Continuous-wave cascaded-harmonic generation and multi-photon Raman lasing in lithium niobate whispering-gallery resonators
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Continuous-wave cascaded-harmonic generation and multi-photon Raman lasing in lithium niobate whispering-gallery resonators

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We report experimental demonstration of continuous-wave cascaded-harmonic generation and Raman lasing in a millimeter-scale lithium niobate whispering-gallery resonator pumped at a telecommunication-compatible infrared wavelength. Intensity enhancement through multiple recirculations in the whispering-gallery resonator and quasi phase-matching through a nonuniform crystal poling enable simultaneous cascaded-harmonic generation up to the fourth-harmonic accompanied by stimulated Raman, two-photon, three-photon, and four-photon Raman scattering corresponding the molecular vibrational wavenumbers 632 and 255 cm\(^{-1}\) in z-cut lithium niobate at pump power levels as low as 200 mW. We demonstrate simultaneous cascaded-harmonic generation and Raman lasing by observing the spectrum of the scattered light from the resonator and by capturing the image of the decoupled light from the resonator on a camera. © 2011 American Institute of Physics.

Wavelength-conversion through various nonlinear processes is an effective approach to extend the emission wavelength of laser sources to wavelengths that are difficult to achieve through standard solid-state laser sources and are in demand for various spectroscopy and microscopy applications. The high pump power levels required for generating sufficient nonlinear optical response are generally achieved by ultra-short pump pulses. This limits the effectiveness of a number of applications such as imaging crystals and molecular states with sub-atomic-scale resolution, which will benefit from a continuous-in-time source.

Whispering-gallery resonators enable strong enhancement of various nonlinear phenomena because of their capability to confine light energy for long periods of time within small volumes. Thus, significant reduction in the required pump power level for parametric oscillations, third, fourth-harmonic generation, Raman lasing, Erbium-lasing, Brillouin-lasing, and optomechanical vibrations have been reported in high-quality-factor whispering-gallery resonators.

In this work, we experimentally demonstrate continuous-wave cascaded-harmonic generation up to the fourth-harmonic accompanied by multi-photon Raman scattering from a lithium niobate whispering-gallery resonator pumped at a telecommunication-compatible infrared wavelength and at pump power levels as low as 200 mW. Our results could benefit high-harmonic and Raman studies from pulsed to continuous-wave.

We use lithium niobate for our whispering gallery resonator nonlinear medium due to its second-order optical nonlinearity and transparency from IR to UV wavelengths, enabling demonstration of cascaded-harmonic generation up to the fourth-harmonic and multi-photon Raman scattering up to the four-photon Raman scattering in whispering-gallery resonators. 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. Quasi phase-matching to conserve momentum for the high-harmonics is achieved by using a non-uniform poling-period and facilitated by a combination of high order transverse resonance modes. Lithium niobate poling is made in stripes such that the azimuthally propagating light sees different periods as it circulates around the resonator circumference and the quasi phase matching requirements are satisfied.

A schematic of the experimental setup is shown in Fig. 1(a). The whispering-gallery resonator was fabricated from a 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. The infrared pump beam is evanescently coupled to the millimeter-scale lithium niobate whispering-gallery resonator via a diamond prism. The highest whispering gallery resonator quality factor is measured to be \(2 \times 10^7\) in the 1550 nm pump wavelength range (Fig. 1(b)), implying up to 400-times enhancement in a circulating pump power inside the 3 mm-diameter whispering-gallery resonator.

The scattered light from the resonator is collected by a multimode optical fiber and analyzed by three spectrum analyzers which cover the infrared to ultraviolet band. The wavelength of the spectral lines of the measured spectrum indicate the molecular vibrational wavenumbers 632 and 255 cm\(^{-1}\) in z-cut lithium niobate crystal domains can be engineered by using a non-uniform poling-period. Lithium niobate poling is made in stripes such that the azimuthally propagating light sees different periods as it circulates around the resonator circumference and the quasi phase matching requirements are satisfied.

The emission spectrum of the whispering-gallery resonator at the pump wavelength of 1536 nm and pump power of 200 mW is shown in Fig. 2. The measured spectrum indicates 2nd (Fig. 2(b)), 3rd (Fig. 2(c)), and 4th (Fig. 2(d)) harmonic generation at 768, 512, and 384 nm, respectively. Moreover, all three harmonics display wide tunability within the pump wavelength range of 1535–1545 nm, and the nth...
Raman lines at 536, 547, 567, and 588 nm match with the three-photon lines, respectively. The observed visible spectral lines at 529, 536, 547, 567, and 588 nm match with the three-photon Raman lines $2\omega_p - \omega_{R1} - \omega_{R2}$ and $2\omega_p - 2\omega_{R2}$, where $\hbar\omega_{R1}$ and $\hbar\omega_{R2}$ are the phonon energy levels of the 255 and 632 cm$^{-1}$ Raman lines, respectively. The observed visible spectral lines at 529, 536, 547, 567, and 588 nm match with the three-photon Raman lines $3\omega_p - \omega_{R2}$, $3\omega_p - \omega_{R1} - \omega_{R2}$, $3\omega_p - 3\omega_{R2}$, and $3\omega_p - 4\omega_{R2}$. The observed ultraviolet spectral lines at 375, 393, 397, 404, 414, 419, 425, 430, 437, 449, 462, 468, and 476 nm match with the four-photon Raman lines $4\omega_p + \omega_{R2}$, $4\omega_p - \omega_{R2}$, $4\omega_p - \omega_{R1} - \omega_{R2}$, $4\omega_p - 2\omega_{R2}$, $4\omega_p - 3\omega_{R2}$, $4\omega_p - \omega_{R1} - 3\omega_{R2}$, $4\omega_p - 4\omega_{R2}$, $4\omega_p - \omega_{R1} - 4\omega_{R2}$, $4\omega_p - 5\omega_{R2}$, $4\omega_p - 6\omega_{R2}$, $4\omega_p - 7\omega_{R2}$, $4\omega_p - \omega_{R1} - 7\omega_{R2}$, and $4\omega_p - 8\omega_{R2}$. We believe that both hyper-Raman$^{19}$ and cascaded Raman processes$^{16}$ are involved in the observed multi-photon Raman scattering that extends the spectrum of the pump and the 2nd, 3rd, and 4th harmonics to a train of lines that are separated by $\hbar\omega_{R1}$ and $\hbar\omega_{R2}$.

Further evidence for the nature of the observed nonlinear processes is achieved by monitoring the whispering-gallery resonator out-coupled light on a color CCD camera (Fig. 3). Spectral filters are employed to prevent saturation of the camera by the 2nd and 3rd harmonics, and the ultraviolet light is observed by coating the CCD with a UV fluorescent ink. The spatial distribution of the beams observed on the CCD correspond to the expected positions of the 2nd, 3rd, and 4th harmonic wavelengths (769, 512.7, and 384.5 nm, respectively for a pump wavelength of 1538 nm).
at a pump power of 200 mW. The spatial separation of the near-infrared, visible, and ultraviolet light is due to dispersion in the prism, as well as chromatic and waveguide-mode dispersion in the resonator. Apart from the cascaded harmonic spots, whenever a Raman line was seen on the spectrum analyzer, a corresponding color (e.g., yellow, orange, and shades of green) was showing up in the proper region on the CCD picture. This serves as another verification for the Raman process.

In conclusion, we experimentally demonstrate continuous-wave cascaded-harmonic generation up to the fourth-harmonic accompanied by stimulated Raman, two-photon, three-photon, and four-photon Raman scattering corresponding to the molecular vibrational wavenumbers 632 and 255 cm$^{-1}$ in z-cut lithium niobate at pump power levels as low as 200 mW. By excitation of higher order harmonics one can push the cascaded-harmonic and multi-photon Raman emission to deep-ultraviolet. However, such demonstration is not possible for our current device due to the high absorption of deep-ultraviolet light in lithium niobate. The continuous progress in developing ultra-high-quality-factor resonators suggests the possibility of extending the continuous-wave high-harmonic generation demonstrated here to over 4-octaves of frequency band. In this regard, high-quality-factor whispering-gallery resonators made of crystals with $\chi^{(3)}$ effect, such as CaF$_2$, are very promising due to their broadband transparency window (0.18–8 $\mu$m for CaF$_2$).

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