Optical Nano-Imaging of Metallic Nanostructures

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ABSTRACT

Metallic Nanostructures are giving rise to a great deal of attention from a broad scientific community, ranging from physicist and electrical engineers to biologists. The interest is growing rapidly in finding novel devices for future applications that allow using metallic waveguides for optical signal transmission and processing. In this contribution, we investigate some of the fundamental phenomena that take place in these systems. Also the extraordinary transmission of light though sub-wavelength holes in a metal is investigated, keeping in mind various potential biophotonics applications. In this paper, we demonstrate an optical nano-imaging technique that is particularly well suited to characterize the near-field interaction of light with metallic nanostructures: coherent near-field microscopy. This technique allows the total characterization of the near-field by giving full access to its amplitude and its phase. Its application to the characterization and study of plasmonic nanostructures is illustrated using several systems, the coherent near-field optical measurements of light transmission though sub-wavelength holes drilled in a gold thin film and surface plasmons propagating on a metal film and its interaction at a metal-air interface.

Keywords: Near-field optics, surface plasmon, PSTM, SNOM, heterodyne interferometry, nanoholes, nanostructures, nano-fabrication.

1. INTRODUCTION

Plasmon-resonant structures are playing an increasingly important role in the miniaturization of complex photonic circuits. On the one side, the strong field associated with plasmon-resonant nanoparticles can enhance and localize chemical interaction at the nanoscale1, thereby providing new functionalities for biophotonic circuits2. Plasmon resonances of individual particles also show complex resonances spectra which cover a broad wavelength range and produce extremely localized fields3, which could be used for optical data storage4. On the other side, surface plasmons propagating on metallic nanowires open new perspectives for realizing optical circuits fully compatible with silicon technology. Finally, surface plasmon propagating on metallic films can be used to strongly localize light, hence, breaking the diffraction limit for applications such as nanolithography5.

Surface plasmon polaritons (SPP’s) are electromagnetic modes (in the visible or near-infrared ranges) existing at the interface of a metal thin film with a dielectric material (SiO₂, air, etc.). SPP’s, or simply surface plasmons, can propagate on a metal layer over a finite length (several tens of microns) depending on the Ohmic properties of the metal. Growing potential applications utilize this extraordinary near-field behavior. In this paper, the propagation of the plasmons is studied in several different metallic structures with the aim to investigate fundamental properties such as propagation length, attenuation, radiation losses, and bouncing effects. Since SPP’s are evanescent and propagate at the surface of a metal only, their direct detection can only be performed using a Scanning Near-field Optical Microscope (SNOM)6. In our case, the plasmonic nano-imaging is performed with a PSTM7 (Photon Scanning Tunneling Microscope) which collects the near-field light tunneling towards the probe (collection mode SNOM). In our unique set-up, we combine a heterodyne interferometer with a PSTM which enables measuring the near-field optical amplitude and phase in the visible range. It allows full characterization of the optical field as well as the propagation of surface plasmons in nanophotonic circuits. Indeed, the measurement of the phase of the field in these structures gives access to the full description of the electromagnetic field, thereby furthering our understanding of light propagation and interactions in plasmonic systems.

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In addition to plasmonic waveguides, tremendous interest for metallic nanostructures such as individual and arrays of nanoholes surrounded with periodic corrugation has grown recently. This new attention is motivated by the extraordinary transmission with small angular divergence through sub-wavelength holes in a metal, demonstrated by Ebbesen et al.\textsuperscript{8,9,10}. Indeed, the amount of transmitted incident light for a periodic nanoholes array can be enhanced at resonant conditions (depending on the number of holes in the array, their shape and illumination wavelength). This phenomenon has prompted many scientists to study this effect and its potential applications (e.g. in Biophotonics). One promising application of such effect being the localized spectroscopy of single molecules\textsuperscript{11}. We also investigate in this paper light transmission through nanohole arrays with our coherent PSTM in a similar set-up than for the plasmon experiment. Different array configurations are tested with varying hole dimensions, shape, period, etc. As a matter of fact, it has been pointed out that a nanohole with a low symmetry geometry exhibits a more interesting spectral signature\textsuperscript{3}.

2. EXPERIMENTAL SET-UP

The experimental setup to detect SPPs and light emerging through nanoholes is described in Fig. 1. Both experiments use the same system. The detection scheme is based on a Photon Scanning Tunneling Microscope (PSTM) combined with a heterodyne interferometer in order to measure simultaneously the atomic force microscopy (AFM) topography, the amplitude and phase of the optical near-field\textsuperscript{12,13,14,15}. The heterodyne detection is shot noise limited. It offers an elegant way to increase a weak electronic signal up to a point where the shot noise dominates the thermal noise of the detector. Moreover, the heterodyne interferometer is a powerful tool for measuring the optical phase with high resolution\textsuperscript{16}. Phase information provides essential supplemental knowledge for the optical near-field, like for example the direction of propagation of an evanescent wave on a surface.

SPPs are excited using resonance conditions. This resonance is achieved by choosing the correct wavelength and angle of incidence, such that the projection of the \( k \)-vector parallel to the surface of the metal matches that of the plasmon on the metal. The most common way to excite a SPP at a metal-air interface consists in illuminating a metal film (evaporated and structured onto a dielectric media such as glass) in total internal reflection (TIR) condition. The sample is mounted on a prism meanwhile a refractive index matching gel. An illumination system, working at different wavelengths (\( \lambda = 532 \text{ nm}, 658 \text{ nm}, 690 \text{ nm} \)), is used to illuminate the sample by controlling the incident angle with a precise goniometer, the incident polarization and the focalization of the Gaussian beam (with a diameter down to 5-10 \( \mu \text{m} \)). The same configuration has been used to study the light emerging from the nanoholes.

![Figure 1](image-url)  
**Figure 1.** Experimental set-up of a coherent Photon Scanning Tunneling Microscope (PSTM) for surface plasmon polaritons detection.

The evanescent part of the plasmon and the near-field light coming out through the nanoholes are detected by a Scanning Near-field Optical Microscope working in collection mode and exploiting the optical tunneling effect (PSTM).
A SNOM fiber tip (coated with Chromium, in order to prevent supplemental plasmon effects at the tip apex) is brought close to the surface using a commercial stand-alone SNOM (Solver SNOM, from NT-MDT) operating in shear-force mode using a tuning fork at a resonance frequency around 195 kHz. As soon as the tip approaches within tens of nanometers of the surface, the resonance frequency of the tuning fork oscillations changes according the Van der Waals interaction between the tip and the surface atoms. The AFM electronic feedback can therefore control the sample-tip distance within a few nanometers (typically 10 nm). The tip scans the surface (up to a range of $70 \times 70 \mu m^2$) with a height resolution of about 5 nm in topography. At the same time, the evanescent light of the plasmon (or light emerging from nanoholes) created at the surface of the metallic structure is collected by the fiber probe, interfering with the light from a reference arm (coherent to the light coming from the tip) of the heterodyne interferometer. A beat signal, due to frequency shift induced by acousto-optic modulators in the reference part at 70 kHz is then detected by an silicon photodetector. A lock-in amplifier separates then the amplitude and the phase from the output signals. The signal is demodulated at the beat frequency within a narrow bandwidth. The topography, amplitude and phase are mapped simultaneously$^{12-15}$. The phase accuracy, for measurement over a long time (up to tens of minutes), strongly depends on the interferometer stability (mechanical, thermal and air fluctuation).

Nano-fabrication of plasmonic metallic structures (like strip waveguides) is quite a challenging task, where metal roughness is key to limiting radiation losses. Conventional lift-off technique following laser and e-beam lithography was used to produce the structures investigated. The nanohole fabrication was performed using a focused ion beam (FIB) to directly mill the holes into the metal.

### 3. LIGHT TRANSMISSION THROUGH NANO-APERTURES

Light transmission through nanoholes is investigated here by mapping the optical near-field amplitude and phase emerging from the structures, according to their shape, size, and period. In particular, the transmission enhancement of irregularly shaped nanoholes is emphasized$^{17,18}$ and will be studied in order to understand the influence of the surface plasmon excitation on the enhanced optical transmission$^{19,20}$. In the future, these measurements will be completed with spectral measurements. In this section, we present two kinds of nanohole arrays, one with a circular shaped hole and the other one with triangular shapes. The nanoholes are drilled through a gold thin film using a FIB, Focused Ion Beam (from FEI Nova 600 Nanolab dual beam (SEM/FIB) system with 30 keV Ga ions, 5 nm nominal spot diameter).

In Fig. 2, an array of $5 \times 5$ circular nanoholes is shown (350 nm diameter, 1 $\mu$m period, gold film 230 nm thick deposited on a quartz slide). The sample is shined by a laser light at $\lambda = 532$ nm in transverse electric (TE)-mode (electric field parallel to the surface plane) in TIR with an angle of incidence of $47^\circ$. The time exposition to the focused ion beam differs for each raw of holes (Fig. 2a, raw A to E), resulting in a variation of milling depth (Fig. 2b) and thus output intensity (Fig. 2c). These experiments are important from a nano-fabrication point of view, as they provide precise measurement of the time required to mill the metal through. Over-milling, in addition to damaging the substrate, can lead to important Ga ion implantation, which leads to artifacts in the optical measurements.

![Figure 2](image-url)

**Figure 2.** (a) 350 nm diameter nanohole array topography (AFM) with different depths of milling (going from A to E, A being deeper than E), (b) inverted topography of (a) pointing out the holes depth, (c) normalized resulting measured optical near-field intensity.
The mechanisms leading to extraordinary transmission through metallic nanoholes are quite complex, with both SPPs propagating on the metallic surface and in the aperture contributing to the phenomenon. To investigate these mechanisms, we brought our particular interest to the extraordinary transmission of irregular shaped nanohole arrays, as resonant modes associated with such a low symmetry geometry should be very strong. In Fig. 3 we present, a 4×4 holes array of triangular apertures (200 nm side dimensions) with 5 µm period drilled in a 150 nm thick gold film. The presence of a supplemental hole at the top-right corner is due to a tip crash during the measurement. The topography is not shown here because of the small size of the holes compared to the total scan size. A single nanohole AFM topography image is shown in Fig. 4b. The sample has been illuminated by TIR with a laser beam (about 3 mW power) at λ = 532 nm in transverse electric (TE) polarization (or s-polarization, Fig. 3a and b) and in (TM) polarization (or p-polarization, Fig. 3c and 3d where the electric field is parallel to the plane of incidence) with an angle of incidence of 47°. The period (5 µm) has been chosen quite large in order to minimize the coupling between neighboring holes. The near-field clearly shows one main peak, surrounded with three smaller ones, emphasizing the role of the triangular shape of the nanohole (Fig. 3a).

\[\text{Figure 3. (a) Normalized near field intensity transmitted through a 4×4 nanohole array with triangular apertures (TE illumination polarization) and (b) corresponding phase surface plot mapping \([-\pi, \pi]\). (c) Similar measurements as in (a), but for a 3×4 array under TM illumination, (d) corresponding phase distribution.}\]

In TE-mode illumination (or s-polarization), we observe the diffraction of the light by individual sub-wavelength hole only (Fig. 3a). The individual point scatterers exhibit an airy-shape pattern and almost no interaction is observed between neighboring holes. This is clearly evidenced in Fig. 3(b) by the circular phase distribution surrounding each hole. In Fig. 4 (a), we present the profile plot along the first aperture raw in x-direction of Fig. 3(a). The full width at half maximum (FWHM) of the individual main peak is about 390 nm. The FWHM of the neighboring peaks “A” is 325 nm and “B”, 265 nm, respectively. This sub-wavelength field fine structure around an individual hole is due to the interferences of the evanescent orders of diffraction with the zero-order of the main peak, and with each another. This phenomenon may also be at the origin of the extraordinary transmission through the holes\(^{19}\).
In *TM*-mode, the intensity and phase distributions are quite different from *TE* illumination. An interaction coupling caused by multiple scatterers and/or coupled interactions seems clearly visible and appears as multiple interferences in the \(\gamma\)-direction, which is also the direction (projected on the surface plane) of the incident excitation (Fig. 3d). It is possible that this mode is associated with surface plasmons propagating on the metal surface, thus creating a near-field interaction between neighboring holes (one should however keep in mind that the metal is quite thick to support a “regular” surface plasmon mode).

In Fig. 4 (b), the AFM image of one single triangular-shape nanohole is shown in detail with the corresponding depth profile along the \(\gamma\)-direction.

![Figure 4](image_url)

**Figure 4.** (a) normalized intensity profile plot corresponding to the first aperture raw map of Fig. 3a) (*TE* polarization). (b) Topography (nm) of a single triangular nanohole and corresponding profile cut.

### 4. PLASMON PROPAGATING ON METALLIC STRUCTURES

One of the main interests for surface plasmons is the ability of a good metal, such as gold or silver, to conduct simultaneously an electrical current and an optical wave on the same wire. The conventional way to create a surface plasmon at a metal-air interface is the excitation of the metal film in TIR; SPPs getting into resonance when the projection of the incident \(k\)-vector onto the surface plane matches the plasmon excitation wave vector\(^{21}\). In our experiment, this is achieved by choosing the correct angle of incidence using a precise goniometer device. Other crucial conditions are the incident polarization, which must be transverse magnetic (*TM*, electric field parallel to the plane of incidence), and an appropriate wavelength (e.g. close to the near-infrared for gold, such as \(\lambda = 690\) nm). In our experiment, the Gaussian beam has a focus diameter down to 5–10 \(\mu m\) at the surface of the metal layer.

The plasmonic structures are fabricated using a standard lift-off lithography technique. First, a photoresist is spin-coated on a dielectric substrate (both float and pyrex glass). The structures to be written are exposed using a laser-written mask and developed. The metal (40 to 60 nm thick) is then deposited directly by evaporation on the entire wafer and the unwanted area is lifted-off. Few atomic layers of Chromium or ITO are often used to help the deposition stickiness of the metal on the glass but we believe this reduces drastically the plasmon effect. Therefore no such adhesion layer was used in our experiment. Also, silver would be a better candidate for plasmon excitation than gold but its rapid oxidization renders its use non-practical. A more advanced lift-off process with two photoresist layers has been used for metallic waveguide fabrication. First, an 800 nm thick sacrificial layer is deposited, followed with a 1.5 \(\mu m\) imaging photoresist. The first sacrificial layer creates an undercut after development so that the edges of the metallic film are well defined.
4.1 Plasmon excitation on infinite thin film

Our first test experiment is naturally the plasmon excitation on a 40 nm thick gold metal layer with (quasi-) infinite lateral extension. The metal layer is illuminated in TIR with a 45° angle of incidence and a wavelength of 690 nm. We investigate the plasmon free propagation on the metal with the PSTM and apply our sensitive heterodyne detection system. In Fig. 5, we show the normalized amplitude and the corresponding phase of a SPP. The topography is flat with no defects. The FWHM of the incident spot size in the lateral x-direction is 5.8 µm. Near the focus spot of the plasmon (at about y = 50 µm), the FWHM is about 2.5 µm. Since the decay of the plasmon is exponential, the propagation extension strongly depends on how far our system is able to detect the light. In Fig. 5(a) top, the profile of the plasmon shows that its detection can be extended over our maximum scanning range, i.e. over 70 µm.

![Figure 5](image.png)

The propagation length L_{spp} of the plasmon is defined to be at the distance where the plasmon amplitude is (1/e) of the incidence amplitude. The exponential fit gives therefore a L_{spp} of about 45 µm, which is in good agreement with the theoretical value obtained for this geometry. The phase of the plasmon is reported here for the first time in Fig. 5(b) together with its profile (unwrapped phase) along the y-direction. The slope of the unwrapped phase gives therefore the k-vector of the plasmon (−Δφ(y)/δy) and thus the SPP wavelength λ_{spp} = 670 nm, which is in good agreement with the theoretical value obtained for the geometry. Note that λ_{spp} strongly depends on the permittivity constant of the gold thin film, which is different from the bulk one, and the thickness of the layer.

4.2 Plasmonic effects at interfaces

We performed a simultaneous topography, optical amplitude and phase measurement of a surface plasmon propagating towards a metal-dielectric (air) interface (Fig. 6). Interesting plasmon field enhancement phenomena are observed. The intensity of the scattered light can reach several times that of the incident plasmon intensity (up to 18× in Fig. 6b). As a consequence, the reflected plasmon is very weak (about 80× smaller than the incident intensity). By measuring the intensity only, the rebounce of the plasmon might be hidden by the strong scattering at the edge of the metallic film. However, the phase clearly emphasizes the reflection phenomenon at the interface. This is also visible in the intensity surface plot once the intensity has been over saturated: the reflected part of the plasmon becomes then visible (Fig. 6d). The behavior of the phase at the interface is very surprising. Indeed, if the field would only be scattered, we should get circular interferences pattern, which is not the case here. The wavefront does not seem to be perturbed by the interface, except for the apparition of some phase singularities where the amplitude is zero (Fig. 6c, white circles), indicative of multiple waves (at least three) interferences. These data illustrate the power of heterodyne near-field measurement to reveal complex near-field phenomena in plasmonic systems.
Figure 6. (a) Metal structure interface topography (Height in nm), (b) incident normalized intensity map and profile (dashed line cross-cut), (c) phase mapping with some phase singularities (white circles) and (d) intensity saturated image enhancing the reflected plasmon.

Another interesting effect is the propagation of a plasmon toward a sharp metallic corner $23$ (Fig. 7a). The field is enhanced like in the previous case (Fig. 7b) but here, the field enhancement is only twice the incident field (cf. profile along the plasmon-tip direction in Fig. 7c). In the edge interface case, the distance between the plasmon initial excitation and the interface was twice as small as here. It is still not clearly understood whether this is merely a tip effect or a plasmon confinement at the interface. To check this latter suggestion, we intend to fabricate sharper tip structures in order to verify if the isophase lines become tighter $24$ (shorter wavelength) when reaching the tip end, which is not the case here (Fig. 7d).

Figure 7. (a) Topography of a metallic gold corner on a glass substrate, (b) normalized near-field intensity of the incoming plasmon, (c) profile line plot of (b) and (d) phase mapping ($[-\pi, \pi]$).

5. CONCLUSION

In this paper, we have presented experimental near-field optical measurements of light emerging from nano-holes in metallic films and SPPs propagating on various types of metallic films. These measurements reveal some of the
phenomena that take place in the near-field of these structures. We have shown the different optical field distributions (amplitude and phase) emerging from triangular shaped nanoholes in TE- and TM-mode, pointing out the coupling/non-coupling conditions that may occur depending on the illumination polarization. The sub-wavelength field fine structure around an individual hole is a signature of a low symmetry geometry, enhancing therefore the optical transmission. We have also demonstrated the ability of our detection system to visualize the surface plasmon propagation over a large range, i.e. over 70 µm. Moreover, the phase information of the surface plasmons, e.g. at metal-air interfaces (flat or sharp), presented here for the first time, reveal its crucial importance in near-field nano-imaging.

Our results show that near-field optical microscopy is a unique tool to investigate the excitation, propagation and scattering of surface plasmons in complex metallic structures. The combination of PSTM and SPP techniques have already shown the way towards new applications in domains as diverse as optical communications, optical computing, biophotonics, optical data storage, etc. To develop these new devices, it is essential to further our understanding of the interaction of light with metals at the nanoscale. Heterodyne PSTM measurements, as illustrated in this article, allow a complete description of the electromagnetic field, including its amplitude and phase. The latter is essential to reveal subtle effects, such as plasmon reflection and refraction, which otherwise disappear in the strong field associated with plasmon scattering at geometrical interruptions. Acting on the phase of the plasmon opens new venues for the development of highly integrated photonic devices, where the information flows as optical surface wave.

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