Broadband highly efficient nonlinear optical processes in on-chip integrated lithium niobate microdisk resonators of Q-factor above $10^8$

Renhong Gao$^{1,2,10}$, Haisu Zhang$^{3,4,10}$, Fang Bo$^1$, Wei Fang$^6$, Zhenzhong Hao$^7$, Ni Yao$^6$, Jintian Lin$^{1,2,*}$, Jianglin Guan$^{3,4}$, Li Deng$^{3,4}$, Min Wang$^{3,4}$, Lingling Qiao$^{1}$ and Ya Cheng$^{1,2,3,4,7,8,9,*}$

1. State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics (SIOM), Chinese Academy of Sciences (CAS), Shanghai 201800, People’s Republic of China
2. University of Chinese Academy of Sciences, Beijing 100049, People’s Republic of China
3. XXL—The Extreme Optoelectromechanics Laboratory, School of Physics and Electronic Science, East China Normal University, Shanghai 200241, People’s Republic of China
4. State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai 200062, People’s Republic of China
5. The MOE Key Laboratory of Weak Light Nonlinear Photonics, TEDA Applied Physics Institute and School of Physics, Nankai University, Tianjin 300457, People’s Republic of China
6. State Key Laboratory of Modern Optical Instrumentation, College of Optical Science and Engineering, Zhejiang University, Hangzhou 310027, People’s Republic of China
7. Collaborative Innovation Center of Extreme Optics, Shanxi University, Taiyuan 030006, People’s Republic of China
8. Collaborative Innovation Center of Light Manipulations and Applications, Shandong Normal University, Jinan 250358, People’s Republic of China
9. Shanghai Research Center for Quantum Sciences, Shanghai 201315, People’s Republic of China
10. Authors to whom any correspondence should be addressed.
11. These authors contributed equally to the work.

E-mail: jintianlin@siom.ac.cn and ya.cheng@siom.ac.cn

Keywords: microcavities, lithium niobate, nonlinear optics

Abstract

Microresonators of ultrahigh quality ($Q$) factors represent a crucial type of photonic devices aiming at ultra-high spectral resolution, ultra-high sensitivity to the environmental perturbations, and efficient nonlinear wavelength conversions at low threshold pump powers. Lithium niobate on insulator (LNOI) microdisks of high $Q$ factors are particularly attractive due to its large second-order nonlinear coefficient and strong electro-optic property. In this letter, we break through the long standing bottleneck in achieving the $Q$ factors of LNOI microresonators beyond $10^8$, which approaches the intrinsic material absorption limit of lithium niobate (LN). The ultra-high $Q$ factors give rise to a rich family of nonlinear optical phenomena from optical parametric oscillation (OPO) to harmonics generation with unprecedented characteristics including ultra-low pump threshold, high wavelength conversion efficiency, and ultra-broad operation bandwidth. Specifically, the threshold of OPO is measured to be only 19.6 $\mu$W, and the absolute conversion efficiency observed in the second harmonic generation reaches 23%. The record-breaking performances of the on-chip ultra-high $Q$ LNOI microresonators will have profound implication for both photonic research and industry.

1. Introduction

Optical microresonators have long been playing a crucial role in a broad range of fields including nonlinear optics [1, 2], cavity optomechanics [3, 4], quantum information [5, 6] and lightwave communications [7]. Generally, these applications are highly in favor of the sustained storage of light for a long duration of time in the microresonator, which is characterized by its optical quality ($Q$) factor. The highest $Q$ factors to date...
have been demonstrated with whispering gallery mode (WGM) microresonators where the loaded light is well confined by the continuous total internal reflection at the resonator interface, necessitating (1) the use of high-transparency dielectrics with ultra-low intrinsic absorption, and (2) the capability of creating highly smooth dielectric interfaces in the quest of ultrahigh-Q (UHQ) WGM microresonators [8–14]. Chip-compatible silica WGM microresonators have been widely employed due to the ease of fabrication via dry etching and heat reflow, featuring ultra-smooth interface and thus UHQ factors above $10^9$ [9, 10]. Dielectric crystalline microresonators with high second-order $\chi^{(2)}$ nonlinear coefficients are preferable in diverse applications, though the UHQ performance of $\chi^{(2)}$-microresonators are only attainable by mechanically polishing bulk crystals into the thickness of several tens of micron [11–13]. Such an UHQ factor has been out of reach in chip-scale $\chi^{(2)}$-microresonator so far mainly due to the fragility of crystals and difficulty in dry etching [15, 16].

To overcome the challenge, thin-film crystals have been developed via ion slicing using ion implantation, relaxing the etching depth requirement and transforming the geometry of microresonators from three-dimensional structures to 2D structures [15, 16]. Therefore, dry etching techniques, such as reactive ion etching [7, 17–19], focused ion beam milling [20, 21], and photolithography assisted chemo-mechanical etching (PLACE) [22, 23], have been utilized successfully for fabrication of on-chip crystalline microresonators with high $Q$ factors on the order of $10^7$, which is still one order of magnitude away from the $Q$ factors determined by the intrinsic material absorption (i.e. for lithium niobate (LN), $2 \times 10^8$ at 1550 nm wavelength band [11]). Specifically, it has been confirmed that the surface roughness achieved with the PLACE fabrication technique is around 0.1 nm, indicating that the $Q$ factors limited to $10^7$ is a result of material absorption property instead of the scattering loss left behind by the fabrication process [23]. Notably, the internal crystal lattice damage induced by the ion slicing fabrication of the LNOI has long been recognized which represents the ultimate limit of the $Q$ factors achievable in the commercially available LNOI [24]. A fundamentally different fabrication strategy has to be developed to realize on-chip LN microresonators with $Q$ factors beyond $10^8$.

In this work, for the first time to the best of our knowledge, we experimentally demonstrate on-chip LN microresonators with $Q$ factors above $10^8$, i.e. approaching the intrinsic material absorption limit. Such an UHQ factor is ensured by ion-free preparation of LN thin film combined with chemo-mechanical etching for nanostructuring of LN, together of which essentially alleviate the lattice damage and interface roughness during the ion slicing as well as the subsequent chemo-mechanical etching processes [15, 16]. Furthermore, the thickness of the microresonator is only 600 nm, which is $\sim 2$ orders of magnitude thinner than the counterparts produced with bulk crystal [11–13], leading to ultra-small mode volumes. Microdisks of sizes ranging from several tens of micron to 3 mm have been demonstrated using our fabrication technique. The combination of UHQ factors, small mode volumes and flexible sizes in single microresonators is of vital importance to push forward the frontier of nonlinear optics such as optical parametric oscillation (OPO) [25, 26], single photon nonlinearity [27], $\chi^{(2)}$ frequency comb generation [28], quantum light sources [5, 29–32], and so on. For these applications, only weak pump light is often allowed, therefore the conversion efficiency is intrinsically low. The conversion efficiency can be boosted by promoting the ratio between the $Q$ factor and mode volume, justifying the necessities of using UHQ microresonators of small mode volumes. In the current primary investigation, we have observed broadband frequency conversions including second harmonic generation (SHG), third harmonic generation (THG), fourth harmonic generation (FHG) as well as OPO, all generated in the same LN microresonator with ultrahigh conversion efficiencies.

2. Experiment

2.1. Fabrication of the microresonators

The LN microdisk resonators were fabricated on the LN thin film wafer prepared by crystal cutting, polishing and bonding. Femtosecond laser direct writing and chemo-mechanical polishing were then utilized in sequence for patterning the LN thin film into free-standing microdisk resonators [details in the fabrication process can be found in the supplemental materials (https://stacks.iop.org/NJP/23/123027/mmedia)]. As a matter of fact, recent works on LN photonic integrated components and circuits exclusively use the thin film LN on insulator (LNOI) as the material platform [15, 16]. Here, we chose to use the mechanically thinned LN thin film but not ion sliced LNOI for the reason as follow [33]. LNOI is obtained by the ion-slicing of submicron-thickness films from the single-crystalline LN bulk, where the implantation layer of helium ions (He$^+$) defines the cleavage plane in the monocristalline LN [15, 16]. The crystal quality of the ion-sliced LN thin film has been examined using Rutherford backscattering, revealing an inferior quality as compared to the LN bulk crystal [24]. Moreover, the quality of the ion-sliced film can
only be partially recovered to the pristine bulk crystal after high-temperature annealing above 1000 °C [24]. In practical photonic applications, the LN thin film is typically bonded on a silica buffer layer for high refractive index contrast and optical isolation. Unfortunately, the annealing temperature at 1000 °C will spoil the bonding between the LN thin film and the underneath silica layer [16, 34]. Therefore, a relatively low temperature of 500 °C is chosen for annealing, leading to inferior optical property of LNOI to the bulk LN crystal. This problem becomes more severe with the development of fabrication techniques to allow for generation of ultra-smooth surface finish like PLACE, because otherwise the $Q$ factor is dominated by the scattering loss at the sidewall surface but not the weak intrinsic material absorption.

The scanning-electron-microscopy (SEM) image of the fabricated LN microdisk resonator with a diameter of 1030 µm is presented in figure 1(a), showing an ultra-smooth surface with an average surface roughness less than 0.5 nm [22, 23]. The wedge angle of the microdisk sidewall is 8°, rendering the thickness of the microdisk resonator being varied from the submicron-scale at the edge to 3.5 µm near the center, as shown in figure 1(b). The fundamental WGM is simulated and plotted in the top inset of figure 1(b), with the center of the mode located 12 µm horizontally from the microdisk edge where the local LN thin film is only 600 nm thick, leading to a mode volume two orders of magnitude smaller than previously reported LN bulk resonators of the same diameter [11–13]. The distance between the SiO$_2$ pedestal and the microdisk edge was chosen to be 100 µm (see optical micrograph in the bottom inset of figure 1(b)) by controlling the HF acid etching time, which is sufficient to protect the WGMs from the scattering loss induced by the rough SiO$_2$ pedestal. The UHQ factors combined with the small mode volumes of the fabricated micro-resonators are ideal for efficient frequency conversions as shown below.

2.2. Characterization of the $Q$ factors

The $Q$-factor of the LN resonator was first measured by sweeping the wavelength of a tunable laser (linewidth < 200 kHz, Model: TLB 6728, New focus Inc.) injected into the resonator using a tapered fiber with a diameter of 0.9 µm. The relative position between the tapered fiber and the resonator was finely
adjusted to fulfill the critical coupling condition using an XYZ piezo stage with a positioning resolution of 20 nm, which was monitored in real time by a charge coupled device (CCD) mounted above an objective lens with numerical aperture of 0.3. A low input laser power of only 5 μW was used in the measurement to avoid the thermo-optic and nonlinear optical effects. The signal was coupled out of the resonator by the same tapered fiber, sent into a high-speed photo detector with a bandwidth of 30 GHz, and real-time recorded with an oscilloscope. The measured transmission spectrum as plotted in figure 1(c) was fitted with a Lorentzian-shaped resonance profile centered at ~1551.52 nm as shown in the inset of figure 1(d), indicating a loaded Q factor $Q_L$ of $7.5 \times 10^7$ and a transmission $T_F$ of 6%. The intrinsic Q factor $Q_i$ was determined to be $1.20 \times 10^8$ by the equation $Q_i = 2 \times Q_L/(1 + T_F)$. Afterwards, a cavity ring-down measurement was conducted for measuring the Q factor and compared with the Q factor determined by the transmission spectrum measurement. In this way, the uncertainty in the precise measurement of such high Q factors can be safely excluded. The ring-down measurement was carried out by repeatedly scanning the laser with a high-speed Mach–Zehnder modulator into the targeted resonance, and the result is shown in figure 1(d). The retrieved photon lifetime is 64.3 ns, from which the intrinsic Q factor is derived to be $1.23 \times 10^8$. The Lorentzian fitting and the cavity ring-down measurement agree very well with each other. Importantly, the mode volume is only $5.2 \times 10^4 \mu$m$^3$ in the UHQ microresonator. The intrinsic Q factor at 777.6 nm wavelength is measured to be $7.75 \times 10^7$, the detail of which is provided in the supplemental materials. Some smaller microdisks with diameters of 130 μm and 240 μm were also fabricated, both showing intrinsic Q factors around $9.9 \times 10^5$. (The details of the results and the statistics on the Q factors of microdisks with different diameters from 100 μm to 3 mm are provided in the supplemental materials). Since the bending losses are exponentially inverse proportional to the diameters of the resonators, the tiny variance ($<30\%$) of the Q-factors for microdisk resonators with diameters from ~1 mm to ~100 μm indicates that the measured Q factors in the UHQ microdisks are solely determined by the intrinsic material absorption.

2.3. Study of the nonlinear optical processes

Frequency conversions in the UHQ LN microdisk resonators were investigated to explore the new regime of nonlinear interactions facilitated by the UHQ factors and small mode volumes [11–13, 25]. It is well known that phase-matching between the pertinent waves underpins efficient and low-threshold nonlinear frequency conversions in WGM-resonators [2]. Modal phase-matching (MPM) by manipulating geometric dispersions and quasi phase-matching (QPM) are frequently employed for frequency conversions in on-chip micro-resonators [35–40]. However, MPM suffers from a relatively low second order nonlinear coefficient, whereas QPM suffers from extra complexity in the fabrication process as well as the spectral fluctuation and optical loss caused by the imperfection in the domain poling [40–43].

We chose another QPM scheme for our nonlinear frequency conversion experiments, in which X-cut LN microdisk resonators were employed for achieving the natural quasi phase-matching (NQPM) as revealed recently [44]. The inherent cyclic phase-matching for transverse-electric (TE) WGMs circulating along the circumference of the $\chi^{(2)}$ micro-resonator with in-plane optical axis naturally facilitates broadband and efficient frequency conversions without resting on specific modal dispersion and nonlinearity patterning. By controlling the polarization of the injected pump light to excite the TE mode in the X-cut LN resonator with a diameter of 1030 μm, strong SHG and THG signals were observed when the wavelength of the pump light was set at 1555.2 nm. The spectrum was recorded by an optical spectrum analyzer (OSA, detected wavelength range of 600–1700 nm) and a fiber spectrometer (detected wavelength range of 300–1000 nm) as shown in figure 2(a). The SHG and THG signals were both TE polarized as well. The conversion efficiency of SHG grows linearly with the in-coupled pump power with a very high normalized conversion efficiency $T$ as high as 23% at a saturation in-coupled pump power $P_{sat}$ of ~0.11 mW, as shown in figure 2(b). The absolute conversion efficiency is also greater than the 15% conversion efficiency of the periodically poled LN (PPLN) microring at similar pump powers [37]. The theoretical value of the absolute conversion efficiency can be calculated by a theoretical modeling considering the pump depletion [37, 45], which is

$$T = \frac{Q_{L,F}}{Q_{C,F}} \frac{Q_{L,SHG}}{Q_{C,SHG}} = 25\%.$$  

(1)

Here, $Q_{L,F}$, $Q_{C,F}$, $Q_{L,SHG}$, $Q_{C,SHG}$ are the loaded (L) Q factors and the coupled (C) Q factors of the fundamental wave (F) mode and the second harmonic (SHG) mode, respectively, which can be experimentally extracted from the transmission spectra. The measured Q factors of the fundamental wave are provided in the supplemental materials. And the coupled Q factor of each mode is calculated by an equation $Q_C = 1/(1/Q_L - 1/Q_i)$. Therefore, the experimental result 23% agrees well with the theoretical prediction 25%. And the saturation in-coupled pump power is determined by $P_{sat} = 167/T_n = 0.85$ mW,
indicating a higher saturation power than the experimental observation. And such a discrepancy is common in WGM LN resonators [11, 13, 37], resulting from the enhanced photorefractive effect [46] with the high in-coupled power, particularly in UHQ microresonators. Besides, the normalized conversion efficiency of THG was measured to be 30%/mW², outperforming the best reported values as well [39, 44, 47], as shown in figure 2(c).

The ultrahigh conversion efficiencies are a result of synergetic contribution from several critical characteristics in the fabricated on-chip X-cut LN microdisk resonator including the UHQ factor, the small mode volume \( V \), the high nonlinear optical coefficient (\( d_{33} \) for LN) and the implementation of NQPM by choosing the proper polarization configuration. Meanwhile, due to the dense spectral mode distribution of the millimeter–diameter resonator, broadband NQPM can be anticipated for a plethora of WGM pairs. To check this hypothesis, the transient output powers of harmonics were detected by continuously tuning the pump wavelength from 1530 nm to 1570 nm with a tuning rate of 0.5 nm s⁻¹, and the output powers of SHG and THG at each pump wavelength are shown in figure 2(d). The spectral resolution of the measurement curves was limited by the power meter of a sampling rate of 10 Hz. Remarkably, both SHG and cascaded THG were observed in the spectrum spanning over the entire telecom C-band with high and relatively stable conversion efficiencies insensitive to the pump wavelength thanks to the choice of the broadband NQPM scheme.

When the in-coupled pump power raised to more than 0.11 mW, higher-order nonlinearities such as Raman scattering and Kerr effect emerged. These processes impeded the power increase in the harmonics generation. Figure 3(a) shows clearly a visible light emission from the resonator, which was captured by a visible CCD camera. The spectrum ranging from 385 to 1700 nm wavelength recorded by the OSA (red line) and the fiber spectrometer (black line) is plotted in figure 3(b), when the in-coupled power was 3.8 mW. Here, FHG from the microdisk without sophisticated dual-periodically polling [39] was also detected at 388.8 nm wavelength with an output power as high as 3.16 \( \mu \)W. Meanwhile, Raman assisted frequency comb generation was observed around the pump wavelength with a comb line spacing of 7.9 THz with Raman shift of 238 cm⁻¹ [48], as shown in the rectangular box II in figure 3(b). Cascaded Raman assisted frequency comb generation was also detected around second harmonic wavelength for the first time, which is shown in the rectangular box I in figure 3(b). Each comb line in box I was generated by sum frequency generation process between the fundamental light and the comb line in box II. There is also a spectrum of cascaded Raman scattering lines [18, 22] recorded in the spectral range from 388 nm to 1700 nm, where the spectrum beyond 1700 nm was unable to measure as limited by the detection range of
Figure 3. (a) Clearly visible optical emission with near infrared pump of 1555.2 nm wavelength. Inset: optical micrograph of the visible emission from the edge of the resonator from the sideview. (b) Spectrum of the emission.

Figure 4. (a) The spectrum of OPO, inset: optical micrograph of the microdisk pumped with 770.4 nm wavelength laser. (b) Power dependence of OPO, showing a threshold pump power of $\sim 19.6 \mu W$ and a gain rate of 20.0%. Inset: the spectrum of the pump laser. (c) Power dependence of the degenerated OPO when pumping at 777.6 nm, showing a threshold pump power of $\sim 20.0 \mu W$ and a gain rate of 19.8%. Inset: the spectrum of the degenerated OPO. (d) The Fourier components of second order nonlinear coefficient $d_{\text{eff}}$, which are $\pm 1942 \, (m^{-1})$ and $\pm 5825 \, (m^{-1})$. Inset: the variation of $d_{\text{eff}}$ of azimuth angle $\theta$.

the OSA (Model: AQ6370D, Yokogawa Inc.). Thus, with only one pump laser operated at 1555.2 nm wavelength, an ultra-broad spectrum from ultraviolet to infrared can be generated with decent conversion efficiencies, indicating the unique nonlinear characteristics of the UHQ LN microresonator.

When the pump laser operating at the telecom wavelengths was replaced with a near-visible tunable laser (765–781 nm) above certain threshold power, broadband non-degenerated OPO signals were observed. Figure 4(a) shows such an OPO spectrum detected by the OSA when being pumped at 770.4 nm wavelength with only 20.6 $\mu W$ in-coupled power. The signal and idler waves at wavelengths of 1517.2 nm and 1565.2 nm were generated thanks to the satisfactory phase matching and triply resonant condition among the signal, idler and pump waves. The detected output power of OPO increases linearly with the
increasing pump power above certain threshold power, as shown in figure 4(b). The threshold power was determined to be only 19.6 μW, which is the lowest among all the OPO investigations in on-chip LN micro-resonators, to the best of our knowledge [26]. The gain rate of OPO was determined to be 20% by linear fitting. Since the NQPM scheme allows the broadband phase matching, the degenerated OPO wave was also detected when the pump wavelength was tuned to 777.6 nm at a slightly higher threshold power of 20.0 μW, as evidenced in figure 4(c).

The versatility of broadband NQPM for a wide range of nonlinear optical processes, including SHG, THG, FHG, and OPO, realized on the same microdisk resonator is quite appealing for further investigations. Previous works have unveiled the indispensable role of the natural ‘poling’ of the effective nonlinearity in X-cut LN microdisk on mitigating the wave-vector mismatch (Δk) which is also oscillating due to the anisotropic refractive indices along the periphery of the microdisk [44, 49]. Specifically, the oscillation of Δk relaxes the stringent requirement by QPM at unique poling period. Besides, NQPM inherently contains four periods of nonlinearity for QPM, and the periods are determined by the diameter of the microdisk which can be easily controlled (see figure 4(d)). Moreover, since the spectral mode density of the microdisk depends on its diameter as well, the millimeter-scale microdisk employed in this work can provide plenty sets of WGMs with suitable wave-vector mismatches covered by the small momentum (long period) of the nonlinearity poling, as corroborated by the simultaneous observations of efficient and broadband frequency conversions arising from various nonlinear processes with distinct phase-matching criteria. It should be noted that although NQPM entails the use of high-order WGMs which reduces the effective nonlinearity, its advantage in the UHQ micro-resonators can easily compensate for this defect by significantly enhanced resonance and achieve ultrahigh efficiencies for multiple nonlinear optical processes.

3. Conclusion

To summarize, the on-chip UHQ factor in excess of 10^8 on pristine LN thin film wafer is experimentally demonstrated for the first time. Broadband SHG, THG, and FHG with ultrahigh normalized conversion efficiencies and low-threshold OPO were observed at the same LN microdisk resonator without introducing domain inversion. The combination of UHQ photonic devices and excellent nonlinear optical property of LN opens promising possibilities for applications ranging from integrated quantum information processing, modern communication to cavity quantum electrodynamics.

Acknowledgments

The work is supported by the National Key R & D Program of China (Grant No. 2019YFA0705000), the NSFC (Grants Nos. 11734099, 62122079, 11874375, 11874154), Key Research Program of Frontier Sciences (QYZDJ-SSWSLH010), Shanghai Municipal Science and Technology Major Project (2019SHZDZX01), Science and Technology Commission of Shanghai Municipality (No. 21DZ1101500), Higher Education Discipline Innovation Project (B07013), and the Youth Innovation Promotion Association of Chinese Academy of Sciences (Grant No. 2020249).

Data availability statement

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

ORCID iDs

Ya Cheng https://orcid.org/0000-0001-8269-5907

References

[1] Zhang X, Cao Q-T, Wang Z, Liu Y-X, Qiu C-W, Yang L, Gong Q and Xiao Y-F 2019 Nat. Photon. 13 21
[2] Lin G, Coillet A and Chembo Y K 2017 Adv. Opt. Photon. 9 828
[3] Shen Z, Zhang Y-L, Chen Y, Zou C-L, Xiao Y-F, Zou X-B, Sun F-W, Guo G-C and Dong C-H 2016 Nat. Photon. 10 657
[4] Kippenberg T J and Vahala K J 2008 Science 321 1172
[5] Ma Z, Chen J-Y, Li Z, Tang C, Su Y M, Fan H and Huang Y-P 2020 Phys. Rev. Lett. 125 263602
[6] Fülöp A et al 2018 Nat. Commun. 9 1598
[7] Guarino A, Poberaj G, Rezzonico D, Degl’Innocenti R and Günter P 2007 Nat. Photon. 1 407
[8] Vahala K J 2003 Nature 424 839

7
