Resonantly tunable second harmonic generation from lithium niobate metasurfaces

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Second harmonic generation (SHG) is a coherent nonlinear phenomenon that plays an important role in laser color conversion. However, the SHG efficiency from natural materials is low and uncontrollable. As a result, large material volume and intense laser are needed, which hinder the miniaturization of SH devices. We experimentally demonstrate efficient SHG from nonlinear lithium niobate (LN) metasurfaces. We find that as a result of the resonant local field enhancement, the SHG at resonance is about two orders of magnitude stronger than the off-resonant situations. Furthermore, we manage to tune the SHG efficiency dispersion by changing the structural parameters of the metasurfaces. Our results would pave a way for developing novel compact coherent light sources for biosensing and optical communications, et al.

Keywords: Lithium niobate, Metasurface, Second harmonic generation, Resonance

Nonlinear optical effects, such as optical harmonic generation, form the bases for extending spectral range of light source. Motivated by goals of developing ultracompact optical devices, nonlinear nanophotonics has become a prominent research area in recent decades.1–3 The nonlinear properties of nanostructures rely on strong light-matter interaction in the subwavelength structures due to the resonant enhanced localized electric fields. The famous examples include metallic plasmonic and all-dielectric metasurfaces. In plasmonic nanostructures, due to the high losses of metals, light fails to penetrate deeply, and the enhanced nonlinear response is dominated by the electric field confinement close to the surface.4–9 On the other hand, dielectric metasurfaces show great promise for enhancing nonlinear optical processes due to their larger mode volumes. And due to the very low intrinsic losses, the dielectric nanostructures can sustain much higher optical powers and ultimately provide orders of magnitude higher frequency conversion efficiency in comparison with their plasmonic counterparts.10–12

In the recent years, dielectric semiconductor nonlinear metasurfaces made of, such as Si,13–15 Ge,16 GaAs,17,18 GaP,19,20 have been reported. However, those semiconductor devices show relatively narrow band gap, and the fundamental frequency (FF) cannot enter the visible range.

Lithium niobate (LiNbO3, LN) is an important nonlinear crystal. It shows a large band gap of 4 eV, and is transparent over a spectral range of violet to mid-infrared. In the meanwhile, LN provides a large quadratic nonlinear susceptibility.22,23 The second harmonic generation (SHG) from LN nanostructures have been a subject under continuous research, including LN nanowires,24 photonic crystals,25–28 waveguides,29–31 hybrid LN-plasmonic nanopillars,32 nanocubes33,34 and most recently micro-ring35 or micro-disk36,37 resonators. In recent years, the design of LN nonlinear metasurfaces have been theoretically studied.38–40 However, no experiments to date have realized the LN nonlinear metasurfaces showing resonantly tunable SHG properties.

In this paper, we experimentally demonstrate resonant tunable SHG from nonlinear LN metasurfaces. We fabricate subwavelength nanograting metasurfaces based on LN films. Due to the distinct resonances in the metasurfaces, the enhanced SHG is observed. And the spectral properties of the SHG enhancement are proved to be tunable by changing the geometric parameters. Our results would pave a way for novel nanoscale LN nonlinear light sources applied in next generation photonic devices, including efficient nonlinear holograms and quantum-light sources.

In our experiment, a LNOI wafer (NANOLN, Jinan Jingzheng Co., Ltd) with a x-cut 220 nm thick LN film on top of a silica insulator was adopted. And we fabricated the nanograting metasurfaces with different periods (D) using focused ion beam. The duty cycle (percentage of LN ridge width d over one period) of each structure remains 62.5%. The direction of the grating is perpendicular to the optical axis of the LN film (x-axis). The representative scanning electron microscopy (SEM) image of nanograting metasurface is shown in Fig. 1(b). We give the simulated transmission of metasurfaces in Fig. 1(c,d) under normal incidence for polarization perpendicular (along x-direction) and parallel (y) to the ridges for structures with D = 500, 600, and 700 nm.
The SHG of the LN metasurface is related to the nonlinear polarization $P^{(2)}$ induced by the FF, which could be calculated using the second-order nonlinear susceptibility $\chi^{(2)}$ through $P_i^{(2)} = e_0 \chi_{ijk}^{(2)} E_j E_k$ (indices $i, j, k$ refer to the Cartesian components of the fields). The $P_i^{(2)}$ possesses the doubled frequency of the FF wave and acts as the radiation source for the SH polarized along the $z$-direction. The tensor nature of $\chi^{(2)}$ implies that one FF polarization component could interact with multiple $\chi_{ijk}^{(2)}$ elements and may lead to SH with multiple polarization components. For the SH propagating along $z$-direction, only $P^{(2)}_z$ along $x$- and $y$-directions play roles $P^{(2)} = \sqrt{P^{(2)}_x^2 + P^{(2)}_y^2}$. And considering that all the polarization components exists in the metasurface layer as a result of scattering interaction of light inside, the component of $P^{(2)}$ can be expressed as

$$
\begin{align}
P^{(2)}_x &= \chi_{ecx}^{(2)} E^2_x + \chi_{xoe}^{(2)} E^2_y + \chi_{oxc}^{(2)} E^2_z \\
P^{(2)}_y &= -\chi_{xoe}^{(2)} E^2_x + \chi_{oxc}^{(2)} E^2_y + 2\chi_{coo}^{(2)} E_y E_x \\
\end{align}
$$

The nonzero elements of $\chi^{(2)}$ are related to the crystal symmetry of the LN. And based on the $\chi^{(2)}$ values measured in Ref. 32, we numerically predict the SH conversion efficiency of the LN metasurfaces, as shown in Fig. 2(a) and (b). For $x$-polarized FF, the SH conversion efficiency has strong resonance enhancement at 385 nm, 465 nm, and 510 nm for $D = 500, 600, 700$ nm, respectively. For $y$-polarized FF, the SH conversion efficiency has strong resonance enhancement at 360 nm, 435 nm, and 505 nm. This shows the spectral dispersion of SHG efficiency are highly sensitive to the resonance of the metasurface, and could be tuned efficiently by geometric parameter adjustment.

In our experiments, we use a commercial microscopic spectrometer (IdeaOptics Technologies) to measure the transmission spectra of LN metasurfaces with different periods. As shown in Fig. 3(a, b), measured transmission spectra show similar trends as the simulation results. However, due to the fabrication infection, the experimental transmission spectra are blue shifted compared with the simulation results. For the SHG measurements, we use a tunable linear polarized femtosecond laser (Maitai, Spectra physics) as FF light to excite LN samples. The FF beam is focused to about $5 \mu m$ in radius with a peak intensity of about $3.8$ GW/cm$^2$. And the generated SH is collected by another $\times20$ UV objective (0.4 N.A.). The FF light is filtered out from the SH by BG40 colored glass filters. The SH signal is recorded using a fiber coupled spectrometer. We define the SH conversion efficiency as $\eta = \frac{P_{SH}}{P_{FF}}$. Figure 3(c) and (d) show the SH conversion efficiency of the LN metasurfaces under two orthogonally polarized FF. For $x$-polarized FF, the SHG efficiency is boosted by 133 times at the resonance compared with the

![Fig. 1. Linear spectral properties of LN nanograting metasurfaces.](image1)

![Fig. 2. Numerical simulated SH conversion efficiency.](image2)
FIG. 3. Experimental measured transmission spectra and SH conversion efficiency of metasurfaces. (a, b) The experimental spectra for the LN metasurfaces for orthogonal polarizations. (c, d) The experimental SH conversion efficiency for the LN metasurfaces under orthogonal FF polarizations. The red, green, and blue lines correspond to $D$ of 500, 600, and 700 nm.

non-resonant wavelength. By increasing the grating period constant ($D$), the resonance peaks of the SH conversion efficiency for LN metasurfaces suffer monotonically red-shift. For the $y$-polarized FF, the SH conversion efficiency of the metasurface is lower, implying that the involved second order nonlinear susceptibility elements are smaller and weaker local field enhancement, as shown in Fig. 3(d). Similarly, we can tune the resonance peaks for the SH conversion efficiency of LN metasurfaces to the red spectral range simply by increasing the grating period constant $D$.

In conclusion, we experimentally realize the efficient SHG from LN metasurfaces. Due to the local field enhancement, the SHG from LN metasurface is significantly enhanced. And we manage to tune the SHG efficiency spectra by changing the geometric parameters of the LN metasurfaces. Experimental results are reasonably consistent with the numerical predictions. Such a platform can also be applied for other nonlinear processes such as four-wave mixing, sum frequency generation, parametric down conversion, and so on, which may find wide applications in biosensing, optical communications, and display, and so on.

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