Plasmonic mode conversion and second harmonic imaging of tilted plasmonic nanocones

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Abstract

Plasmonic nanocones offer strong, highly localized near-fields at the cone apex that can be utilized for applications in microscopy and sensing. However, the tip mode can only be excited by electric field components parallel to the cone axis. To enable the excitation of the tip mode under vertical illumination, tilted cones are fabricated. Their linear optical properties are investigated and simulated, and their nonlinear optical properties are illustrated by second harmonic imaging.

1. Introduction

Near-field enhancement and confinement of the electric field of electromagnetic waves are promising features in plasmonics and nano-photonics. Therefore specially designed optical antennas are needed, which have broad applications, for example in surface-enhanced Raman spectroscopy or single molecule detection [1,2]. Nanocones are well-suited for such applications as they can easily be fabricated and have a very sharp tip with a radius smaller than 10 nm [3]. For an efficient plasmon excitation along the vertical axis the electric field vector of the exciting external electromagnetic wave should have a significant component parallel to the vertical axis [4]. This is only the case for certain laser modes or the illumination of the cones from the side. Therefore the excitation of many cone tips at once under vertical incidence is not possible. If the electric field is oriented perpendicular to the cone axis, no excitation of the tip will take place [3]. But for many potential applications it would be beneficial if the electric field of the light was enhanced at the tips of many cones simultaneously.

It was recently shown that this difficulty can be circum-vented by breaking the symmetry of the nanocone. In one case, wing protrusions were added on one side of the cone bases, leading to a conversion of the in-plane excitation of plasmons in the winged base to an excitation of tip plasmons through the asymmetric geometry [5]. This approach was further pursued through the investigation of randomly distributed tilted nanopillars, whose axes were forming defined angles with the substrate plane [6,7].

2. Discussion

In the present work we introduce two strategies for the fabrication of gold nanocones with a pre-defined tilt of the cone axis [8]. In the first process, circular oxide nanodiscs are prepared as hard masks on top of a gold film by electron beam lithography and a lift-off. The masks are subsequently transferred into the gold layer by argon ion milling, where the substrate is tilted relative to the ion beam by the amount of the tilting angle. In the second process, electron beam lithography is performed under an angle, such that the resulting nanoholes in the resist are tilted relative to the substrate. Afterwards, metal is evaporated under the same tilting angle, leading to tilted nanocones remaining on the surface after the lift-off. Examples of a symmetric vertical vs. an asymmetric tilted nanocone are shown in Figure 1.

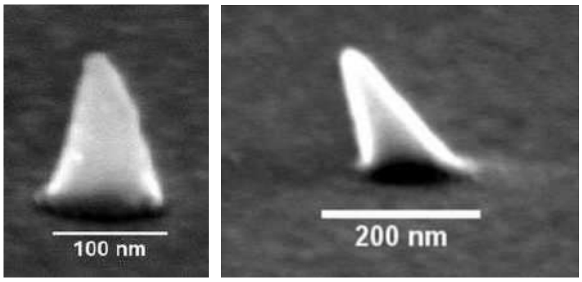

Figure 1: (left) vertical gold nanocone, tilting angle 0°; (right) tilted gold nanocone.

The asymmetric geometry supports the transformation of a transversal (parallel to the substrate) electric far-field to a longitudinal (perpendicular to the substrate) plasmonic excitation. Extinction spectra obtained by transmission spectroscopy and corresponding simulations will be shown, in which cones with a variety of tilting angles are illuminated under a variety of illumination angles, thus systematically changing the ratio of the electric field components oriented parallel vs. perpendicular to the respective cone axes. Furthermore second harmonic generation microscopy of tilted cones will be shown. The cones are scanned through tightly focused radially or azimuthally polarized cylindrical vector beams (CVBs), and for each relative position between the cone and focus center the second harmonic intensity generated by the interaction is recorded. The expected simulated results are shown in Figure 2 and will be discussed in comparison with the corresponding experiments [8].

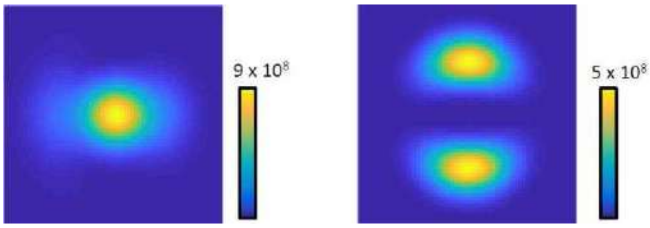

Figure 2: Simulated second harmonic imaging patterns of (left) a tilted nanocone scanned through the focus of a radially polarized CVB, and (right) a tilted nanocone scanned through the focus of an azimuthally polarized CVB.

3. Conclusions

Two processes for the nanofabrication of tilted gold nanocones with defined tip displacements are presented. Under vertical illumination, plasmon resonances are excited in the base plane of the nanocones, as is also observed for symmetrical nanocones. In contrast to symmetrical cones, the symmetry breaking leads to a conversion of the base mode into a tip mode, such that an enhanced near-field can be observed at the tip apex even under vertical illumination. These findings are confirmed by the nonlinear optical properties observed in second harmonic imaging of the nanocones gained by scanning through radially and azimuthally polarized focused cylindrical vector beams.

Acknowledgements

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