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ABSTRACT
Vacuum ultraviolet (VUV) light plays a crucial role in various scientific and technological fields, such as nanolithography and biomedical treatments. However, the inherent nonlinear optical coefficient of nonlinear optical crystals is typically very low, and increasing the action length is often necessary to improve the nonlinear conversion efficiency. This makes it challenging for these materials to achieve high-density optoelectronic integration at the micro-/nano-scale. In this study, we propose a design for generating coherent VUV radiation close to 175 nm using second harmonic generation (SHG) with an absolute efficiency exceeding 1.2 mW/lo. This is achieved by merging multiple bound state in the continuum modes in a free-standing photonic crystal slab. Even with fabrication imperfections at a level lower than 10% disorder, the SHG efficiency of the samples remains robust, maintaining an efficiency of at least 2 mW/lo. This research provides a beneficial platform for generating efficient VUV light in the nanoscale.

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Vacuum ultraviolet (VUV) light is an electromagnetic wave with the wavelength range of 100–200 nm, which is utilized in a wide range of applications, from biomedical procedures to nanolithography and attosecond pulse generation application, due to its high photon energy. Achieving coherent VUV light primarily relies on nonlinear optical processes using solid-state laser sources in nonlinear optical crystals. However, higher-efficiency upconversion processes in nonlinear optical crystals, such as second harmonic generation (SHG), are limited by the phase-matching of VUV and fundamental frequency (FF) waves, as well as optical absorption. In addition, because of its low inherent nonlinear optical coefficient, the nonlinear conversion efficiency of nonlinear optical crystals is typically boosted by increasing the action length or periodically poled grating structure, posing challenges for their application in integrated micro-/nano-optoelectronic devices.

Recently, there has been extensive research on the generation of coherent VUV radiation from nanostructures, including nanodisks with anapole modes, gratings with periodic structures, and magnetic dipole resonance. Nanostructures offer high compactness, allowing them to focus incident light into nanoscale hotspots and generate harmonic light without the need for phase matching. By designing nanostructures, both wavelength and intensity of VUV radiation can be flexibly regulated and controlled. These simple and compact VUV sources are ideal for promising applications, including multi-point scanning systems for imaging and materials analysis, high-resolution nanolithography, and microscopy, resulting in improved capabilities.

Bound states in the continuum (BIC) in periodic nanostructures have recently garnered significant attention in the field of nonlinear optics. These structures exhibit infinite quality factors and are particularly relevant in photonic crystal slabs (PCS). In PCS, BICs display topological vortex center of polarization vectors in far-field, which ensures their robustness of BICs due to the existence of conserved topological charge. These BICs have found applications...
micro-laser,\textsuperscript{17,18} circularly polarized light,\textsuperscript{19,20} and nonlinear nanophotonic.\textsuperscript{21} To overcome the limitations of ideal BICs, quasi-BICs are created by breaking the in-plane symmetry or oblique incident excitation to obtain better interaction with incoming waves. However, the Q factor of isolated quasi-BICs may degrade due to structural asymmetry and fabrication imperfections. An attempt to increase Q factor has been the topologically merging multiple BICs in the reciprocal system.\textsuperscript{22,23} Merged BICs exhibit high-Q resonances that are resilient against structural disorder, providing an effective means to enhance VUV radiation.

In this Letter, we present a simple and robust approach for generating efficient VUV light using merged BICs in a PCS made of periodic circular air holes. By gradually varying the lattice constant and membrane thickness of the structure, we merge nine BICs in the Brillouin zone while maintaining a resonant wavelength of 350 nm. The merged BICs possess ultra-high Q factor and enhanced electric field, surpassing those of the isolated and accidental BICs. Consequently, the SH conversion efficiency improves at oblique incident angles. We achieve 3.35\% SHG conversion efficiency of lithium niobate (LN) excited by a fundamental wavelength, with the electric field mainly concentrated inside the PCS, resulting in an enhanced interaction between the LN and FF field.

The LN thin film is a promising material for coherent radiation with optical frequency conversion.\textsuperscript{24} It possesses its high nonlinear susceptibility, transparency in the wavelength range below 350 nm, and the potential for miniaturization in optoelectronic devices. The LN nanostructures, such as LN waveguide\textsuperscript{25} and metasurface,\textsuperscript{26,27} have been extensively utilized to enhance second harmonic nonlinear processes, leading to increased conversion efficiency with tightly localized fundamental light.

A PCS consists of a suspended LN thin film with periodic circular air holes, as illustrated in Fig. 1(a). The geometry parameters of a unit cell are characterized by the thickness of membrane (h), periodicity (a), and radius of circular holes (r). An x-cut LN is chosen to ensure that the direction of the maximum second-order nonlinear coefficient (d\textsubscript{33}) lies within the plane. Linearly polarized light enters from the x direction with polarization in the z axis. Therefore, despite LN being an anisotropic material, the incident plane wave is still considered with linear polarization. In the simulation using COMSOL Multiphysics software, the refractive index of LN was taken from the ordinary refractive index. In the simulation using COMSOL, we calculated the near-field distribution of three types of BICs on the upper surface (parallel to the xy plane) and the side (parallel to the xz plane) at an angle of 0.009 rad. Compared to both accidental and isolated BICs, merging BICs exhibit a stronger field enhancement at the fundamental wavelength, with the electric field mainly concentrated inside the PCS, resulting in an enhanced interaction between the LN and FF field.

When the BICs are merged, the angle-resolved transmittance spectra of TE A band of structure under linear polarized incidence are simulated in Fig. 2(a). The merging BIC mode cannot be excited only where \( \theta \) is the angle between z axis and field polarization. The \( \theta \) in our simulation is zero; thus, we only take account of nonlinear coefficient \( d_{33} \) and z component \( P_{z}^{2} \) of polarization intensity. The nonlinear conversion efficiency can be significantly improved by strengthening the local electromagnetic field.

In order to construct merging BICs, we start by identifying accidental BICs at the off \( \Gamma \) point. The black curve in Fig. 1(c) shows that the Q factor of transverse electric (TE) A photonic band (red line in Fig. 1(b)) becomes infinity at \( k = 0 \) and 0.07\( \pi/a \), which is in accordance with the expression\textsuperscript{22} \( Q \propto 1/(k - k_{BIC})(k + k_{BIC})^{2} \). Therefore, TE A band preserves both symmetry-protected BIC (k = 0) and accidental BICs (k = 0.07\( \pi/a \)) when the PCS has \( h = 200 \) nm, \( a = 189 \) nm, and \( r = 80 \) nm. The two-dimensional (2D) color map of Q factors in Fig. 1(d) reveals eight symmetrically located accidental BICs around the center of Brillouin zone. The corresponding far-field polarization vector maps shown as the white arrows in Fig. 1(d) demonstrate that each BIC behaves as a topological vortex point, possessing an integer topological charge of either +1 or -1. Owing to the symmetry of the structure, one BIC is fixed in the center of the first Brillouin zone, while the other eight off center BICs can be shifted by varying system parameters.\textsuperscript{22} To achieve FF resonant BIC modes at approximately 350 nm, we adjust both the lattice \( a \) and thickness \( h \) of the structure, resulting in photonic bands of accidental BICs, merged BICs, and isolated BIC as illustrated in Fig. S1. As illustrated in Fig. 1(d), when \( a \) increases from 189 to 189.1 nm and \( h \) decreases from 210 to 206.8 nm, the nine BICs merge into a single BIC with a charge of +1. Further changing to \( a = 200 \) nm and \( h = 92 \) nm, the positive and negative topological charges annihilate around \( \Gamma \) point, leaving only an isolated BIC.

Figure 1(c) illustrates that the Q factor of the isolated BIC decreases quadratically with momentum k (blue curve), while this scaling formula changes to Q \( \propto 1/k^{2} \) in the merged BIC (red line), thus preserving its larger Q factor relative to k. Meanwhile, the merged BIC has very high Q values over a wider range of k space when compared to isolated BIC. Figure S3 illustrates the relationship between the mode volume and wavelength of three types of BIC at vertical incident angle. It can be seen that with an FF wavelength of 350 nm, the mode volumes for accidental BIC, merged BIC, and isolated BIC are 6.35 \( \times 10^{5} \), 6.18 \( \times 10^{5} \), and 7 \( \times 10^{5} \) nm\(^{3}\), respectively. At an oblique incidence angle of 0.009 rad, the FF wavelength is 356.6 nm. In this scenario, the mode volume is 8.96 \( \times 10^{5} \) nm\(^{3}\) for the accidental BIC, 6.53 \( \times 10^{5} \) nm\(^{3}\) for the merged BIC, and 8.17 \( \times 10^{5} \) nm\(^{3}\) for the isolated BIC. The mode volumes for three types of BIC modes are roughly \( 10^{6} \) nm\(^{3}\), and they are almost identical. The Q factor of merged BIC is significantly larger than that of the other two types of BIC near the \( \Gamma \) point, as shown in Fig. 1(c). Thus, the interaction between light field and lithium niobate is mainly determined by Q factor. As depicted in Fig. S2, we calculated the near-field distribution of three types of BICs on the upper surface (parallel to the yz plane) and the side (parallel to the xy plane) at an angle of 0.009 rad. Compared to both accidental and isolated BICs, merging BICs exhibit a stronger field enhancement at the fundamental wavelength, with the electric field mainly concentrated inside the PCS, resulting in an enhanced interaction between the LN and FF field.

The Q factors of merged BICs are calculated by

\[
Q = \frac{1}{\kappa k} \quad \text{and} \quad \kappa = \frac{\omega}{c} \quad \text{for} \quad k \neq 0.
\]
at normal incidence (gamma-point). All angles excluding 0 degree have to result in the excitation of quasi-BIC resonance. Nevertheless, when the incidence angle lies between $-0.008$ and $0.008$ rad, no transmittance spectra are detected. From Fig. 1(c), it can be seen that the Q factor of the merging BIC mode is greater than $10^6$ within this range, leading to an extremely narrow full width at half maximum (FWHM) of the transmittance spectra. The sweep step size of our numerical simulation was set to $0.02$ nm, which may have led to poor resolution of Fig. 2(a) in the vicinity of gamma-point. Black curve in Fig. 2(b) displays one-dimensional (1D) transmittance spectrum at $0.01$ rad oblique incidence. Its resonance dip is positioned at $356.62$ nm, with a FWHM of around $0.03$ nm.

In the SHG simulation, the refractive index is set to $2.54$ for the fundamental wavelength, and $1.665$ (real part) and $1.186$ (imaginary part) for the SH frequency, taking into account the loss in the LN. The nonlinear polarization can be expressed as $P_{2\omega} = e_0 d_{33} E_{\omega} E_{\omega}$. The $d_{33}$ of LN is estimated to be $41.7$ pm/V with the pump wavelength linearly polarized at around $350$ nm. The SHG intensity is
directly proportional to $P(2\omega)$, meaning that we can control SHG intensity by the tuning local electric field of FF. The incident FF light resonates with the merged BIC mode, thereby enhancing the FF electric field intensity and effectively strengthening the corresponding SH electric field.

The SHG conversion efficiency $\eta$ is defined as $P_{SH}/P_{FF}$. Here, $P_{SH}$ is the total radiated power at SH, and $P_{FF}$ is the pump power incident at the fundamental frequency FF. The parameters set in the COMSOL Multiphysics software electromagnetic waves frequency domain module for the specific simulation calculation, include an FF wavelength, an input average power of 30 mW, and an oblique incidence angle. The output SH power is acquired through closed surface integration of the Poynting vector. By dividing the SH power by the input power, the SH conversion efficiency can be calculated. The red line in Fig. 2(b) shows that the SHG efficiency reaches 2.2% at the resonant dip when the incidence angle is 0.01 rad. Figure 2(c) illustrates the SHG conversion efficiency of LN PCS for varying incident angles. The peak value of $\eta$ reaches 3.35%, occurring at 0.009 rad in response to a fundamental wavelength of 356.6 nm. Compared to previous LN nanostructures generating VUV radiation, our result demonstrates significantly higher SHG at low pump power, as shown in Table I. As shown in Fig. 2(d), the SHG conversion efficiency is plotted as a function of the average power of the excited light, ranging from 10 mW to 110 mW. It is clear that the SHG conversion efficiency is linearly proportional to the pump power, leading to the normalized conversion efficiency reaching 1.2 mW/C0.

Table I displays the near-field distributions and far-field patterns of LN PCS for merged BICs at 356.6 and 178.3 nm wavelengths under 0.009 rad incident angle. The near-field distribution at 356.6 nm in the yz plane [Fig. 3(a)] and xy plane [Fig. 3(c)] presents that the electric field is symmetrically located at the interface between LN and air within the plane, with the maximum electric field enhancement at the

<table>
<thead>
<tr>
<th>Year</th>
<th>Structure type</th>
<th>Pump power/peak intensity</th>
<th>FF (nm)</th>
<th>$\eta = P_{SH}/P_{FF}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>2007</td>
<td>PPLN</td>
<td>29 mW</td>
<td>1068</td>
<td>0.01138</td>
</tr>
<tr>
<td>2015</td>
<td>LN resonator</td>
<td>10 mW</td>
<td>785–815</td>
<td>1.35 × 10^{-4}</td>
</tr>
<tr>
<td>2018</td>
<td>LN nanodisk</td>
<td>5.31 GW cm^{-2}</td>
<td>351.3</td>
<td>1 × 10^{-4}</td>
</tr>
<tr>
<td>2019</td>
<td>LN thin film</td>
<td>22.8 mW</td>
<td>840</td>
<td>1.6 × 10^{-5}</td>
</tr>
<tr>
<td>2020</td>
<td>LN metasurfaces</td>
<td>0.88 mW/4.3 GW cm^{-2}</td>
<td>1550</td>
<td>1 × 10^{-6}</td>
</tr>
<tr>
<td>2020</td>
<td>LNGW</td>
<td>25 mW</td>
<td>1064</td>
<td>4.6 × 10^{-7}</td>
</tr>
<tr>
<td>2021</td>
<td>LNGW</td>
<td>0.22 mW/1.33 GW cm^{-2}</td>
<td>690</td>
<td>8.13 × 10^{-5}</td>
</tr>
<tr>
<td>2022</td>
<td>LN metasurfaces</td>
<td>35 mW</td>
<td>951</td>
<td>2 × 10^{-4}</td>
</tr>
<tr>
<td>2023</td>
<td>LN metasurfaces</td>
<td>2 kW cm^{-2}</td>
<td>965</td>
<td>0.01</td>
</tr>
<tr>
<td>2023</td>
<td>This work</td>
<td>30 mW</td>
<td>356.6</td>
<td>0.0335</td>
</tr>
</tbody>
</table>
The SH near-field distribution is mainly concentrated on the surface of the LN structure, as shown in Figs. 3(b) and 3(d). The far-field distribution shows that the FF electric field [Fig. 3(e)] propagates within the yz plane, while the SH electric field [Fig. 3(f)] propagates along the x axis. Despite adsorption loss in LN within the VUV range, Fig. 3(f) demonstrates efficient strengthening of the SHG electric far-field intensity at merged BIC resonance mode.

Real fabricated samples exhibit disorder and imperfections, which lead to increased loss channels of resonance modes, thereby restricting the maximum attainable Q factor. Figure 4(a) depicts a typical manufacturing error, where a round hole is replaced with an elliptical hole having different short axes, determined by the asymmetry parameter $\Delta r$. The blue curve in Fig. 4(b) displays the SHG conversion efficiency is kept $2\%$ for $\Delta r$ smaller than 8 nm, indicating a robust field enhancement even with sample disorder. This is due to the fact that the merging BICs resonance in a sample with fabrication defect has the weaker radiation field and the higher Q value than those of isolated BIC. The red line in Fig. 4(b) displays a redshift in fundamental wavelength as a function of $\Delta r$, with a slope of 2.1, indicating that the SH wavelength shifts by roughly 1 nm for every 1 nm of perturbations. As a result, the merging BICs stabilize considerable SHG enhancement even in structural disorder.

In conclusion, the design of LN freestanding PCS aims to greatly enhance the efficiency of SHG through merged BICs. When pump source at 356.6 nm is incident at 0.009 rad, the absolute conversion efficiency at the SH of 178.3 nm can reach up to $1.2 \cdot 10^{-3}$ mW$^{-1}$. This represents a significant improvement compared to that of previous LN nanostructures. In the far-field region, the FF electric field has been enhanced through the merging BICs resonance mode.
strengthened within the plane, while SH electric field has been intensified out of the plane. By replacing circle holes with ellipse holes featuring asymmetry parameters, SHG efficiency is found to remain robust, even with up to 10% fabrication imperfections. Overall, this work establishes the foundation for realizing efficient VUV source at the micro-/nano-scale, which can facilitate integration of nanophotonic devices.

See the supplementary material for the supplementary figures about photonic band structures of accidental BICs, merged BICs and isolated BICs, the near-field distributions, and mode volumes of these types of BICs.

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**AUTHOR DECLARATIONS**

**Conflict of Interest**

The authors have no conflicts to disclose.

**Author Contributions**

Jianmei Li and Wenyao Chang contributed equally to this work.

**Jianmei Li:** Conceptualization (lead); Formal analysis (equal); Funding acquisition (equal); Investigation (lead); Methodology (equal); Project administration (equal); Supervision (lead); Validation (equal); Writing – original draft (equal); Writing – review & editing (equal).

**Wenyao Chang:** Data curation (equal); Formal analysis (equal); Investigation (equal); Methodology (equal); Project administration (equal); Supervision (lead); Validation (equal); Writing – original draft (equal); Writing – review & editing (equal).

**Zirui Guo:** Formal analysis (equal); Methodology (equal); Software (equal); Visualization (supporting).

**Pinxu Li:** Software (equal); Formal analysis (equal); Visualization (equal); Validation (equal).

**Ziyi Fu:** Methodology (supporting); Writing – review & editing (supporting).

**Yanxue Hou:** Funding acquisition (equal); Project administration (equal); Supervision (equal); Validation (equal); Writing – review & editing (equal).

**Changzhi Gu:** Funding acquisition (equal); Project administration (equal); Supervision (equal).

**DATA AVAILABILITY**

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

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