Near-field polarization shaping by a near-resonant plasmonic cross antenna

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The optical phase in the feed gap of a plasmonic dipole antenna shows a transition from in-phase to counter-phase response, when its length is varied across the resonance length. We exploit this behavior in an asymmetric cross antenna structure, constituted of two perpendicular dipole antennas with different lengths, sharing the same feed gap, in order to shape the local polarization state. As an application of this concept, we propose a λ/4 nanowaveplate, able to shape and confine linearly polarized propagating waves into circularly polarized fields localized in the feed gap.

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I. INTRODUCTION

The remarkable development of plasmonics over the last decade has been motivated by applications in biosensing, nonlinear optics, quantum optics, and optical circuits.[1] Illumination of noble metal nanoparticles with well-designed geometry by means of visible and near-infrared light leads to resonantly increased polarizabilities, resulting in largely enhanced and confined near fields. The resulting nanometer-scale modal volumes can be used to achieve efficient coupling to nanometer-scale emitters, like single molecules and nanocrystals, and to implement high-resolution optical microscopy.[2] Optical antennas have gained particular relevance in this respect. So far, mainly linear antennas have been studied and realized,[3–9] which allow only one field component along the antenna axis to be present in the feed gap. However, a whole range of applications in quantum optics[10] and solid state physics (e.g., ellipsometry[11] or magneto-optical effects)[12,13] require control over the polarization state of photons.

Recently, symmetric cross antennas have been proposed,[14] constituted by two identical, but perpendicular dipole antennas sharing a common feed gap. Using cross antennas, arbitrarily polarized optical fields can be efficiently confined and enhanced in the feed gap, while maintaining their polarization state with high fidelity.

Symmetric cross antennas faithfully translate the polarization state of an incoming polarized wave into a subwavelength spot in the antenna near field. This is in contrast to plasmonic scatterers reported so far, which often show changes in the polarization state of light, so-called depolarization effects. So far, such depolarization has only been exploited to tailor the polarization of the far-field scattered light. Drezet *et al.* proposed a miniature plasmonic waveplate, where birefringence in the extraordinary transmission through a bull's eye structure is achieved by introducing a uniaxial distortion in the grating surrounding the hole.[15] Strong optical rotation has also been observed in the extraordinary transmission through *L*-shaped hole arrays.[16] Finally, polarization conversion by an array of vertically aligned nanorods has been demonstrated due to differences in transmission for *s* and *p* polarized light.[17]

In this Brief Report, we propose a concept for deterministic near-field polarization shaping, based on the phase shift in the local field due to the resonant response of noble-metal antennas. A dipole plasmonic antenna shows to a first-order approximation a harmonic oscillator behavior, which means that its electric near field exhibits a phase shift with respect to the driving field, changing from 0° to 180° when the length is varied from below to above the resonance length for a given illumination wavelength. Therefore, by properly tuning the geometry of the antenna arms, i.e., by choosing a proper antenna length at fixed wavelength, one can tune the phase of the electric near field.

In particular, we study an asymmetric cross antenna, constituted by two perpendicular dipole antennas of different length. We are thus able to deterministically encode the desired polarization state of light that is produced in the common feed gap by a specific choice of the structure's geometrical parameters. As a relevant case, we implement a λ/4 nanowaveplate, which upon illumination by a linearly polarized propagating wave creates circularly polarized, highly confined and enhanced fields in the feed gap.

II. PHASE RESPONSE OF A LINEAR PLASMONIC ANTENNA

We first consider a linear dipole antenna, consisting of two aligned gold nanorods separated by a small gap. As the length of the two arms is increased, the resonance energy of the antenna is red-shifted, accordingly with the behavior of the plasmonic resonance supported by nanorods when their aspect ratio is varied. Therefore, it is expected that the phase shift between the driving field and the plasmon field goes from 0° to 180° when the antenna length is varied from well below to well above the resonance length, in qualitative agreement with the response of a harmonic oscillator. A quantitative assessment of this behavior is obtained with a first set of simulations, based on the finite-difference time-domain method,[18] where we study a dipole Au antenna with squared cross section of $30\times30\ \text{nm}^2$ and gap size of 30 nm, with tapered rod ends at the gap side [Fig. 1(a)].[19] A three-dimensional discretization mesh of $0.5\times0.5\times0.5\ \text{nm}^3$ is used. We consider a focused Gaussian beam illumination

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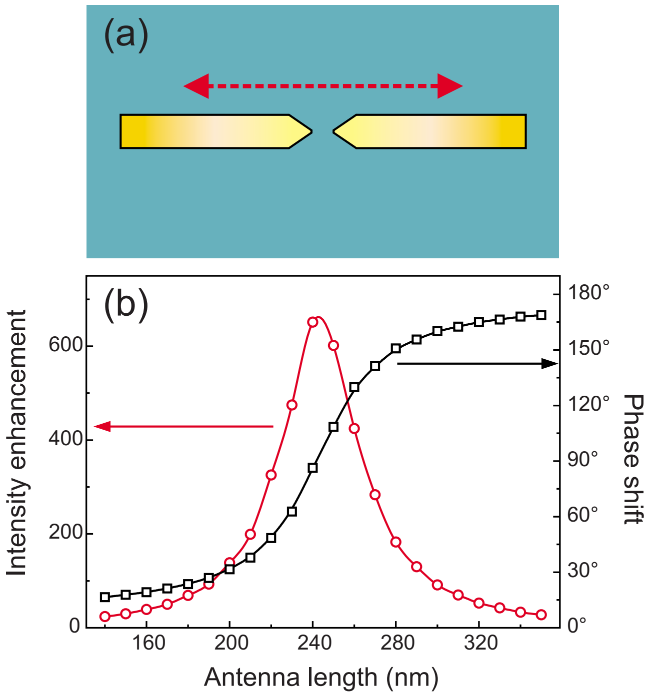

FIG. 1. (Color online) Linear dipole antenna. (a) Sketch of the antenna geometry under consideration (dashed arrows indicate the direction of linear polarization for the excitation field); (b) intensity enhancement (red circles) and phase shift (black squares) of the electric field in the middle of the antenna feed gap, using the intensity and the phase of the excitation field without the antenna as a reference.

(0.5 numerical aperture) at a wavelength of 800 nm, linearly polarized along the antenna axis, with direction of incidence perpendicular to the substrate plane. Our results hold independently of the illumination wavelength provided the antenna geometry is properly adapted, albeit gold can achieve very good enhancing performances only in the red and near-infrared part of the visible spectrum, while other plasmonic materials may be preferred to optimize the performance in the green-blue region. The antenna is residing on a fused-silica substrate ($\varepsilon=2.11$), while the upper half space is vacuum. Permittivity of gold at 800 nm is $\varepsilon=-23.72+i1.13$.[20]

To observe the resonance, the antenna length l is varied from 140 to 350 nm, in steps of 10 nm. For each l, we determine the field in the center of the feed gap and calculate its relative amplitude and phase with respect to the driving field in the absence of the antenna. The results are displayed in Fig. 1(b). The field amplitude clearly reveals the resonance behavior of the antenna, with a peak at $l \simeq 240$ nm. At this length, the accumulated phase shift is roughly 90°, as expected, while it approaches 0° (180°) for $l \ll 240$ nm ($l \gg 240$ nm).

It is worth noting that even if the linear polarization of the illuminating beam is not parallel to the antenna axis, the linear antenna will selectively enhance the field projection along its own axis due to the strong resonance.[21] This argument is valid as long as the illumination wavelength is much closer to the longitudinal resonance than to the transverse plasmonic resonance of the antenna (which for gold in the treated geometries appears far away in the green spectral region).

III. LOCAL POLARIZATION SHAPING WITH A CROSS PLASMONIC ANTENNA

We now consider a cross antenna structure, constituted by two perpendicular dipole antennas sharing a common feed gap [Fig. 2(a)]. Upon far-field illumination with a wavelength close to their longitudinal resonances, each of the two antennas will enhance the field component parallel to its own axis. Considering Fig. 1(b), it is clear that by a proper choice of the length of each antenna, one can adjust the relative amplitude and phase of the two perpendicular electric field components in the feed gap. These two field components, shaped by the two antennas, will add up coherently in the feed-gap volume, so as to build localized and enhanced fields that exhibit the polarization state encoded in the antenna lengths.

Using this principle, by properly shaping and orienting a cross antenna, it is possible to obtain the desired in-plane polarization state in the feed gap, starting from a well-characterized (but otherwise arbitrary) polarization of the propagating wave. An asymmetric cross antenna can thus act as a nanowaveplate, able to control the main axis and ellipticity of polarization in its feed gap.

One may envisage to fabricate such an antenna on top of a flat atomic-force microscopy tip.[22] This will allow scanning it over a sample of interest and placing an emitter close to its feed gap. On the one hand, it should be stressed that, to this aim, fields right above the antenna feed gap give a better description of the average field experienced by the sample. On the other hand, fields inside the feed gap, although experimentally harder to access, can be relevant when an emitter is placed directly inside the feed-gap volume.

IV. NANOWAVEPLATE ANTENNA

As a relevant example, we tune the response of an asymmetric cross antenna in order to achieve quarter-wave behavior. Such a nanowaveplate antenna, when illuminated by a wave linearly polarized at 45° with respect to the antenna axes, should build circularly polarized fields in the feed-gap area. In order to achieve this result, we choose the two antenna lengths in such a way that they provide the same local field enhancement, but induce a 90° phase shift between the two field components. This can be obtained by making one linear antenna longer and the other shorter than the resonance length, in order to keep the same amplitude response for both antennas but also accumulate a positive (negative) phase shift with respect to the phase at resonance. By looking at Fig. 1(b), we infer that this can be achieved by choosing the two antenna lengths to be about 220 and 265 nm, because for these lengths the field intensity enhancement is roughly the same (about 325), while the two phase shifts are about 48° and 138°, respectively, thus yielding a 90° phase difference overall.

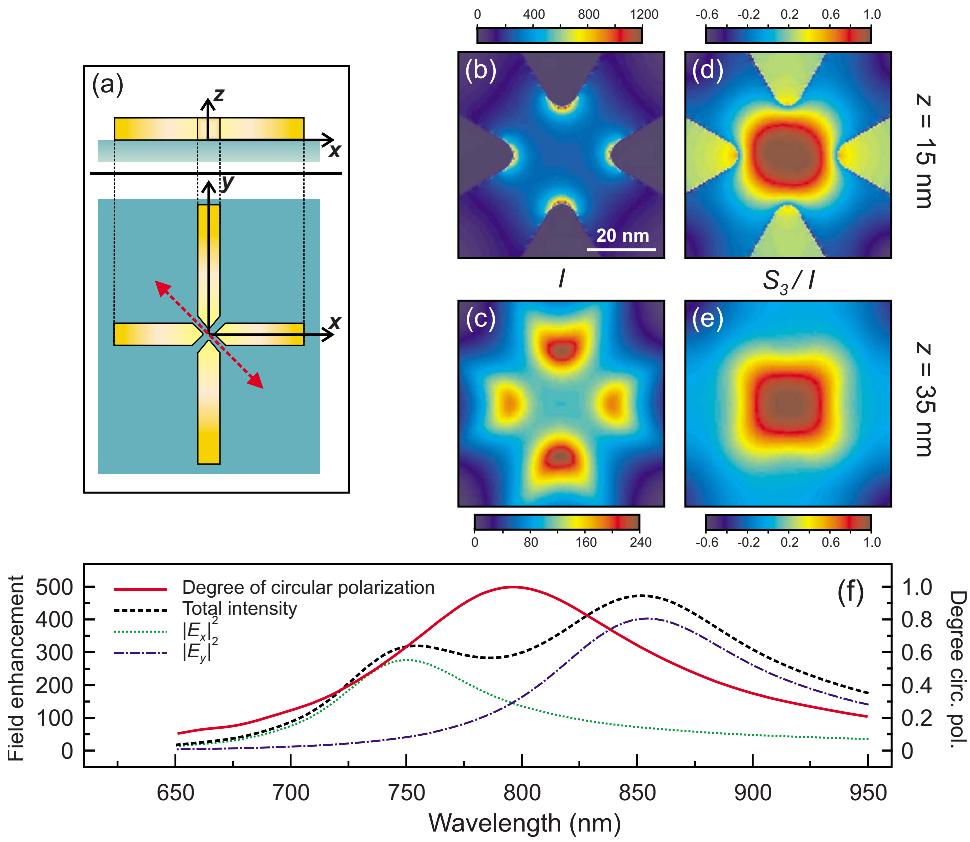

FIG. 2. (Color online) Asymmetric cross antenna. (a) Sketch of the antenna geometry (dashed arrows indicate the direction of linear polarization for the impinging field); (b) and (d) intensity enhancement and degree of circular polarization for the fields in a plane at mid-height in the feed gap ($z=15$ nm); (c) and (e) intensity enhancement and degree of circular polarization for the fields in a plane right above the upper antenna surface ($z=35$ nm); (f) spectral response of the field intensity enhancement in the middle of the feed gap for the simulated antenna: black dashed line is the total field intensity $|E_x|^2+|E_y|^2+|E_z|^2$, red solid line the degree of circular polarization, while dotted and dashed-dotted lines represent the intensity of the x and y electric field components, respectively. Note that Stokes parameters are defined everywhere, even in regions of extremely low field. Therefore, intensity maps always need to be considered as well.

We now discuss the resulting field distribution for this asymmetric cross antenna, when illuminated by a focused Gaussian beam linearly polarized at 45° with respect to the antenna axes [Fig. 2(a)]. Maps of the simulated total field intensity I are shown in Figs. 2(b) and 2(c), calculated in the two relevant planes mid-height inside the feed gap ($z=15$ nm) or right above the upper antenna surface ($z=35$ nm), respectively.

When dealing with polarization in the near field of metal nanostructures, one should consider modified Stokes parameters[23] in order to take the presence of longitudinal field components into proper account. In our case, it is sufficient to consider the three parameters related to the xy plane, $S_1=\langle E_x(t)^2-E_y(t)^2\rangle$, $S_2=\langle E_x(t)E_y(t)\cos(\delta_x-\delta_y)\rangle$, and $S_3=\langle E_x(t)E_y(t)\sin(\delta_x-\delta_y)\rangle$. We can then define three degrees of polarization as S_1/I (predominance of x linearly polarized over y linearly polarized light), S_2/I (predominance of +45° linearly polarized over −45° linearly polarized light), and S_3/I (predominance of right circularly polarized over left circularly polarized light). Spatial maps of S_3/I are shown in Figs. 2(d) and 2(e), providing evidence that a fairly uniform and almost unitary degree of circular polarization is obtained over the area inside the gap as well as above the gap. The proposed antenna geometry can therefore be used as an efficient and confined source of circularly polarized near-field photons. Maps for the degrees of polarization related to the other Stokes parameters are presented as Supplementary Material.[24]

It should be noted that boundary conditions force the fields to be mainly linearly polarized close to the metal structures, while the desired circular polarization is achieved in the center of the gap. Such unwanted linearly polarized components, which cannot be avoided when metal nanoparticles are used, will result in a background-related lower sensitivity for measurements involving circularly polarized photons.

It is also worth considering that the possibility to infer the geometrical parameters of the cross waveplate antenna from the simulations of isolated linear dipole antennas fails when the gap size is further reduced, and therefore the coupling between the two perpendicular antennas gets stronger. In this case, the presence of polarizable material from one antenna in the near field of the perpendicular one causes a red shift in the resonance frequency, requiring further iterative simulations in order to fine-tune the response of the cross structure.

In view of experimental setups making use of asymmetric cross antenna structures as λ/4 nanowaveplates, a relevant issue is their sensitivity to changes in parameters such as (i) the arm length or (ii) the illuminating wavelength. The first issue is particularly important because of possible imperfections occurring during nanostructuring. In this respect, we find by simulations that a change of 5 nm in the length of one of the two perpendicular antennas, with all other parameters fixed, determines a drop in the degree of circular polarization smaller than 2%, as calculated in the center of the feed gap. To address the second issue, we have calculated the spectral response, after broad-band excitation with the same antenna and illumination geometry as before, by evaluating the field in the middle of the feed gap as a function of the illuminating wavelength [Fig. 2(f)]. We find that the degree of circular polarization remains larger than 0.9 all over a bandwidth of 40 nm. This is compatible with the use of the nanowaveplate in combination with ultrashort 100-fs laser pulses.

From the results plotted in Fig. 2(f), one can also infer the two resonances of the system, located roughly at 750 and 855 nm, which can be selectively excited with two perpendicular linearly polarized illuminations. In perspective, this spectral response suggests that the antenna might also be used for applications involving two-color processes, e.g., pump-and-probe experiments (with the two resonances tuned to the pump and the probe wavelengths) or single-emitter photoluminescence (with the two resonances tuned to the absorption and emission wavelengths).

V. CONCLUSIONS

In conclusion, we have demonstrated the concept for polarization shaping of localized fields by exploiting the phase response of plasmonic resonators. In particular, we introduced an asymmetric cross antenna, constituted by two perpendicular dipole antennas with common feed gap but differing lengths. By tuning the response of each antenna, the desired polarization state can be synthesized in the feed-gap area, starting from a polarized propagating wave. As a relevant example, we have analyzed the behavior of a $\lambda/4$ nanowaveplate, able to transform a linearly polarized propagating wave into circularly polarized localized fields. Other polarization transformations can be obtained by a different choice of the arm lengths.

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[24] See EPAPS Document No. E-PRBMDO-80-064939. Figure 1: plot of the degrees of polarization S_1/I and S_2/I. For more information on EPAPS, see http://www.aip.org/pubservs/epaps.html.