Second harmonic generation enhancement by polarization-matched nanostructures -INVITED

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Abstract. Frequency conversion plays an important role in both fundamental and applied nano-optics. Doubling the frequency of light by second harmonic generation (SHG) is a vital process e.g. in laser optics or high-resolution microscopy. SHG can be created through symmetry breaking at plasmonic nanostructures, or the local high electric near-fields of plasmonic nanoantennas can be utilized to further enhance the SHG e.g. from nonlinear crystals. Examples of SHG microscopy using cylindrical vector beams in combination with tilted nanocones and radially symmetric oligomers are shown as well as enhancement studies of the SHG from nonlinear crystals decorated with polarization-matched nanostructures.

1 Introduction

In the interaction of light with nanostructures many intricate effects can be observed. For light with high local intensities also nonlinear effects, such as second harmonic generation (SHG), can be observed [1,2]. In plasmonic nanostructures, which are most often created from gold, silver or aluminium, collective oscillations of the free electron density (so-called localized surface plasmon polaritons) are excited, leading to high scattering and absorption cross-sections, a high local concentration of optical energy near the nanostructures, strong electric near-fields, and resonances that depend on the geometry, material, arrangement, and dielectric environment [3,4]. Advanced microscopy and spectroscopy techniques are being developed to further probe these interactions.

One possibility for investigating optical nanoantennas is the use of cylindrical vector beams (CVBs) [5]. These CVBs can exhibit either azimuthal or radial polarization with rotationally-symmetric electric or magnetic field components. In a tight focus they contain either exclusively in-plane or predominantly out-of-plane components. In the present work SHG microscopy is performed by scanning plasmonic nanostructures through the focus of a CVB and recording the resulting SHG intensity for each relative position of the focus and the nanostructure [6,7]. The resulting scanning images give clear indication of the polarization-dependent SHG induced by the nanoantennas, where maximum intensity can be found for polarization-matched nanostructure arrangements.

For non-linear applications that require larger volumes for SHG, non-linear crystals such as lithium niobate ($LiNbO_3$) are commercially available. For applications, typically high signal intensities are required, whereas the SHG efficiency is typically limited. To further enhance the overall SHG intensity, the non-linear crystals can be decorated with plasmonic nanoparticles. The polarization direction of maximum near-field enhancement should be matched to the extraordinary axis of the crystal. In this work, the interaction of individual gold nanodiscs with y-cut crystals is investigated under excitation with in-plane polarized light [8] as well as the SHG enhancement by self-assembled gold nanospheres on a z-cut crystal in the focus of a radially polarized CVB [9,10].

2 Results and Discussion

For the first sample type (see Fig. 1, left), a strategy for the fabrication of gold nanocones with a pre-defined tilt of the cone axis was developed [6]. The asymmetric geometry supports the transformation of a transversal electric far-field (oriented parallel to the substrate) to a longitudinal plasmonic excitation (perpendicular to the substrate). Nanocones with a range of different tilting angles were examined by angle-dependent extinction spectroscopy. This way the ratio of the electric field components oriented parallel vs. perpendicular to the respective cone axes was systematically changed, as could be observed in the respective experimental and simulated spectra. The cones were imaged by SHG microscopy using tightly focused either azimuthally or radially polarized CVBs, and the data compared to simulations. It could be demonstrated in both the linear and the non-linear optical measurements that an excitation of the cone tip is achieved even with purely in-plane polarized excitation fields.

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For the second sample type, oligomers consisting of gold nanorods that were either radially (Fig. 1, center) or azimuthally (Fig. 1, right) arranged were lithographically fabricated. The longitudinal plasmon resonance of the nanorods was designed to coincide with the fundamental laser wavelength (1060 nm laser with 140 fs pulses and a repetition rate of 80 MHz), and the oligomer geometry was carefully designed to match the electric field distributions of the focused CVBs to maximize interaction and analyse the ensuing collective effects [7]. Maximum SHG was observed for radial oligomers excited by radial, and for azimuthal oligomers excited by azimuthal CVBs. Whereas for radial oligomers, the SHG intensity increased with the number of nanorods, the SHG intensity for azimuthal oligomers went through a maximum, with increasing silencing effects for increasingly small gaps.

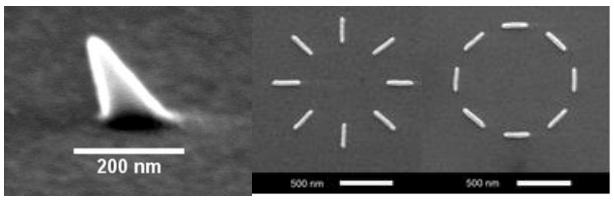

Fig. 1. Tilted gold nanocone (left), radial oligomer (center) and azimuthal oligomer (right) that are imaged by SHG imaging microscopy with cylindrical vector beams.

For the third non-linear study, SHG enhancement was pursued by nanofabricating plasmonic nanostructures on top of nonlinear $LiNbO_3$ crystals. Depending on the cut of the crystal, the extraordinary optical axis can be oriented in different crystal directions. The gold nanoparticles were designed to be resonant at the fundamental harmonic of the exciting pulsed laser (774 nm laser with 110 fs pulses and a repetition rate of 89 MHz), and the cut was chosen for the extraordinary axis to be oriented parallel to the respective dipolar localized plasmon excitation of the nanoparticle. The two systems outlined in Fig. 2 were investigated numerically, as well as with linear dark-field and non-linear SHG spectroscopy. Individual nanodiscs were lithographically prepared on a y-cut crystal and examined under linearly polarized excitation [10], and dense arrays of self-assembled gold nanospheres [8] were created on a z-cut crystal and analyzed in the focus of a radially polarized CVB [9]. While the SHG of the nano-structures themselves proved negligible compared to the SHG intensity from the crystal, 3-fold SHG enhancement could be demonstrated for a single disc on $LiNbO_3$, and 60-fold SHG enhancement for the dense arrays on $LiNbO_3$, due to the interaction of the hybrid systems.

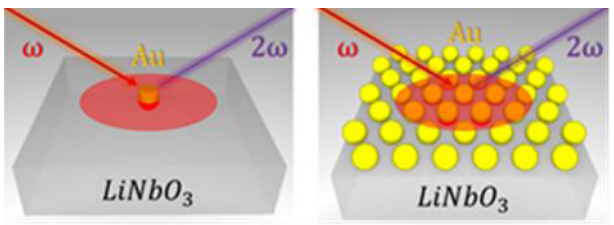

Fig. 2. SHG enhancement by a single gold nanodisc (left) and an array of gold nanospheres from diblock-copolymer micellar lithography (right) on non-linear $LiNbO_3$ crystals (from [11]).

3 Conclusions

SHG from nanostructures that are designed to exhibit localized surface plasmon resonances at the fundamental harmonic of the pulsed excitation laser and are matched to the polarization direction of the electric field components in the laser focus is investigated. In a first example, SHG scanning images are recorded of tilted nanocones, which prove that the hotspots at the tip apex can be excited even with purely in-plane polarized illumination. From SHG scanning images of radially and azimuthally oriented oligomers of gold nanorods it can be shown that collective modes are excited when the orientation of the nanorods in the oligomer matches the orientation of the electric field components in the focus. In a third example, strong local near-field enhancement is created by matching the resonance wavelength of gold nanodiscs or nanospheres that are positioned on the surface of a non-linear crystal to the fundamental of the incident laser beam, leading to up to 60-fold SHG enhancement factors within the laser focus.

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