















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

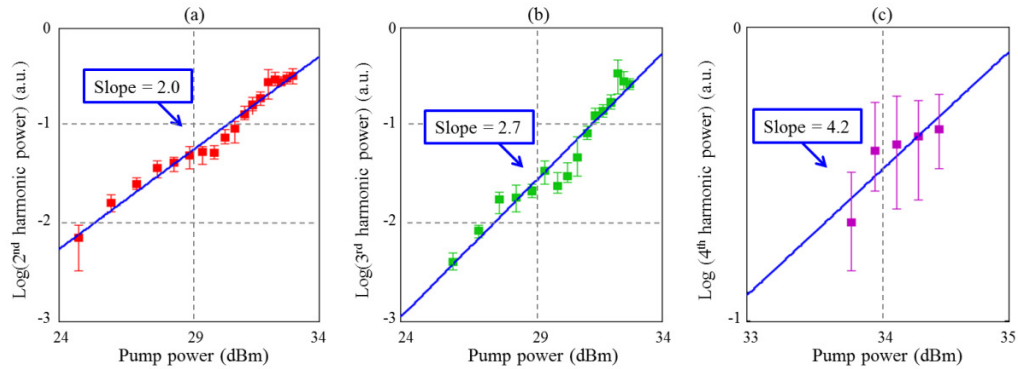


Fig. 7. Measured power of the generated 2<sup>nd</sup>, 3<sup>rd</sup>, and 4<sup>th</sup> harmonics at a pump wavelength of 1550nm, as a function of the pump power are illustrated in d, e, and f, revealing nearly quadratic, cubic and power-of-4 dependency for the 2<sup>nd</sup>, 3<sup>rd</sup>, and 4<sup>th</sup> 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 3<sup>rd</sup> harmonic is only observed simultaneously with the 2<sup>nd</sup> harmonic. Similarly, the 4<sup>th</sup> harmonic is only observed simultaneously with both the 2<sup>nd</sup> and 3<sup>rd</sup> harmonics. This suggests that the 3<sup>rd</sup> and 4<sup>th</sup> 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 2<sup>nd</sup>, 3<sup>rd</sup>, and 4<sup>th</sup> 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].

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