Digital plasmonics: from concept to microscopy

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ACTIVE CONTROL OF PLASMONIC FIELDS

We control the wavefronts of Surface Plasmon Polaritons (SPP) on nanohole arrays using a digital spatial light modulator. Optimizing the plasmonic phases via feedback we focus SPPs at a freely pre-chosen point on the surface of the array with high resolution. Digital addressing of SPPs without mechanical motion will enable novel interdisciplinary applications of advanced plasmonic devices in cell microscopy, optical data storage and sensing.
2.1 Introduction

Positioning and focusing waves in transparent media requires fine tuning of the phase profile so that waves converge and constructively interfere at a point. A conventional lens uses refraction to redirect the waves to the focus and a well-designed lens shape to align the phase vectors of these waves. Due to the fixed geometric shape of the lens, the position of the focus can only be controlled by mechanically moving the lens or changing the angle of incidence of the incident beam. Focusing and controlling the position where waves constructively interfere in complex structures require new methods that are more versatile [67]. Optical wavefront shaping has become a popular method that allows to focus light even inside completely disordered materials [58, 68].

The field of plasmonics [25] offers a complementary route to control light fields with metallic nanostructures through the excitation of Surface Plasmon Polaritons (SPP's) [69, 70]. These surface waves, bound to a metal dielectric interface, tightly confine electromagnetic energy [71]. Plasmonics has applications in sensing [33], photovoltaics [72], quantum communication [73, 74], nano circuitry [29, 30], metamaterials [75, 76] and super-resolution microscopy [77].

Positioning and focusing SPP waves in a controlled way is important for nanophotonic applications. To date plasmonics offers only limited flexibility in the control of light fields: as with the conventional lens the geometry is typically fixed, so for a given optical frequency the locations of optical field enhancement are also fixed. Recently, some breakthroughs have been made on active control in which the intensity of the light fields is influenced in time, either through pump-probe [49, 78, 79, 80] or through coherent control [81]. Only in specific cases does this control also lead to spatial selectivity [82, 83, 84]. However, in these experiments the spatial selectivity is limited to a few modes predefined by the sample structure.

2.1.1 Our contribution

We demonstrate here a new level of control of SPP wavefronts. This control allows us to tune any SPP interference phenomenon with unprecedented flexibility. Specifically, we show that we can generate and focus SPPs at any location on a nanohole array with an electronically controlled spatial light modulator and standard helium neon laser. Because the light-to-SPP conversion process is coherent, the structured optical wavefront is projected onto
the SPP wavefront. This conversion gives us full phase control of the SPPs, allowing us to shape the SPP wavefronts digitally. Because we use optimization loops to determine the necessary wavefront, our method is applicable to any plasmonic structure. Such flexible and digital control of SPPs is a large step forward towards interdisciplinary applications of advanced plasmonics.

### 2.2 Experimental configuration

#### 2.2.1 The nanohole array(s)

The sample is a metallic nanohole array with a rich electromagnetic behavior [85]. Subwavelength gratings offer potential for super-resolution either for dielectric [86] or metal-dielectric [87] configuration. Our sample is composed of a 200 nm of gold film deposited on top of 1 mm BK7 glass substrate. The array covers an area of 30 x 30 $\mu m^2$ and the hole period ($a_0$) is 450 nm. Square holes were milled (focused ion beam) with sides of 177 nm. Other arrays with different hole periods will also be used.

![SEM image of the sample: a nanohole array on gold. The hole period is 450 nm while the hole side is 177 nm](image)

The electromagnetic behavior close to subwavelength corrugations on metal films is very rich and can be described in terms of a hybrid wave [88] arising from SPPs and quasi-cylindrical waves [89]. The same description also holds for nanohole arrays. Nevertheless, it is generally well accepted that for optical frequencies SPPs are very efficiently excited on hole arrays due to their resonant nature and thus dominate the electromagnetic field.

The SPP wavelength at the gold-air interface from incident radiation of $\lambda_0 = 633$ nm is given by
\[ \lambda_S = \lambda_0 \text{Re} \sqrt{\frac{\varepsilon_m + \varepsilon_d}{\varepsilon_m \varepsilon_d}}, \] 

(2.1)

with \( \varepsilon_m \) and \( \varepsilon_d \) the dielectric constants of gold and air, respectively. Using tabulated bulk values for \( \varepsilon_m \) [53] we found \( \lambda_S = 600 \text{ nm} \).

**Figure 2.2:** Experimental Setup. The Spatial Light Modulator (SLM) is projected onto the sample via the two-lens imaging system \( L_1 \) and \( L_{\text{OBJ}} \). The demagnification is 650 times. Every image point on the sample is formed with a different average angle of incidence (shown for three pixels). The amplitude and phase of each pixel of the SLM are independently controlled with a computer. The sample is a nanohole grating engraved on a gold film. The blue arrows illustrate the propagation of Surface Plasmon Polaritons (SPP) launched from two pixels of the SLM. The amplitudes and phases of the SPPs are effectively clamped to those of the launching pixels. The surface of the sample is imaged onto the camera via \( L_{\text{OBJ}} \) and \( L_2 \).
2.2.2 The setup

Our aim is to digitally control the amplitude and phase of SPPs locally on the surface of the sample. This control is achieved by imaging a Spatial Light Modulator (SLM) onto the surface of the sample thus mapping each unit cell (pixel) of the SLM to a corresponding area on the sample. Amplitude and phase control of the SLM is achieved by grouping pixels into superpixels. We independently control amplitude and phase of 32 x 32 superpixels. The image of each superpixel locally excites surface waves [90] (to which SPPs are the dominant contribution).

A diagram of the setup is given in Fig. 2.2. Based on SPP momentum conservation we designed the imaging system such that plasmons are launched toward the center. The light reflected from the sample is imaged on the detector. This light includes both the direct reflection of the illuminating beam and the scattered light from SPPs. Thus the resulting image is a combination of both the SLM amplitude pattern and the generated SPPs.

2.3 SPP amplitude experiments

2.3.1 Preparation of the SPP arena

To separate plasmonic from optical effects we spatially design the amplitude of the incident light to define four bright plasmon launching areas and one central dark arena. Any intensity detected inside the arena is purely plasmonic. The designed amplitude profile for focusing experiments is a four-block pattern of fully “on” (A=1) superpixels on an “off” (A=0) background. The resulting illuminated areas on a bare gold substrate are visible in Fig. 2.3a. The overall phase is constant. Each “on” block is 10 x 8 superpixels in size. Because no SPPs are launched on bare gold due to momentum mismatch, the image of Fig. 2.3a is used as a background reference and measure of contrast ratio between the “on” and “off” areas. The observed contrast is nearly three orders of magnitude, confirming that no photons enter the SPP arena.

2.3.2 SPP propagation in the arena

When the designed amplitude profile is projected onto the hole array, SPPs propagate into the central dark arena. The nanohole array has a dual role: it is used to launch SPPs (bright rectangles in Figs. 2.3b-2.3d) and to visualize the launched SPP’s through their out-of-plane scattering in the central arena.
The SLM projection and the resulting surface plasmons for uniform phase profile (no optimization). In each image the bright rectangle(s) are the illuminated (amplitude=1) SPP launching areas. The SPPs are observed in the dark (amplitude=0) central arena. (a) Bare gold reference (no SPPs launched). The dashed lines demarcate the SLM area. (b)-(d) SLM projected on the 450 nm hole array. (b) SLM image plus white light illumination to observe the hole array. (c) SPPs launched toward the central arena. (d) Vertical polarization of incident light (horizontal polarization for the other images).

The SPPs are launched only along the direction of the incident polarization as seen in Figs. 2.3c and 2.3d, consistent with expectations [91, 92]. In Fig. 2.3b, the sample is illuminated both by the structured amplitude profile and an additional white light source, revealing both the hole array grating itself (the fast amplitude modulation) and the laser light. This figure also demonstrates the optical resolution of the setup: sufficient to resolve the presence of the hole array pattern but not the shape of holes.

2.3.3 Formation of the SPP standing pattern

In Fig. 2.4a and 2.4b a SPP plane wave with exponentially decaying amplitude is launched from a single bright area towards the SPP arena. The measured propagation length is $\ell_S = 3.4 \pm 0.1 \mu m$ and the observed direc-
2.4 SPP phase experiments

Figure 2.4: (a)-(b) Exponentially decaying SPP plane waves launched in opposing directions. (c) SPP fringe formation via counter propagating waves. (d) SLM projected on the 400 nm hole array results in a different fringe pattern.

Tionality is consistent with our oblique illumination scheme.

When two counter propagating SPP waves interfere, a standing wave pattern of intensity is created. The observed period of the fringe pattern is clearly not half the SPP wavelength, as is expected for SPPs propagating on an ideally smooth and non-corrugated sample. Actually, the fringe period of Fig. 2.4d is found to be sample dependent. We measured fringe periods of $1 \pm 0.05 \, \mu m$, $0.85 \pm 0.05 \, \mu m$ and $0.7 \pm 0.05 \, \mu m$ for grating pitches of 450 nm, 425 nm, and 400 nm respectively. The standing pattern detected on the 400 nm grid is presented in (Fig. 2.4d). We attribute the fringe patterns to a Moiré effect between the true standing SPP wave and the periodicity of the arrays.

2.4 SPP phase experiments

Now we present experiments of SPP focusing with digital phase control. These experiments are performed on the nanohole array with 450 nm grating
pitch. The achieved SPP focusing is shown in (Fig. 2.5). We use a phase optimization loop [93] to focus SPPs at a pre-chosen target. This loop yields the optimal phase ($\tilde{\phi}$) for each superpixel as well as the relative contribution ($C$) to focus. The amplitude profile is the same as for the bare gold case with four launching areas and a central dark arena where only SPPs can propagate. The incident polarization is diagonal with the grating lines so that all bright superpixels contribute to the focus, thereby maximizing the NA and resolution.

### 2.4.1 SPP focusing and scanning

Successful focusing at the center of the SPP arena is shown in Fig. 2.5a. The structured SPP wavefront produces an intensity in the designated target that is at least 20 times higher than the average SPP background of an unstructured SPP wavefront (uniform phase profile).

Figure 2.5: Digital focusing of SPPs. (a) The relative phases of the superpixels are optimized to focus SPPs in the center of the SPP arena. The intensity in the target spot is purely plasmonic and 20 times higher than the average background of an unstructured plasmonic wavefront. The focus size is diffraction limited by the detecting optics. (b) and (c) Demonstration of SPP focusing on freely chosen targets in the SPP arena. (d) Background reference of an unstructured SPP wavefront (uniform phase profile).
unstructured wavefront. The measured size of the plasmonic focus is 410 nm, consistent with the diffraction limit of our optics. The flexibility of the method is demonstrated in Fig. 2.5b and Fig. 2.5c which show the SPP focus relocated without mechanical motion to controlled positions in the plasmonic arena.

### 2.4.2 Interpretation and consequences

We interpret SPP focusing in terms of Green’s functions connecting the electric fields at any two points. We idealize every “on” superpixel \( n \) with a light source positioned at \( \mathbf{r}_n \) and with phase \( \phi_n \) and strength \( A_n = 1 \). The amplitude of the electric field (normalized to the incident field) at the target \( \mathbf{r}_0 \) due to these sources is

\[
E(\mathbf{r}_0, \{\phi_n\}) = \sum_{n=1}^{N} g(\mathbf{r}_0, \mathbf{r}_n) \exp[i\phi_n],
\]

(2.2)

where \( g(\mathbf{r}_0, \mathbf{r}_n) \) is the Green’s function connecting each source to the target, and the sum runs over all the “on” superpixels of the amplitude profile. The target field is maximal when all source contributions are in phase. The optimal phase for superpixel \( n \) is \( \tilde{\phi}_n = -\arg[g(\mathbf{r}_0, \mathbf{r}_n)] \). Assigning this phase to the superpixel yields an intensity increase of \( C_n = |g(\mathbf{r}_0, \mathbf{r}_n)| \).
Combining together the amplitude and phase of the Green’s function, we have

\[ g(\mathbf{r}_n, \mathbf{r}_0) = C(\mathbf{r}_n, \mathbf{r}_0) \exp \left[ i\tilde{\phi}(\mathbf{r}_n, \mathbf{r}_0) \right]. \tag{2.3} \]

Equation (2.3) implies that when a focus is achieved in the SPP arena, the recorded optimal phases and relative contributions of the superpixels Fig. 2.7 give the Green’s function for a plasmonic source located at that exact focus point. Thus the superpixels of the SLM effectively behave as amplitude and phase sensitive detectors. These results are valid for any Green’s function or nanostructure and can be extended to the time domain [94] and the transfer matrix approach [66].

For a perfectly smooth sample with no corrugations the SPP Green’s function is simply a cylindrical wave in two dimensions (the Hankel function \( H_0^{(1)}(Kr) \) with \( K \) the complex-valued SPP momentum). Our digitally measured Green’s function includes the light-to-SPP coupling and therefore presents much more complexity.

### 2.5 The phase optimization routine

In this section we will show how the optimal phase of each superpixel is measured [93]. We idealize every “on” superpixel \( n \) with a light source positioned at \( \mathbf{r}_n \) and with phase \( \phi(\mathbf{r}_n) \) and strength \( A(\mathbf{r}_n) = 1 \). The amplitude of the electric field (normalized to the incident field) at the target \( \mathbf{r}_0 \) due to these sources, already shown in Eq. (2.2), is
2.5. The phase optimization routine

\[ E(\mathbf{r}_0, \{\phi(\mathbf{r}_n)\}) = \sum_{n}^{N} g(\mathbf{r}_0, \mathbf{r}_n) \exp[i\phi(\mathbf{r}_n)], \]  

(2.4)

where \(g(\mathbf{r}_0, \mathbf{r}_n)\) is the Green’s function connecting each source to the target, and the sum runs over all the “on” superpixels of the amplitude profile. We use an optimization loop to focus at a chosen target: the phase of one single superpixel is varied from 0 to \(2\pi\) causing an intensity oscillation at the target location due to interference with the static field of the other superpixels (all other superpixels have zero phase). The field at the target as a function of the phase of the single superpixel \(n\) is:

\[ E[\mathbf{r}_0, \phi(\mathbf{r}_n)] = \sum_{m \neq n}^{N} g(\mathbf{r}_0, \mathbf{r}_m) + g(\mathbf{r}_0, \mathbf{r}_n) \exp[i\phi(\mathbf{r}_n)] \equiv S_n + g_{0,n} \exp(i\phi_n), \]  

(2.5)

where \(S_n\) is defined as the static background of all other superpixels and we exchanged in the formula the spatial dependence \((\mathbf{r}_n)\) with a matrix index \(n\). The measured intensity oscillation due to the field of Eq. (2.4) is

\[ I_0(\phi_n) = |S_n|^2 + |g_{0,n}|^2 + 2|S_n||g_{0,n}| \cos[\arg(S_n) - \arg(g_{0,n}) - \phi_n]. \]  

(2.6)

The amplitude of the intensity oscillation is \(C_n = 2|S_n||g_{0,n}|\) and the optimal phase of the superpixel is \(\tilde{\phi}_n = \arg(S_n) - \arg(g_{0,n})\). One iteration is completed by repeating the procedure for all other superpixels. Finally we make the assumption that the contribution of a single superpixel is small compared to the sum of all other superpixels. Under this assumption we can consider \(S_n\) as a constant independent of the index \(n\) and thus we set its phase to zero. This yields the desired result

\[ g(\mathbf{r}_n, \mathbf{r}_0) = \text{constant} \times C(\mathbf{r}_n, \mathbf{r}_0) \exp\left[i\tilde{\phi}(\mathbf{r}_n, \mathbf{r}_0)\right]. \]  

(2.7)

Equation (2.7) confirms that via our algorithm we are effectively measuring the plasmonic Green’s function as intuitively expected from eq. (2.3). In conclusion, through achieving SPP focusing, our optimization algorithm measures the amplitude and phase Green function for a source in the exact focal spot.
2.6 Conclusions

With digital plasmonics we demonstrate the first plasmonic system that operates as a “black-box”, requiring the user only to input the desired location of the nanoscale plasmonic focus which is the system’s output. Specifically, we focus SPPs on hole arrays and locally scan the focus freely over a field-of-view (SPP arena) without any mechanical translation. In achieving such dynamic focusing we recorded amplitude and phase Green’s functions. These digital records, which contain the full complexity of the Green’s function, are used as self-calibrated inputs. The method can be extended to the time domain and to any plasmonic structure, thus focusing light pulses in time and space for any given nanostructure. By proper tailoring the nanostructure of the SPP arena will be possible to confine and manipulate the achieved focus beyond the diffraction limit. This digital plasmonic workbench is anticipated to enable interdisciplinary applications in microscopy, optical data storage and in bio-sensing.

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