Advanced Tip-Enhanced Nanoscopy (TEN) Using Optically Resolved Scanning Probe Tips

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Abstract

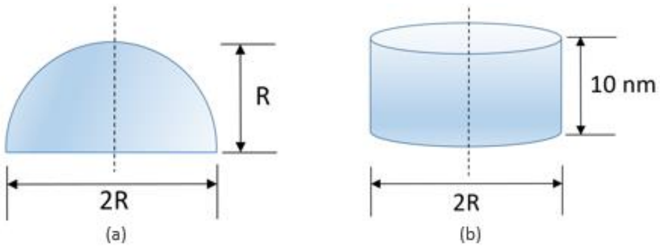
In this article we take the first steps towards next-generation TEN by demonstrating the fabrication and modelling of specialized TEN probes with known optical properties. The proposed framework is highly flexible and can be easily adjusted to for use in various TEN techniques; probes with known optical properties could potentially enable faster and more accurate imaging via different routes such as direct signal enhancement or novel signal modulation strategies. We consider that the reported development can pave the way for a vast number of novel TEN imaging protocols and applications, given the many advantages that it offers.

Introduction

A thorough understanding of biological species and of emerging nanomaterials requires, among other efforts, their in-depth characterization through optical techniques capable of nano-resolution. Nanoscopy techniques based on tip-enhanced optical effects have gained tremendous interest over the past years given their potential to obtain optical information with resolutions limited only by the size of a sharp probe interacting with focused light, irrespective of the illumination wavelength. Although their popularity and number of applications is rising, tip-enhanced nanoscopy (TEN) techniques still largely rely on probes that are not specifically developed for such applications, but for Atomic Force Microscopy. This limits their potential in many regards, e.g. in terms of signal-to-noise ratio, attainable image quality, or extent of applications.

Methods

In order to implement enhanced tuning via excitation of localized surface plasmons (LSP) at the tip extremity, an AFM tip was modified by the addition of a gold nanoparticle at the apex. According to both LSP theory and observations [1], the nanoparticle will display plasmon resonance at a specific wavelength that depends on its material and size/geometry.



Comsol layers concept for half-sphere model (3D)

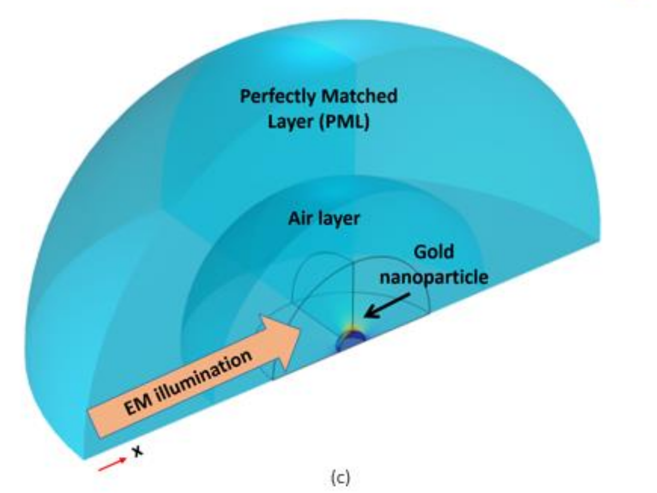


Fig. 1. Diagram of the two models of nanoparticle: (a) Diagram of half-sphere; (b) Thin-cylinder. (c) Comsol Multiphysics: simulation diagram of the half sphere model.

In the quasi-static approximation, the scattering cross-section of the nanoparticle is given by:

$$\sigma_{sca} = \frac{k^4}{6\pi}|\alpha|^2 = \frac{8\pi}{3}k^4 a^6 \left|\frac{\varepsilon(\omega)-\varepsilon_d}{\varepsilon(\omega)+2\varepsilon_d}\right|^2 \qquad (1)$$

Where $\alpha = 4\pi a^3 \frac{\varepsilon(\omega)-\varepsilon_d}{\varepsilon(\omega)+2\varepsilon_d}$, $\omega -$ frequency of the planar wave illumination, $\varepsilon -$ permittivity of the gold nanoparticle, $\varepsilon_d -$ permittivity of the dielectric (air).
The resonance occurs when the Fröhlich condition (2) is fulfilled:

$$\mathrm{R}_e[\varepsilon(\omega)] = -2\varepsilon_d \qquad (2)$$

Two different geometries – a half-sphere (Fig. 1(a)) and a thin cylinder (Fig. 1(b)) – were used to model the nanoparticle, and their plasmon resonance was simulated using the Comsol Multiphysics software. Fig. 1(c) depicts the setup of the simulation. The nanoparticle is surrounded by concentric air and PML layers, and is illuminated by a plane wave propagating along the x-axis. Different radii were simulated for both models in order to find the one for which resonance occurs at the desired wavelength (532nm).

Results

The scattering cross-section resonance for both models are shown in Fig. 2. The half-sphere model exihibits a resonance peak for radii between 30nm to 40nm (Fig.2(a)), while for the cylinder model peaking occurs between 75nm to 90nm (Fig.2. (b)).

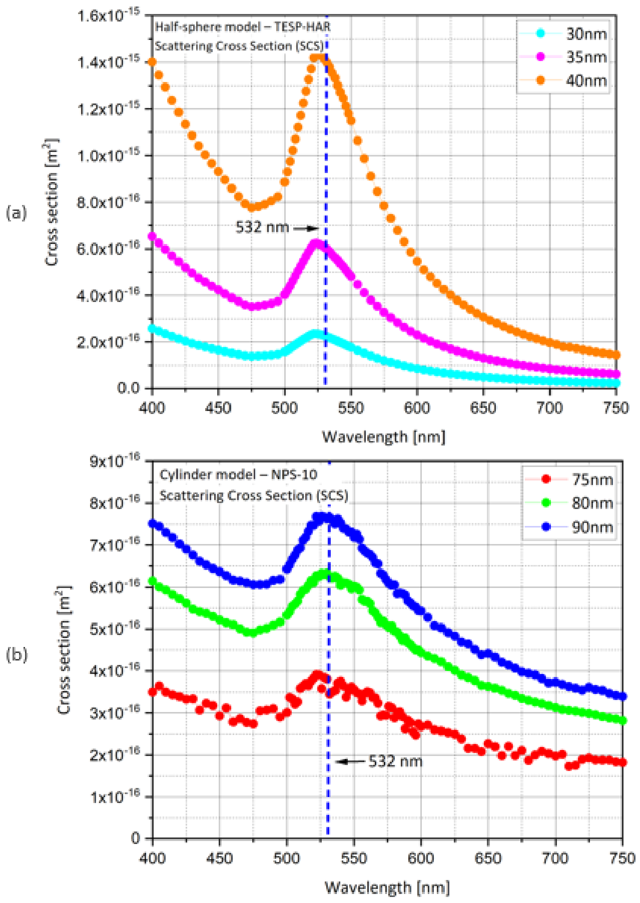


Fig. 2. COMSOL results on the scattering cross-section using the two models. (a) Graph of the scattering using the half-sphere model for different radii; (b) Graph of the scattering using the thin cylinder model for different radii;

Once we have found the nanoparticles' charateristic sizes (i.e. radii), fabrication of the enhanced tip can begin. A thin gold layer was deposited on a standard silicon-based AFM probe (optionally, this layer can be preceded by a chromeone in order to reinforce the gold bonding). The layer thickness was dictated by the desired height of the nanoparticle. Afterwards, the gold layer was removed from most of the tip using a focused ion beam (FIB), leaving only the nanoparticle present on the apex.

The fabricated, optically resolved scanning probe is shown in Fig 3(a). One can observe the gold nanoparticle at the tip apex.

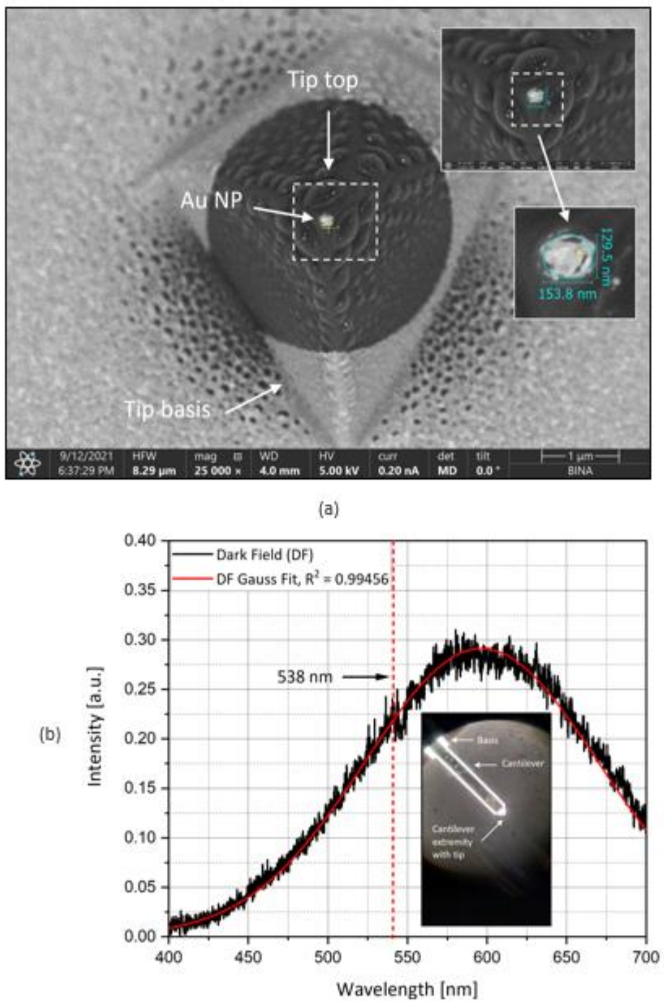


Fig. 3.(a) Fabricated optically resolved scanning probe, with a nanoparticle radius suitable for a predicted 532nm resonance peak (half-sphere model); (b) Spectrometry analysis of a fabricated probe, using dark field microscopy.

The probe resonance peak calculated using the simulations can be then verified via spectrometry analysis. The light scattered from the tip can be collected using dark field observation, and the resonance peak can then be extracted from the scattered light background.

Conclusions

Optically resolved scanning probe tips for tip-enhanced nanoscopy can be designed and caharacterized using numerical methods. Fabrication of such probes is possible via a standardized process, and the finished product can be analyzed using spectrometry. Many applications of this method in optical enhanced nanoscopy, are eagerly awaited.

Main References

1. Maier, S. A. (2007). *Plasmonics: fundamentals and applications* (Vol. 1, p. 245). New York: springer.

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