Interleaved difference-frequency generation for microcomb spectral densification in the mid-infrared

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With their compact size and semiconductor-chip-based operation, frequency microcombs can be an invaluable light source for gas spectroscopy. However, the generation of mid-infrared (mid-IR) frequency combs with gigahertz line spacing as required to resolve many gas spectra represents a significant challenge for these devices. Here, a technique referred to as interleaved difference-frequency generation (iDFG) is introduced that densifies the spectral line spacing upon conversion of near-IR comb light into the mid-IR light. A soliton microcomb is used as both a comb light source and microwave oscillator in a demonstration, and the spectrum of methane is measured to illustrate how the resulting mid-IR comb avoids spectral undersampling. Beyond demonstration of the iDFG technique, this work represents an important feasibility step towards more compact and potentially chip-based mid-IR gas spectroscopy modules.

1. INTRODUCTION

Optical microresonator-based frequency combs (microcombs) have received increasing interest over the past decade [1–4]. They can operate with low power consumption and offer compact form factors. Of special importance, soliton formation in microresonators provides an elegant method to generate fully coherent frequency combs [3,5]. These soliton microcombs have been tested in a wide range of applications including spectroscopy [6–8], optical frequency synthesis [9], and optical clocks [10]. Since microcombs are fabricated on the wafer scale [see Fig. 1(a)], a large number of portable functional modules can in principle be produced efficiently.

For spectroscopy and gas sensing, the mid-infrared (mid-IR) bands have been of keen interest for conventional frequency comb development [11]. And mid-IR microcomb-based sensor modules, on account of their compact form factor, could potentially be used in the food industry [12], for human breath and health analysis [13], for detection of chemical threats [14], and for monitoring of greenhouse gases (such as CO₂, CH₄) [15,16]. Microcomb-based spectroscopy has been demonstrated in both the near-IR [6,7] and the mid-IR [8]. However, monolithic microcombs presently feature very wide comb line spacings [3,17] in the mid-IR bands. While such wide spacings can increase the acquisition rate in dual-comb spectroscopy (DCS) [18], they also lead to undersampling of gas spectra and require separate frequency tuning of the comb to fill in the spectrum [19]. Reducing microcomb line spacing has been a priority for opto-electrical interface in self-referenced microcomb systems [9], and ultra-high-Q resonator platforms can overcome the increased pumping volume of the narrower line spacing combs. However, narrow gigahertz-rate line spacings have been possible only in the near-IR [20,21]. Apart from this issue, the development of complex self-referenced microcomb systems in the mid-IR presents an even greater challenge. And while there has been remarkable progress on mid-IR comb formation using quantum cascade lasers, these devices so far also feature wide comb line spacings [22,23].

For these reasons, the method of difference-frequency generation (DFG) presents an appealing alternative for generation of microcomb-based coherent combs in the mid-IR. DFG conveniently leverages more mature visible and near-IR signal sources to generate a wide range of mid-IR spectra and has an extensive application history. An early use of DFG was to generate 3.39 µm comb light to create a methane optical clock [24]. Generation of mid-IR comb light for the earliest demonstration of DCS also relied on DFG by mixing Ti:sapphire lasers in GaSe crystals [25,26]. The use of fiber laser combs either alone (distinct spectral portions of a single comb) [27] or in conjunction with a CW source [28] for generation of mid-IR comb light was soon followed by demonstration of spectroscopic measurements of methane [29]. When combined with DCS, it later enabled remarkably precise spectral measurements of methane and other gases [30–33]. All
of these works used periodically poled lithium niobate (PPLN) as the mixing crystal. More recently, super-octave mid-IR comb spectra have been generated using fiber-comb intrapulse DFG in GaP crystals [34]. These results dramatically extend the spectral reach of the DFG technique, and intrapulse DFG has also been applied using PPLN [35]. However, all comb DFG methods demonstrated to date utilize narrow line spacing table-top combs. It is therefore interesting to consider ways to generate mid-IR combs with gigahertz line spacing using available near-IR wide line spacing microcombs (10s of GHz). Such mid-IR combs would avoid spectral undersampling while retaining a high acquisition rate for DCS.

Here, we introduce interleaved DFG (iDFG) to generate gigahertz line spacing mid-IR comb light at 3.3 μm. Similar to conventional DFG, iDFG occurs between two combs, but now having different repetition rates. The rates are specially chosen so that a new mid-IR comb is formed with a line spacing equal to an integer fraction of the repetition rate of either original comb (i.e., the mid-IR comb spectrum is densified relative to the original combs). In the work, a silica soliton microcomb [36] acts as both a lightwave source and a microwave source [see Fig. 1(a)] and is mixed with an electro-optic frequency comb (EO-comb) [37] to produce mid-IR comb light. iDFG enables agile adjustment of the mid-IR comb line spacing, and the generated 3.3 μm comb is used to perform methane spectral measurement.

2. RESULTS

A. Interleaved Difference-Frequency Generation

In the measurement, a 1.5 μm soliton microcomb [36] and a 1 μm EO-comb [37] having different repetition rates are mixed for mid-IR comb generation. Due to their repetition rate difference (Δfr, subject to conditions described below; see the Appendix A), mixing of the comb pulses repeats after 1/Δfr [Fig. 1(a)], thereby creating a temporal interleaving effect. Thus, iDFG not only converts the near-IR combs into the mid-IR, but also reduces the repetition rate (line spacing) of the mid-IR comb spectrum. The line spacing is also tunable by adjustment of Δfr, and to demonstrate this feature, a range of mid-IR comb line spacings (lowest 0.7 GHz) is generated. The soliton microcomb having a comb line spacing of approximately 22 GHz uses a 3 mm diameter ultra-high-Q wedge silica microresonator [36,38]. Its spectrum [Fig. 1(b)] features a smooth and sech²-like envelope with a 3 dB bandwidth of 1.5 THz (~70 lines within the 3 dB bandwidth). As shown in the Appendix A, a uniform line spacing of the iDFG-generated mid-IR comb requires that Δfr = mfr/n, where m, n are mutually prime. To ensure this condition, the soliton stream is detected to generate a 22 GHz microwave signal fr and is then electrically processed to create the EO-comb drive signal frequency equal to fEO = (N − 1)fr/N [Fig. 1(a) and inset of Fig. 1(b)]. This ensures Δfr = fr/N, where N is an integer (typically 16, 32) and guarantees a strict frequency (and phase) relationship between
the EO-comb and soliton comb repetition rates. Significantly, the approach also leverages the excellent microwave stability of the soliton microcomb \[36,39–41\] to replace a bulk microwave source that is normally required to drive the EO-comb. As described elsewhere \[37\], the EO-comb consists of cascaded phase and intensity modulators. Figure 1(b) contains a representative optical spectrum of the generated EO-comb. Current progress in on-chip EO modulators suggests that integrated solutions to this EO-comb will be possible in the near future \[42\]. Alternatively, it is also possible to replace the EO-comb with a 1 \(\mu\)m soliton microcomb using fabrication methods demonstrated elsewhere \[43\].

### B. Mid-IR Combs Generated by iDFG

The mid-IR comb at 3.3 \(\mu\)m is generated by pumping a 4 cm long PPLN crystal (NTT Corporation) with the above soliton and EO-combs. Integrated waveguide PPLN devices can offer much smaller form factors \[44,45\]. When setting \(N = 16\), a mid-IR comb spanning about 80 nm can be generated [spectrum shown in Fig. 2(a)]. The center frequency of the mid-IR comb can be shifted by changing the temperature of the crystal to thereby adjust the phase-matching condition. Due to the limited spectral resolution of the mid-IR spectrometer (Horiba iHR 550), a discrete comb structure is not resolvable in the spectrum. Therefore, to test that the generated comb has a uniform line spacing that has been reduced to \(\delta f/16\) through the iDFG process, a fast mid-IR detector is used to measure its line spacing by photodetection of the comb. The electrical spectrum of the detected 3.3 \(\mu\)m comb shows a single peak at \(f_c/16 \approx 1.4\) GHz [see Fig. 2(b)]. There are no additional peaks in the spectrum, consistent with a uniform line spacing and also showing reduction of the comb line spacing to 1.4 GHz from that of the 22 GHz soliton microcomb. The resulting mid-IR comb consists of more than 1500 lines. The measured phase noise spectrum of the detected mid-IR comb signal is shown in the inset of Fig. 2(b) and verifies excellent repetition-rate stability.

To further study the iDFG system, we replaced the soliton-based microwave drive signal by an independent microwave signal source (Agilent PSG). This allowed examination of the effect of non-optimal drive frequencies on the detected mid-IR microwave spectrum. As shown in Fig. 2(b), by using a non-optimal frequency of \(15 f_c/16 + \delta f (\delta f \approx 48\) MHz) to drive the EO-comb, the detected mid-IR electrical spectrum features peaks at the frequencies of 168 \(\delta f\) and \(f_c/16 - 178\) \(\delta f\) in addition to the main peak at \(f_c/16 - \delta f\). This drive frequency arrangement thereby induces a non-uniform line spacing in the mid-IR comb and is consistent with our analysis in Appendix A. This non-uniformity is undesirable, as it would increase the signal processing complexity in applications such as DCS. As an aside, the ability to observe the additional peaks also verifies that the mid-IR comb densification occurs without the appearance of spectral gaps (i.e., there are \(N\) mid-IR lines generated for every 22 GHz spacing in the mid-IR). Otherwise, these tones would not appear.

By adjusting the division ratio \(N\), the iDFG system allows generation of other mid-IR combs with different line spacings. For example, when dividing \(f_c\) by \(N = 32\), a mid-IR comb with a line spacing of \(f_c/32 \approx 0.7\) GHz can be generated. The corresponding optical spectrum is shown in Fig. 2(c) and is similar to the spectrum in Fig. 2(a). The repetition rate of 0.7 GHz is verified in the inset of Fig. 2(c). Other comb line spacings can also be readily generated, but are not demonstrated here due to the limitations of the electrical band pass filter employed. Hence, the line spacing of the 3.3 \(\mu\)m comb can be selected in an agile way for different applications. Residual gaps are believed to occur in the comb densification for the \(N = 32\) case. This is, however, not a fundamental limitation but rather one caused by the limited phase-matching bandwidth of the PPLN crystal as discussed in Section 2.C.

### C. Methane Measurement

As a simple demonstration, the 3.3 \(\mu\)m iDFG comb was used for absorption measurement of methane. This also tested its ability to avoid spectroscopic undersampling in the mid-IR. Methane spectra were measured by passing comb light from the 1.4 GHz mid-IR comb through a 5 cm long single-pass gas cell containing a mixture of 200 Torr methane and 560 Torr nitrogen, followed by spectral analysis using the Horiba spectrometer. The absorption spectrum is shown in Fig. 3(a). Six branches \([P(2)\) to \(P(7)]\) in the \(v_1\) band can be observed in the spectrum and match the methane absorption lines (included for comparison in the figure). Because only two phase modulators and one intensity modulator were used for EO-comb generation in this measurement (one phase modulator was damaged), the mid-IR comb is spectrally narrower than the one in Fig. 2, where three phase modulators and one intensity modulator were used. Even with this narrower spectrum, three methane absorption branches are observed in a single measurement. By tuning the temperature of the PPLN crystal it was possible to extend the spectrum over additional branches. Similar to Fig. 2(a), comb lines and fine absorption features are not resolved due to the limited resolution of the spectrometer.

To illustrate the benefit of the densified line spacing, a 22 GHz mid-IR comb was also generated by conventional DFG of the 1.5 \(\mu\)m soliton with a CW 1 \(\mu\)m laser. Using these widely spaced comb lines for measurement of the \(P(4)\) and \(P(5)\) branches, the methane lines are spectrally undersampled [see Fig. 3(b)]. Moreover, the resulting 22 GHz mid-IR comb is narrower than that shown in Fig. 2. This is because the PPLN crystal provides a broader phase-matching bandwidth at 1 \(\mu\)m versus phase matching at 1.5 \(\mu\)m \[46\]. Thus, a broader comb is obtained when having a broadband 1 \(\mu\)m input. It is interesting to note that the CW 1 \(\mu\)m laser mixes with only \(~19\) soliton microcomb lines within the PPLN phase-matching bandwidth. To avoid the presence of spectral gaps in the mid-IR comb, this number needs to be larger than \(N\). As a result, the \(N = 32\) mid-IR comb is believed to contain spectral gaps.

The mid-IR comb power generated here is about 200 \(\mu\)W starting with \(~100\) mW 1.5 \(\mu\)m soliton power and \(~100\) mW 1 \(\mu\)m EO-comb power as inputs. Only a portion (less than 20\%) of the soliton spectrum contributes to iDFG due to the abovementioned phase-matching bandwidth. iDFG should decrease the conversion efficiency compared to conventional DFG with identical repetition rates, since it reduces the overall temporal overlap between the two near-IR combs. However, the iDFG efficiency and output comb power can be enhanced relative to the current results by using a thin-film PPLN waveguide, as the confinement is higher \[44,45\], and dispersion engineering can be employed to provide a more optimal design \[42,47,48\]. For example, significant enhancement of second-harmonic generation (SHG) efficiency in thin-film PPLN waveguides compared to conventional PPLN waveguides has been reported \[45\]. These waveguides have also enabled SHG over a bandwidth exceeding 10 THz via dispersion engineering \[48\]. By replacing the EO-comb with a 1 \(\mu\)m soliton microcomb \[43\], an increased efficiency can also result from the
Fig. 2. Mid-IR frequency combs generated by iDFG. (a) Optical spectra of the iDFG generated mid-IR comb. The center wavelength of the comb can be shifted by changing the temperature of the PPLN crystal to vary the phase-matching condition. Due to the limited resolution of the spectrometer, the individual comb lines (spaced by 1.4 GHz) are not resolved. (b) Electrical spectrum of the photodetected mid-IR comb in panel (a) showing a repetition rate of $f/16 = 1.4$ GHz (red line) resulting from driving the EO-comb at $15f/16$. When driving the EO-comb at a frequency slightly offset from $15f/16$ by an independent microwave oscillator, there will be additional peaks in the electrical spectrum (green spectral peaks). The inset shows the phase noise of the generated mid-IR comb at 1.4 GHz. (c) The line spacing of the mid-IR comb generation can be varied. Here, a line spacing of $f/32 = 0.7$ GHz, half of that shown in panel (a), is generated by driving the EO-comb at a frequency of $31f/32$. The line spacing is verified by spectral analysis of the detected comb (inset).

Fig. 3. Methane absorption measurement using the 3.3 μm comb. (a) Measured absorption spectrum of methane over six branches [P(2) to P(7) in the $\nu_3$ band] using the 3.3 μm comb with a line spacing of 1.4 GHz. The absorption spectrum is obtained by normalizing the spectrum measured upon transmission through the gas cell with the incident comb spectrum. Single comb lines are not resolved due to the limited resolution of the spectrometer (Horiba iHR550). (b) When using the 22 GHz line spacing 3.3 μm comb to measure the P(4) and P(5) branches, the absorption features are spectrally undersampled.
higher peak pulse power relative to that afforded by the EO-comb. However, a microcomb has a relatively fixed repetition rate, and this will limit mid-IR line spacing tuning agility compared to the EO-comb. To overcome this tuning limitation, multiple 1 µm soliton microcombs, each having different repetition rates, could be integrated on a single chip to allow discrete repetition rate tuning. The repetition rate of a single 1 µm microcomb would be locked to the \((N - 1) f_s / N\) frequency by either active feedback or injection locking [41]. This would require precise microfabrication control to obtain the desired free-spectral-range (FSR) so as to facilitate locking. Control at the level of 1:20,000 of FSR has been demonstrated [38], and the remaining fine tuning of FSR could be accomplished using microheaters [49].

The absorption measurement demonstrated here is currently limited by the resolution of our spectrometer. Moreover, the spectrometer itself is large and bulky and employs a mechanical scanning mechanism. These undesirable features of the measurement can be eliminated by adding one mid-IR comb so as to implement a DCS approach [6,18]. DCS can also leverage the advantage of gigahertz mid-IR combs in terms of increased acquisition rate.

3. DISCUSSION

The iDFG method was introduced and applied to generate mid-IR frequency comb light from two near-IR combs: one a soliton microcomb and the second an EO-comb whose drive frequency is derived from the soliton microcomb. The iDFG method enabled comb line densification of the mid-IR comb relative to the near-IR combs so that spectral measurement of methane in the 3.3 µm band was possible. This method should be easily extended to produce denser combs at other spectral regions. Also, interleaved sum-frequency generation (isFG) is possible. Moreover, densification of the sparse microcomb spectra is potentially useful in dual microcomb lidar as a way to increase the ambiguity range [50,51].

The demonstrated iDFG module still consists of fiber-based components. However, current progress towards complex chip-integrated systems using soliton microcombs [9] as well as significant progress in the area of integrated lithium niobate components [42,45,47] bodes well for development of compact iDFG modules. Moreover, soliton microcombs based entirely upon lithium niobate and operating in the 1.5 µm band have recently been demonstrated [52], so that all mid-IR comb generation components can in principle be monolithically integrated. Other compact waveguide DFG (SFG) materials are also possible. For example, DFG to 3 µm has been demonstrated in GaAs waveguides [53], and there has been impressive progress in second-harmonic mixing using heterogeneously integrated GaAs structures [54].

Mid-IR microcombs could also function as compact instrument calibration sources in the field. Also, the iDFG microcomb approach can provide a range of mid-IR comb wavelength bands by leveraging the wide operational wavelength band of near-IR microcombs (currently 780 nm [43] through 2000 nm [55]). The soliton microcomb range between 1000 nm and 1550 nm, in particular, can be applied to generate mid-IR iDFG combs for chemical threat detection. Finally, densified 3.3 µm microcomb and methane measurement can be used to distinguish the biological and abiotic contributions to methane formation on Mars [56].

APPENDIX A

Selection of \(\Delta f_s\) in iDFG. We write the near-IR EO and soliton combs as \(\nu_{EO} = n_1 f_s^{EO} + f_0\), \(\nu_S = n_2 f_s + f_0\), respectively. The repetition rate of the soliton microcomb is denoted as \(f_s\), to be consistent with the main text. The mid-IR comb generated in iDFG can be written as

\[
v_{MIR} = \sum_{n_1, n_2} (n_1 f_s^{EO} - n_2 f_s) + (f_0^{EO} - f_0^S).
\]

We further define \(f_0^{MIR} = f_0^{EO} - f_0^S\) (the mid-IR comb offset frequency) and \(\Delta f = f_s - f_0^{MIR} \equiv m f_s / n + \delta f\), where \(m, n\) are mutually prime, and \(\delta f\) is zero or a small frequency compared to \(f_s\) (\(\delta f / f_s\) is an irrational number).

To analyze the condition for uniform line spacing generation in iDFG, we focus on the virtual comb lines near the offset frequency of the mid-IR comb (i.e., the comb does not extend to this range) between \(f_0^{MIR}\) and \(f_0^{MIR} + f_s\). Also, we consider separating the overall mid-IR comb into sub-combs created by mixing one of the comb lines of the EO-comb with the comb lines of the soliton comb. The offset frequency for the \(k^{th}\) sub-comb is

\[
f_0^{MIR}(k) = f_0^{MIR} + \text{mod} \left[ - (k - 1) \Delta f_s, f_s \right] = f_0^{MIR} + \text{mod} \left[ - (k - 1) \left( m + \frac{n \delta f}{f_s} \right), f_s \right] / n.
\]

When \(\delta f = 0\), all possible frequencies in the modulo term are \(n_1 f_s / n\) (\(n_1\) is an integer within \([0, n - 1]\)). In other words, the offset frequencies of different sub-combs will be spaced by \(f_s / n\), and a uniform line spacing of \(f_s / n\) in the mid-IR can be obtained. Otherwise, non-zero \(\delta f\) will create additional line spacings.

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