Continuous-wave ultraviolet emission through fourth-harmonic generation in a whispering-gallery resonator

Jeremy Moore, Matthew Tomes, Tal Carmon, and Mona Jarrahi*

Department of Electrical Engineering and Computer Science, University of Michigan, Ann Arbor, 1301 Beal Ave, Ann Arbor, Michigan, 48109, USA
*mjarrahi@umich.edu

Abstract: We experimentally demonstrate continuous-wave ultraviolet emission through forth-harmonic generation in a millimeter-scale lithium niobate whispering-gallery resonator pumped with a telecommunication-compatible infrared source. The whispering-gallery resonator provides four spectral lines at ultraviolet, visible, near-infrared and infrared, which are equally spaced in frequency via the cascaded-harmonic process and span a 2-octave frequency band. Our technique relies on a variable crystal poling and high transverse order of the modes for phase-matching and a resonator quality factor of over 10^7 to allow cascaded-harmonic generation up to the fourth-harmonic at input pump powers as low as 200mW. The compact size of the whispering gallery resonator pumped at telecommunication-compatible infrared wavelengths and the low pump power requirement make our device a promising ultraviolet light source for information storage, microscopy, and chemical analysis.

OCIS codes: (190.0190) Nonlinear optics; (230.0230) Optical devices.

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1. Introduction

Basic physics imposes the condition that the required energy for a conventional laser scales as $1/\lambda^5$, where λ is the laser wavelength [1]. On the other hand, high-harmonic generation is not subject to this limit and therefore allows extending the emission wavelength of a pump laser

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to produce coherent ultraviolet light, unrestricted by the $1/\lambda^5$ relation. However, to date such short-wavelength sources have required very high pump power levels that could generally be achieved only by ultra-short pump pulses [2–6]. Instead of using ultra-short pump pulses, whispering-gallery resonators can be used to provide the high field intensities required for various nonlinear phenomena. This is because high quality factor whispering-gallery resonators enhance the intensity of light continuously in time via multiple recirculations, resulting in large light-matter interaction distances [7–9]. This intensity enhancement mechanism has enabled various nonlinear phenomena, including optomechanical vibrations [10–14], parametric oscillations [15–18], Raman-lasers [19–21], Erbium-lasers [22], Brillouin-lasers [10–13], and continuous-wave second- and third-harmonic generation [23–29].

In this work, we experimentally demonstrate continuous-wave harmonic generation up to the fourth-harmonic, enabled by multiple-recirculation intensity enhancement in a lithium niobate whispering-gallery resonator. This could potentially transform high-harmonic studies from pulsed to continuous-wave. Specifically, we generate continuous-wave near-infrared, visible, and ultraviolet light from a telecommunication-compatible infrared pump through cascaded-harmonic generation in a whispering-gallery resonator at a record-low pump power of 200 mW [2–6]. Further, our millimeter-scale emitter is simple, as the polished lithium niobate resonator comprises both the nonlinear medium and the mode-confining resonator. Finally, a non-uniform poling of lithium niobate [30] and existence of higher order transverse modes [24] provides the required quasi phase-matching between the infrared pump and the corresponding near-infrared, visible, and ultraviolet harmonics.

2. Whispering-gallery resonator design

Resonance calculation for the pump and its corresponding harmonics in a 3mm lithium niobate whispering-gallery resonator is done using finite element method simulation in COMSOL [31] and is shown in Fig. 1 (TM modes are considered for this analysis). The electric field profiles indicate that the infrared pump and the corresponding 2nd (near-infrared), 3rd (visible), and 4th (ultraviolet) harmonic modes are confined inside the nonlinear lithium niobate medium with a considerable mode overlap. The modes are expected to resonate at integer multiples of the pump frequency, in order to conserve energy as required by coupled-mode theory [32]. However, structural and material dispersion cause the modes' propagation constants to be scaled differently as a function of frequency. This implies that momentum is not *a priori* conserved for the high-harmonic processes in the bare resonator. We compensate for the deviation from momentum conservation by quasi phase-matching, as shown below.

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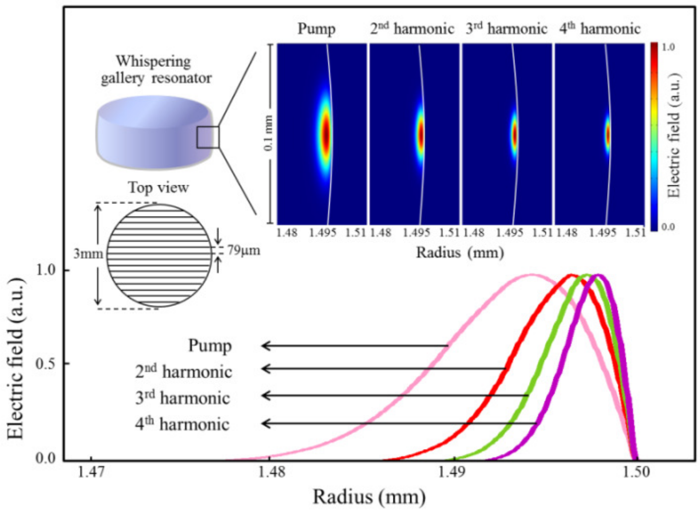

Fig. 1. Radial electric field profiles for a 1550 nm pump and the corresponding 2nd, 3rd, and 4th harmonics, which are numerically calculated for the 3mm lithium niobate whispering-gallery resonator (TM modes).

Quasi phase-matching to conserve momentum for the harmonics and pump is achieved by periodic poling of lithium niobate [30,33] and facilitated by a combination of high order transverse resonance modes [24]. For momentum conservation, the optimum poling period for each three-photon interaction harmonic generation process is given by $\Lambda = \left(n_1/\lambda_1 + n_2/\lambda_2 - n_3/\lambda_3\right)^{-1}$, where λ_1 and λ_2 are the wavelengths of the input photons, λ_3 is the wavelength of the generated photon, n_1 and n_2 are the mode index of the input photons and n_3 is the mode index of generated photon. A major challenge in quasi phase-matching is that different poling periods are required to compensate for the momentum mismatch of different harmonics. For example, the optimal poling periods for 2nd, 3rd, and 4th harmonic generation for a 1550nm pump are 19 µm, 7 µm, and 2.13µm, respectively. This problem has previously been solved by using a non-uniform effective poling-period [33]. The lithium niobate is poled in a striped configuration, as in Fig. 1(a) (left inset), such that the azimuthally propagating light sees a spectrum of effective poling periods as it circulates around the resonator circumference. Plotting the envelope function of the Fourier coefficients for the poling pattern seen by the azimuthally propagating mode provides information about the relative efficiency of phase matching for different processes. The efficiency of a nonlinear process is directly proportional to amplitude of the Fourier coefficient at the optimum poling period for that process.

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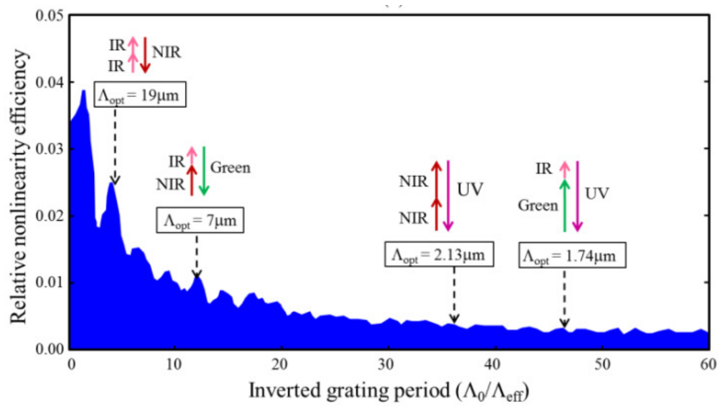

Fig. 2. The amplitude of the Fourier coefficients for the poling pattern seen by the azimuthally propagating mode, for our 3mm diameter resonator with Λ_0 = 79 µm striped poling, confirming that the energy-momentum condition can be satisfied for all three harmonic-generation processes simultaneously.

Figure 2 shows the amplitude of the Fourier coefficients as a function of inverse grating period for our 3mm diameter resonator with 79 µm striped poling, confirming that the energy-momentum condition can be satisfied for all three harmonic-generation processes simultaneously. Additionally, the existence of high-order modes facilitates quasi phase-matching over a broad pump wavelength range [34,35]. It should be noted that poling configurations with shorter periods will have a much better phase-matching performances than the PPLN wafer available for this experiment. We anticipate that shortening the grating period will greatly improve efficiency in the next-generation devices.

3. Experimental results and discussion

The experimental setup (Fig. 3(a)) is based on a crystalline whispering-gallery resonator that is polished to reduce optical losses introduced by scattering. Lithium niobate was chosen for our whispering-gallery resonator nonlinear medium for its second-order optical nonlinearity and its transparency from infrared to ultraviolet wavelengths. Additionally, due to its ferroelectric properties, lithium niobate crystal domains can be engineered by electrical poling to achieve quasi-phase matching of diverse optical modes. The whispering-gallery resonator was fabricated from a commercial periodically-poled, z-cut lithium niobate substrate. A 3-mm disk was cut from the wafer and the edge was mechanically polished to a spherical profile.

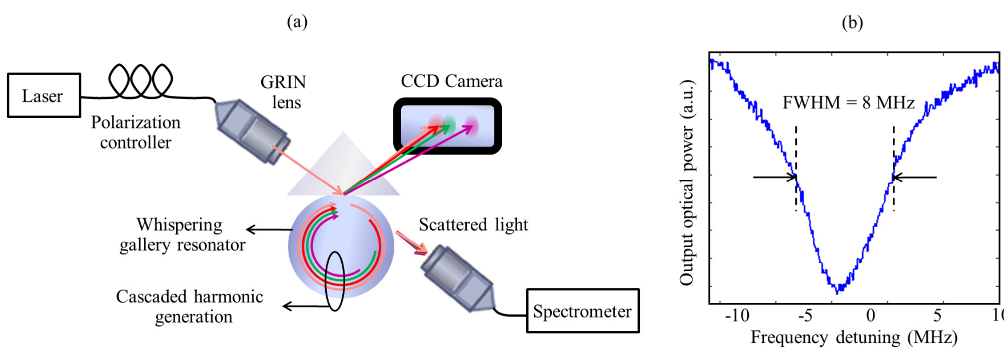

Fig. 3. (a) Experimental setup for demonstrating cascaded-harmonic generation in the periodically poled lithium niobate resonator. (b) Measured whispering-gallery resonator resonance, implying a quality factor of 2×10^7 at 1540nm.

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The pump beam, tunable from 1535 to 1545nm, is evanescently coupled to the cavity modes via a diamond prism [36]. A quality factor on the order of 10^7 was measured by monitoring transmission through the prism while scanning the IR pump frequency through the optical resonances (Fig. 3(b)). Measuring the quality factor in the UV is challenging because of the lack of narrow linewidth tunable lasers for the UV band, as well as the lack of spectrum analyzers with resolution in the order of 10MHz. Compared to the IR pump, absorption losses for the UV 4th harmonic will be higher in lithium niobate. However, loss via tunneling [37] decreases at shorter wavelengths. Additionally and by definition, quality factor is inversely proportional to wavelength, assuming that other losses are held constant. We therefore estimate that Q for the 4th harmonic is of the same order as for the IR pump. Also, the power used in this experiment is not high enough to distort the Lorentzian shape of the absorption line, indicating a lack of thermal bistability in this experiment [38]. Emitted light is collected by a CCD camera as well as a multimode optical fiber for analysis. In the first case, light is directly detected from the prism coupler, and in the second case the signal is collected from the residual Rayleigh scattering to the sides of the resonator.

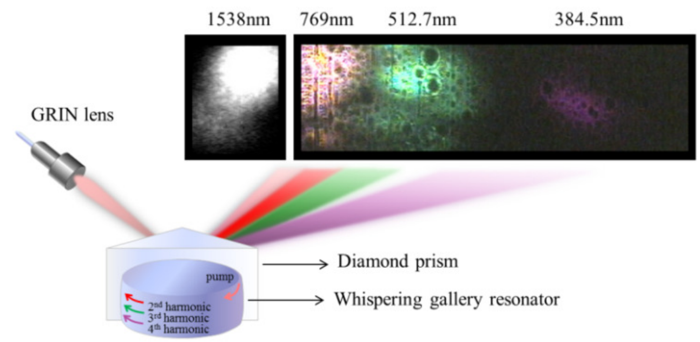

Fig. 4. Visual verification of cascaded-harmonic generation: The pump beam is recorded with an infrared CCD camera, and the harmonics are observed on a color CCD coated with ultraviolet fluorescent ink. The photograph is taken at a pump wavelength of 1538 nm and a pump power of 200mW.

Experimental visualization of the cascaded-harmonic generation process is achieved by photographing spatially resolved spots on a color and IR CCD camera, with spot separations corresponding to the infrared pump and its 2nd, 3rd, and 4th harmonics (Fig. 4). Spectral filters are employed to prevent saturation of the camera by the 2nd and 3rd harmonics, and the 4th harmonic is observed by coating the CCD with a fluorescent ink that is sensitive to ultraviolet. The noncircular shape of the spots suggests that high order transverse modes are involved in this process. We emphasize that this picture describes a continuous-wave emission for all of the generated harmonics.

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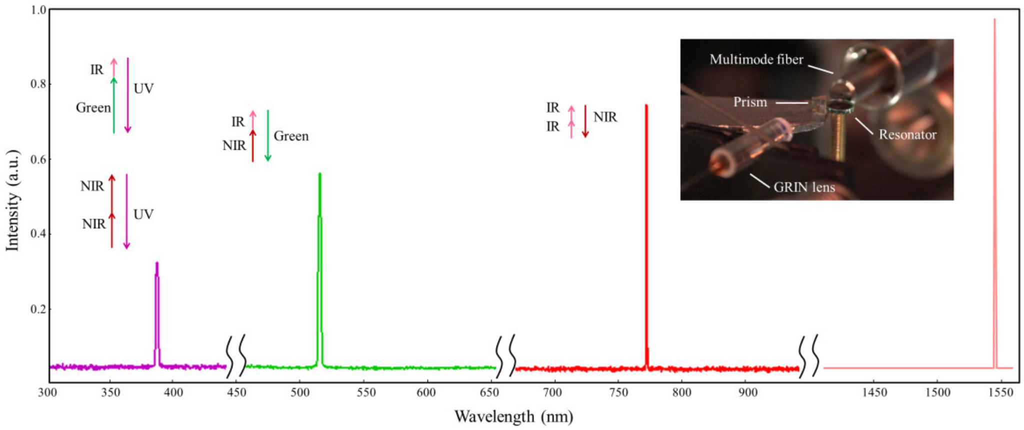

Fig. 5. Measured emission spectrum at 1546nm pump wavelength, indicating generation of the 2nd, 3rd, and 4th harmonics. Harmonics are measured using three different spectrum analyzers and are plotted at different intensity scales.

Measuring the harmonics wavelengths is done by three spectrum analyzers which cover the infrared to ultraviolet band. The n^{th} harmonic is expected to be at the (*pump wavelength*)*/n.* The experimentally measured 2nd, 3rd, and 4th harmonic lines for the pump wavelength of 1546nm are at 773 nm, 515 nm, and 387 nm, respectively (Fig. 5). These measured wavelengths lay within the 2nm error margin of our spectrum analyzers.

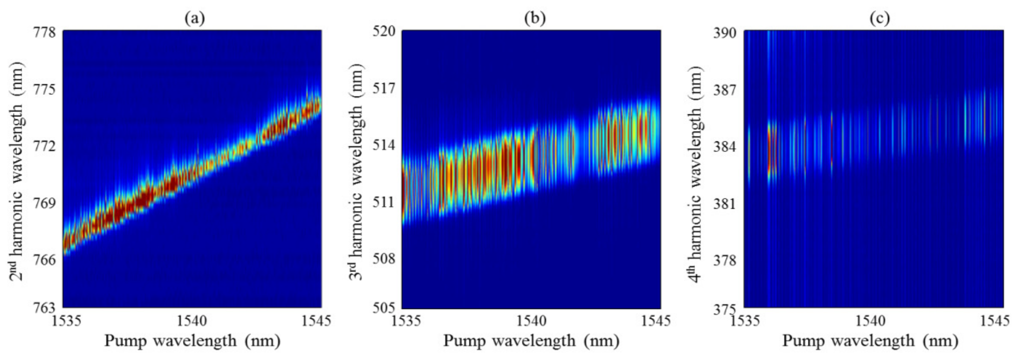

Fig. 6. Measured spectrograms of the generated 2nd, 3rd, and 4th harmonics at a pump wavelength range of 1535-1545nm are illustrated in a, b, and c. Colors stand for intensity. All three harmonics display wide tunability within this wavelength range.

Tuning the harmonics wavelengths is possible in our experimental setup by sweeping the pump wavelength through the very dense infrared resonance modes of the whispering-gallery resonator to reveal a nearly continuous tuning capacity. We experimentally demonstrate continuous tuning of the 2nd, 3rd, and 4th harmonics wavelengths while sweeping the pump wavelength between 1535nm and 1545nm (Fig. 6). All three harmonics wavelengths are observed to track the expected values as the pump wave length is varied.

Measuring the harmonics' power as a function of the input pump power is performed to confirm the cascaded-harmonic generation process. Inherently, the n^{th} harmonics power should scale as (*pump power*)^n, which is verified via a logarithmic fit of the 2nd, 3rd, and 4th harmonic power as a function of the pump power level (Fig. 7). This measurement was done by scanning the pump wavelength through several whispering-gallery resonances to record an average output power for each harmonic at a given pump power. As it is evident from the measured harmonics power, the cascaded-harmonic process improves its efficiency as pump power increases. This is expected from the (*pump power*)^n scaling of the n^{th} harmonic power.

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This efficiency will, of course, stop increasing when limiting effects such as pump depletion become evident.

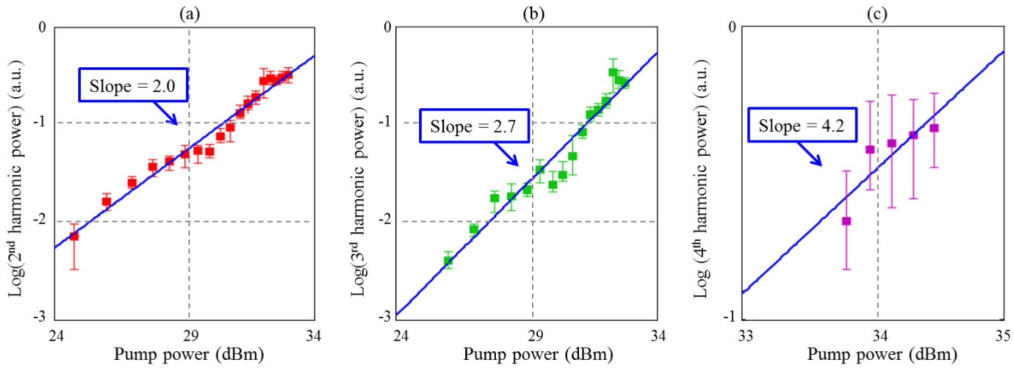

Fig. 7. Measured power of the generated 2nd, 3rd, and 4th harmonics at a pump wavelength of 1550nm, as a function of the pump power are illustrated in d, e, and f, revealing nearly quadratic, cubic and power-of-4 dependency for the 2nd, 3rd, and 4th order processes

We have experimentally confirmed that the mechanism responsible for the observed harmonic generation is cascaded-harmonic generation via $\chi^{(2)}$ processes. This is because the 3rd harmonic is only observed simultaneously with the 2nd harmonic. Similarly, the 4th harmonic is only observed simultaneously with both the 2nd and 3rd harmonics. This suggests that the 3rd and 4th harmonics arise from cascaded $\chi^{(2)}$ processes, as opposed to $\chi^{(3)}$ and $\chi^{(4)}$ effects. This observation is further supported by the fact that third and fourth order nonlinear coefficients are many orders of magnitude smaller than the second order coefficient for lithium niobate [39]. In order to further validate the effectiveness of the employed quasi-phase matching technique, a second lithium niobate whispering-gallery resonator with no crystal poling was fabricated and tested using the same experimental setups. Harmonic generation was not observed in the similar experimental conditions, confirming the significant role of the employed non-uniform poling in providing quasi-phase matching for 2nd, 3rd, and 4th harmonic generation processes.

In conclusion, we experimentally demonstrate continuous-wave cascaded harmonic generation up to the fourth harmonic in a millimeter-scale whispering gallery resonator, allowing four spectral lines which are equally spaced in frequency and span a 2-octave frequency band. Many challenges exist, but we believe this work can be extended toward continuous-in-time extreme nonlinear optics where the electron is repeatedly torn from and recombines with the atom. These challenges include phase matching and concentration of light in the gaseous region near the evanescent tail of the modes discussed here. Still, the first steps in this journey, demonstrated here, can be followed toward the extreme by adding structures such as in [40] as suggested in [41].

Acknowledgments

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