Research Article

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Si metasurface supporting multiple quasi-BICs for degenerate four-wave mixing

https://doi.org/10.1515/nanoph-2024-0128
Received March 11, 2024; accepted May 22, 2024; published online June 5, 2024

Abstract: Dielectric metasurfaces supporting quasi-bound states in the continuum (qBICs) enable high field enhancement with narrow-linewidth resonances in the visible and near-infrared ranges. The resonance emerges when distorting the meta-atom’s geometry away from a symmetry-protected BIC condition and, usually, a given design can sustain one or two of these states. In this work, we introduce a silicon-on-silica metasurface that simultaneously supports up to four qBIC resonances in the near-infrared region. This is achieved by combining multiple symmetry-breaking distortions on an elliptical cylinder array. By pumping two of these resonances, the nonlinear process of degenerate four-wave mixing is experimentally realized. By comparing the nonlinear response with that of an unpatterned silicon film, the near-field enhancement inside the nanostructured dielectric is revealed. The presented results demonstrate independent geometric control of multiple qBICs and their interaction through wave mixing processes, opening new research pathways in nanophotonics, with potential applications in information multiplexing, multi-wavelength sensing and nonlinear imaging.

Keywords: nanophotonics; dielectric metasurfaces; bound states in the continuum; nonlinear flat-optics

1 Introduction

In recent years, metasurfaces supporting resonances originating from bound states in the continuum (BICs) have enabled strong enhancement of electromagnetic fields with high quality factors \(Q = \nu/\Delta \nu\), with \(\nu\) the resonance frequency and \(\Delta \nu\) its linewidth) in the visible and near-infrared spectral regions. BICs, by nature, are perfectly confined states that cannot couple to the radiation continuum. However, by distorting the meta-atom’s shape or orienting a metasurface supporting a symmetry-protected BIC [1]–[3], the dark state can be turned into an accessible high-Q quasi-BIC (qBIC). Among many different design choices of unit cells, such as rods [4]–[6], disks [7], [8], blocks [9], [10], and rings [11], [12], that have allowed quality factors as high as \(10^4\), the dimer of tilted elliptical cylinders has shown great promise thanks to its fabrication robustness [13]. Initially devised for realizing optomechanically induced chirality [14], it was later adapted for surface enhanced molecular detection in the mid-infrared region [15] and efficient second harmonic generation with continuous wave laser sources [16].

Devices incorporating nonlinear metasurfaces could be the next step in the emerging industry of flat-optics [17]–[21], where some uses like nonlinear beam steering [22], directional emission switching [23], nonlinear lensing and imaging [24], and nonlinear holography [25] have already been demonstrated. The applications are, however,
restricted by the inherent low efficiency of nonlinear effects in nano-sized media, as the subwavelength interaction volume prevents exploiting phase-matching processes [26]. Dielectric nanostructures that support qBIC resonances could overcome this problem as they allow for high enhancement of the near fields [27], [28] and exhibit low absorption and high nonlinear susceptibilities in the visible and near-infrared ranges, making them a very interesting platform to further develop the area. QBIC-supporting metasurfaces have demonstrated their capabilities for second-[16], [29] and third- [4], [9], [30], [31] harmonic generation, while more complex nonlinear processes involving multiple frequencies, like four-wave mixing (FWM), have been explored only with Mie-like modes [32], [33], or combining qBIC with Mie resonances [34], but not by mixing different qBICs.

Typically, a single qBIC resonance is present for a given metasurface design [4]–[10]. Having various of these states could potentially improve the metasurface performance and expand the scope of planar opto-devices. Recently, theoretical investigations have predicted multiple qBICs in dielectric metasurfaces by means of symmetry breaking [35]–[37] and period doubling [38]. We further highlight that both the refractive index contrast between the dielectric and its surrounding and the height of the array need to be carefully optimized to achieve multiple qBICs in a single metasurface [37]. The linear excitation of two symmetry-breaking qBICs has been experimentally demonstrated [39], while stacked metasurfaces have been needed to excite more than two of these states [40]. In this work, we design and fabricate a metasurface of amorphous-Si (a-Si) elliptical cylinder dimers on silica, as shown in Figure 1A. Two geometric distortions are exploited, one involving the tilt angle \( \alpha \) of the meta-atoms, and the other the change of intra-dimer separation, \( d \times P/2 \), with \( d \) a dimensionless parameter and \( P \) the array period (see Figure 1A and B). We show that two qBIC resonances can be excited for each of these distortions, with all four states present when both asymmetries are introduced (Figure 1C). We further study the third order process of degenerate FWM (DFWM), which involves two incident light sources, to exploit the different qBIC resonances of our metasurface, an effect that has so far only been addressed numerically [37], [41]. DFWM combines two photons of frequency \( \omega_1 \) with one photon of frequency \( \omega_2 \) to create a photon of frequency \( \omega_s = 2\omega_1 - \omega_2 \). The experimental results are validated by linear and nonlinear numerical simulations, showing very good agreement.

2 Results

Figure 1 illustrates the unit cell design of the studied a-Si metasurface on silica, fabricated by electron beam lithography (details on the fabrication process can be found in the Section 4). The height \( H \) is 160 nm, the short \( (D_x) \) and long \( (D_y) \) diameters of the elliptical cylinders are 130 and 290 nm, respectively, and the periodicity \( (P) \) is 485 nm. These dimensions were chosen to generate resonances in the near-infrared region. The tilt angle \( \alpha \) and the intra-cell distance \( d \times P/2 \) are varied to excite the qBIC resonances. (c) Schematic illustration of the transmission spectra with all four resonances present and the process of degenerate four-wave mixing exploiting two of them.

Figure 2A displays the experimental and simulated transmittance spectra of the metasurfaces by varying \( d \) at \( \alpha = 0^\circ \) (see Section 4 for measurement and simulation specifics). The topmost curve \( (d = 1) \) shows a broad Mie-like resonance below 700 nm, arising from a magnetic dipole...
contribution of the individual meta-atom, and a high flat transmission level above 720 nm, within the transparency window of the dielectric. When \( d \) is decreased, two narrow resonances appear around 740–750 nm due to the introduced asymmetry, which comes from the difference between the center-to-center distance of meta-atoms within a unit cell and that across neighboring unit cells [42], [43]. A similar scenario develops in Figure 2B, which presents the response when changing \( \alpha \) at \( d = 1 \). As the tilt angle increases, two resonances emerge around 760 and 820 nm, respectively. In all cases, the resonance linewidths increase when further deviating from the symmetric conditions, with quality factors ranging from 50 to 200.

Next, we demonstrate that the in-plane rotation and lateral displacement of the meta-atoms can be combined to simultaneously generate the two pairs of qBICs, as can be seen in Figure 3A and B by setting \( d = 0.8 \) and \( \alpha = 10^\circ \). The average field enhancement inside the dielectric is presented in Figure 3C, showing an enhancement factor around 3 at resonant conditions. To characterize this light confinement, we later performed DFWM experiments by exciting two of the most prominent resonances, at 741 and 816 nm (740 and 800 nm in simulations, respectively), marked with asterisks in the graphs. Their corresponding field distributions at different cross-sections of the unit cell can be seen in Figure 3D. At 800 nm (right column), an electric dipole pointing in the \( x \)-direction allows \( x \)-polarized light to excite the qBIC, due to the tilt distortion. This state, which has been extensively observed in tilted elliptical cylinders metasurfaces [15], [44], [45], is characterized by an out-of-plane magnetic dipole and an in-plane electric quadrupole that cancel each other out in the far-field when \( \alpha = 0^\circ \); increasing the tilt angle adds an electric dipole component that couples the state to external radiation. As for the other selected qBIC, at 740 nm wavelength (left column in Figure 3D), the excitation of a magnetic dipole in the \( y \)-direction, originating from the asymmetry parameter \( d \), creates an out-of-plane electric field circulation, as has been previously observed in arrays of dimers of circular [35], [46] and D-shaped [47] meta-atoms. The two remaining states (at 730 and 760 nm) correspond to the magnetic counterparts of the described resonances. A detailed mapping of the field distributions for all four resonances with their multipolar decompositions can be found in Figure S2 of the Supplementary Materials. In Figure S3 of the Supplementary Materials, we show the transmission response of other possible combinations of \( d \) and \( \alpha \). In Figure S4 of the Supplementary Materials, we demonstrate how all resonances can be tuned across a 100 nm window at a fixed height of the metasurface by scaling the diameters of the elliptical cylinders and the array periodicity. The same figure also reveals that a minimum height of the structure is needed to support the multiple modes, consistent with our previous findings [37].

The DFWM experiments were performed by using two tunable pulsed laser sources, from now on labeled as ‘Pump 1’, adjustable from 720 to 750 nm, and ‘Pump 2’, from 800 to 840 nm. Figure 4A shows representative spectra of the pump pulses alongside the signal of the generated nonlinear light at \( \omega_s = 2\omega_1 - \omega_2 \). By comparing with the response of an unstructured a-Si film, a nonlinear enhancement factor...
Figure 3: Combining tilt angle and dimer distance distortions. (a) Experimental and (b) simulated transmittance spectra of a metasurface with $d = 0.8$ and $\alpha = 10^\circ$ for $x$-polarized incoming light. (c) Corresponding average field enhancement inside the dielectric $\langle E/E_0 \rangle$. (d) Electric field distributions for different planes of the unit cell at the two resonant conditions highlighted with asterisks in A–C (740 nm, left, and 800 nm, right). These two states were selected for the DFWM experiments.

Figure 4: Degenerate four-wave mixing performance. (a) Pump lasers spectra and nonlinear signal spectrum of the metasurface characterized in Figure 3. The signal of an a-Si film is shown for reference. The nonlinear process diagram is illustrated as an inset. Arrows at the bottom mark the corresponding scale of each spectrum. (b) Dependence of the generated nonlinear light power, $P_{NL}$, on the pump powers $P_1$ and $P_2$. When changing $P_1$ and $P_2$ is fixed at 0.76 mW, and a quadratic dependence is obtained. When varying $P_2$ at $P_1 = 0.86$ mW, the relationship is linear.
of 600 times is observed, an order of magnitude above that reported for a dimer-hole a-Si metasurface mixing a qBIC and a Mie resonance [34]. To further characterize the nonlinear nature of the process we independently varied the pump power of both lasers \( (P_1 \) and \( P_2 \)) and monitored the collected nonlinear power \( (P_{NL}) \), as shown in Figure 4B. As expected, when increasing \( P_1 \) while keeping \( P_2 \) constant, \( P_{NL} \) shows a quadratic dependence, while a linear relationship is obtained when changing \( P_2 \) at fixed \( P_1 \). With this data, we compute a normalized efficiency of the process of \( (1.1 \pm 0.2) \, \text{W}^{-2}\% \). We attribute this relatively low value to the reduced vectorial overlap between the excited modes. As the nonlinear polarization in the material is maximum when the fields are aligned with one another (see Section 4), having mostly in-plane and out-of-plane electric field distributions for the different states, respectively, weakens the nonlinear effect. It is also important to note that these experiments were performed using \( x \)-polarized light. Results by varying the polarization are present in Figure S5 of the Supplementary Materials, revealing a further decrease in the signal, as expected from numerical simulations.

To assess the nonlinear signal dependence on the resonant conditions, both pump wavelengths were independently swept. In Figure 5A, on the right side, \( \lambda_1 \) is kept fixed at 741 nm, and \( \lambda_2 \) is varied, showing a well-defined peak at the resonant state. On the other hand, fixing \( \lambda_2 = 816 \, \text{nm} \) and tuning \( \lambda_1 \) (left side) reveals a less defined peak, which can be described by the contribution of 3 nearby resonances (see Figure 3A), convoluted with the spectral width of the laser, which is around 7–8 nm, same as the studied resonances. Figure 5B exhibits the corresponding nonlinear simulations, showing good agreement with the measurements. Sharper responses are obtained as the calculation assumes monochromatic pumping wavelengths. Given that the measured efficiency was performed in a transmission configuration, the simulated radiation in Figure 5B considers only the cone matching the numerical aperture of our collection objective (see inset), so that the relative height of the peaks can be compared to the measurement.

The presented results show that multiple qBIC resonances can be attained in one metasurface design. We further demonstrate that these states can be tailored to select the enhancement of a specific third-order wave mixing process. This concept could be extended to new mixings based on second-order nonlinearities by using noncentrosymmetric materials, such as GaP, GaAs, and AlGaAs. To improve nonlinear efficiency, fabrication quality as well as spatial and vectorial overlap of the engineered states need to be optimized.

### 3 Conclusions

In summary, we have designed an a-Si on silica metasurface with an elliptical cylinder dimer unit cell that can support two pairs of qBIC resonances, depending on the imposed geometric variations, which are the tilt angle \( \alpha \) and the intra-cell distance parameter \( d \). The four qBICs have quality factors ranging from 50 to 200 and can all be present when both asymmetries are introduced. We selected a metasurface with \( \alpha = 10^\circ \) and \( d = 0.8 \) to study the third order nonlinear process of DFWM by pumping two qBIC resonances. The obtained normalized efficiency is \( (1.1 \pm 0.2) \, \text{W}^{-2}\% \) with incident average powers below 1 mW for both laser sources. Our work demonstrates that multiple qBIC resonances can be controlled independently in a metasurface, showing promise for nanoscale wave mixing phenomena. This could enable new research pathways for sensing, nonlinear imaging, and information multiplexing applications of metasurface-based opto-devices.
4 Methods

Metasurface fabrication: A 160-nm thick amorphous silicon layer was deposited onto a fused silica substrate by plasma-enhanced chemical vapor deposition (PECVD). The sample was spin-coated with an adhesion layer of SurPass 4000 that was washed away with isopropyl alcohol (IPA) before coating with photoresist ZEP 520A and a highly conducting polymer ESpacer 300Z. The patterning was done via electron-beam lithography (Raith eLINE Plus) with a dosage of 100 μC/cm². After development for 60 s in 3:1 MIBK:IPA solution, a hard-mask consisting of 30 nm Cr was deposited by electron-beam evaporation before lifting-off the photoresist with a bath of Microposit Remover 1165 for 2 h at 80 °C. At this point, the remaining Cr pattern allows transferring the design by etching the uncovered Si by means of reactive ion etching (RIE). Finally, the Cr layer was removed with Sigma-Aldrich-651826 Standard chromium etchant. The presented SEM images in the Cr layer was removed with Sigma-Aldrich-651826 Standard Si by means of reactive ion etching (RIE). Finally, the Cr layer was removed with Sigma-Aldrich-651826 Standard chromium etchant. The presented SEM images in Figure S1 of the Supplementary Materials were taken with the same e-beam lithography equipment.

Transmittance characterization: A supercontinuum light source from NKT Photonics was used to measure the metasurfaces transmittance spectra. The illumination and the transmitted light collection were done with a 10X, NA = 0.25 and a 60X, NA = 0.70 microscope objectives, respectively.

Nonlinear measurements: Two pulsed sources were provided by a COHERENT Ti:Sapphire laser pumping an OPO module of the same company. ‘Pump 1’ was selected from the second harmonic of the OPO signal (converting 1000–1600 nm to 500–800 nm) and ‘Pump 2’ was obtained directly from the Ti:Sapphire laser (700–1000 nm). The pulse width is around 170 fs and the repetition rate 80 MHz. To ensure that both laser pulses arrived at the same time on the sample, a delay line was added on the ‘Pump 2’ arm. The lasers were focused onto a 10–12 μm spot using a 4X, NA = 0.1 microscope objective. The generated nonlinear light was collected in transmission with a 40X, NA = 0.6 objective. The excitation laser beams were filtered with low pass filters for the nonlinear signal measurements. For more details refer to the schematic in Figure S6 of the Supplementary Materials.

Numerical simulations: Linear and nonlinear calculations were performed using the Wave Optics module of the COMSOL Multiphysics software [48]. The Si structures composing the unit cell were placed on a SiO₂/Air interface in a square prism domain geometry with periodic boundary conditions in the four lateral faces and a perfectly matched layer (PML) at the top and bottom faces. The refractive index of the materials was obtained from ellipsometry data; the obtained values for n and k of Si and SiO₂ are presented in Figure S7 of the Supplementary Materials. A non-zero extinction coefficient value (k = 0.02) was added to the high-index dielectric to better approximate the experimental results, effectively accounting for non-radiative losses coming from surface roughness.

The nonlinear simulations were performed under a perturbative approximation. First, the linearly excited fields within the nanostructures were computed to define a third-order polarization in the material, which was then used as a source at the DFWM frequency to obtain the nonlinear field. The third-order susceptibility tensor of Si has 21 nonzero components with three independent elements [49]; all were set to \( \chi^{(3)} = 2.8 \times 10^{-18} \text{ m}^2/\text{V}^2 \). With this, the nonlinear polarization has the form:

\[
\mathbf{P}(2\omega_1 - \omega_2) = 6\varepsilon_0 \chi^{(3)}(\mathbf{E}(\omega_1) \cdot \mathbf{E}(\omega_2))\mathbf{E}(\omega_1) \\
+ 3\varepsilon_0 \chi^{(3)}(\mathbf{E}(\omega_2) \cdot \mathbf{E}(\omega_1))\mathbf{E}(\omega_2)
\]

The multipole decompositions shown in Figure S3 of the Supplementary Materials were calculated using the equations found in reference [51]. The near-to-far field transformation used to calibrate the collected signal in Figure 5B was computed using the open-source software package RETOP [52], for which we created an artificial 15 × 15 array using the solution of a single unit cell as the near-field solution.

Research funding: This work was partially supported by PICT 2021 IA 363, PICT 2021 GRF-TI-00349, PIP 112 202001 01465, UBACYT Proyecto 2002022000078BA, and UBACYT Proyecto 2002190200296BA. GQM acknowledges 2023 ‘Ale-Arg’ research grant, from the Argentine Ministry of Education and the Deutscher Akademischer Austauschdienst (DAAD, German Academic Exchange Service). AVB acknowledges the Alexander von Humboldt Foundation’s generous support. We also acknowledge funding and support from the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) under Germany’s Excellence Strategy – EXC 2089/1 – 390776260, the Bavarian program Solar Energies Go Hybrid (SolTech), the Bavarian program Enabling Quantum Communication and Imaging Applications (EQAP), the Center for NanoScience (CeNS), the Lee-Lucas Chair in Physics, and the European Union (ERC, METANEXT, 101078018). Views and opinions expressed are however those of the authors only and do not necessarily reflect those of the European Union or the European Research Council Executive Agency. Neither the European Union nor the granting authority can be held responsible for them.
Author contributions: GQM carried out sample design, optical measurements, data analysis, and manuscript writing. TW and GQM performed metasurface fabrication. TP and LM assisted with optical measurements. EC, LM, AVB, SAM, AT and GG provided overall research leadership. All authors revised the manuscript.

Conflict of interest: Authors state no conflicts of interest. Stefan Maier is an Editor of the Nanophotonics journal and was not involved in the review and decision-making process of this article.

Data availability: The data supporting this study’s findings are available from the corresponding author upon reasonable request.

References


Supplementary Material: This article contains supplementary material (https://doi.org/10.1515/nanoph-2024-0128).