Supplementary Materials for

High-harmonic generation from a subwavelength dielectric resonator

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Supplementary Text
Figs. S1 to S8
ABSTRACT

Supplementary Materials include complementary finite-difference time-domain (FDTD) simulation results of ultrashort laser pulse propagation through AlGaAs subwavelength resonator and layers of Al₂O₃, ITO, and glass as well as additional information on their fabrication. First section compares the field enhancements, laser-induced carriers produced inside the resonator in Figures S1 and S2, and the transmitted spectra for resonant and off-resonant irradiation conditions in Figure S3. For the resonant case, the HHG spectra and the conversion efficiencies for the harmonics of interest are compared for different incident laser power in Figures S4 and S5. Finally, the nonlinear optical response produced by unstructured bulk sample is shown, and the perturbative and non-perturbative contributions related to laser-induced carriers to the harmonic yields are identified in Figure S6. Figure S7 shows the additional data for optimization of the thickness of a spacer between ITO layer and the resonator. The last figure S8 shows schematics of fabrication steps of the resonators.

Resonant and Off-Resonant Excitation

In order to verify that the subwavelength structure is very sensitive to the resonant position and the resonant geometry or laser excitation conditions are favorable to HHG, the simulations were performed for different values of nanodisk diameter and laser wavelengths. The laser power is fixed to $P = 50$ mW everywhere except different values are mentioned, pulse duration is $\theta = 522$ fs, the nanodisk height is fixed to $H = 1.38 \mu m$. We show 2D distributions of nonlinear intensity and laser-induced carriers inside the resonator in the propagation excitation plane in Figures S1 and S2. In both situations, stronger fields and higher electron densities are confined on the front side of the nanostructure. As a result, more pronounced HHG spectra are obtained with the comparison provided in Figure S3.

We note additionally that the field enhancement predicted in the nonlinear Maxwell simulations might deviate here from the analytical predictions of the resonant mode. One of the reasons is the spatial resolution limited to $\Delta x = 10$ nm, for instance, the considered resonant height of the nanodisk $H = 1.38 \mu m$ deviates from the optimal position of the resonance peak ($H = 1.384 \mu m$). The second reason is related to the transient optical properties of AlGaAs during ultrashort laser pulse propagation defined by laser-induced carriers that would modify the maximum field enhancement and would slightly shift the resonant position. Nevertheless, the numerical experiment shows that the resonant geometry and laser irradiation conditions correspond to a stronger nonlinear signal, which is close to its maximum and decreases significantly if the diameter is increased/decreased or laser wavelength is blue- or red-shifted.

Power Dependence

For the resonant geometry/excitation case, the simulations have been performed for different laser powers with the transmitted HHG spectra shown in Figure S4. Here, the numerical model includes such effects as the spectrum broadening and the blueshift of the individual harmonics due to laser-induced plasma, pronounced for laser powers $P > 60$ mW. The scaling of the calculated
**Figure S1.** Theoretical calculations of light intensity and electron density distributions inside the resonators. (a-c) Nonlinear intensity. (d-f) Maximum electron density distributions in subwavelength resonator. The diameters are varied: (a, d) $D = 1.6 \mu m$, (b, e) $D = 2.05 \mu m$, and (c, f) $D = 2.15 \mu m$. Laser wavelength is centered at $\lambda = 3.75 \mu m$. Wave-vector $\vec{k}$ indicates the propagation direction of laser pulse.

**Figure S2.** Theoretical calculations of light intensity and electron density distributions inside the resonators. (a-c) Nonlinear intensity. (d-f) Maximum electron density distributions in subwavelength resonator. The excitation laser wavelengths are varied: (a, d) $\lambda = 3.5 \mu m$, (b, e) $\lambda = 3.75 \mu m$, and (c, f) $\lambda = 4 \mu m$. The diameter of the nanodisk is fixed to $D = 2.05 \mu m$. Wave-vector $\vec{k}$ indicates the propagation direction of laser pulse.
Figure S3. Theoretical calculations of high harmonic spectra from subwavelength resonator. Fields transmitted into the substrate for resonant and off-resonant excitation conditions. (a) The diameters are varied $D = 1.6 \mu m$ (blue), $D = 2.05 \mu m$ (resonant, red), and $D = 2.15 \mu m$ (magenta). (b) Central laser wavelengths are varied $\lambda = 3.5 \mu m$ (blue), $\lambda = 3.75 \mu m$ (resonant, red) and $\lambda = 4 \mu m$ (magenta).

Contributions to HHG

It is interesting to compare the HHG spectra from unstructured bulk material (the thickness of AlGaAs layer is taken equal to the nanodisk height) and from a single subwavelength resonator shown in Figure S6(a). The graph shows that the amplitudes of high harmonic signal (harmonics after the third one) from the unstructured sample are lower by many orders of magnitude than the ones amplified by resonator and would be undetectable in the experiment.

High-order harmonics can be produced by frequency mixing after propagation of second- and third-order nonlinearities or by non-perturbative effects, which are incorporated in the simulation model via the intraband current driven by laser-induced carriers in AlGaAs. To evaluate the contribution of these carriers to HHG spectra, it is possible to perform the simulations with and without carriers. The results are shown in Figure S6(b), underlying that the carriers are responsible from one to two orders stronger harmonic yields for odd harmonics. As a result, the estimations based solely on the constant nonlinear susceptibilities in a perturbative regime would strongly underestimate the nonlinear effects in resonant structures.

Optimization of the Substrate Thickness

Schematics of the Fabrication Process
Figure S4. Theoretical calculations of high harmonic spectra from subwavelength resonator. (a) High harmonic spectra of the transmitted fields from subwavelength resonator into glass sample for different laser powers $P = 10 - 100$ mW and resonant excitation conditions (laser wavelength centered at $\lambda = 3.75\mu m$ and the diameter is $D = 2.05\mu m$). (b) The fifth and (c) the seventh harmonics are shown separately.
Figure S5. Theoretical calculations of 5\textsuperscript{th} and 7\textsuperscript{th} harmonics power versus pump power. At 50 mW pump power the calculated conversion efficiencies are 10\textsuperscript{-7} and 10\textsuperscript{-9}.

Figure S6. Theoretical comparison of harmonics generation for a resonator vs an unstructured film, and comparison of perturbative vs nonperturbative calculations. (a) High harmonic spectra of the transmitted fields in case of subwavelength resonator (red line) and unstructured sample (blue line) of the same thickness as the disk height \( H = 1.38 \mu m \). (b) Comparison of the harmonic spectra produced by both perturbative second- and third-order nonlinearities via cascade effects and intraband current of laser-excited carriers (non-perturbative, red) and by perturbative nonlinearities alone (perturbative, blue).
Figure S7. Theoretical linear calculations of the resonator’s Q-factor as a function of the substrate coating layer thickness.

Figure S8. Schematics of fabrication process. (A) Electron-beam lithography for the definition of a PMMA mask. (B) Dry etching for the formation of a vertical pillar structure. (C) PMMA mask removal with O₂ plasma. (D) Wet etching of the Al₀.₅₁In₀.₄₉P sacrificial layer by HCl solution. (E) Picking up the Al₀.₂Ga₀.₈As nanodisks using the PPC-coated PDMS stamping method. (F) Transferring the nanodisks to the ITO substrate by heating the hot chuck to 90°C to separate the PPC from the PDMS.