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Characterization of plasmonic nanoantennas by Holographic Microscopy and Scanning Near-field Microscopy

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ABSTRACT

We present a comparison between the optical intensity distributions above a scattering nanohole chain antenna obtained using two different imaging techniques: wide-field digital heterodyne holographic (DHH) microscopy and scanning near-field optical microscopy (SNOM). We show that these techniques have complementary possibilities and limitations but can both deliver accurate measurements of the light distribution in and across the plane of the sample in the near- to far-field transition region, at distances up to a few wavelengths around the nanostructure. The easy access to phase measurements using DHH allows for a deeper insight on the nanoantenna scattering behaviour.

1. Introduction

In analogy to their longer wavelength counterparts in the radio or microwave frequency range, nanoantennas are able to establish an efficient coupling between the optical near- and far-fields, channelling freely propagating electromagnetic radiation to the near-field zone of the nanoantenna or, conversely, releasing energy from the near- to the far-field zone [1]. The characterization of the electromagnetic field distribution around these nanoantennas is thus of interest in itself, aiming to ease the way to a better understanding of their properties as well as to improve their design for multiple applications, mainly to enhance light-matter interaction [2]. However, while most studies focus either on the non-radiative near-field distribution or in the antenna's far-field directivity, the mesoscale transition region between the near- and far-field zones has often been overlooked. Nevertheless, the power density radiated by the nanostructure in this intermediate region is still high and significantly contributes to its interaction with the surrounding medium.

In the optical as well as in the Hertzian frequency range, antenna's properties are highly improved when using a sub-wavelength structuration of the device and therefore of the fields surrounding it. Thus, characterization techniques providing sub-wavelength resolution are desirable. Besides, the distinctive photonic and electronic hybrid character of optical antenna resonances has suggested analysis techniques using either photons, electrons or a combination of both. Techniques involving electrons such as photoemission electron microscopy (PEEM) [3] offer high spatial resolution but require vacuum environments. Among the different existing optical techniques, the most extensively used for the characterization of optical nanoantennas are scanning near-field optical microscopy (SNOM) techniques. Many different configurations exist [4–6], but the common idea is to use a local probe that is raster-scanned extremely close to the sample under study (or, alternatively, many local probes can be previously fixed at different positions onto the sample [7]). The probe can therefore reveal the evanescent fields confined at the sample surface, allowing their measurement in the far-field with subdiffraction resolution. However, these techniques are essentially restricted to imaging features near the surface. Likewise, since SNOM probes are in close interaction with the system, perturbative effects on the system under study are possible [8].

Conversely, far-field techniques are non-contact and non-perturbative, but only give access to diffraction-limited features. In some cases, the angular radiation pattern of the nanoantenna can be accessed, as in back focal plane microscopy [9]. However, although Fourier-space analyses can reveal directionality, they cannot deliver a full 3D reconstruction of the field since the optical phase is not measured.

We recently demonstrated that digital heterodyne holography (DHH) is a suitable technique for the 3D mapping of the field

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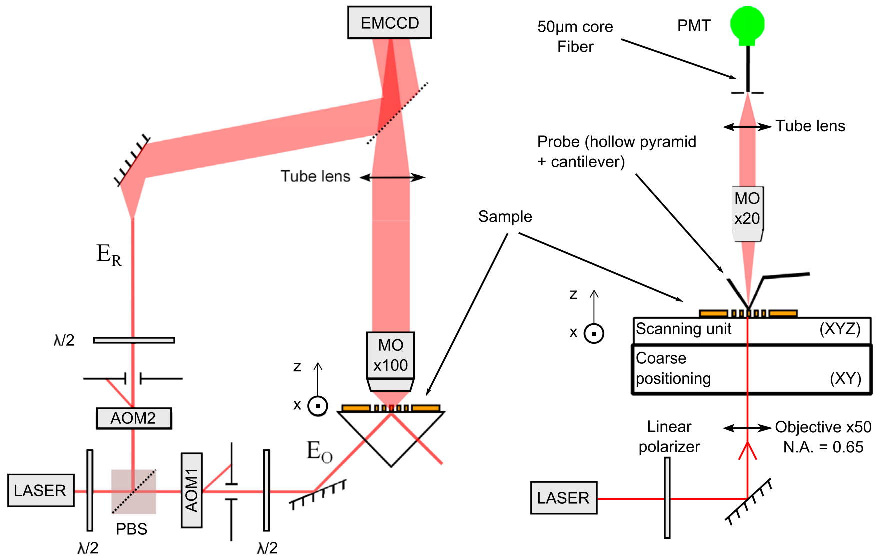

Fig. 1. Left: Digital Heterodyne Holographic microscope with TIR illumination of the sample. Right: aperture SNOM based on a modified commercial Witec microscope.

scattered by plasmonic nanodisc chains [10]. Despite its diffraction-limited resolution, this interferometric far-field technique allows for the back propagation of the scattered field to any plane, including that of the nanoantenna. It is also relatively easy to implement, non-invasive and non-destructive. Here, we propose a comparison between a DHH and a SNOM measurement (see the two setups in Fig. 1) for both in-plane and cross-plane characterization of linear chains of disc-like nanoholes on a thin gold film, with a double purpose. On the one hand, to further prove the validity of DHH as a technique capable of characterizing resonant plasmonic nanostructures by comparing it with a well-established technique such as SNOM. On the other hand, we aim to validate both techniques at mesoscale with respect to the wavelength, i.e. at distances of a few wavelengths around the sample.

2. Samples and experimental setups

In the same way as classical gold nanodisc linear chains, series of disc-like nanoholes can also present characteristic nanoantenna features. The samples under study were fabricated by thermal evaporation of a 2 nm Cr layer for adhesion prior to a 40 nm gold deposition forming a thin, partially transparent layer. The nanoantenna pattern was defined by electron beam lithography in a poly-(methylmethacrylate) resist and transferred to the metal layer by ionic beam etching. A set of different chains was fabricated by varying three parameters, in order to find nanostructures resonating at our laser wavelength (660 nm): the number of holes, the holes' diameter and the edge-to-edge distance between holes. This was the case for chains composed of 17 nanoholes, 150 nm diameter, spaced by 20 nm gaps, for an incident polarization along the chain axis. For a polarization perpendicular to the chain axis, the nanostructure is non-resonant and the extinction cross-section at our excitation wavelength is much weaker (see the far-field spectra in Fig. 2(b), under white light excitation). The total length of the nanoantenna measures around 3 µm. We note that this scattering spectrum is very similar to the one obtained in previous works [10] for a nanodisc chain antenna of exactly complementary geometry and dimensions. This could be expected, in agreement with the Babinet principle, although more studies are clearly necessary in order to validate this principle in the near-field: nanoholes and nanodiscs strongly differ at this scale since the localized surface plasmons (LSP) excited at the nanoholes can decay

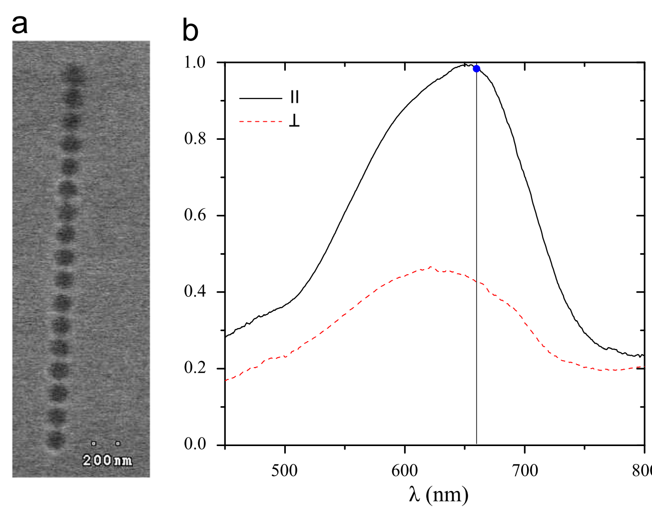

Fig. 2. (a) SEM image of a Au nanohole chain consisting of 17 discs of 150 nm diameter and Δ = 30 nm edge-to-edge spacing. (b) Corresponding far-field transmission scattering spectra measured on a single antenna with an incident polarization along the chain axis (solid curve) and perpendicular to it (dashed curve). The vertical line indicates λ=660 nm.

through surface plasmon polariton (SPP) emission [11].

In both experimental setups depicted in Fig. 1, the light source is a single longitudinal mode laser diode (λ=660 nm, P_{max} = 120 mW). For the holographic measurement, the illumination beam is separated in two paths, the reference (E_R) and the object arms (E_O), which are frequency-shifted by two acousto-optic modulators (AOM1, AOM2) with typical working frequencies f_{AOM1} and f_{AOM2} in the MHz range. To avoid blinding the camera with the direct illumination and masking the weak light scattered by the nano-objects, dark field illumination is essential. The sample is illuminated under total internal reflexion (TIR) using a glass prism with an illumination beam at a 45° incidence angle. Thus, only the field scattered by the nanostructures is collected by the microscope objective (100 × magnification, NA=0.8) and reaches the EMCCD camera (Andor iXon3 885), where it interferes with the reference beam in an off-axis configuration. The resulting interference pattern is modulated at a beating frequency $\Delta f = f_{AOM1} - f_{AOM2}$ = 4 Hz while the acquisition rate is set to f_{CCD} = 4Δf = 16 Hz in order to perform a frequency filtering by