Resonant harmonic generation in AlGaAs nanoantennas using cylindrical vector beams

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Abstract: We use second- and third-harmonic generation with cylindrical vector beams to investigate AlGaAs nanodisks. The nonlinear emission is found to depend strongly on the interplay between tensorial nonlinearities, focal-field symmetries and resonant multipolar excitations.

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1. Introduction

Semiconductor nanostructures with high refractive index provide new opportunities to shape nonlinear optical effects through Mie-type electric and magnetic resonances [1]. Earlier works on such structures, however, have relied on multiple measurements using different plane waves or focused beams with homogenous states of polarization (e.g., linear) requiring several successive measurements by tuning the polarization of the incident beams. Therefore, new and simple nonlinear optical techniques are highly desirable to investigate and understand in detail the nonlinear optical responses of such structures. In this work, we investigate for the first time, the second- (SHG) and third-harmonic generation (THG) microscopy of individual AlGaAs nanodisks with *cylindrical vector beams* (CVB). We demonstrate strong interplay between the tensorial nonlinearities of the nanodisk material, symmetry of the focal-field distributions, and multipolar excitations of the nanodisks.

2. Materials and methods

The samples consist of periodically-arranged AlGaAs nanodisks (height 300 nm and diameter ranging from 327 to 716 nm) that are embedded in a transparent benzocyclobutene substrate [2]. The nanodisk exhibits a zinc blende crystal structure with known crystalline axes (Fig. 1a).

The nanoantennas were studied using a custom-built point-scanning nonlinear microscope [3] that is powered by a pulsed laser (pulse duration 140 fs, repetition rate 80 MHz, and wavelength 1060 nm). To examine a single nanodisk, a microscope objective (numerical aperture 0.8) was used. The scattered nonlinear signals from the samples were collected in reflection and directed to cooled photomultiplier tubes. To create an image, the signals were collected pixel-by-pixel in a scanning mode, i.e., as a function of the sample position with respect to the beam focus. To distinguish SHG and THG, optical filters were used. A mode-converter was used to generate CVBs.

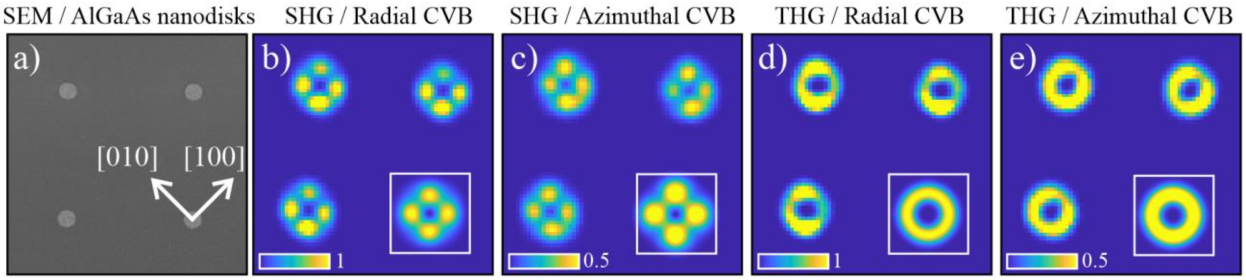

Fig. 1. a) Scanning electron micrograph (SEM) of the fabricated AlGaAs nanodisks (diameter 585 nm, period 5 µm). The crystalline axes [100] and [010] are indicated by white arrows. b-e) Experimental far-field SHG and THG scanning images of the AlGaAs nanodisks (diameter 585 nm) using b,d) radial and c,e) azimuthal CVBs. The SHG images were acquired using an input power of 1 mW and a pixel dwell time of 50 ms. The layout and crystal orientation is the same as in Fig. 1a. Image size: 9.6 × 9.6 µm^2. Insets: calculated far-field SHG and THG scanning images of the nanodisks using the corresponding CVBs. The parameters used in the calculations closely resemble the experiment. Image size: 3 × 3 µm^2. All SHG and THG images are separately normalized for comparison.

The method of moments (MoM) was used to simulate SHG and THG microscopy images from the nanoantennas where the nonlinearities are dominated by the bulk response, despite the strong absorption of the AlGaAs material at the wavelengths of the second and third-harmonic waves. We considered a nanodisk that is made of AlGaAs and embedded in a homogenous medium (n = 1.44). The physical dimensions of the nanodisk were taken from SEM measurements. The far-field scanning images of the nanodisk were calculated at the SH and TH frequencies using parameters that closely resemble the experiments. Finite element method (FEM) was also used to simulate the resonance behavior of the nonlinear emission from the nanodisks.

3. Results and discussions

Due to the structure of the quadratic nonlinear susceptibility tensor of zinc blende crystals, the SHG from the nanodisk is expected to be efficient whenever the impinging local polarization is at 45° to the crystalline axes. As a consequence, the SHG microscopy images of the nanodisk always exhibit a four-fold symmetric intensity pattern (Figs. 1b,c). These hot spots are observed whenever the transversal electric field component of the CVB illuminates the nanodisk. These complementary results strongly suggest that the technique is very sensitive to the crystallographic orientation of the nanodisk. In addition, the crystal orientation of the disks can be identified using a single scanning image without the need for several successive measurements as is traditionally done with linear input polarizations. In contrast, we did not find significant variations in the corresponding THG. This confirms that the THG process in these structures is inherently isotropic. These experimental results are in good qualitative agreement with our MoM calculations (Inset Figs. 1b-e, surrounded by white square outlines).

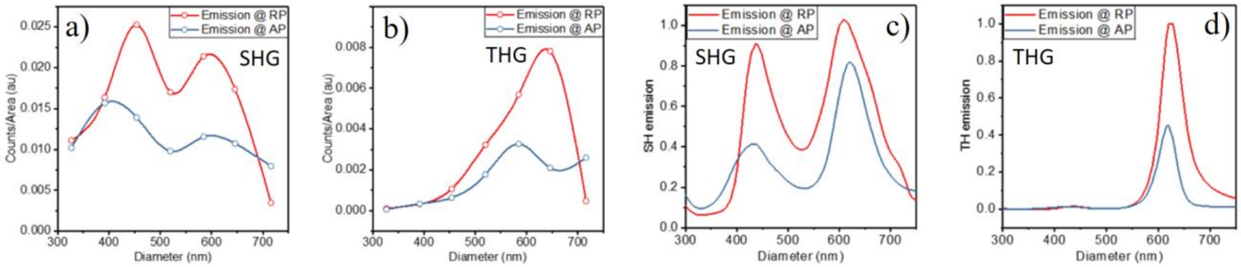

Fig. 2. a,b) Measured and c,d) simulated SHG and THG intensities when the nanodisk is excited by radial (red) and azimuthal (blue) CVBs. In a,b, the SHG and THG signals from a single disk were integrated and normalized by the disk area.

We also found a strong resonance behavior of the SHG and THG signals from nanodisks of different diameters. SHG was resonantly excited for nanodisks with a diameter around 420 nm and 600 nm (Fig. 2a). THG, on the other hand, was resonantly excited only for nanodisks with diameter around 600 nm (Fig. 2b). Again, these experimental data are in good qualitative agreement with our FEM calculations (Figs. 2c,d). Overall, the nonlinear emissions around the resonances due to a radial CVB are higher than that with an azimuthal CVB. These findings are supported by multipolar decomposition of the nonlinear responses of the nanodisks using CVBs.

4. Conclusions and outlook

We used SHG and THG microscopy with cylindrical vector beams (CVB) to investigate individual AlGaAs nanodisks. The technique was found to be very sensitive to the crystallographic orientation of the nanostructure. Furthermore, we found a strong resonance behavior in the nonlinear responses from these structures despite the strong absorption of the second and third harmonic fields. We found good qualitative agreement between our experimental results and simulations. Our work supports the ever-growing utility of imaging techniques using CVB for nonlinear nanophotonics applications. This study is therefore relevant in the development of efficient all-optical characterization platforms for coupling light into nanostructures and in the tailoring of nonlinear optical effects in the nanoscale.

5. References

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