Resonant harmonic generation in AlGaAs nanoantennas using structured light

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Abstract: We employ structured light to study resonantly-enhanced second- and third-harmonic emission from AlGaAs nanoantennas. We demonstrate correlation between nonlinear emissions with the pump polarization state and Mie-resonant excitation.

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1. Introduction

High-index dielectric and semiconductor nanostructures, supporting Mie-type electric and magnetic resonances can enhance the efficiency of nonlinear optical effects due to the strong field enhancement in the volume of the nanostructure at the resonant wavelength. Second-harmonic generation (SHG) and its polarization-dependence have been studied recently in semiconductor nanostructures, where it was shown that a rotating linearly-polarized pump across the nanostructured substrate can be employed to perform crystallographic characterization [1]. However, several consecutive measurements are usually required to understand the SHG polarization-dependence, which hinders the practical use of this technique. Here, we demonstrate the use of structured light to study the SHG polarization-dependence and analyze the resonant properties of the process. In particular, we perform SHG and third-harmonic generation (THG) scanning microscopy of individual AlGaAs nanodisks, where the SHG can determine the crystalline orientation of the AlGaAs nanodisks through a single measurement. It was observed that both nonlinear processes are resonantly driven when the disk size is resonant with the incident light.

2. Materials and methods

The nanoantennas consist of periodically arranged [100] grown AlGaAs nanodisks with different diameters and constant height h (periodicity = 5 μm, h = 300 nm), embedded in a transparent benzocyclobutene substrate [2]. Radially- and azimuthally-polarized beams were obtained from a femtosecond laser beam at 1060 nm, through a commercial polarization converter (ARCoptix, S.A.). The resulting structured pump beam (Fig. 1a) was focused in the AlGaAs nanoantennas by an infinity corrected microscope objective (50×, 0.8 NA). A point-scanning microscope, operating in reflection, was utilized by shifting the sample along the XY plane and collecting the backscattered signal pixel-by-pixel in a scanning mode, with the same microscope objective [3].

The calculations to simulate the SHG and THG scanning images were performed at the SH and TH frequencies using the method of moments (MoM), where the nonlinearities are dominated by the bulk response of the AlGaAs. The simulations considered a single AlGaAs nanodisk with the same dimensions as the SEM measurements, embedded in a homogeneous medium (n = 1.44). The finite element method (FEM) solver by COMSOL Multiphysics was used to simulate the nonlinear behavior of the AlGaAs nanodisks.

3. Results and discussions

The SHG and THG scanning images show similar features for the excitation with both structured light beams – radial and azimuthal. In the case of the SHG images, the intensity distribution shows a four-lobe symmetric emission, indicating a polarization-dependent process (Fig. 1b). Meanwhile, the THG images show a symmetric doughnut-shaped emission suggesting an isotropic and not polarization-dependent process (Fig. 1c). Higher SHG emissions from the nanodisks are observed when the local polarization of the structured beam is at 45° to the crystalline axis (white arrows shown in Fig. 1b), as compared to a polarization parallel to the crystalline axis [2]. This explains the four-lobe-symmetry emission, observed in the corresponding scanning images (Fig. 1b). Therefore, the crystal orientation of the nanodisks can be identified using a single scanning image. The

measurements are in good agreement with the MoM calculations shown by the images surroanded by the white square.

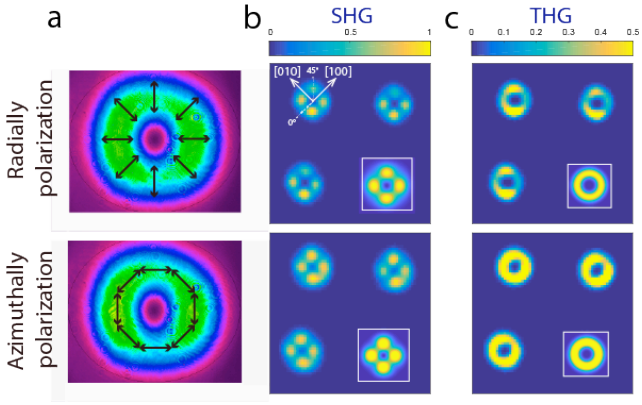

Fig 1. a) Spatial profile of the structured beams, black arrows indicate the polarization state of the beams. b) SHG and c) THG scanning images of AlGaAs nanodisks (d = 485 nm) excited by structured pump beams. White arrows indicate the crystalline axis orientation of the nanodisks. The calculated SHG and THG scanning images of the nanodisks using the corresponding pump beam are indicated by the white square.

We further analyze the resonant behavior of nonlinear emissions by measuring the SH and TH nonlinear emission intensity for different disk diameters, as shown in Fig. 2. Resonantly enhanced SHG emissions are obtained for disk diameters around 450 nm and 600 nm (Fig. 2a). Meanwhile, THG was resonantly enhanced for a nanodisk with a diameter around 600 nm (Fig. 2b). The experimental results are in good agreement with the performed FEM simulations, shown in Figs. 2 c,d.

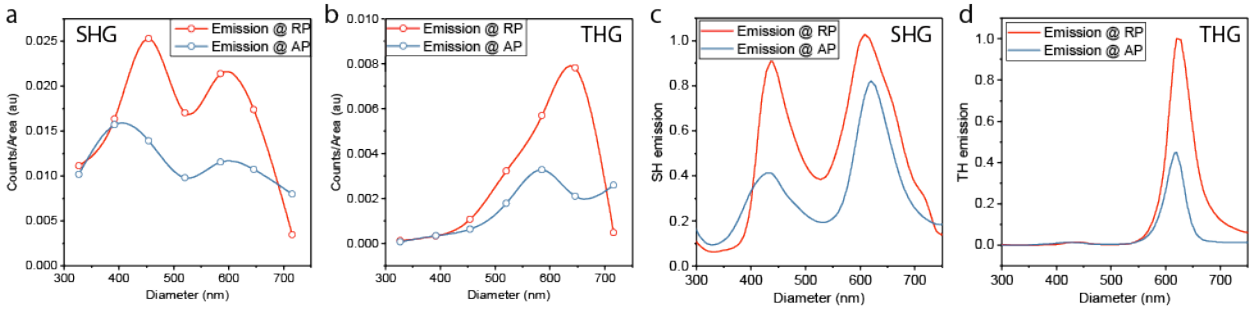

Fig 2. Measured (a,b) and simulated (c,d) second- (a,c) and third- (b,d) harmonic backward signal excited by a radially (red) and an azimuthally (blue) polarized pump, as a function of the nanodisk's diameter. In a) and b) the SHG and THG signals were integrated and normalized by the disk area.

4. Conclusions

SHG and THG microscopy with structured light was performed on individual AlGaAs nanodisks. The imaging technique showed the possibility to map the polarization-dependence of both nonlinear processes. This non-invasive microscopy technique allows the determination of the crystalline orientation of the AlGaAs nanoantennas through a single measurement. Additionally, it was shown that both nonlinear processes are resonantly-driven despite the strong absorption at the second- and third-harmonic wavelengths. The experimental results show good agreement with our numerical simulations.

5. Acknowledgments

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6. References

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