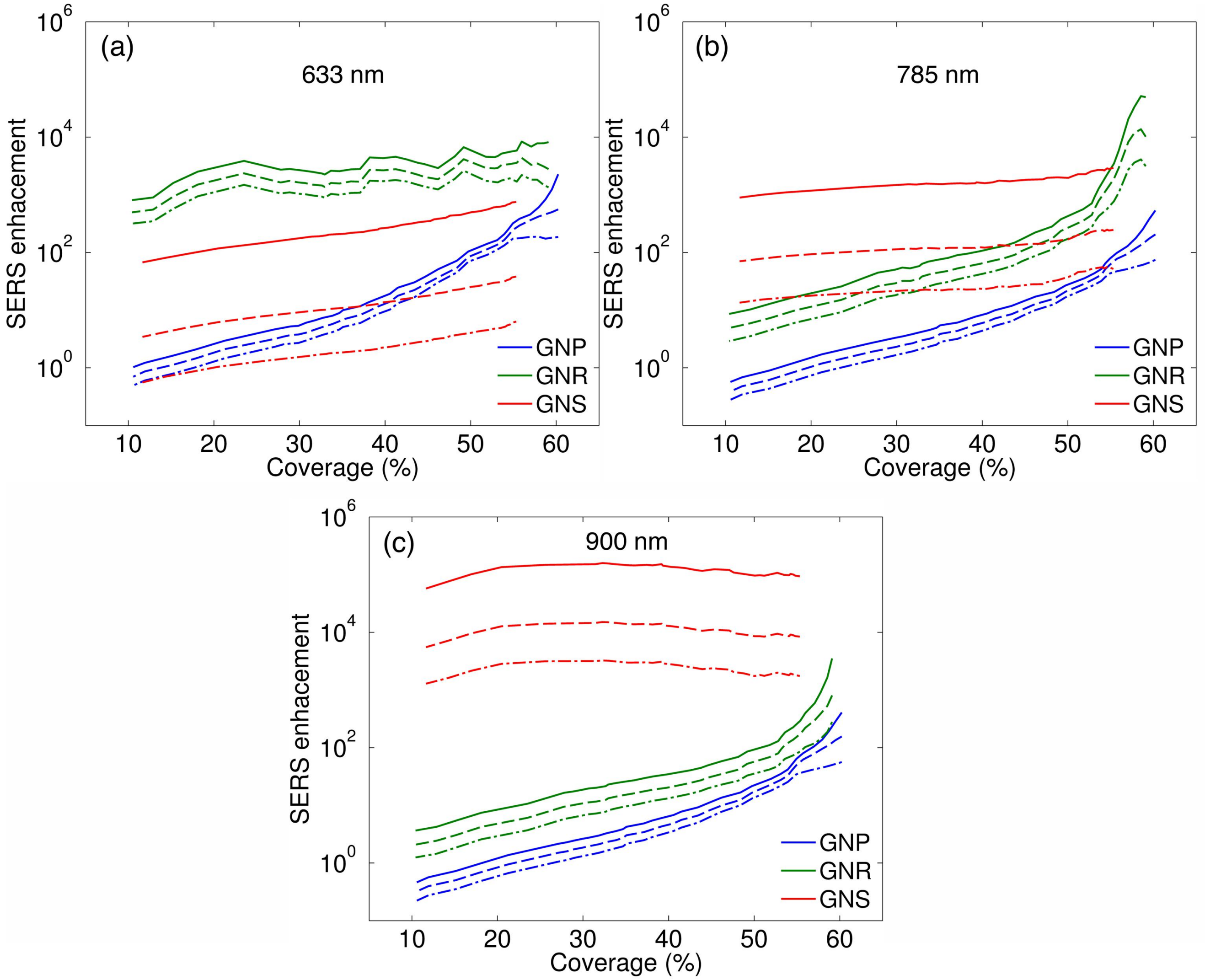

Figure S7. Same as Figure 5 of the main paper for different light wavelengths and molecule-surface separations. We consider skin-type molecular coverage with a molecule-surface distance of 1 nm (solid curves), 2 nm (dashed curves), or 3 nm (dash-dotted curves). The light wavelengths are indicated by labels.

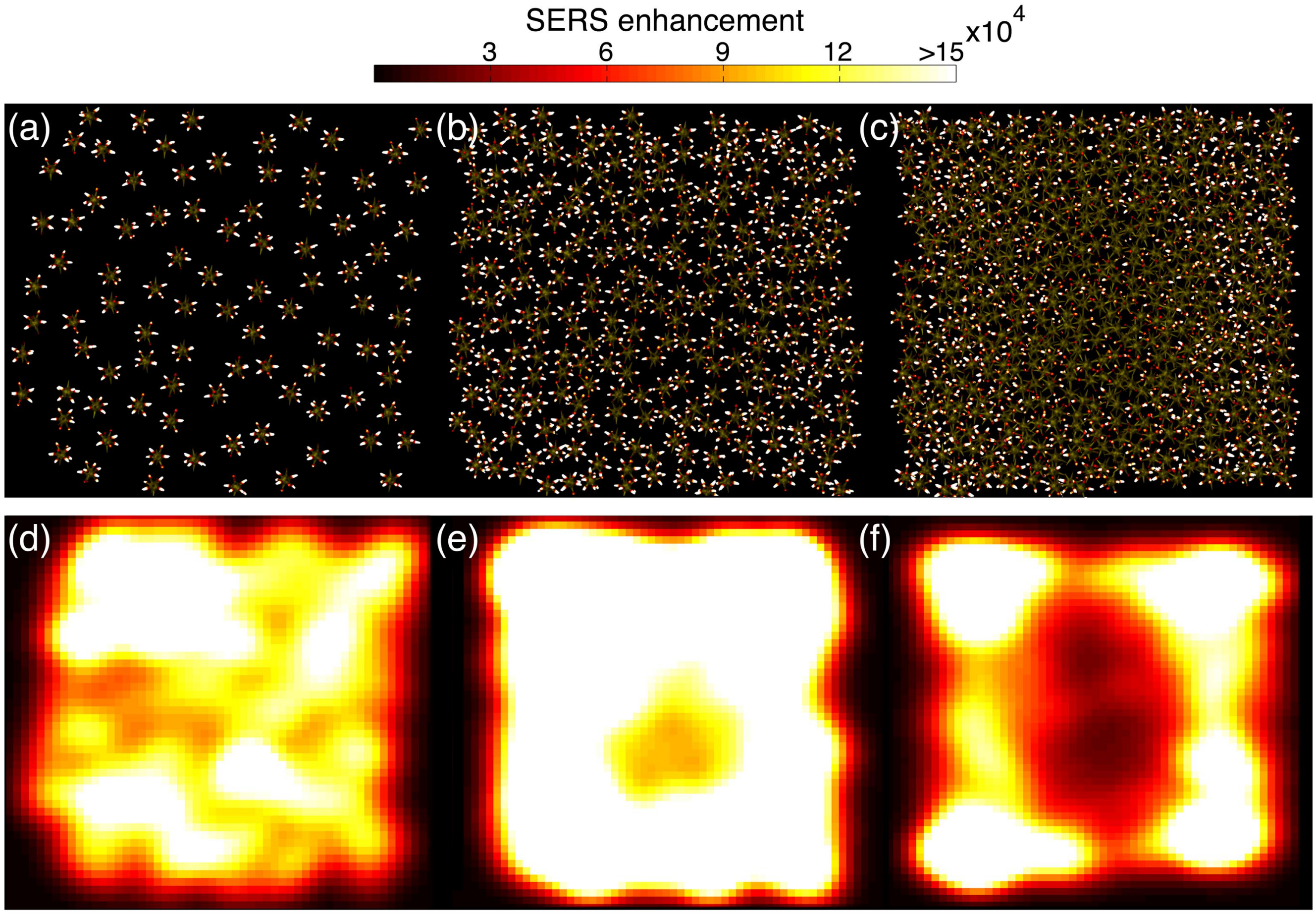

Figure S8. Same as Figure 6 of the main paper for linearly polarized light with polarization along the vertical axes of the plots.

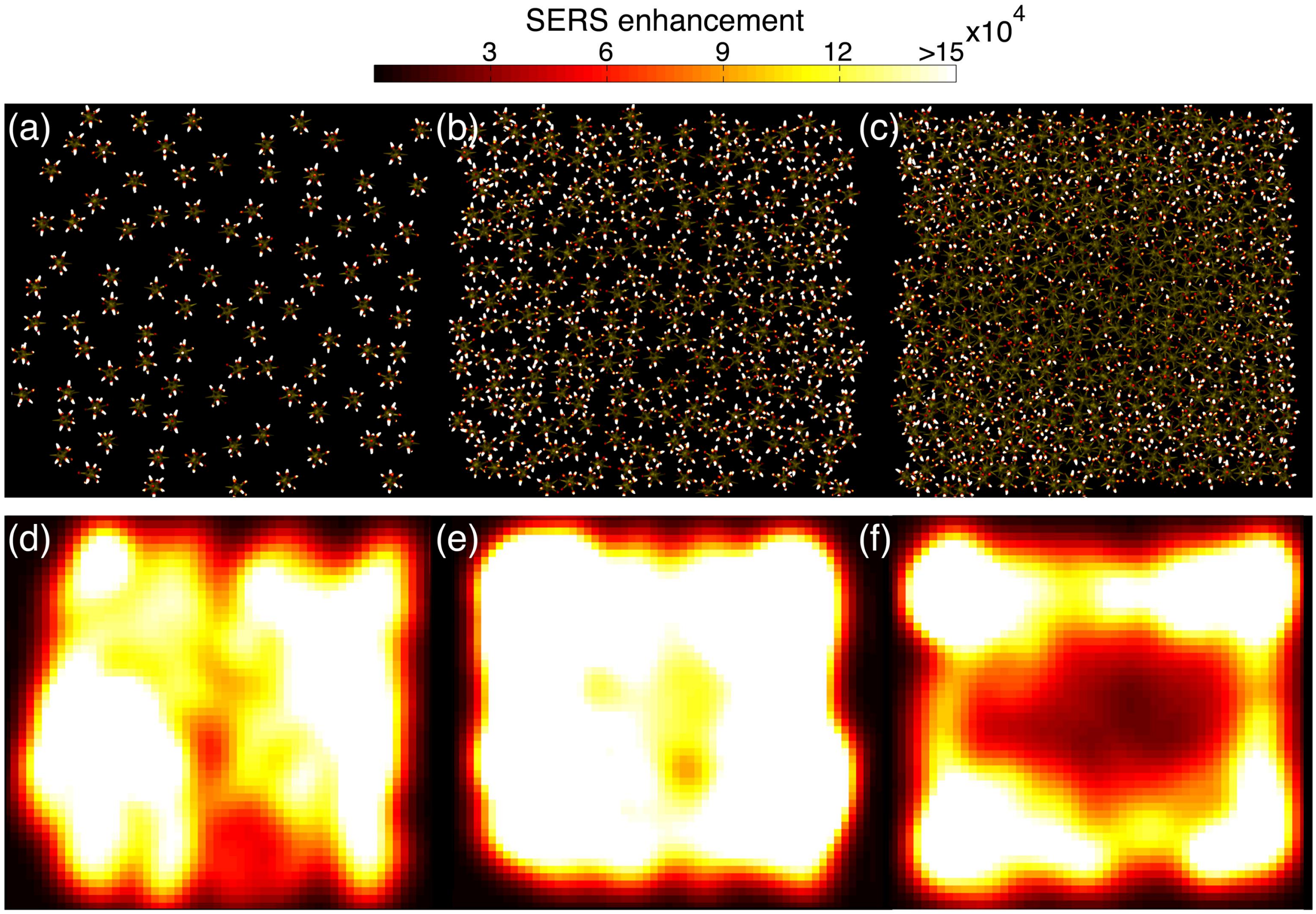

Figure S9. Same as Figure 6 of the main paper for linearly polarized light with polarization along the horizontal axes of the plots.