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Silicon Metasurfaces with Bound States in the Continuum for High-Harmonic Generation

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ABSTRACT

Bound states in the continuum (BICs) represent dark modes trapped in the radiation continuum. BICs received significant attention in optics and photonics as a simple tool to achieve giant quality factors by transforming them into quasi-BICs. Here, we report the observation of high-harmonic generation in dielectric metasurfaces hosting BICs. The metasurface is composed of a square lattice with parallel Si bars of a slightly different width placed on a transparent substrate. The structure is engineered to support a quasi-BIC in the mid-IR with a high quality factor. We tune the metasurface asymmetry to enable the optimal coupling condition that provide the highest high-harmonic generation efficiency. In the experiment, we demonstrate the generation of odd optical harmonics from the 3rd to the 11th order in the BIC regime and study their polarization dependence. We measure the dependence of the high-harmonic signal on the input intensity. The concept of metasurfaces with highly localized light boosted by BIC resonances provides a new degree of freedom to control experimentally strong nonlinear optical response.

Keywords: bound states in the continuum, metasurfaces, nonlinear optics, high-harmonic generation

1. INTRODUCTION

Photonic BIC[1-5] and quasi-BICs find a variety of applications in optics. Recently it was shown that for metasurfaces composed of subwavelength meta-atoms with broken in-plane symmetry, the Q factor of quasi-BICs can be unambiguously controlled by variation of meta-atom asymmetry parameter[6]. The variation of the asymmetry parameter allows to achieve the optimal coupling condition that maximizes the fields inside the metasurface via matching the rates of radiative and non-radiative losses.

Lower-order harmonics (such as 2nd and 3rd) have been observed in a variety of subwavelength systems starting from plasmonic nanoparticles, as well as in all-dielectric nanostructures with substantially enhanced efficiencies due to Mie resonances[7-8]. The maximization of field enhancement in asymmetric metasurfaces supporting quasi-BICs was investigated for enhancement of second- and third-order nonlinear response[9-10]. High-harmonic generation (HHG) was originally studied for gas[11] and solid-state[12] systems, while in nanostructures its observation is challenging due to low conversion efficiencies and has been little studied.

Here, we report the observation of HHG in dielectric metasurfaces hosting BICs. The metasurface is composed of a square lattice with parallel Si bars of a slightly different width placed on a transparent substrate. The structure is engineered to support a quasi-BIC in the mid-IR with a high Q factor. We tune the metasurface asymmetry to enable the optimal coupling condition that provide the highest high-harmonic generation efficiency. In the experiment, we demonstrate the generation of odd optical harmonics from the 3rd to the 11th order in the BIC regime and study their polarization dependence.

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2. RESULTS AND DISCUSSION

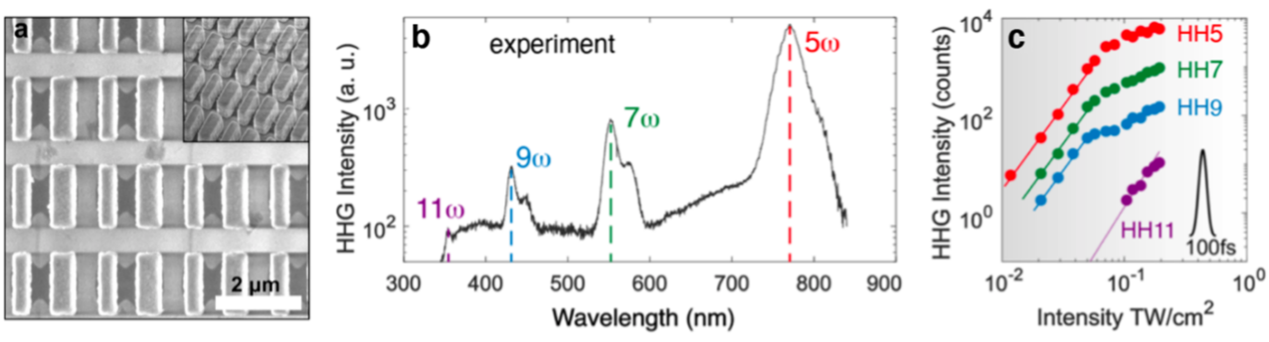

Figure 1. Resonant enhancement of harmonic generation efficiency by the BIC resonance. (a) SEM image of the fabricated metasurface. (b) Spectral measurements of the generated signals up to 11th optical harmonic. (c) Measured HHG intensity versus the pump intensity for 5th to 11th optical harmonics. Lines shows the fit with a power dependence.

The fabrication process of the metasurfaces hosting BICs begins with the deposition of an amorphous Si film on a sapphire wafer with plasma-enhanced chemical vapor deposition. The metasurface is composed of a square lattice with parallel Si bars of a slightly different. The metasurface pattern is defined via electron-beam lithography and then the pattern is transferred onto the material using a reactive ion etching. The scanning electron microscopy image is shown in Fig. 1a.

Next, we measure the HHG spectra for various setups and pulse parameters. More specific, 3rd, 5th and 7th harmonics were studied using 2 ps pulses of a mid-infrared laser system consisting of a femtosecond laser (Ekspla Femptolux), and an optical parametric amplifier (MIROPA Hotlight Systems). When the metasurface was pumped at 3830 nm wavelength with 2 ps pulses at 5MHz repetition rate, it produced the signal spectrum. The pump wavelength in the region 3500-4100 nm was scanned while monitoring the intensity of the 5th harmonic and the enhancement of the generation efficiency in the vicinity of the BIC resonance was observed. Next, the 9th and 11th optical harmonics were detected using shorter mid-IR pulses of duration 80fs and a repetition rate of 1 kHz from a Ti:Sa (Coherent Astrella) pumped optical parametric amplifier (TOPAS). The spectrum of high-harmonic intensity is shown in Fig. 1b. The power-to-power dependence on the incident intensity is shown in Fig. 1c.

The dependence of harmonic intensity on the pump power in Fig. 1c is non-perturbative. This can be explained by generation of free carriers by multiphoton absorption and tunneling mechanism. We calculate the electron density n in the conduction band as:

$$\frac{\partial n}{\partial t} = R(t, \omega, |\mathbf{E}(t)|) \tag{1}$$

Here the strong field excitation rate R(t, λ, E) is calculated using the Keldysh expression

$$R(t,\omega,|\mathbf{E}(t)|) = 2\frac{2\omega}{9\pi}\left(\frac{m^*\omega}{\hbar}\frac{\sqrt{1+\gamma^2}}{\gamma}\right)^{\frac{3}{2}} Q\left(\gamma, \frac{\Delta_{\text{eff}}}{\hbar\omega}\right) \times \exp\left[-\pi\left\langle\frac{\Delta_{\text{eff}}}{\hbar\omega}+1\right\rangle\frac{\mathcal{K}(\phi)-\mathcal{E}(\phi)}{\mathcal{E}(\Theta)}\right], \tag{2}$$

where

$$Q(\gamma,x) = \sqrt{\frac{\pi}{2\mathcal{K}(\Theta)}} \times \sum_{n=0}^{\infty} \exp\left[-\pi\cdot\frac{\mathcal{K}(\phi)-\mathcal{E}(\phi)}{\mathcal{E}(\Theta)}\cdot n\right]\cdot\Phi\left[\sqrt{\frac{\pi(\langle x+1\rangle - x + n)}{2\cdot\mathcal{K}(\Theta)\cdot\mathcal{E}(\Theta)}}\right], \tag{3}$$

K and E are complete elliptic integrals of first and second kind, respectively, with dimensionless derivatives

$$\Theta = \frac{1}{1+\gamma^2}, \tag{4}$$

and

$$\phi = \frac{\gamma^2}{1+\gamma^2}, \quad (5)$$

$\gamma = \frac{\omega\sqrt{m^*\Delta}}{e|\mathbf{E}(t)|}$ is the Keldysh parameter, Δ is the intrinsic band gap, m* is the reduced mass of the effective electron and hole masses, ω is the laser frequency, *e* is the elementary charge and |**E**(t)| is the amplitude of the total electric field inside the metasurface enhanced by the quasi-BIC resonance. Also,

$\Phi(x) = \int_0^x \exp(\xi^2 - x^2) d\xi$ is the Dawson integral, and $\Delta_{\text{eff}} = \frac{2}{\pi}\Delta \cdot \left[\frac{\sqrt{1+\gamma^2}}{\gamma}\mathcal{E}(\Theta)\right]$ is the bandgap value modified due to the Stark shift. The parameters of silicon are $m^*=0.18m_e$ and direct bandgap Δ=3.4 eV. For the simulations we use a pulse with the peak field

$$E_p^{(0)} = \left(\frac{2I_p}{c\varepsilon_0}\right)^{1/2}. \quad (6)$$

The peak intensity is $I_p = 3\times10^{11}$ W/cm^2 (pulse energy 0.8 μJ), which gives $E_p^{(0)} = 1.5\times10^9$ V/m. The value of the Keldysh parameter for $|\mathbf{E}(t)|=E_p^{(0)}$ is γ = 0.6. The pulse is resonant with the quasi-BIC wavelength 3.9 μm, $\tau_{FWHM} = 100$ fs, τ = 85 fs. The losses of quasi-BICs are given by γ_0 = 5THz, which corresponds to the mode quality factor of 50.

The numerical simulations are shown in Fig. 2. They demonstrate that the field strength of the laser pulse enhanced by the quasi-BIC reaches 3×10^9 V/m, which allows the density of carriers of $n_p = 6.8\times10^{19}$ cm^{-3}. Since we evaluated the field enhancement at the quasi-BIC electric hotspot, the value of $n_p=6.8\times10^{19}$ cm^{-3} gives us the upper bound for the electron-hole plasma density. The lower bond in given by the density of carries induced solely by the incident laser pulse, which is 1×10^{17} cm^{-3}.

To conclude, we have engineered Si metasurfaces supporting BIC resonances in the mid-IR spectral range. Using the optimal coupling principle, we have designed and fabricated metasurfaces that allow the observation of the high optical harmonics up to 11th order. The concept of metasurfaces with highly localised light which is achieved by employing BIC resonances provides a new degree of freedom to control experimentally the nonlinear optical response.

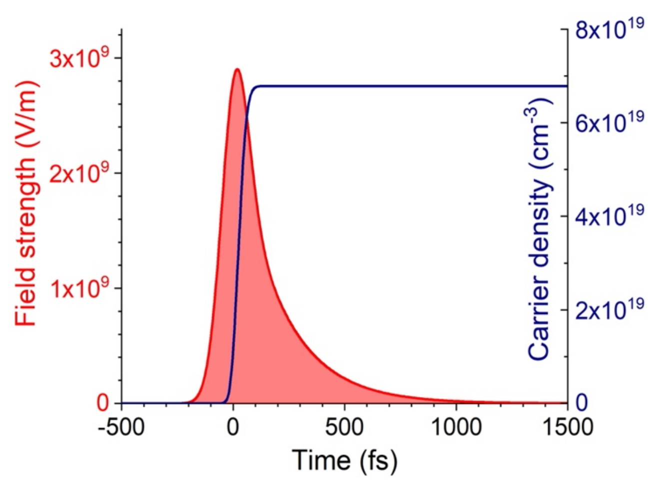

Figure 2. Calculated resonant field strength |**E**(t)| and electron-hole plasma carrier density for the 0.8 μJ pulse.

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