Controlling the Optical Near Field of Nanoantennas with Spatial Phase-Shaped Beams

Giorgio Volpe,† Sudhir Cherukulappurath,† Roser Juanola Parramon,† Gabriel Molina-Terriza,†‡ and Romain Quidant*†,‡

ICFO-Institut de Ciencies Fotoniques, Mediterranean Technology Park, 08860 Castelldefels, Barcelona, Spain, and ICREA-Institució Catalana de Recerca i Estudis Avançats, 08010 Barcelona, Spain

Received June 8, 2009; Revised Manuscript Received July 16, 2009

ABSTRACT

We report on a novel approach, based on sub-wavelength spatial phase variations at the focus of high-order beams, to reconfigure the optical near field distribution near plasmonic nanostructures. We first show how the introduction of phase jumps in the incident field driving a gap nanoantenna strongly affects its near field response. Beyond, we demonstrate the feasibility of exploiting this approach to selectively switch on and off hot-spots sites within a complex antenna architecture.

Plasmonic nanostructures such as antennas,1−4 metal−insulator−metal stacks5 or tapered wires6 have been designed to confine light in truly sub-wavelength (sub-λ) volumes opening new opportunities to enhance the interaction of light with small quantities of matter down to the molecular level. Beyond confining light at fixed locations, imposed by the structure geometry, there is a need for dynamical spatial control of such hot-spots, for instance to achieve selective optical addressing of different nearby nanoobjects. Several strategies borrowed from the field of coherent control have recently been suggested to reach this goal. A first approach relies on temporally shaping the phase and amplitude of an ultrashort laser pulse illuminating the nanostructures.7 By combining pulse shaping with a learning algorithm, Aeschlimann et al. have recently demonstrated experimentally the feasibility of generating user-specified optical near field response of a starlike silver object.8 Experimental control of the local optical response of a metal surface was also achieved by adjusting the temporal phase between two unshaped ultrashort pulses.9 Alternatively, the idea of time reversal has been lately proposed by Li and Stockman.10 In this approach, a femtosecond optical nanosource is locally coupled to the surface plasmon oscillations of a complex plasmonic system leading to the subsequent radiation of electric field in the far zone. Time-reversing the later and sending it back to the system as an excitation wave thus provides the right illumination conditions for concentrating light at the initial local source location.

Here we propose a novel approach based on continuous light flows which aims at achieving a deterministic control of plasmonic fields by using the spatial polarization inhomogeneities of high order beams such as Hermite−Gaussian (HG) and Laguerre−Gaussian (LG) beams. A prior study has shown that azimuthal and radially polarized beams with different polarization singularities are transmitted through sub-λ apertures in metal film with different efficiencies.11 Here we show both experimentally and numerically that spatial phase shaping of the illumination field provides an additional degree of freedom to drive nano-optical antennas and consequently control their near field response. Furthermore, the potential of this approach to deterministically confine light at specific locations of a more complex metallic nanostructure is also demonstrated.

As a first test structure to demonstrate the proposed concept, we consider a coupled antenna formed by two adjacent gold nanowires forming an air nanogap. It is now well-known that driving the overall antenna with a field linearly polarized across the gap leads to a strong gradient of surface charges responsible for a dramatic enhancement of the field within the gap region. In this study, we are interested in a configuration where the incident field polarization is not constant over the whole antenna extension. In particular, the overall near field response of the antenna is expected to be dramatically changed when driving out of phase each of the constitutive bars forming the antenna. Such illumination conditions can be provided by high-order modes
such as HG or LG beams. These two families of optical fields form two different bases which can independently describe any paraxial beam. The HG beams possess Cartesian symmetry, i.e. the HG functions are separable on the transverse Cartesian coordinates \((x, y)\). They are labeled with two non-negative discrete indices, \(HG_{l,m}(x,y)\), corresponding to the intensity nodes of the functions along the two spatial coordinates. The intensity nodes arise from the field inversion associated to a \(\pi\)-phase jump. The LG modes have a cylindrical geometry and are also described with two discrete indices: \(LG_{l,p}(\rho,\phi)\). In this case, the non-negative index \(p\) corresponds to the intensity nodes of the functions along the radial direction (with \(\rho > 0\)). The index \(l\) can take negative values and is proportional to the orbital angular momentum of the mode in the direction of propagation. These two sets of modes can be directly linked with the nonparaxial multipole solutions of the Maxwell equations. In practice, any LG or HG beams can be generated from a Gaussian beam by either using patterned phase masks or a spatial light modulator (SLM).

In order to evaluate the influence of high-order beams on the local optical response of optical antennas, mode mapping experiments were performed using two-photon-induced luminescence (TPL) microscopy. We have recently shown that TPL mapping over a gold gap antenna provides direct information about its actual mode distribution. Gap antennas formed by two adjacent \((500 \times 100 \times 40 \text{ nm})\) gold bars and forming a 50 nm air gap were fabricated on a ITO-coated glass substrate by combining e-beam lithography with lift-off. The fabricated sample was mounted on a 3D piezoelectric scanner lying in the sample plane of an inverted optical microscope equipped with a 100× (NA = 1.25) immersion objective lens. The expanded beam from a tunable fs-pulsed Ti:Saph laser (~150 fs pulse width) operated at 750 nm was sent on a removable phase mask before being focused on one of the antennas. Underfilling the entrance aperture of the objective to get to an effective numerical aperture of about 0.4 enabled us to both minimize depolarization effects at the focus and generate an incident beam (~2 \(\mu\)m waist) commensurable with the whole antenna size.

Figure 1. Artistic view of the optical configuration where a Gaussian beam from a near-infrared laser is transformed into a higher-order beam using a phase mask before being focused onto a single gold optical antenna (top inset).

While accurate nanopositioning of the antennas with respect to the optical axis was done using the piezoelectric scanner, mapping the TPL at each point of the sample surface was performed by raster scanning the confocal detection volume using an automated steering mirror included in the detection path.

Figure 2 shows TPL maps recorded over a single gap antenna illuminated by three different beams, all linearly polarized along the \(x\)-axis. For reference, we first consider, in Figure 2(a), the case of a Gaussian beam (HG\(_{00}\)), without any phase mask. The TPL distribution is dominated by emission maxima around the gap region and the outer extremities of the antenna. It is interesting to note that, despite the significant differences in the excitation/collection scheme used in ref 15, very similar maps were found. A drastic change of the antenna near field response was observed when switching the illumination from a Gaussian to a HG\(_{10}\) beam whose phase jump coincides with the antenna gap location (\(x = 0\)). In particular, unlike the Gaussian case, the gap region now leads to an emission minimum. Such drastic inversion in the near field contrast is attributed to the \(\pi\)-phase shift in the polarization of the field driving each of the two antenna arms. A first intuitive interpretation can indeed be that, by driving each of the arms in phase opposition, the surface charge gradient across the gap vanishes thereby canceling the associated electric field. Further data acquired for different relative positions between the \(\pi\)-phase shift of the incident HG\(_{10}\) beam and the antenna gap show that the minimum of TPL emission in the gap region is observed within a range of about 100 nm around the gap center. Any lateral shift within this range only alters the symmetry of the TPL distribution along the overall antenna. Beyond the change within the gap region, driving the antenna with an HG\(_{10}\) beam instead of a Gaussian beam also leads to a displacement of the TPL emission from the outer extremities.

Figure 2. (a–c) Experimental TPL maps recorded on a single gap antenna (located by the white rectangles) when driven by three different incident beams (\(\lambda = 750\) nm): (a) HG\(_{00}\) (Gaussian), (b) HG\(_{10}\) and (c) LG\(_{20}\). The color scale gives the TPL intensity in photon counts. In map (b), the vertical dashed line locates the \(\pi\)-shift position of the HG\(_{10}\) beam. (d–f) Associated computed intensity distribution of the incident field (scale bar = 500 nm). Each of the three beams was linearly polarized along the \(x\)-axis. The black arrows give the relative polarization orientation across the beam.
Figure 3. (a–c) Convoluted TPL maps computed in the half-plane of a gap antenna for the three types of beams considered in the experiments of Figure 2. In (b) the dashed line locates the position of the π-shift. (d–f) Associated surface charge distribution (scale bar = 100 nm).

In what follows we investigate the feasibility of using HG beams to deterministically control the confinement of light in more complex plasmonic architectures. As a first geometry we consider a double gap antenna formed by three 500 nm aligned gold bars forming two identical 50 nm air gaps separated by 500 nm. For reference, we first record the TPL map when driving the whole antenna with a Gaussian beam linearly polarized along the x-axis. As expected, a field concentration was observed in both gaps. Figure 4b and Figure 4c show TPL maps recorded when the π-phase shift of a HG$_{10}$ beam coincides successively with the right and left gaps. These data demonstrate how a suitable positioning of the phase jump over the double antenna enables us to selectively switch on and off one of the two hot-spot sites.

While our results provide a first experimental evidence of the feasibility of using spatial phase-shaped beams for controlling the near field distribution of plasmonic systems, it is important at this stage to evaluate the potentiality of this approach to achieve spatial light control within a sub-λ region. For this purpose, we performed numerical simulations on a 3 × 4 array of 100 nm gold pads separated by 30 nm.
when illuminated by different HG beams. This structure covers a surface area that is commensurable with the diffraction limited focus (375 nm for a wavelength of 860 nm) delimited by a dashed circle. Figure 5 shows the evolution of the computed electric near field intensity distribution at its collective plasmon resonance (860 nm) for different \((m,n)\) sets.

For reference, Figure 5a displays the near field map when illuminating the array with a 500 nm waist Gaussian beam linearly polarized along the \(x\)-axis. Under these conditions, an intensity maximum is observed at each of the gaps oriented perpendicular to the incident field. By switching to a HG\(_{01}\) whose phase jump coincides with the central row, the field along the later is inhibited and only the gaps of the outer rows remain bright. Conversely, the use of a HG\(_{04}\) enables a reverse situation where the light is exclusively confined within the gaps of the central row. This way we demonstrate that the minimum distance between two hot spots that can be selectively switched on and off is of the order of 100 nm, i.e. three times shorter than the diffraction limit at the considered wavelength.

We have shown both numerically and experimentally that spatial phase shaping in the focus of high order beams can be exploited to control the near field of plasmonic nanostructures. While, in our experiments, control is performed manually by introducing a fixed phase mask and adjusting the relative position between the generated beam and the nanostructure, dynamical control can be achieved by using SLM-generated phase-shaped beam.\(^{18}\)

Acknowledgment. This work was supported by the Spanish Ministry of Sciences through Grants TEC2007-60186/MIC and CSD2007-046-NanoLight.es and Fundació CELLEX Barcelona.

References


NL901821S