Interactions in optical nanoantennas for field-enhanced spectroscopy and microscopy

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Optical antennas are nanoscale metallic structures which act as effective receivers, transmitters and receivers of visible light. These nanoatennas show the ability to focus electromagnetic radiation into tiny spots of nanometer-scale dimensions allowing for more effective field-enhanced visible spectroscopies such as in surface-enhanced Raman spectroscopy (SERS). A brief review on the basics of the optical response of these optical nanoantennas will be presented, with examples of the optical response in different canonical nanostructures such as metallic nanorings [1], nanorods [2], nanowires [3], dimers [4] or nanoshells [5] which are commonly used as optical nanoantennas.

We will address the use of optical nanoantennas in a variety of spectroscopy and microscopy techniques. In particular, the use of $\lambda/2$ nanorod-like gold nanoantennas will be described in detail. By engineering the length of the rod-like nanoantennas, it is possible to extend the field enhancement capability into the infrared range of the spectrum (as shown in Fig. 1 (a) for a micron-sized nanoantenna) to perform direct surface-enhanced infrared absorption (SEIRA) [6]. With use of this concept, we show that it is possible to obtain direct IR spectral information of a few thousand molecules deposited on the antenna (see Fig. 1(b)). Another option to engineer the optical response of a nanoantenna relies on the manipulation of the antenna gap. We show theoretically and experimentally the modification of the optical response of nanoatennas as a function of the thickness of the antenna gap, bridging together concepts of optics and circuit theory [7].

The interaction between tip and sample in scattering-type near field optical microscopy (s-SNOM) can also be understood as an antenna effect due to the interaction of tip and sample. This near-field interaction allows for direct mapping of near-field patterns with nanoscale resolution with use of radiation from the visible to the Terahertz [8]. Examples of nanoscopy for each range of the spectrum will be presented. Another spectroscopy where the role of plasmonic resonances plays an important role is Raman-Brillouin scattering of single metallic nano-objects. The interaction between the vibrations of a metallic nano-object and the plasmons induced on it determine the activation and deactivation of certain vibrational modes in the Raman scattering. We analyse in detail how the presence of geometrical indentations and cavities in optical nanoantennas localizes the electromagnetic fields at the indentations (see Fig. 2 (a)). Following the variations of the near-field for a particular vibrational mode (Fig. 2(b)), we can address the modulation of the near-field (Fig. 2(c)), and determine how strongly the field in the cavities and in the indentations is modulated. For certain vibrational modes such as the breathing-like mode in silver nanocolumns, these “acousto-plasmonic hot spots” produce breaking of Raman selection rules with activation of anomalous vibrational modes in Raman spectroscopy.

To illustrate the wide range of applications of plasmonic interactions in totally different systems, we will conclude by analysing the forces originated from the excitation of plasmons by the fast electron beam in Scanning Transmission Electron Microscopy (STEM). Our model calculations show that metallic nanoparticles experience attractive or repulsive forces as a function of the position of the electron beam. This ability to manipulate the forces on the particles can be used in gold nanoparticles for example to produce coalescence.

From the overview and the examples shown here, it is straightforward to conclude that an understanding of the interactions occurring at the optical nanoantennas in such a variety of systems, and the knowledge on the electromagnetic response occurring in the different spectroscopy and microscopy configurations are crucial to engineer and design plasmonic devices for improved detection and controlled optical response.

References:

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Figures:

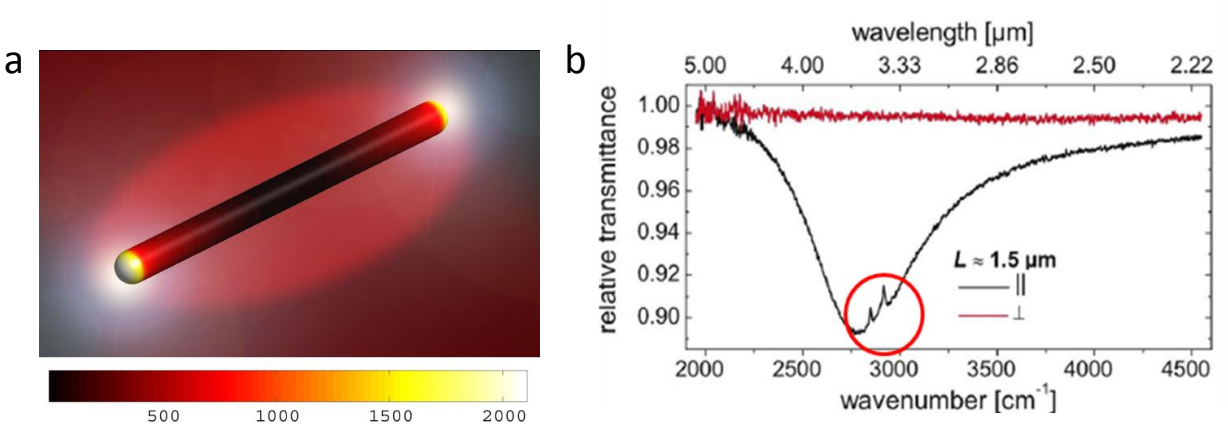

Fig. 1. (a) Near-field around an infrared nanoantenna of length L=1.3 μm when illuminated resonantly with wavelength λ=3.41 μm. (b) Transmission spectroscopy of two molecular fingerprints (marked as a red circle), when the molecules are deposited on top of an antenna similar to that in (a).

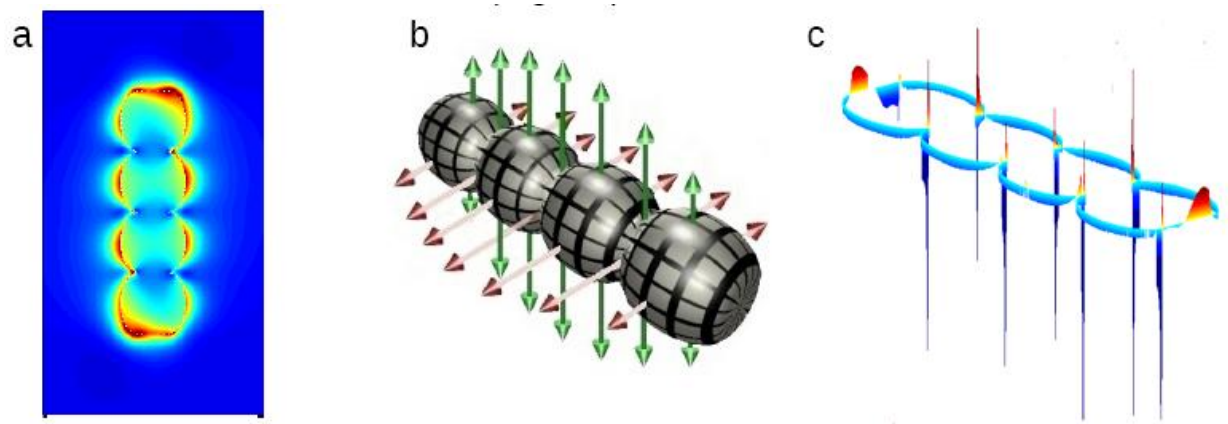

Fig. 2. (a) Near-field map around a silver nanocolumn presenting indentations.(b) Breathing-like vibrational mode of the same nanocolumn, and (c) Modulation of the near-field around the nanocolumn surface for the breathing-like vibrational mode in (b). Strong "acousto-plasmonic hot spots" can be observed at the indentations, producing Raman selection rules breaking. The nanocolumn is 10 nm long, 2 nm wide and the wavelength of the incident light is λ=413nm.