Supplementary Information

Single-molecule Detection and Radiation Control in solutions at high concentrations via a heterogeneous Optical Slot Antenna

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Plasmonic mode hybridization of the Au-OSA:

Fig. S1 shows the wavelength dependence of the near field variation at the origin of an Au-OSA with water or objective immersion oil as the surrounding medium, respectively. The whole system is symmetric about the gold film when using immersion oil as superstrate. There are two resonant peaks for the Au-OSA with water (centered at λ =600 and 750 nm) or oil superstrate (centered at λ =626 and 808 nm). The two resonant peaks, red shifts with superstrate of higher refractive index, are results of plasmonic hybridization between the upper (water- or oil-Au interface) mode ω_u and lower (glass-Au interface) mode ω_l of the Au-OSA as schematically shown in the inset of Fig. S1a. The upper mode ω_u and lower mode ω_l are the intrinsic plasmonic modes at the top and bottom of the Au-OSA when the thickness of the gold film is infinity. As the film thickness decreases, the two modes begin to overlap and interact with each other, resulting in two new modes ω_+ and ω_-. The higher energy mode ω_+ corresponds to the resonant peak at λ =600 nm (626 nm) with water (immersion oil) superstrate. The lower energy mode ω_- corresponds to the resonant peak at λ =750nm (808 nm) for water (immersion oil) superstrate. The mode hybridization was further verified by changing the metal film thickness as shown in Fig. S2.When the gold film thickness is 300 nm, the two modes no longer overlap and only the lower intrinsic mode ω_l is excited since the incident light illuminate the structure from glass substrate resulting in one peak centered at λ =680 nm. Fig. S1b shows the near-field distribution for the two modes of the Au-OSA with oil superstrate. The instantaneous current flow direction along x axis J_x is marked by green arrows and the resulting instantaneous charges are also marked as positive (+) or negative (-) depending on the current flow direction. We define the symmetric mode if the current flow direction is the same on both surface, otherwise, it is anti-symmetric. The symmetric mode $\omega_-$and anti-symmetric mode ω_+ of the Au-OSA with oil superstrateare excited at λ = 808 and 626 nm, respectively. The corresponding charges accumulated at the edge of the Au-OSA shown in Fig. S1b are with the same (opposite) signs at top and bottom of the Au-OSA for the symmetric (anti-symmetric) mode. The fields from the two opposite charge oscillations result in destructive interference at middle of the Au-OSA as shown in Fig. S1b.

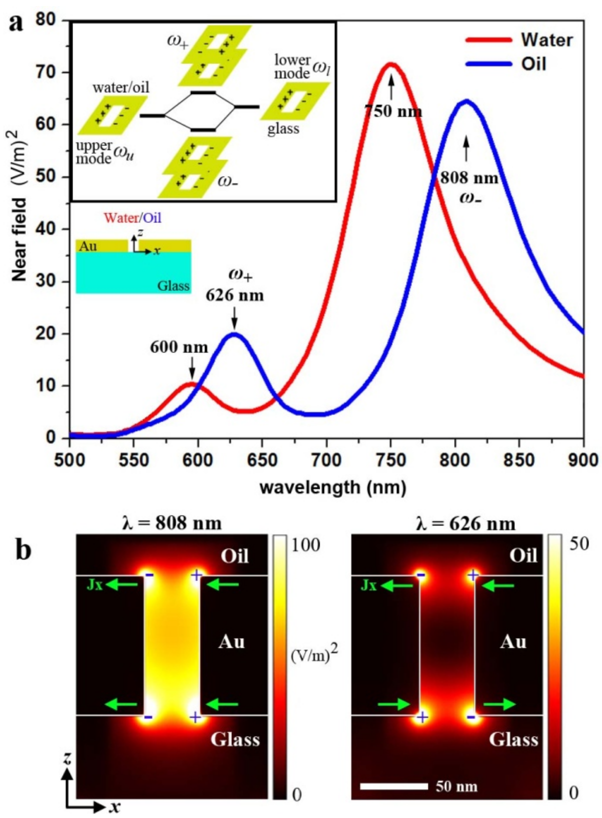

Fig. S1. a. Wavelength dependence of the near-field intensity of a Au-OSA with water or oil as the surrounding medium. The inset shows the OSA structure within the xz plane. The inset in the black box schematically shows the plasmonic mode hybridization. **b.** Field distribution for the symmetry and anit-symmetry mode in the xz planes. The green arrows shows the instantaneous direction of the x compenent J_x of the curren flow and the corresponding sign of the charges are also marked.

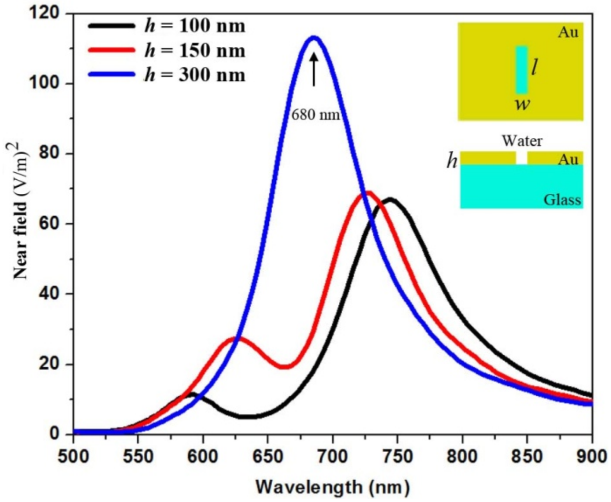

Fig. S2. Wavelength dependence of the near-field intensity for an Au-OSA with three different gold film thickness (h = 100, 150, and 300 nm). Inset schematically shows the structure of the Au-OSA. Near-field intensity was recorded at origin (x=0, y=0, z=0 nm).

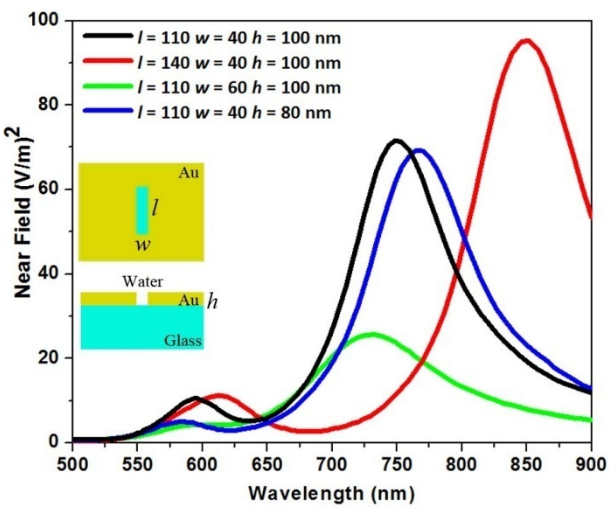

Fig. S3. Plasmonic resonance variation as a function of the width, length, and depth of the Au-OSA. The black solid curve corresponds to the plasmonic resonant peak of an Au-OSA with parameters w = 40, l = 110 and h = 100 nm, the same curve shown in Fig. 3a in the main text.

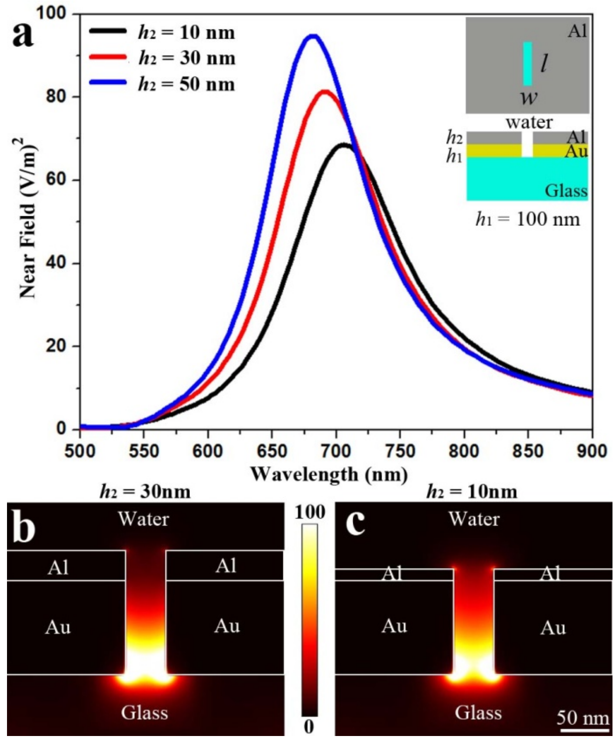

Fig. S4. **a.** Wavelength dependence of the near-field intensity for a heterogeneous OSA with different Al film thickness (h_2 =10, 30, and 50 nm). The thickness of the gold film is kept constant at h_1= 100nm. Inset schematically shows the structure of the heterogeneous OSA. Near-field intensity was recorded at origin (x=0, y=0, z=0 nm). **b.** Field distribution for a heterogeneous OSA (Au thickness h_1 = 100nm, Al thickness h_2 = 30nm) in the xz plane at its plasmonic resonant wanvelength λ = 700 nm. **c.** Field distribution for a heterogeneous OSA (Au thickness h_1 = 100nm, Al thickness h_2 = 10nm) in the xz plane at its plasmonic resonant wanvelength λ = 710 nm.

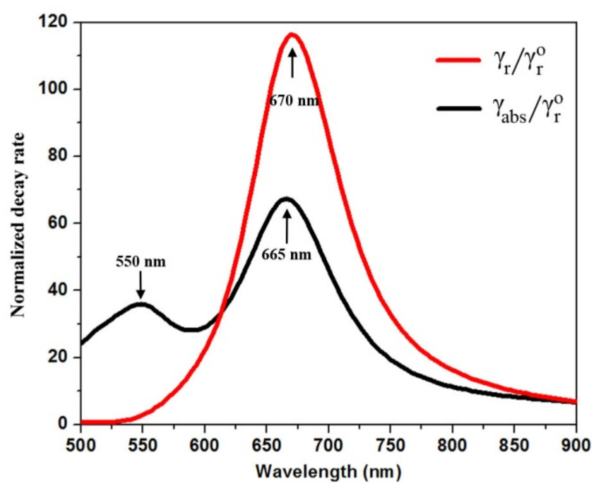

Fig. S5. Wavelength dependence of the normalized radiative and nonradiative decay rate for a molecule located at bottom of a heterogeneous OSA (x=0, y=0, z=10 nm) 10 nm above the glass substrate.