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Enhanced second-harmonic generation with structured light in AlGaAs nanoparticles governed by magnetic response

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High-index dielectric nanoparticles can support both electric and magnetic Mie-type resonances in the visible and near-infrared spectral ranges [1], and they can easily be tailored by the nanoparticle geometry. The optically-induced magnetic response of subwavelength dielectric structures is expected to complement or even replace the electric response of plasmonic components for potential applications at the nanoscale. The Mie-resonant silicon nanoparticles have recently received considerable attention for applications in nanophotonics and metamaterials [1–3] including optical nanoantennas, wavefront-shaping metasurfaces, and nonlinear frequency generation.

In the last few years, the role of nonlinear magnetic response in nanophotonics was intensively addressed. In particular, it was demonstrated [4, 5] that the nonlinear response from silicon nanodisks prevails over the harmonics generated from the bulk silicon, and it possible to reach conversion efficiency high enough for the generated visible light to be observed by naked eye. Generation of different localized Mie-resonant modes can reshape completely the physics of nonlinear effects at the nanoscale [6]. Generated multipoles are closely related to the vectorial nature of the pump beam, and can be distinguished in the far-field region by their polarization and modal composition [7].

Focused and structured vector beams have already been employed for the harmonic generation, and they have been used in nanophotonics as a versatile tool, in comparison with the linearly polarized light, for all-optical characterization, particularly, for microscopic techniques [8]. Recently, we have studied the third-harmonic generation with radially and azimuthally polarized light [9] and, by tailoring the vectorial structure of the pumping light, we have demonstrated a control of both strength and polarization of the excited harmonic fields, also addressing selectively different types of multipolar Mie resonances.

In this Letter, we employ our earlier approach for third-harmonic-generation experiments and study, for the first time to our knowledge, nonlinear second-harmonic (SH) spectroscopy of individual Mie-resonant AlGaAs nanoparticles excited with structured light. Earlier, such problems have been considered only for plasmonic nanoparticles and nanoparticle oligomers [8, 10, 11]. We observe the substantial enhancement of the SH nonlinear signal generated by an azimuthally polarized (AP) pump beam. We supplement our observations by numerical mode-decomposion analysis and reveal the contribution of the modes at the double frequencies, as well as confirm that structured light can be employed as an efficient tool for the enhanced harmonic generation and subwavelength mode control in nanophotonics.

We choose geometrical parameters of an AlGaAs resonator to excite a magnetic multipolar resonance at the fundamental wavelength by an AP pump beam in the considered spectral range and simulate its linear and nonlinear optical response, as well as a disk eigenmode spectrum using the finite-element method. The height of a disk is equal to 650 nm, its diameter is 935 nm. The azimuthal polarization excites magnetic Mie-type multipolar modes which is a typical tendency for subwavelength high-index particles [9, 12, 13]. Contrary to the AP excitation, there are not spectral features for the disk pumped by a radially polarized (RP) vector beam in the linear regime.

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Nanoparticles are made of a custom-designed wafer consisted of GaAs, AlInP and AlGaAs layers by sequential processes of electron beam lithography, dry etching, selective wet-etching, and pillar dropping down to a slide glass substrate. We construct a nonlinear spectroscopy setup based on an optical parametric amplifier. We implement a silicon-based q-plate metasurface [14] to create the vectorial structure of a pump beam. A train of wavelength tunable 300-fs laser pulses is focused from the front sample side to a beam waist size, which is close to a diffraction limit. The SH signal is collected by an objective lens with a numerical aperture 0.9, detected by a visible CCD camera, and normalized over a spectral function of the setup.

Our experimental results on the SH generation spectroscopy of an individual AlGaAs resonator of the specific geometrical parameters with cylindrical vector beams are shown in Fig. 1. There is the enhancement

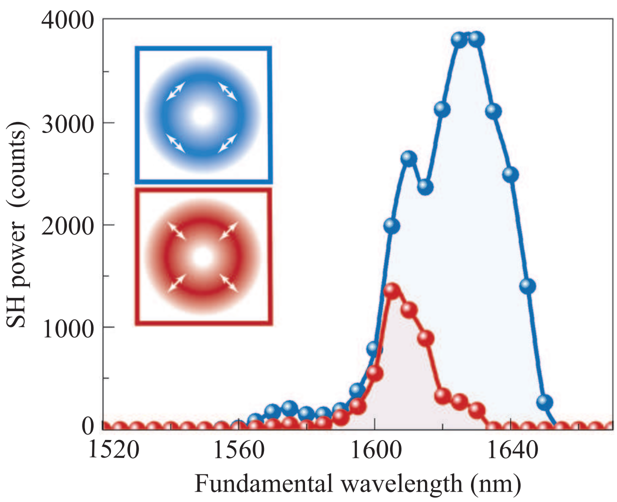

Fig. 1. (Color online) Experimental results. Second-harmonic generation spectroscopy of a single $Al_{0.2}Ga_{0.8}As$ resonator with a diameter of 935 nm pumped by structured light. Blue dotes correspond to the azimuthally polarized pump; red dots – to the radially polarized light, as shown in the inset. White arrows illustrate the polarization distributions in the central cross-section of cylindrical vector pump beams

of the SH signal for the case of a particle which is resonant at the fundamental wavelength and pumped by AP laser light. The additional local peaks of the experimental SH spectrum are associated with the excitation of disk eigenmodes at the SH wavelengths. We observed the four-time less enhancement of the nonlinear response provided by a non-resonant in the linear regime particle excited by a RP laser beam.

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