Observation of Extraordinary SHG from All-Dielectric Nanoantennas Governed by Bound States in the Continuum

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Abstract: We observe record-high efficiency of the second-harmonic generation from AlGaAs nanoantennas fabricated on a transparent substrate and pumped with structured light. The engineered nanoantennas exhibit high-quality optical resonances governed by quasi-bound states in the continuum.© 2019 The Author(s)

OCIS codes: (160.4236) Nanomaterials; (050.6624) Subwavelength structures; (190.0190) Nonlinear optics

1. Introduction

The recent developments of nanoresonators facilitating strong light concentration opened the path towards efficient nonlinear processes on a subwavelength scale [1]. In the recent past many demonstrations have been performed with metallic nanoparticles. Metals were an attractive choice as they exhibit high optical nonlinearities. In addition, plasmon resonances allow for strong light confinement and local field enhancement. However, the efficiency of nonlinear frequency conversion in metal nanostructures remains small, being of the order of $\sim 10^{-11}$ [2]. In plasmonics, it is restricted by Ohmic losses, small mode volumes and low laser damage thresholds. All-dielectric nanostructures have recently been suggested as a promising pathway to improve the nonlinear light conversion beyond the limits associated with plasmonics. High-index dielectric nanoparticles can support nontrivial optical states that increase the efficiencies of nonlinear processes by orders of magnitude, thus offering a paradigm shift in nonlinear optics.

Recently, a concept of bound states in the continuum (BICs) was suggested as a pathway towards efficiency enhancement in nonlinear nanophotonics. BICs represent localized states with energies embedded in the continuous spectrum of radiating waves. BICs were predicted as a mathematical curiosity in quantum mechanics long time ago, and more recently the beautiful physics of BICs was widely used in photonics [3]. In practice, infinite quality factors (Q factors) of BICs are limited by a finite size, material absorption, structural disorder and surface scattering. BICs are manifested as states with a large Q factor (quasi-BICs). Recently, such quasi-BIC were shown to exist in high-index dielectric nanoparticles exhibiting a dramatic increase of their Q factors via the BIC-inspired mechanism [4].



Fig. 1. Theoretical results. (a) Illustration of SHG in a resonant particle under vector beam excitation. (b) Near-field (upper panel) and far-field (lower panel) distributions of the electric field at the pump wavelength for the quasi-BIC. (c) Dependence of the SHG conversion efficiency (solid red curve) and mode quality factor (dashed blue) on the nanodisk aspect ratio *r/h* in the vicinity of the quasi-BIC resonance.

Here we demonstrate sharp enhancement of the second-harmonic generation (SHG) driven by the optically excited quasi-bound state in the continuum (quasi-BIC), which is formed in a subwavelength dielectric resonator via destructive interference of two leaky modes with similar far-field patterns, as suggested previously in Ref. [5].

2. Results and Discussions

Figure 1 summarizes our theoretical results for the SHG enhancement in individual dielectric nanodisks. We choose AlGaAs as a resonator material for its superior second-order nonlinear properties. As shown in Fig. 1a, the resonator is excited by an azimuthally polarized Gaussian beam. It provides perfect mode matching with the high-Q mode field, which distribution is shown in Fig. 1b. A pair of low-Q eigenmodes sustained by the open resonator undergoes strong coupling via continuous tuning of the disk aspect ratio *r/h* that modifies strongly the radiative lifetimes leading to the occurrence of a quasi-BIC state with the significant enhancement of Q factor (see Fig. 1c). Specifically, for 1615 nm wavelength the high-quality mode is formed in an AlGaAs cylinder with 650 nm height and 915 nm diameter. The calculated SHG efficiency exhibits a sharp maximum for the aspect ratio of the quasi-BIC (see Fig. 1c).

We epitaxially grow 650 nm AlGaAs (20% Al) film on a buffer AlInP layer on top of a [100] GaAs wafer. The AlGaAs resonators are fabricated with an electron-beam lithography with chemically-assisted ion beam etching. The resonators are transferred to a glass substrate from GaAs wafer [see Figs. 2(a, b)]. We vary the diameters of the resonators in the range of 875-935 nm around the size of 915 nm. In our experiment, we pump the resonators with 300 fs pulses from an optical parametric amplifier MIROPA-fs-M from Hotlight Systems tuning the wavelength from 1490 to 1700 nm. To couple efficiently to high-Q modes, we employ structured light [6], creating azimuthally or radially polarized pump beam with a home-made silicon metasurface. The beam is then focused by X100 0.7NA infrared objective, and the SH is collected in forward direction by X100 0.9NA visible objective. The harmonic is then spectrally filtered, and it is imaged on a camera Trius 694 with a lens. We use an infrared Ophir power meter to measure the intensity of pump, and a visible Ophir power meter to calibrate the camera measurements.

Figures 2(c, d) show the SHG intensity as a function of both pump wavelength and the resonator diameter, and Figure 2(e) shows the results for the 915 nm resonator supporting the high-Q mode. We observe a pronounced peak in the second-harmonic intensity for the 915 nm resonator at 1615 nm wavelength for the azimuthal polarization of pump. To date, in our experiments we have estimated the overall conversion efficiency of $I_{SH}/I_{pump}=10^{-3}$ in a sub-optimal experimental arrangement. The system holds promise to demonstrate higher efficiency in our ongoing research.



Fig. 2. Experimental results. (a, b) Schematic and electron microscope image of a subwavelength resonator, respectively. (c, d) Experimentally measured intensity of the generated second harmonic field as a function of the pump wavelength and resonator diameter for (c) azimuthally and (d) radially polarized pump beam. Dashed and solid lines show the dispersion of nanodisk modes at the pump frequency and second-harmonic frequency, respectively. (e) SHG intensity as a function of the pump wavelength for a high-Q resonator with the diameter of 915 nm.

3. Conclusions

We have demonstrated experimentally optical resonances governed by bound states in the continuum in an individual sub-micrometer AlGaAs particle. We have observed record-high efficiency of the second-harmonic generation reaching 10⁻³ being empowered by the enhanced field confinement associated with high-Q resonant modes.

The authors acknowledge the Russian Science Foundation (18-72-10140) and collaboration with A. Bogdanov. K.K. acknowledges the Foundation for the Advancement of Theoretical Physics and Mathematics "BASIS" for support.

4. References

- [1] S. Kruk and Y. Kivshar, "Functional meta-optics and nanophotonics governed by Mie resonances", ACS Photonics 4, 2638 (2017).
- [2] D. Smirnova and Y. S. Kivshar, "Multipolar nonlinear nanophotonics", Optica 11, 1241 (2016).
- [3] C.W. Hsu et al., "Bound states in the continuum", Nature Review Materials 1, 16048 (2016).
- [4] M. V. Rybin et al., "High-supercavity modes in subwavelength dielectric resonators", Phys. Rev. Lett. 119, 243901 (2017).
- [5] L. Carletti et al., "Giant nonlinear response at the nanoscale driven by bound states in the continuum", Phys. Rev. Lett. 121, 033903 (2018).
- [6] E. Melik-Gaykazyan et al., "Selective third-harmonic generation by structured light in Mie-resonant nanoparticles", ACS Phot. 5, 728 (2017).