

# Microresonator-Based Scanning Comb Spectroscopy

Mengjie Yu<sup>1,2</sup>, Yoshitomo Okawachi<sup>1</sup>, Austin G. Griffith<sup>3</sup>,  
Michal Lipson<sup>4</sup>, and Alexander L. Gaeta<sup>1</sup>

<sup>1</sup>Department of Applied Physics and Applied Mathematics, Columbia University, New York, NY 10027

<sup>2</sup>School of Electrical and Computer Engineering, Cornell University, Ithaca, NY 14853

<sup>3</sup>School of Applied and Engineering Physics, Cornell University, Ithaca, NY 14853

<sup>4</sup>Department of Electrical Engineering, Columbia University, New York, NY 10027

Author e-mail address: my2473@columbia.edu

**Abstract:** We demonstrate mode-hop-free tuning of a modelocked silicon microresonator frequency comb over 60 GHz. Gas-phase spectroscopy of acetylene is performed with high-spectral-resolution (< 80 MHz) over a bandwidth of 40 THz.

**OCIS codes:** (300.6340) Spectroscopy, infrared; (190.4975) Parametric processes; (190.4390) Integrated optics

Molecular spectroscopy is an invaluable tool for a wide range of applications including environmental monitoring, real-time medical diagnostics and remote sensing. Especially for trace gas detection, a system that has high resolution and high sensitivity over a broad spectral bandwidth would allow for parallel detection of multiple species and accurate measurement in complex environments [1,2]. Optical frequency combs (OFC's) have a broad spectrum that consists of a large number of evenly spaced narrow comb lines, which is suitable for broadband molecular spectroscopy [2-12]. Typically, only OFC's with a line spacing of < 100 MHz are utilized for high-resolution gas-phase spectroscopy due to their narrow absorption features. Recently there has been interest in the development of OFC sources in the mid-infrared (mid-IR) regime where the gas phase has strong fundamental vibrational transitions, enabling high sensitivity spectroscopy [13]. However, the extension of OFC's into the mid-IR has proven difficult and remains under development. Most mid-IR sources require transferring an OFC from the near-infrared to the mid-IR by nonlinear wavelength conversion using either optical parametric oscillation or difference frequency generation [5-11], which may be complex and posts a limitation of comb line power, phase-matching optical bandwidth and compactness. Recently, modelocked mid-IR combs have been directly generated in CMOS-compatible silicon microresonators [14] with a high comb line power, large comb spacing and a broad spectral bandwidth of ten's of THz using a single-frequency pump laser. While microresonator combs have proven to be suitable for spectroscopy in the liquid or condensed phases [15], the large repetition rate (>100 GHz) precludes their use for high-resolution molecular spectroscopy.

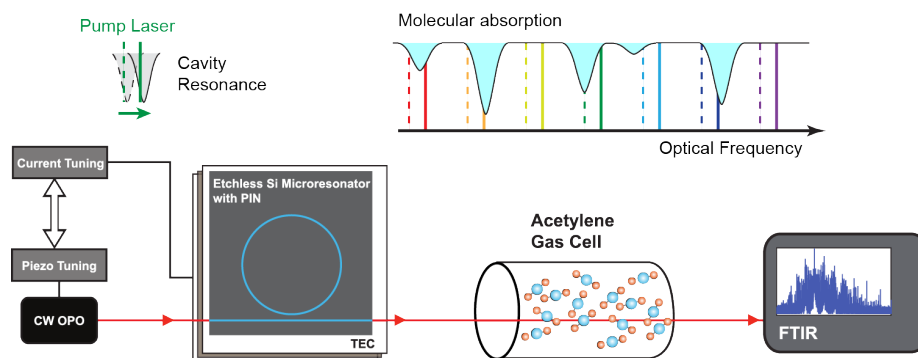


Fig. 1. Scheme for scanning comb spectroscopy. A silicon microresonator is pumped by a continuous-wave optical parametric oscillator. The output is sent through a 2-cm gas cell of acetylene and then recorded by an FTIR spectrometer. A thermoelectric cooler (TEC) is used to control the position of the cavity resonance via tuning temperature. A modelocked comb is scanned via joint control of the pump laser frequency and the TEC current.

In this paper, we report the first demonstration of a microresonator-based scanning OFC spectrometer suitable for gas-phase spectroscopy. We demonstrate mode-hop-free tuning of a modelocked mid-IR frequency comb in a silicon microresonator over 60 GHz ( $2 \text{ cm}^{-1}$ ) via joint tuning of temperature and pump laser frequency. The modelocked comb spans from 75 – 115 THz ( $1333 \text{ cm}^{-1}$ ) with a comb line spacing of 127 GHz ( $4.2 \text{ cm}^{-1}$ ). Multiple absorption features of gas-phase acetylene are captured and an absorption linewidth of 6.4 GHz ( $0.213 \text{ cm}^{-1}$ ) is measured with a spectral resolution of < 80 MHz ( $0.0026 \text{ cm}^{-1}$ ). This technique overcomes the resolution limitation

(typically 10-100 GHz) inherently induced by the ultra-small physical size of the integrated optical device and offers a significant step towards a high spectral resolution, on-chip broadband spectroscopic device.

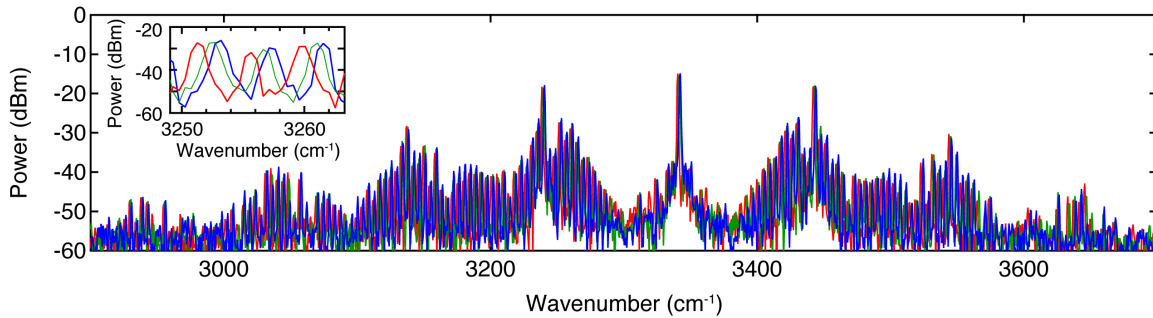


Fig. 2. Optical spectra recorded by an FTIR during the tuning process. A  $2 \text{ cm}^{-1}$  (60 GHz) tuning range is achieved with a TEC current change of 200 mA. Embedded: Zoom-in spectra of three comb lines.

In our experiments, a high-Q ( $\sim 200,000$ ) silicon microresonator with a  $100\text{-}\mu\text{m}$  radius is dispersion engineered to have anomalous group-velocity dispersion beyond  $3 \mu\text{m}$  for the fundamental TE mode, similar to Griffith, *et al.* [16]. As shown in Fig. 1, the microresonator is pumped by a cw OPO ( $<100\text{-kHz}$  linewidth) at  $3 \mu\text{m}$ . Integrated PIN diodes, are operated at a reverse-bias voltage of  $-19 \text{ V}$  for the extraction of free carriers (FC) generated from three-photon absorption (3PA). A thermoelectric cooler (TEC) is used to control the temperature of the silicon device. The output spectrum is monitored using a commercial Fourier-transform infrared spectrometer (FTIR). We generate a modelocked mid-IR frequency comb by tuning the pump laser into the cavity resonance [14], and the spectrum consists of 305 comb lines with a spacing  $f_{rep} = 127 \text{ GHz}$  and spanning  $2.6 - 4.1 \mu\text{m}$ , an atmospheric window important for the detection of the fundamental CH, NH and OH stretching modes in molecules. In order to access the spectral region between the comb lines, we tune both the pump laser frequency and the TEC current, which tunes the cavity resonance, while maintaining the modelocked state. During the process, the DC component of the FC current is carefully kept the same in order to keep the relative pump-cavity detuning unchanged. The relative pump-cavity detuning has been proven to be the key parameter of the modelocking dynamics [17]. Using this method, it is proven possible to keep the low-noise modelocked state while frequency shifting the entire comb and is equivalent to mode-hop-free tuning all 305 comb lines simultaneously. We achieve a mode-hop-free tuning range of  $60 \text{ GHz}$  ( $2 \text{ cm}^{-1}$ ) which is half of  $f_{rep}$ , as shown in Fig. 2. Ideally, a tuning range of one free-spectral range will enable continuous coverage over the entire spectral range. Currently, we are currently limited by the pump power flatness over this larger bandwidth. However this is not a fundamental issue and a  $60 \text{ GHz}$  mode-hop-free tuning range can be applied to a similar silicon microresonator of  $200\text{-}\mu\text{m}$  radius to cover the entire generated spectral range. Furthermore, this technique can be readily extended to other microresonator-based OFC's with repetition rates on the order of 10-100 GHz.

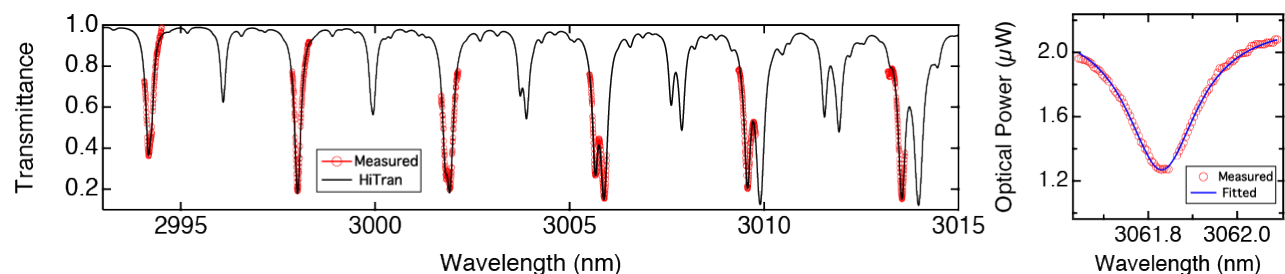


Fig. 3. Absorption measurement of acetylene. (a) Transmittance measured by the selected five comb lines (zoom-in) from the entire OFC which are scanned over  $15 \text{ GHz}$  ( $0.5 \text{ cm}^{-1}$ ). The measured transmittance shows good agreement with the simulated Hi-Tran data under the same condition as our gas cell. (b) The measured acetylene absorption with a half-linewidth of  $3.2 \text{ GHz}$  centered at  $3061.83 \text{ nm}$  (fitted with Lorentz line shape).

Finally we apply the scanning comb for an absorption measurement of gas-phase acetylene. The output is sent through a  $2\text{-cm}$  long single-pass cell which is filled to a pressure of  $40 \text{ Torr}$  of acetylene and  $415 \text{ Torr}$  of nitrogen. The output spectrum is recorded with the FTIR. Using this method, microresonator-based frequency comb spectroscopy can break the resolution limitation of both the  $f_{rep}$  or the FTIR resolution. The spectral resolution of our

system is limited by the comb linewidth in the modelocked state. Since our system is free running, the resolution is primarily limited by the pump laser linewidth, which in our case is  $< 100$  kHz. The instrumental line-shape is negligible, since narrow comb lines sample the molecular spectrum and the FTIR simply acts as a broadband detector for each of the measured comb lines. In our case, the FTIR is operated with a low resolution of 30 GHz, which enables relatively fast acquisition. The pump (and comb) are tuned over 15 GHz ( $0.5 \text{ cm}^{-1}$ ) with a TEC current change of 100 mA. By monitoring the beatnote between the pump laser and a fs modelocked OPO, we finely tune the pump frequency with a precision of  $< 10$  MHz. While tuning, all the optical spectra are captured and used to extract the molecular absorption. Absorption features are clearly observed and its line parameter can be retrieved over the 15 GHz scanning range between  $3 - 3.1 \mu\text{m}$ . Figure 3a shows our transmittance measurement for five selected comb lines. It agrees well with the computed transmittance profile using the HITRAN line parameters and a Lorentzian lineshape. As shown in Fig. 2(b), we have fitted one of the measured absorption features with a Lorentzian profile and have achieved a half linewidth of 3.2 GHz centered at 3061.83 nm. Additional frequency precision can be achieved with a fully stabilized OFC and using a combination of integrated heaters and a monochromator will significantly improve the tuning speed.

In conclusion, our scanning microresonator-based combs offer the promise of capturing multiple gas absorption features within the entire optical spectral range of 40 THz with  $< 10$  MHz resolution and opens up new opportunities for on-chip gas phase spectroscopy.

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