Supporting information for “A Comparative Investigation of Plasmonic Properties between Tunable Nanoobjects and Metallized Nanoprobes for Optical Spectroscopy”

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We describe in details the measurement procedure of the geometrical parameters for Au nanocones (Figure 4 of the Article) and the correction procedure introduced in numerical simulations.

In order to describe the finite apex radius of nanocones, we use a sphere-cone description based on the nanocone half angle θ, base diameter D, and apex radius r. The nanocone geometric properties have been measured experimentally from scanning electron microscopy (SEM) images (example shown in Figure SI-1). Nanocones have been covered for this purpose by a 5 nm layer of Ge to ensure SEM imaging without charging effects. Experimentally, we measured the nanocone half angle, the nanocone nominal height H_0 (extrapolated height assuming an infinitely sharp cone), and the nanocone actual height H. Data for θ, H_0 and H extracted from SEM images are shown in Table SI-1 (left column). The nanocone base diameter D is geometrically derived from θ and H_0, as well as the nanocone apex.

A source of error in these measurements lay in the Ge coating of nanocones and residual blurring of the nanocone edges upon SEM imaging. To account for this, we introduced a correction procedure in the simulations, in which the cone half-angle value has been slightly adjusted in order to fit the wavelength of the main resonance observed in experimental TIR scattering spectra of Figure 5, while keeping constant the values of H_0 and H extracted from SEM measurements. Adjusted values for θ are shown in Table SI-1 (right). They only differ from SEM values by a few percent at most, as well as the affected values of the nanocone base diameter D and nanocone apex radius, as shown in Table SI-1 (right). The corrected values of θ, D and r have been used in numerical simulations shown in Figure 4 of the main Article.

A particular situation is found for the nanocone "NC1", for which corrected values for θ, D and r have not been shown in Table SI-1. This structure is, in reality, closer to a double nanocone as seen from the SEM image shown in Figure SI-1 (right). It has been adequately treated as a double sphere-cone structure in numerical simulations.

	Nanocone parameters derived from SEM					Nanocone parameters used in numerical simulations					
	Parameters measured by SEM			Base diameter *D* (nm)	Apex radius *r* (nm)	Correction		Base diameter *D* (nm)	error	Apex radius *r* (nm)	error
(nm)	θ	H_0 (nm)	*H* (nm)			θ	error				
NC 1	12°	175	107.3	74.4	17.8	-	-	-	-	-	-
NC 2	12.15°	143.6	108.1	61.8	9.5	12.25°	0.8%	62.4	0.8%	9.6	1.0%
NC 3	12.95°	158.7	130.8	73.0	8.1	12.65°	2.4%	71.2	2.5%	7.8	3.0%
NC 4	10.4°	206	158	75.6	10.6	10.75°	2.2%	74	2.3%	10.3	2.7%
NC 5	10.5°	235.1	203.3	87.2	7.1	10.65°	1.4%	88.4	1.4%	7.2	1.7%
NC 6	15.85°	217	195.8	123.2	8.0	16.5°	3.9%	128.6	4.2%	8.4	5.3%
NC 7	13.7°	234.5	213.1	114.4	6.6	14.25°	3.9%	119.2	4.0%	7.0	5.0%
NC 8	13.75°	263.7	243.4	129.0	6.3	13.75°	0.0%	129.0	0.0%	6.3	0.0%
NC 9	11.8°	259	229.8	108.2	7.5	11.5°	2.6%	105.4	2.7%	7.3	3.2%

Table SI-1: Geometrical parameters of nanocones NC1 to NC9 as deduced from SEM imaging. Left columns: the parameters θ, H_0 and H have been measured from scanning electron micrographs (see Figure SI-1, left) and the base diameter D and apex radius r derived from geometrical considerations (see text). The right column shows the parameters used in numerical simulations: the parameter θ has been adjusted in order to fit the main scattering spectrum resonance wavelength (the error indicates the variation with respect to the value of θ measured from SEM); the values of D and r have been recalculated, taking into account the corrected value of θ.

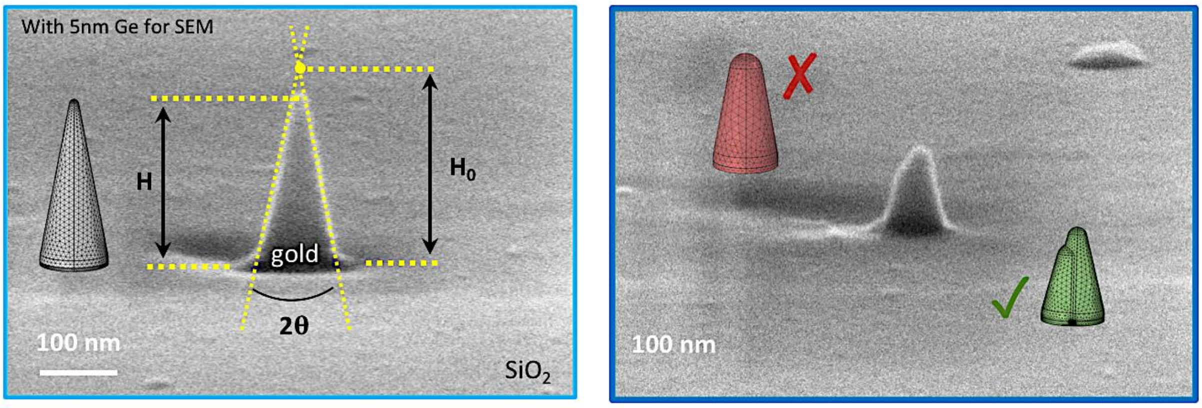

Figure SI-1: Left image: SEM image of a nanocone with the measured parameters θ, H_0 and H. Right image: specific case of the nanocone NC1, appearing as a double nanocone (see text).

Numerical modeling has also been used in this work to confirm that the efficiency of the apertureless SNOM probes proposed at the end of the study, which are expected to demonstrate a high electromagnetic enhancement factor as well as a spectral control of the plasmon resonance. We hence show in Figure SI-2 that the optical efficiency can be drastically improved by considering a metal/dielectric hybrid tip rather than a bulk metallic probe. We consider in Figure SI-2 the near-field response of a hybrid structure consisting of an isolated gold or silver tip at the apex of a semi-finite dielectric tip – with an apparent length no more than 3 µm for calculations memory optimization purposes (Figure SI-2, left). Excited under TIR configuration, several sizes of the metallic apex have been studied (ranging from 80 to 200 nm). The calculation of the electromagnetic field enhancement factor is compared to the case of a perfectly smoothed full metallic tip under the same geometry. Note that the semi-infinite character of the metal bulk tip has been obtained by merging the upper extremity of the tip with the perfect match layer (PML - see methods in the Article), avoiding then any Fabry-Pérot resonance along the 3-µm cavity, and presenting an electromagnetic enhancement only due to the lighting rod effect. No plasmonic resonance can be identified in this reference case, as seen from the blue curves on the graphics ((Figure SI-2, right)), both for Au and Ag. In the case of metal/dielectric hybrid systems, we observe strong plasmonic resonances with a spectral position defined by the size of the metallic apex and the choice of the metal at the tip apex. This numerical approach thus demonstrates the interest of hybrid metal/dielectric SNOM probes with a single plasmonic resonance due to a metallic nanostructure at is extremity with height of 80-200 nm, leading to enhancements factors of about 12000 close to a plasmon resonance tunable between 700 nm and 900 nm.

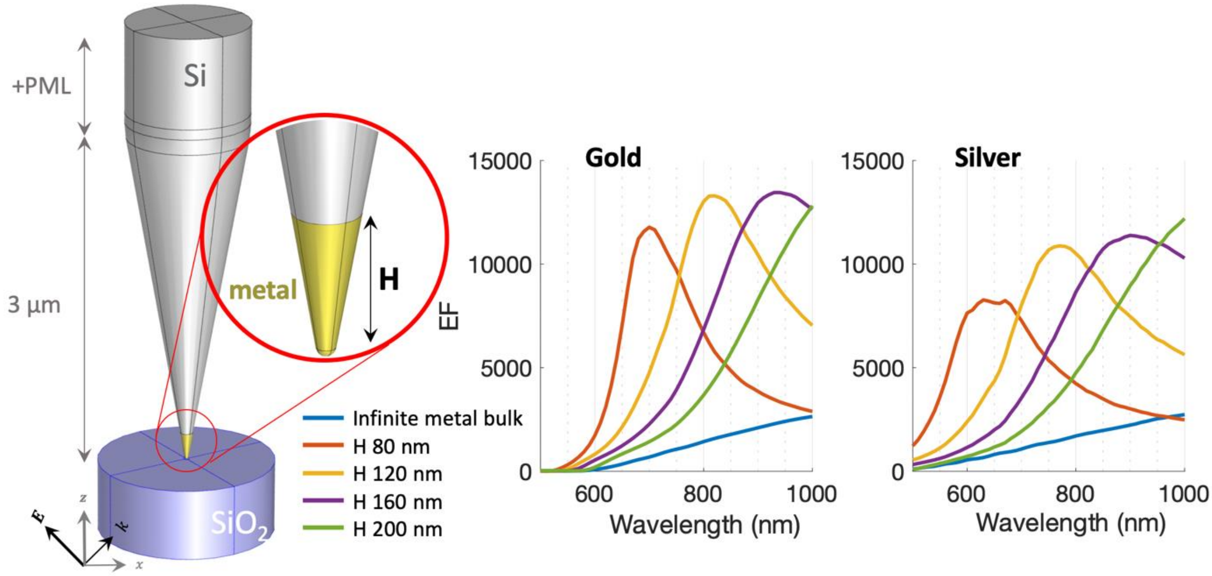

Figure SI-2: Left image: Three-dimensional numerical representation of the 3 µm high metal/dielectric hybrid tip with its upper extremity embedded within the PML (see text). Right graphics: TIR spectra of the electromagnetic enhancement factor between the glass substrate and the tip apex. The blue curves in both graphics correspond to the case of a full (respectively, gold and silver) metal tip, as a reference.