Highly efficient second harmonic generation of thin film lithium niobate nanograting near bound states in the continuum

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Abstract
Bound states in the continuum (BICs) are ubiquitous physical phenomena where such states occur due to strong coupling between leaky modes in side lossy systems. BICs in meta-optics and nanophotonics enable optical mode confinement to strengthen local field enhancement in nonlinear optics. In this study, we numerically investigate second-harmonic generation (SHG) in the vicinity of BICs with a photonic structure comprising one-dimensional nanogratings and a slab waveguide made of lithium niobate (LiNbO3, LN). By breaking the symmetry of LN nanogratings, BICs transition to quasi-BICs, which enable strong local field confinement inside LN slab waveguide to be supported, thereby resulting in improving SHG conversion with lower pump power of fundamental frequency (FW). With a peak intensity of 1.33 GW cm\(^{-2}\) at the FW, our structure features a second-harmonic conversion efficiency up to \(8.13 \times 10^{-5}\) at quasi-BICs. We believe that our results will facilitate the application of LN in integrated nonlinear nanophotonic.

Supplementary material for this article is available online

Keywords: lithium niobate, second harmonic generation, bound states in the continuum

(Some figures may appear in colour only in the online journal)
Q-factor states, lossy materials such as silicon nitride (Si$_3$N$_4$), silicon (Si) have been investigated. Recently, all-dielectric high-Q resonant structures such as symmetry-breaking silicon metasurfaces [10] and silicon nitride nanogratings or photonic crystal [11, 12] have garnered significant interest. Although these structures support quasi-BICs, ultrahigh-Q resonances can still be observed. The excitation of the strong narrow-linewidth resonance results in strong local field enhancement in optical structures and is a dominant feature of light–matter interactions.

Second harmonic generation (SHG) is the most prominent nonlinear effect, converting a fundamental harmonic of $\omega$ into its second harmonic with frequency with $2\omega$ [13]. It has been applied extensively, i.e. in holographic imaging [14], human face and quick response code recognitions [15] and harmonic optical tomography [16], etc. However, because the optical nonlinear response is intrinsically weak, advances in subwavelength-engineered plasmonic and all-dielectric metasurfaces have been studied thereby to enhance optical nonlinear processes in very small cavities, which phase-matching condition seems no long significant [17–19]. Theoretically, the second-order nonlinear polarization can be expressed as $P^{(2)}(2\omega) \propto \int \chi^{(2)}(r)|E_{loc}(r, \omega)|^2dV$, where $\chi^{(2)}(r)$ is the second nonlinear coefficient; $E_{loc}(r, \omega)$ is the local optical field of nanostructure at fundamental frequency (FW). It indicates that these techniques are developed to realize strong local field enhancement of electromagnetic fields in a small volume, which boosts much higher nonlinear conversion efficiencies than in the constituent materials [20]. Hence, significant efforts have been expanded for the local field enhancement. However, high-Q resonances driven by BICs has been developed to improve second- and third harmonic generation from periodic or individual subwavelength resonators with high-refractive index materials such as Si and GaAs.

Among the nonlinear materials, lithium niobate (LN) is an excellent choice for SHG owing to its significant second-order susceptibility ($d_{33} \sim 27\,\text{pm}\,\text{V}^{-1}$) [21, 22] and wide applications in electro-optical modulators [23]. Furthermore it is also a low-loss material with a transparency window spanning from the ultraviolet to the mid-infrared regime, thereby enabling operation in the visible spectral range (VIS). Consequently, significant efforts have been expanded for improving the SH conversion efficiency of LiNbO$_3$-based nanostructures, e.g. optical anapole mode assisted SH egneration in LN nanodisk [24, 25], gold nanoring embedded LN nanocylinder for SH enhancement [26], Fano-resonance structures for high-efficient SHG [27]. In this regard, BICs or quasi-BICs contributing to strong light–matter interactions provide a platform for SHG, thereby enabling potential applications of LN in integrated nonlinear nanophotonics.

In this study, we numerically investigate optical BICs and quasi-BICs in a photonic structure consisting of one-dimensional nanograting and slab waveguide made of LN. This photonic structure can be abbreviated as LNGW. We focused on its ability to upconvert VIS light to near ultraviolet range. In particular, we investigated the generation of BICs or quasi-BICs in LNGW using COMSOL Multiphysics. Next, we exploited its optical characteristics via Lumerical FDTD solution at FW, where the behavior and local field distribution in the LNGW were reflected by changing the asymmetry of nanogratings. Ultrahigh Q-factor resonance at quasi-BICs yield strong local field enhancement, thereby enabling light–matter interaction in SHG to be improved. Furthermore, we comprehensively investigated the SHG of LNGW from BICs to quasi-BICs at visible pump wavelength and demonstrate the dependence between SH conversion efficiency and the asymmetry of LN nanogratings detailedly. Finally, we conclude that our findings and show that the BICs or quasi-BICs supporting LN nanophotonics structures enable highly efficient SH response to be achieved without necessitating a cavity at a low pump power.

2. BICs in LN waveguide-grating optical structure

Theoretical and numerical calculations were performed to better understand the physical phenomenon of BICs. The LNGW under consideration is shown in figure 1(a).

One unit cell of this structure comprises a 360 nm thick LN slab waveguide and two 290 nm high LN gratings placed on a SiO$_2$ substrate in free space, illuminated in an oblique manner with a z-polarized (TE-polarized) plane wave. Although LN is an anisotropic material with extraordinary and ordinary refractive indexes, the latter is considered herein such that the coupling between the LN slab waveguide and two nanogratings can be revealed more easily. Therefore, we set refractive index of LN to 2.2 at 700 nm. To reveal the coupling, we first utilize Helmholtz equation to perform calculations for the dispersion diagrams of the TE-polarized modes inside the LN slab waveguide, as follows:

$$\frac{d^2E_z}{dx^2} + \left[ k_0^2 n_{LN}^2 - \beta^2 \right] E_z = 0. \tag{1}$$

Here $n_{LN}$ is the refractive index of LN, $k_0$ the incident wavevector in free space, $\beta$ the propagation constant of LN waveguide. By considering the boundary condition in equation (1), we can further obtain the following eigenmode:

$$\frac{\omega n_{LN}}{2\pi c} = \frac{1}{n_{LN}} \left[ \frac{\beta h_2}{2\pi} + \frac{\varphi_0 + m\pi}{2\pi} \right]. \tag{2}$$

Here $\varphi_0$ is a phase constant; $m = 0, 1, 2$, which corresponds to modes TE$_0$, TE$_1$, TE$_2$, respectively. The dispersion curves are shown in figure 1(b). When the LN nanogratings are symmetrical in one unit cell, e.g. the pitch is one-half of the entire grating, the diffraction wavevector is expressed as:

$$\frac{k_x h_2}{2\pi} = \frac{k_0 h_2 \sin(0)}{2\pi} - \frac{n h_2}{\lambda} (n = 0, \pm1, \pm2, \cdots). \tag{3}$$
In this study, the incident angle was set to be $\theta = 5^\circ$, $n$ is the order of diffraction wavevector, $k_0$ is the wavevector in free space. To further illustrate this physical phenomenon, we plot the dispersion relationship of waveguide modes (TE$_0$, TE$_1$, TE$_2$), represented by blue, orange and yellow lines, respectively. BICs occurs when two modes satisfy wavevector matching condition, e.g. $\beta = k_{x,m}$. (c) Mode-coupling analysis between gratings and waveguide, marked by red spheres and olive spheres, respectively. BICs will occur around the guided modes resonance. By tuning center-to-center distance of LN nanogratings, one channel is opened to couple with broadband channel supported by the LN slab waveguide. To quantify the asymmetry of the LN nanogratings, we define a factor $\delta$ as follows:

$$\delta = \left( \frac{2d - \Lambda}{2\Lambda - 4w} \right).$$

where $d$ is the distance between two nanogratings with one of them fixed, the other moving in one unit cell. Figure 1(c) demonstrates the wavevector relationship between slab waveguide (red spheres) and nanogratings (olive spheres) when the asymmetry factor $\delta$ is tuned. The wavevector of slab waveguide was maintained, whereas that of nanogratings was varied to perform coupling with slab waveguide; the cross-section point implies the occurrence of BICs. Hence, the results of the eigenmode analysis was confirmed

$$k'_{x,m} = n_{air}k_0 \sin(\theta) - j\pi \left( \frac{1}{\Lambda_1} + \frac{1}{\Lambda_2} \right).$$

where $n_{air}$ is the refractive index of air, $j$ is $-1$ in this study, and $k_0$ is the wavevector in free space. $\Lambda_1$ and $\Lambda_2$ are expressed as $\delta(2\Lambda - 2w) + \Lambda$ and $\Lambda - \delta(2\Lambda - 2w)$,
The Q-factor estimated in this method are represented as a real constant, and respectively (see S1 (available online at stacks.iop.org/ NANO/32/325207/mmedia) in the supporting information for more details). We investigated the reflection spectra via Lumerical FDTD solution with perfect matched layers in x-axis and periodic boundary condition in y-axis. The oblique incident light was TE-polarized with an electric field parallel to the z-axis in Cartesian coordinate. The results are shown in figures 1(d), (e). As shown in figure 1(d), two breakpoints occurred at wavelengths 695 and 825 nm in reflection spectra when the asymmetry factor \( \delta \) was tuned from -1 to 1, i.e. from the asymmetric to symmetric case of two LN nanogratings. In figure 1(d), two breakpoints occurred at \( |\delta| = 0, 0.2, 0.4, 0.6, 0.8 \) and 1 in a wavelength ranging from 670 to 720 nm; a narrow Fano-shape peak that vanishes when the LN nanogratings were symmetric was observed.

The narrow resonances located in the reflection spectra were fitted to a Fano line shape expressed as [28]

\[
R = R_0 + 1 - A \left( \frac{q + \omega - \omega_0}{\gamma} \right)^2 \frac{1}{1 + \left( \frac{\omega - \omega_0}{\gamma} \right)^2}. \tag{6}
\]

Here, \( \omega_0 \) is the central resonant frequency, \( R_0 \) the basic reflection at \( \omega_0 \), \( q \) the asymmetric factor of Fano resonance, \( A \) a real constant, and \( \gamma \) the damping rate of the Fano resonance. The Q-factor estimated in this method are represented as violet spheres in figure 2(a) (see S2 in the supporting information for more details). For confirmation, the Q-factor was calculated as \( \text{Re}(\varepsilon)/2\text{Im}(\varepsilon) \) in COMSOL Multiphysics, where \( \varepsilon \) is eigenfrequency of quasi-BICs (olive line in figure 2(a)). As shown in figure 2(a), the Q-factor approaches infinity when the \( \delta \) is zero, i.e. a transition from quasi-BICs to BICs. In addition, when \( \delta \) is approximately zero, the local resonance modes have ultranarrow resonance widths and these modes are known as quasi-BICs. Figure 2(b) shows the local field distribution at \( |\delta| = 1, 0.8, 0.6 \) and 0.4; it is indicated that strong local field confinement will be realized when parameters of optical structure is tuned close to BICs. In this case, the Q factor remain high, i.e. up to \( 10^4 \) but optical energy will leak from the waveguide. Hence, light–matter interactions in nonlinear optics or the observation of embedded striking physical phenomena must be enhanced.

### 3. Enhancing SH response assisted quasi-BIC

According to the previous studies [29–31], strong local optical field confinement is crucial for improving nonlinear responses in nanophotonic structures. In this study, we first investigated the SHG of the LNGW structure at asymmetry factor \( \delta = 0.2 \) via numerical simulation using Lumerical FDTD solution for simplicity. Additionally, the case of \( \delta = 0.1 \) was considered for comparison. Based on previous studies [32–35], the second-order nonzero susceptibilities of LN are \( d_{33} = 27 \text{ pm V}^{-1} \), \( d_{31} = 4.6 \text{ pm V}^{-1} \), \( d_{22} = 3 \text{ pm V}^{-1} \). In the SHG simulations, the pulse duration and the repetition rate of fundamental pump light source were set as 200 fs, and 80 MHz, respectively. Based on the discussion in section 2, ultrahigh-Q resonances spawned by BICs or quasi-BICs remain at FW \( \sim 700 \text{ nm} \); however, in SH frequency, they will evolve into low-Q resonances with strong radiation in free space. Therefore, both reflected- and transmitted SH power are calculated using the field-power monitor via the integration of Poynting vector [36] under the TE-polarized pump source with an incident angle of \( 5^\circ \) (see S3–S5 in the supporting information for more details). Figure 3 shows the calculated SHG characteristics of LNGW structure. Figure 3(a) shows a comparison of SHG response between LNGW structure (red line) and LN thin film (olive...
Based on the same fundamental pump light source, the LNGW structure indicated an SH intensity enhancement approximately $10^4$, which was higher compared with that of the LN thin-film with the same thickness, owing to strong local field confinement. The fundamental excitation wavelength was at $\sim 690$ nm (SHG signal $\sim 345$ nm) with a peak intensity of $1.33$ GW cm$^{-2}$, corresponding to an amplitude of $1 \times 10^9$ V m$^{-1}$.

For practical application, power dependence between FW and SHG should be investigated. The fundamental excitation power ranges from 0.002 to 0.22 mW, corresponding to peak intensity from 0.12 to 1.33 GW cm$^{-2}$.

We plotted the SHG power as a function of the average power of excited light from 0.002 to 0.22 mW, which corresponded to peak intensity ranging from 0.12 to 1.33 GW cm$^{-2}$, as shown in figure 3(b). Notably, the SHG power indicated a squared relationship with the increasing excited average pump power (in the left inset), showing a linear trend with a slope of $\sim 2$ in the log plot. The results are consistent with the following equation [13, 34]

$$\log(P_{\text{SH\ avg}}^{\text{SH}}) \propto 2 \log(P_{\text{FW\ avg}}^{\text{FW}}).$$

Here, $P_{\text{SH\ avg}}^{\text{SH}}$ is the SH output power, and $P_{\text{FW\ avg}}^{\text{FW}}$ is the power of the excited source. The total SHG power is divided by the FW power to define the conversion efficiency, as follows:

$$\eta = \frac{P_{\text{SH\ avg}}^{\text{SH}}}{P_{\text{FW\ avg}}^{\text{FW}}}$$

Our calculate the SH conversion efficiency showed a maximum value of $1.53 \times 10^{-5}$ with an averaged excited power of 0.22 mW, i.e. a peak intensity of 1.33 GW cm$^{-2}$ when the asymmetry factor $|\delta| = 0.2$ as shown in the right inset of figure 3(b). However, to characterize the intrinsic SHG conversion efficiency, we theoretically quantified the SHG process following the figure-of-merit (FOM) $\zeta$ expressed in

- **Figure 3.** Characteristics of second harmonic wave generated by LNGW optical structure at dissymmetric factor $|\delta| = 0.1$. (a) Normalized amplitude of SHG signal generated from LNGW (red line) and undersigned LN thin-film (olive line), respectively. All the excitation intensity is $\sim 1.33$ GW cm$^{-2}$ with a wavelength of $\sim 690$ nm. (b) Log-plot power dependence for the SHG signals of LNGW structure under the illumination from 0.002 to 0.22 mW with a fitting slope $1.9994$. The inset at right corner shows that SHG conversion efficiency increases as well as average pump power increases. When the peak intensity of pump light is 1.33 GW cm$^{-2}$, corresponding to average power of 0.22 mW, the SHG conversion efficiency reaches to $1.53 \times 10^{-5}$. (c) Normalized optical field distribution at fundamental wavelength of 690 nm and second harmonic wavelength of 345 nm with $|\delta| = 0.1$. (d) SHG conversion efficiency ($|\delta| = 0.1$, marked by red dashed circle) will greatly boost extremely close to BICs marked red dashed line.
In conclusion, we introduced the breaking symmetry of LN nanogratings and a waveguide optical structure to realize an
In particular, our calculation showed that the SH conversion efficiency achieved was 8.13 × 10⁻⁶ when the optical structure has an asymmetric factor δ = 0.2, whereas the FOM was 1.08 × 10⁻⁵ W⁻¹. Based on table 1, our proposed structure performed well in improving SH conversion efficiency. The nonlinear wave conversion efficiency can be improved to a higher level when the asymmetric factor is approximately zero. When it is zero, e.g. at BICs, no optical energy can coupled coupled into the SH field distribution in dielectric gratings

| Structure type                      | Pump power/peak intensity | EUV FOM | €FOM | BICs | LNW | SH | BICs |
|-------------------------------------|---------------------------|--------|-----|------|-----|----|------|
| Monolithic LN metasurface [37]      | 1 GW cm⁻²                 | 5 × 10⁻⁵ | —   | —    | —   | 5.8 × 10⁻⁹ | —    |
| LN nanodisk on Al substrate [25]    | 5.31 GW cm⁻²              | 1.153 × 10⁻⁵ | 7.68 × 10⁻⁷ | —    | —   | 5.8 × 10⁻⁹ | —    |
| LN nanodisk on hyperbolic metamaterial [24] | 5.31 GW cm⁻²              | 5.14 × 10⁻⁵ | 1.23 × 10⁻⁶ | —    | —   | 5.8 × 10⁻⁹ | —    |
| Periodic LN nanoparamid [38]        | 4.3 GW cm⁻²               | ~10⁻⁶ | —   | —    | —   | —   | —    |
| LN nanopillar grating [39]          | 1 GW cm⁻²                 | 5 × 10⁻⁶  | 5.8 × 10⁻⁹ | —    | —   | 5.8 × 10⁻⁹ | —    |
| LNGW structure (this work)          | 0.22 mW (1.33 GW cm⁻²)    | 8.13 × 10⁻⁸ | 1.08 × 10⁻⁵ | —    | —   | —   | —    |

Here, $P_{peak}^{SH}$ and $P_{peak}^{FW}$ are the peak power of SH and FW, respectively. This FOM is independent of the power of pump source and provides a consistent and comparable parameter with other structures [35, 36]. Hence, our calculated FOM showed a maximum value of 9.18 × 10⁻⁶ W⁻¹ at δ = 0.2 with pump peak intensity of 1.33 GW cm⁻². Based on the linear and nonlinear local field distribution in figure 3(c), this result proves that the excited SH intensity has not reached saturation yet when the asymmetric factor is 0.2 (i.e. quasi-BICs). However, based on the discussion above, the transition from BICs into quasi-BICs results in the decay of resonances leading to optical radiation emitting in free space, which has affect the light–matter interaction in nonlinear material. To confirm this speculation, we determined the relationship between SH conversion efficiency and structural asymmetric factor, as shown in figure 3(d). The maximum SH conversion efficiency achieved was 8.13 × 10⁻⁵ in the case where the asymmetric factor $|δ| = 0.1$, which is five times higher than that of the case $|δ| = 0.2$, whereas the FOM was 1.08 × 10⁻⁵ W⁻¹. Based on table 1, our proposed structure performed well in improving SH conversion efficiency. The nonlinear wave conversion efficiency can be improved to a higher level when the asymmetric factor is approximately zero. When it is zero, e.g. at BICs, no optical energy can coupled coupled into the LNGW and hence no light–matter interaction exist. Therefore, we discuss only case where the asymmetric factor $|δ| = 0$

Table 1. Comparison of SHG conversion efficiency at bound state in the continuum.

| Structure type                      | Pump power/peak intensity | EUV FOM | €FOM | BICs | LNW | SH | BICs |
|-------------------------------------|---------------------------|--------|-----|------|-----|----|------|
| Monolithic LN metasurface [37]      | 1 GW cm⁻²                 | 5 × 10⁻⁵ | —   | —    | —   | 5.8 × 10⁻⁹ | —    |
| LN nanodisk on Al substrate [25]    | 5.31 GW cm⁻²              | 1.153 × 10⁻⁵ | 7.68 × 10⁻⁷ | —    | —   | 5.8 × 10⁻⁹ | —    |
| LN nanodisk on hyperbolic metamaterial [24] | 5.31 GW cm⁻²              | 5.14 × 10⁻⁵ | 1.23 × 10⁻⁶ | —    | —   | 5.8 × 10⁻⁹ | —    |
| Periodic LN nanoparamid [38]        | 4.3 GW cm⁻²               | ~10⁻⁶ | —   | —    | —   | —   | —    |
| LN nanopillar grating [39]          | 1 GW cm⁻²                 | 5 × 10⁻⁶  | 5.8 × 10⁻⁹ | —    | —   | 5.8 × 10⁻⁹ | —    |
| LNGW structure (this work)          | 0.22 mW (1.33 GW cm⁻²)    | 8.13 × 10⁻⁸ | 1.08 × 10⁻⁵ | —    | —   | —   | —    |

Here, $P_{peak}^{SH}$ and $P_{peak}^{FW}$ are the peak power of SH and FW, respectively. This FOM is independent of the power of pump source and provides a consistent and comparable parameter with other structures [35, 36]. Hence, our calculated FOM showed a maximum value of 9.18 × 10⁻⁶ W⁻¹ at $δ = 0.2$ with pump peak intensity of 1.33 GW cm⁻². Based on the linear and nonlinear local field distribution in figure 3(c), this result proves that the excited SH intensity has not reached saturation yet when the asymmetric factor is 0.2 (i.e. quasi-BICs). However, based on the discussion above, the transition from BICs into quasi-BICs results in the decay of resonances leading to optical radiation emitting in free space, which has affect the light–matter interaction in nonlinear material. To confirm this speculation, we determined the relationship between SH conversion efficiency and structural asymmetric factor, as shown in figure 3(d). The maximum SH conversion efficiency achieved was 8.13 × 10⁻⁵ in the case where the asymmetric factor $|δ| = 0.1$, which is five times higher than that of the case $|δ| = 0.2$, whereas the FOM was 1.08 × 10⁻⁵ W⁻¹. Based on table 1, our proposed structure performed well in improving SH conversion efficiency. The nonlinear wave conversion efficiency can be improved to a higher level when the asymmetric factor is approximately zero. When it is zero, e.g. at BICs, no optical energy can coupled coupled into the LNGW and hence no light–matter interaction exist. Therefore, we discuss only case where the asymmetric factor $|δ| = 0$

4. Conclusion

In conclusion, we introduced the breaking symmetry of LN nanogratings and a waveguide optical structure to realize an ultrahigh-Q factor via engineering quasi-BIC to realize strong local field enhancement. An SH intensity enhancement of 10², which was higher compared with that of an undersigned LN thin film, and a conversion efficiency of up to $1.5 \times 10^{-5}$ with a pumping intensity of 1.33 GW cm⁻², was obtained when the optical structure has an asymmetric factor $|δ|$ of 0.2. In particular, our calculation showed that the SH conversion efficiency exceeded $8.13 \times 10^{-5}$ at $|δ| = 0.1$. Therefore, we predict that a significant SH response will be realized when the asymmetric factor $δ$ drift approaches zero, i.e. from quasi-BICs to BICs. Although our study was based on theoretical and numerical simulations, the results can be used to promote the widespread applications of lithium niobate in nonlinear integrated nanophotonics.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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