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(54) **METHODS AND APPARATUS FOR GENERATING MID-INFRARED FREQUENCY COMBS**

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G02F 1/35   (2006.01)
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CPC **G02F 1/3501** (2013.01); **G02F 1/3551** (2013.01); **G02F 1/365** (2013.01);
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CPC G02F 1/3501; G02F 1/3551; G02F 1/365; G02F 2001/3503; G02F 2203/56; H01S 3/1671

See application file for complete search history.

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(57) **ABSTRACT**

Apparatus and methods for generating mid-IR frequency combs using intra-pulse DFG. A mode-locked pulse generation laser generates near-IR pulses which are amplified. The amplified pulses are spectrally broadened by a nonlinear element, for example a normal dispersion highly nonlinear fiber (ND-HNLF) to generate broadened pulses. The nonlinear spectral broadening element is a transparent dielectric material having a cubic nonlinear response. Broadened pulses are temporally compressed to generate short, high-power pulses which few-cycle conditioned pulses which are ready for the intrapulse DFG process. The DFG block generates a mid-IR comb by difference frequency generation. It might comprise an orientation patterned GaP (OP-GaP) crystal or a poled lithium niobate (PPLN) crystal.

16 Claims, 7 Drawing Sheets
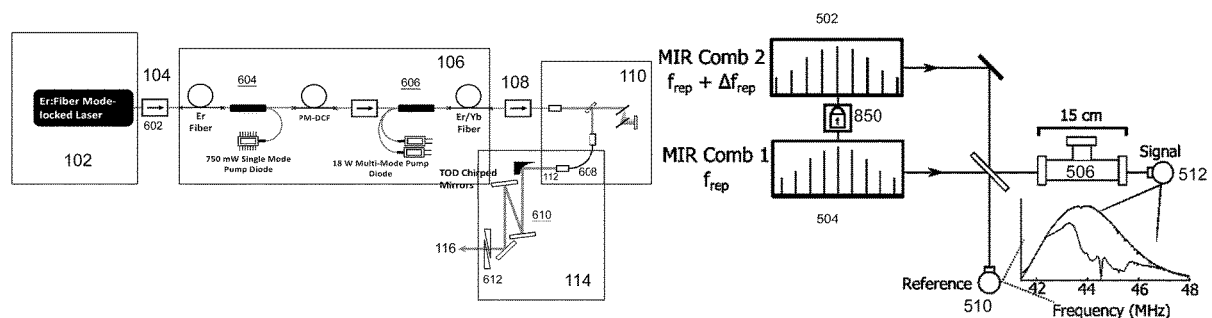
(51) **Int. Cl.**
    G02F 1/365   (2006.01)
    H01S 3/16   (2006.01)

(52) **U.S. Cl.**
    CPC *H01S 3/1671* (2013.01); *G02F 1/3503*
          (2021.01); *G02F 2203/56* (2013.01)

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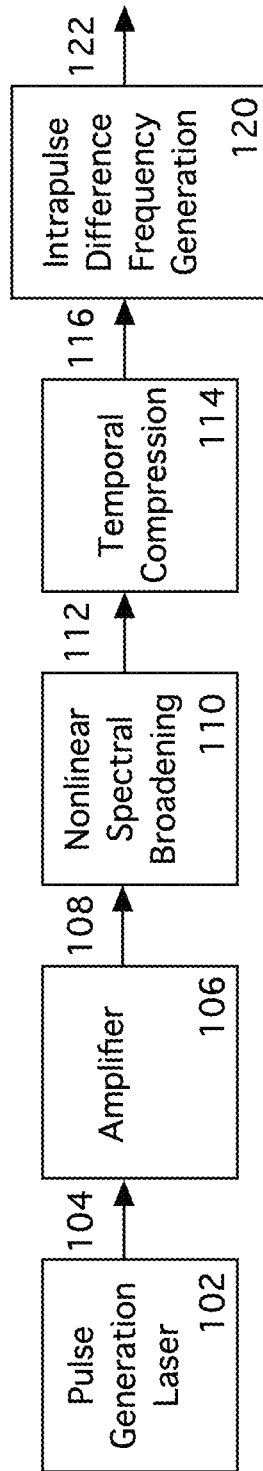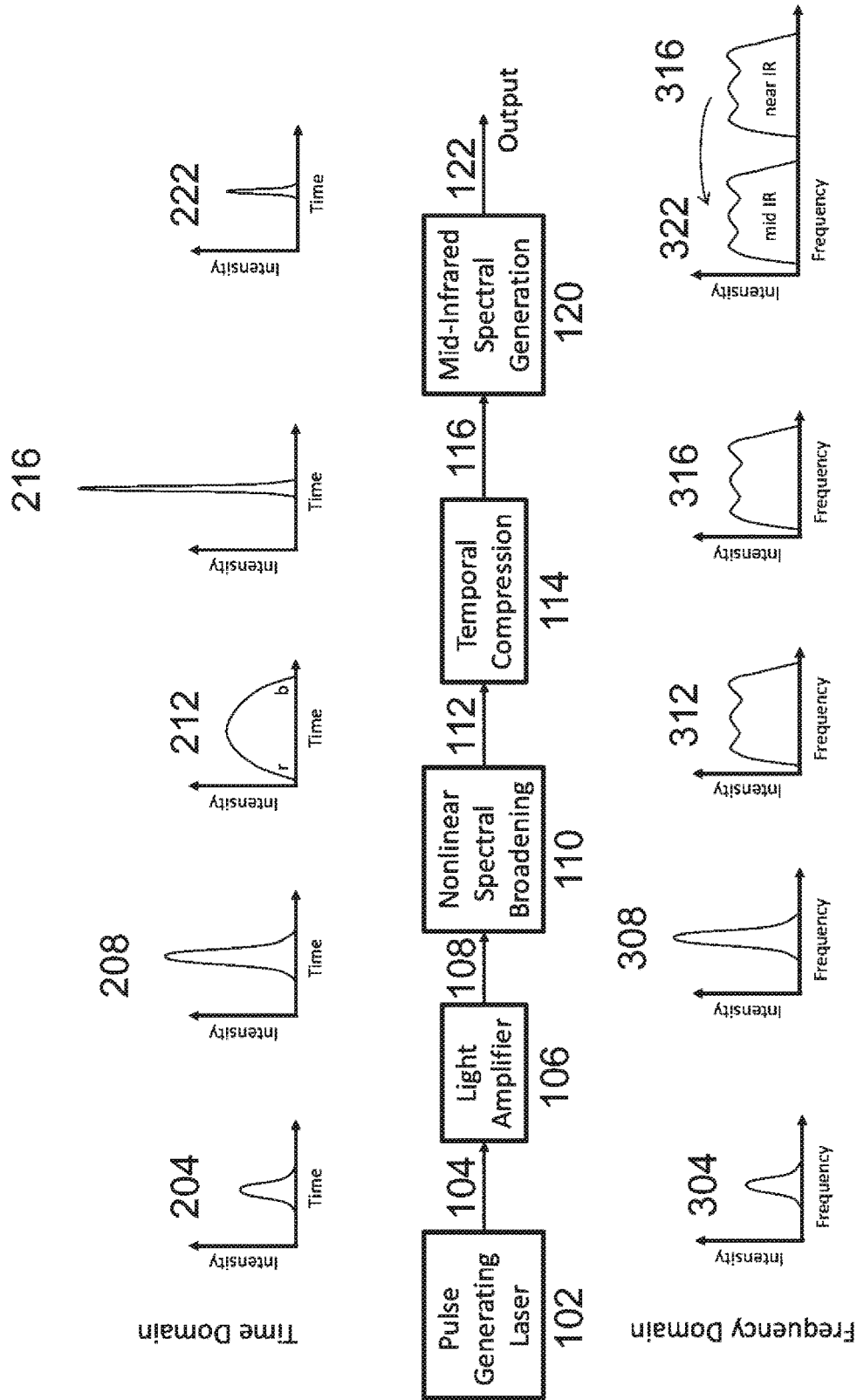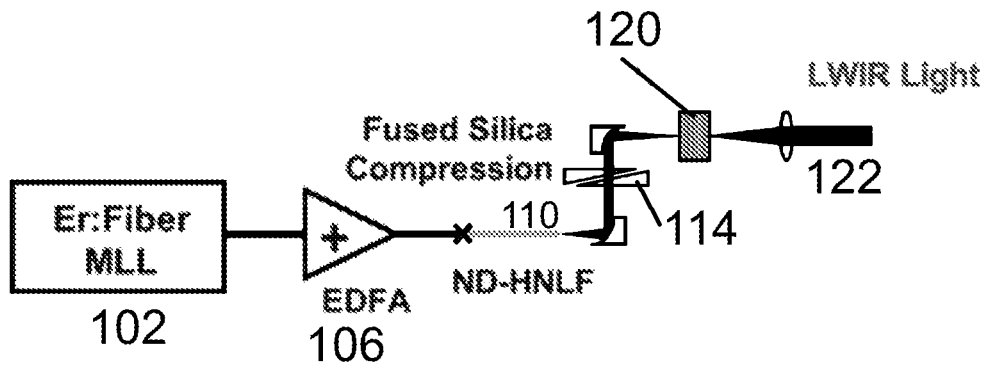| Pulse Generation Laser 102 | 104 → | Amplifier 106 | 108 → | Nonlinear Spectral Broadening 110 | 112 → | Temporal Compression 114 | 116 → | Intrapulse Difference Frequency Generation 120 | 122 → |

Figure 1

Figure 2

Figure 3
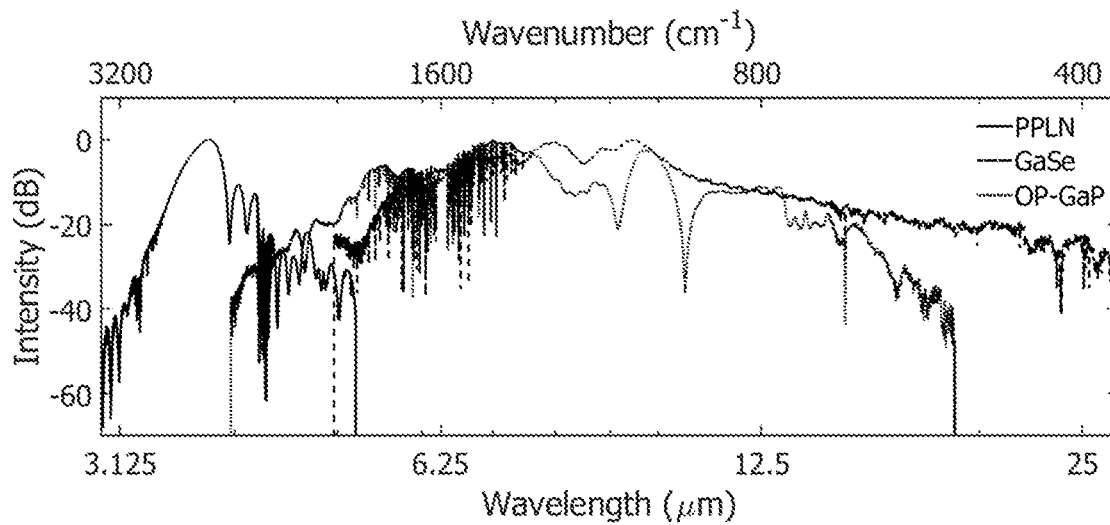
Figure 4

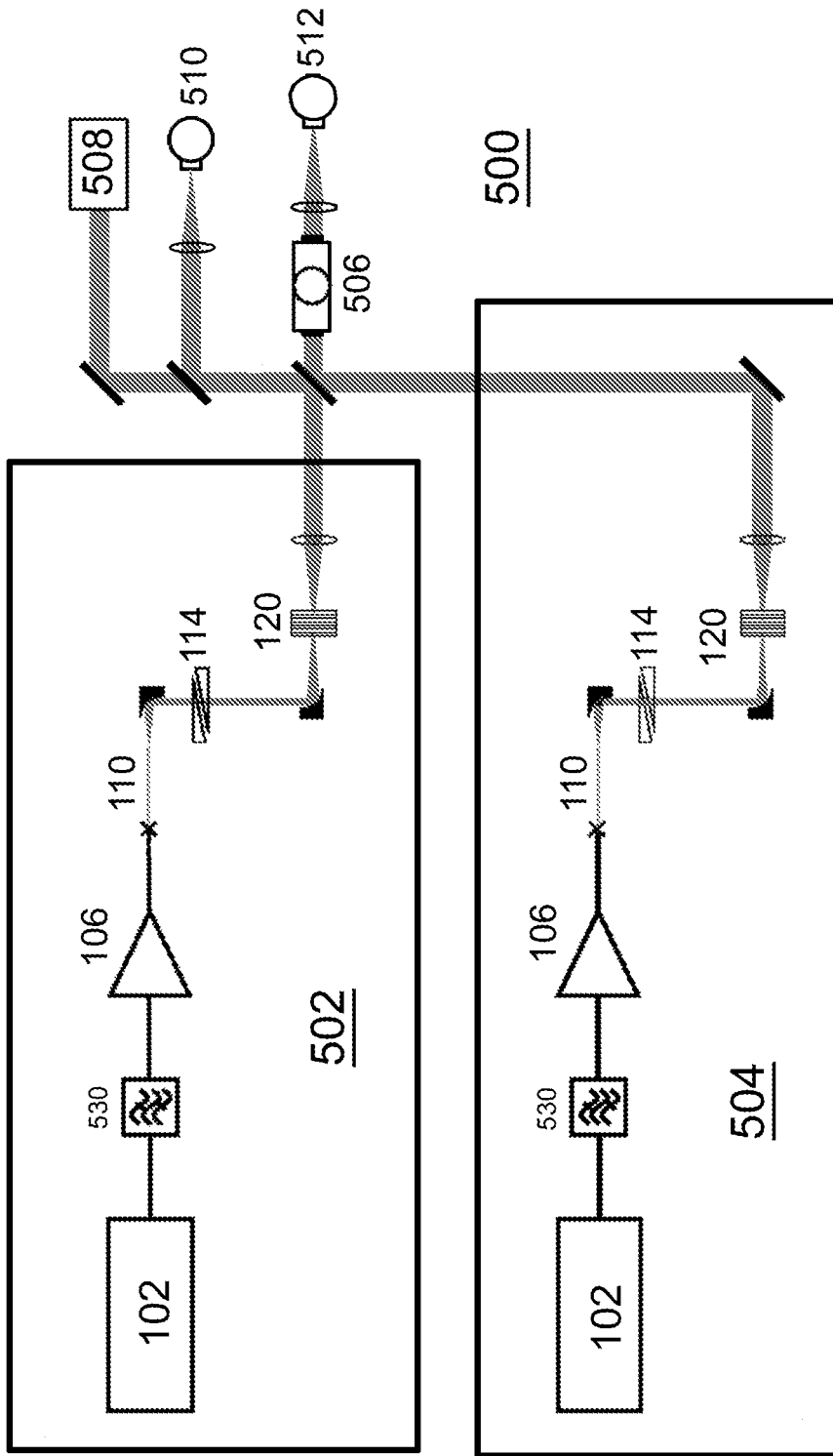Figure 5

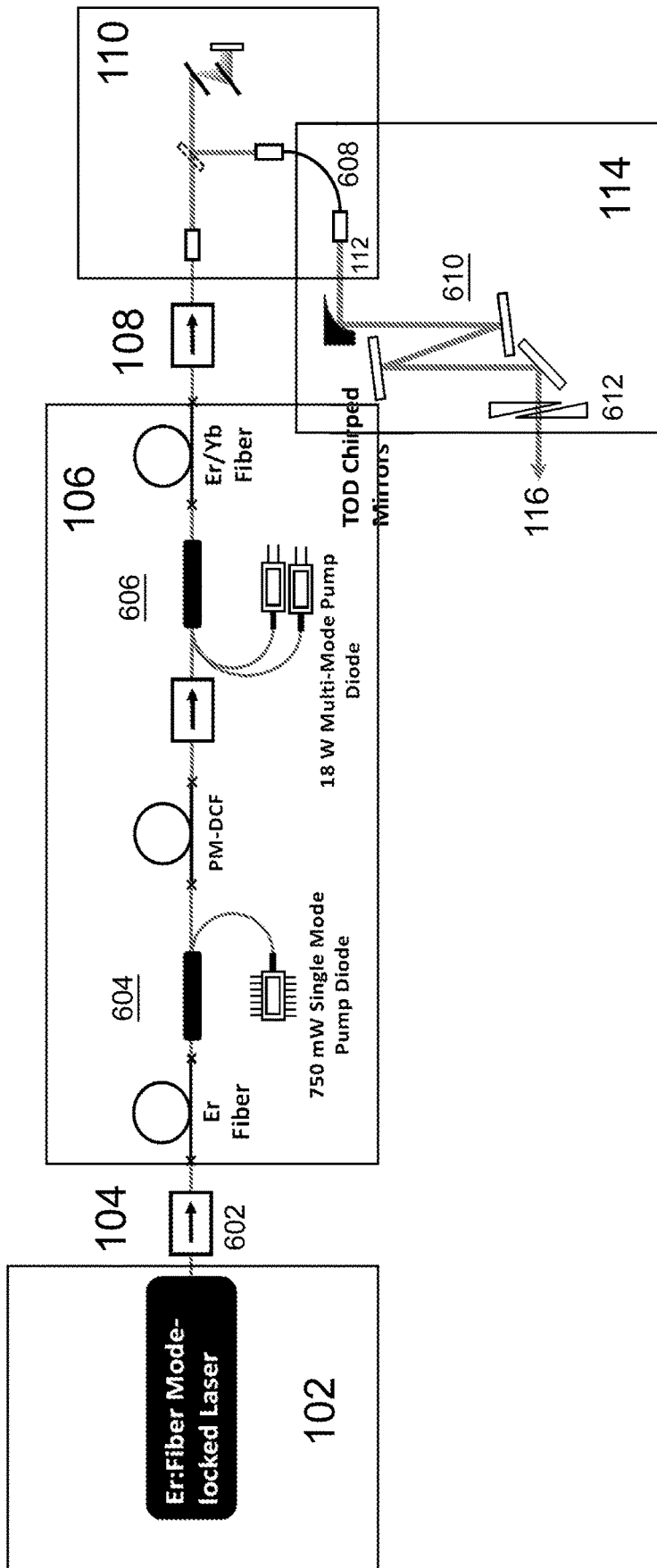Figure 6

800

Ultrashort Pulse Generation
102

116

120

117

810   812

850

814   818

1550 nm CW Laser

803

801   802   804

f_b 820

f_0 808

DCS   806

816

122

Figure 7A
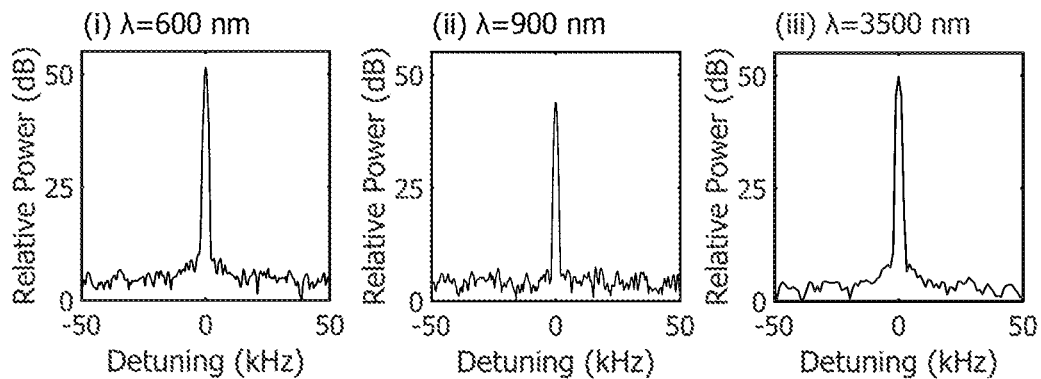

Figure 7B

(i) λ=600 nm   (ii) λ=900 nm   (iii) λ=3500 nm
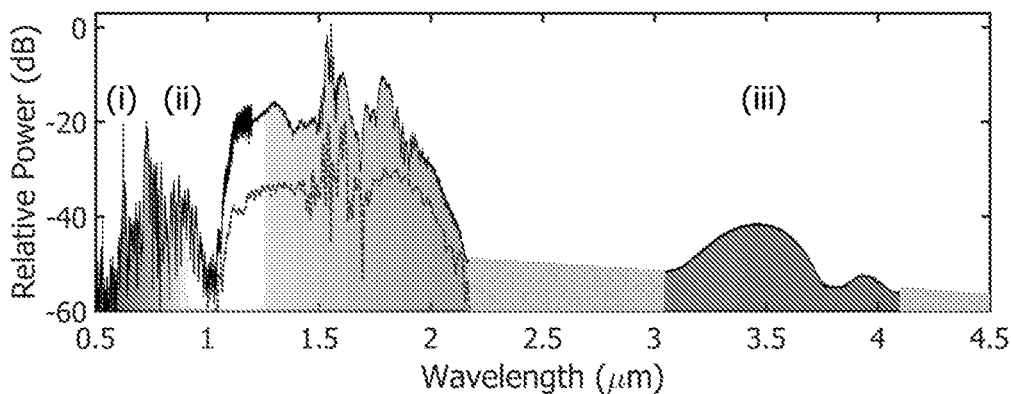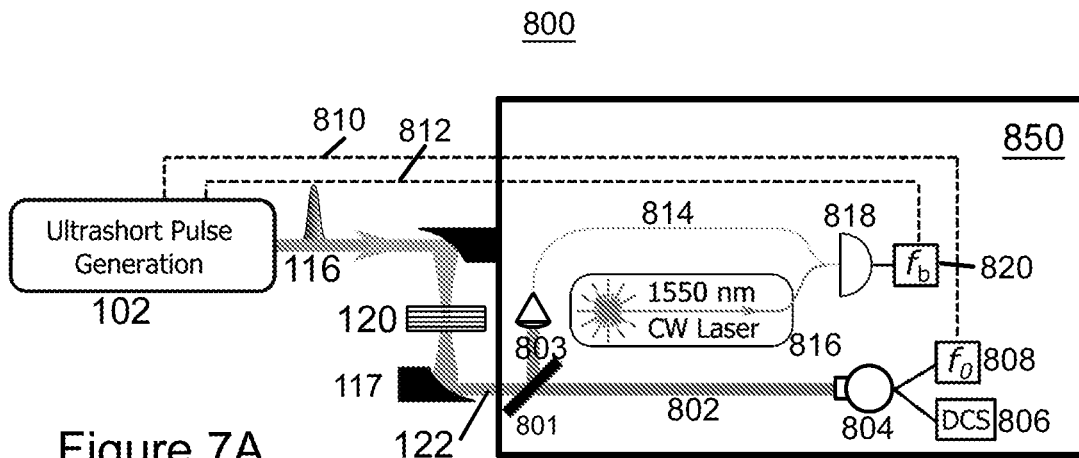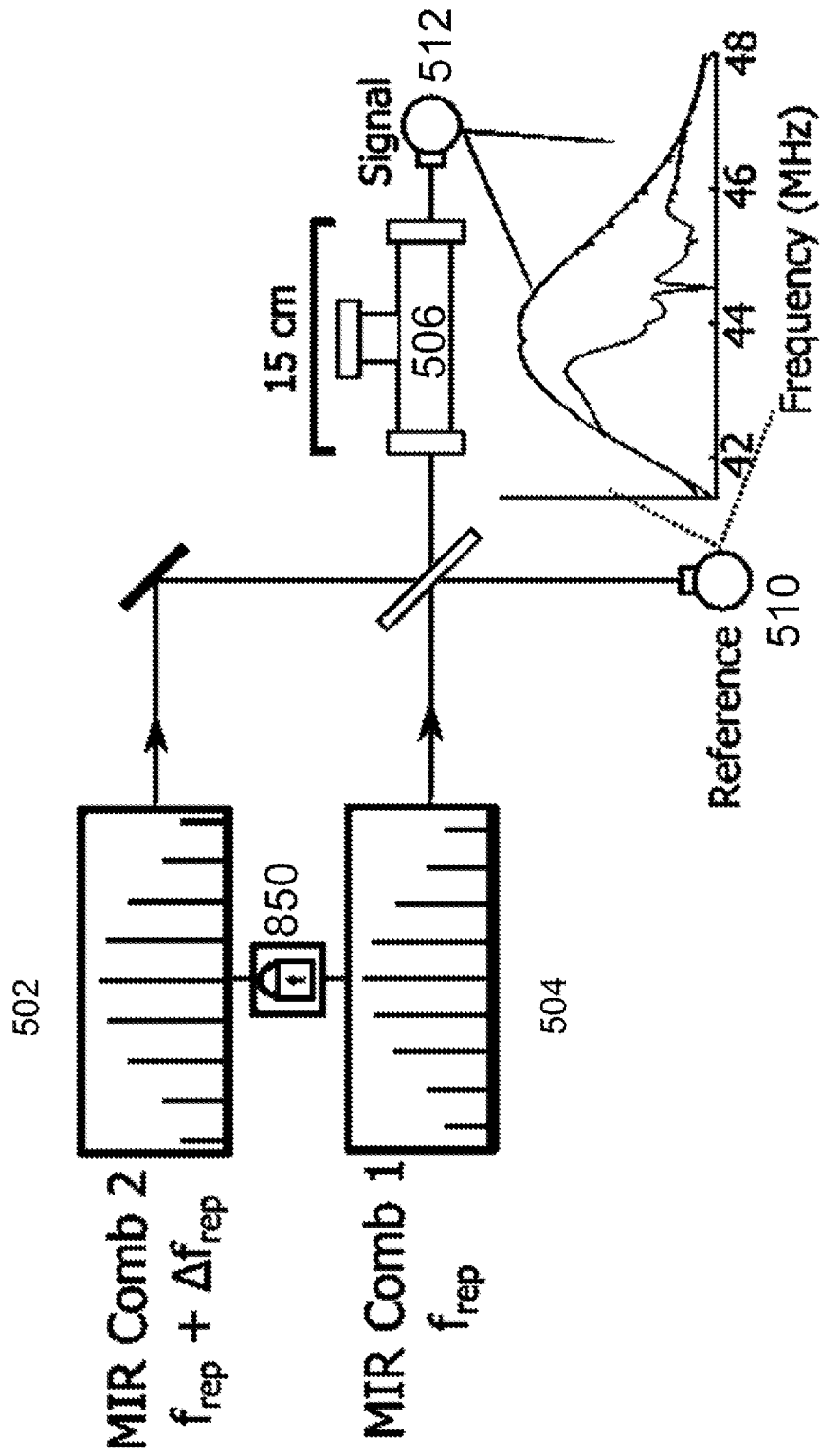
Figure 7C

Figure 7D

METHODS AND APPARATUS FOR GENERATING MID-INFRARED FREQUENCY COMBS

BACKGROUND OF THE INVENTION

Field of the Invention

The present invention relates to the generation of infrared laser frequency combs. In particular, the present invention relates to apparatus and methods for generating mid-infrared laser frequency combs using intra-pulse difference frequency generation (DFG).

Discussion of Related Art

Infrared molecular spectroscopy is a powerful and ubiquitous technique for measuring the chemical makeup and structure in almost any state of matter. In particular, the inter-atomic degrees of freedom within a molecule or compound lead to series of discrete, vibrational states whose resonances are unique identifiers in the mid- to long-wave infrared spectrum spanning the molecular fingerprint region from approximately 6.7 to 20 μm (~1500-500 cm^{-1}). For the past 50 years, Fourier transform infrared spectroscopy (FTIR) using incoherent, thermal light has been a primary tool for determining molecular structure in this spectral region, imparting wide-ranging impact in the physical, chemical, biological, and medical sciences. However, the low brightness of FTIR thermal sources restricts focusability, long-distance propagation, and spectroscopic sensitivity, while the required mechanical delay of FTIR limits the combination of spectral resolution and measurement time. Infrared laser spectroscopy has been pursued over a similar epoch, but simultaneous spectral coverage of the entire fingerprint region remains limited to radiation only available at infrared synchrotron facilities.

Optical frequency comb spectroscopy has been introduced in the past decade as a compelling alternative to FTIR by providing a unique combination of large spectral bandwidth, high frequency precision, and rapid data acquisition that can be integrated with cavity enhancement techniques or long-distance propagation to enhance sensitivity. Significant effort has gone into the development and extension of laser frequency combs deeper into the infrared spectral region beyond 2 μm, with techniques including difference frequency generation (DFG), optical parametric oscillation (OPO), mode-locked quantum cascade lasers, super-continuum generation, and Kerr micro-resonator technology. However, to date, most frequency comb sources have been restricted to wavelengths below 6 μm or have only been able to access discrete portions of the fingerprint region with limited resolution and accuracy. In parallel, there has also been ongoing research to generate broadband multi-terahertz pulses extending up to the mid-IR region through intra-pulse DFG using mode-locked oscillators based on both Ti: sapphire and Er: fiber platforms. However, reported infrared powers from such optically rectified pulses at >10 MHz repetition rates have been limited to the microwatt (μW) scale.

A need remains in the art for apparatus and methods for generating mid-IR laser frequency combs using intra-pulse DFG.

SUMMARY OF THE INVENTION

It is an object of the present invention to provide apparatus and methods for generating mid-IR laser frequency combs using intra-pulse DFG. Such systems include an oscillator configured to generate near-IR mode-locked light pulses, the pulses having a duration under 500 fsec, a normal dispersion broadening element for receiving the near-IR pulses and generating nonlinearly spectral broadened pulses, an anomolous dispersion compression element for receiving the nonlinearly spectral broadened pulses and forming few-cycle, temporally compressed, conditioned pulses and a comb generating element for receiving the conditioned pulses and generating a mid-IR frequency comb by intra-pulse difference frequency generation.

In some embodiments, the oscillator is a Er: fiber laser and the broadening element is a normal dispersion highly nonlinear fiber.

The compression element might be anomalous dispersion fused silica, such as in wedges, or dielectric chirped mirrors.

The comb generating element is generally a $\chi^{(2)}$ Crystal, such as a periodically poled lithium niobate crystal, an orientation patterned GaP crystal, a CSP crystal, or an AgS crystal.

One useful application for mid-IR frequency comb generating systems is dual comb spectroscopy. For these systems a second oscillator, broadening element, compression element, and comb generating element generates a second mid-IR frequency comb, and both combs are directed into an absorption cell containing a sample gas in such a manner as to cause dual frequency comb spectroscopy in the absorption cell. A MCT detector detects the light from the absorption cell.

Apparatus and methods are also provided for locking the frequency combs, using a reference near-IR oscillator for generating a reference near-IR signal, a filter for selecting a near-IR comb tooth from the first mid-IR frequency comb, a heterodyne element for heterodyning the reference near-IR signal and the near-IR comb tooth and generating a heterodyned signal, and a detector for detecting the mid-IR frequency comb. The detector also detects a beat note comprising the carrier envelope offset frequency of the mode-locked laser. A feedback mechanism locks the mid-IR frequency comb using the beat note and the heterodyned signal. This is especially useful in dual comb spectroscopy, where both mid-IR frequency combs are locked according to the beat note and the heterodyned signal.

The present invention enables bright, stabilized mid-IR frequency combs based on intra-pulse DFG using a few-cycle pulse derived from commonplace and robust pulse generation devices (e.g. Er: fiber laser technology). The parametric conversion occurs within a quadratic nonlinear material, for example, periodically poled lithium niobate (PPLN) crystal which employs quasi-phase matching to enhance the light conversion efficiency into the mid-IR regime. Other specific examples of quadratic nonlinear materials are orientation-patterned gallium phosphide (OP-GaP), gallium selenide (GaS), cadmium silicon phosphide (CSP), and silver gallium sulfide (AgS). The invention generates spectra containing up to several mW of average power and spanning 3-27 μm (3300-370 cm^{-1}), with a pathway to coverage across even broader spectral range. The present super-octave bandwidth consists of 500,000 fre-

quency comb modes, providing a spectral resolution down to 100 MHz (0.0033 cm^{-1}). The present invention produces coherent, broadband infrared light exhibiting comparable brightness to an infrared beamline at synchrotron user facilities.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. **1** is a high-level schematic block diagram of an infrared comb source according to the present invention.

FIG. **2** is a schematic diagram illustrating the process of conditioning pulses in the comb source of FIG. **1**.

FIG. **3** is a detailed block diagram of a first embodiment of the comb source of FIG. **1**.

FIG. **4** is a plot showing the performance of the embodiment of FIG. **3** using various χ$^{(2)}$ materials for intrapulse difference frequency generation.

FIG. **5** is a schematic block diagram of a dual comb spectroscopy system based on two comb sources such as those shown in FIG. **3**.

FIG. **6** is a detailed block diagram of the portion of the invention which generates higher power, few-cycle conditioned pulses.

FIG. **7**A is a schematic block diagram of an infrared comb source implementing a beat note as a lock parameter for comb offset frequency. FIG. **7**B is a plot showing the results of the embodiment of FIG. **7**A. FIG. **7**C is a plot showing three carrier envelope offset frequencies from the embodiment of FIG. **7**A. FIG. **7**D is a schematic block diagram of a dual comb spectroscopy systems based upon two infrared comb sources of FIG. **7**A.

DETAILED DESCRIPTION OF THE INVENTION

TABLE 1

Ref. no.	Element
100, 300, 502, 504, 800	Mid-IR comb generating systems
102	Pulse generating laser (near IR)
104/204/304	Near-IR pulses
106	Light amplifier
108/208/308	Amplified near-IR pulses
110	Nonlinear spectral broadening
112/212/312	Broadened pulses
114	Temporal compression
116/216/316	Few-cycle conditioned pulses
117	Off-axis parabolic mirror
120	Intrapulse difference frequency generation
122/222/322	Mid-IR frequency comb
502	First mid-IR comb source
504	Second mid-IR comb source
506	Absorption gas cell
508	FTS
510	MCT, reference spectrum
512	MCT detector
530	Bandpass filter
602	Optical isolator
604	Preamplifier
606	Amplifier
608	ND-HNLF
610	Chirped mirrors
612	Fused silica wedges
800	Mid-IR comb generating system
801	Lowpass filter
802	Comb
804	MCT Detector
806	DCS
808	Offset frequency beat
810	F$_0$ lock parameter
812	Heterodyned lock parameter

TABLE 1-continued

Ref. no.	Element
814	Comb tooth
816	Stable laser signal
818	Photodetector
820	Heterodyned signal
850	Lock dual comb spectroscopy system

Table 1 provides reference numbers and associated elements of the invention.

FIG. **1** is a schematic block diagram of an infrared comb source **100** according to the present invention. Mode-locked pulse generation laser **102** generates near-IR pulses **104**. Amplifier **106** amplifies pulses **104** to generate higher-energy pulses **108**. In general amplifier **106** is utilized after laser **102** because commercially available lasers do not produce high enough energy pulses. As an alternative, amplifier **106** could incorporated into block **102**.

Next, amplified pulses **108** are spectrally broadened by a nonlinear element **110**, for example a normal dispersion highly nonlinear fiber (ND-HNLF) to generate broadened pulses **112**. Nonlinear element **110** is a transparent, dielectric material having a cubic nonlinear response. Next, pulses **112** are temporally compressed by block **114** to generate short, high-power pulses **116**. Pulses **116** are thus few-cycle conditioned pulses which are ready for the intrapulse DFG process (see FIG. **2**). Intrapulse DFG block **120** receives few-cycle conditioned pulses **116** and generates a mid-IR comb **122** by difference frequency generation. Intrapulse DFG block **120** might comprise an orientation patterned GaP (OP-GaP) crystal or a poled lithium niobate (PPLN) crystal.

FIG. **2** is a schematic diagram showing the process of generating mid-IR frequency comb **122** according to the embodiment of FIG. **1**. The top row shows time domain pulses **204-222**, while the bottom row shows frequency domain spectra **304-322**. Thus, for example pulses **104** have a time domain plot **204** and a frequency domain plot **304**. The rest of the reference numbers follow the same pattern.

In one embodiment, the pulse **104** wavelength is around 800 nm-2000 nm. Pulses **104** might be around 100-500 fs. A commercial Er: fiber mode-locked laser generating 1550 nm pulses works well. Generally, the pulses are then amplified by amplifier **106** to generate pulses **108** of around 0.3-10 Watts in energy. Pulses **108** will be about the same length and have about the same spectrum as pulses **104**, just higher energy.

Nonlinear spectral broadening block **110** broadens pulses **112** with normal dispersion as shown in plot **212**, wherein the higher frequency portions of the spectrum are separated from the lower-frequency portions in time. Plot **312** shows that pulses **312** are broader in frequency as well. Nonlinear spectral broadening block **110** is a transparent, dielectric material having a cubic nonlinear response, for example a normal dispersion highly nonlinear fiber (ND-HNLF).

Temporal compression block **114** compresses pulses **112** to form pulses **116**, so that the pulses are short (plot **216**) but still broad in frequency (plot **316**). Temporal compression block **114** is anomalously dispersive and compensates for the normal dispersion and self-phase modulation imparted by nonlinear element **110**. Pulses **116** are thus few-cycle conditioned pulses **116** which are ready for the intrapulse DFG process **120**.

Finally, block **120** generates the mid-IR frequency comb **122** by intrapulse difference frequency generation. This process may be accomplished with an OP-GaP block, a

PPLN crystal, or GaSe. FIG. 4 shows resulting mid-IR frequency combs obtained using each of these elements. FIG. 3—OP-GaP. FIG. 7A—PPLN.

FIG. 3 is a more detailed block diagram of a specific example of a mid-IR frequency comb generation system. Here, pulse generation laser 103 comprises an 100 MHz Er: fiber mode-locked laser generating femtosecond pulses. Amplifier 106 is an Er-doped fiber amplifier (EDFA) which amplifies the pulses to a pulse energy of 3.5 nJ, corresponding to an average power of 350 mW.

The compressed output of amplifier 106 is contained in optical fiber which is spliced directly to nonlinear spectral broadening block 110, in this case 4 cm of normal dispersion highly nonlinear fiber (ND-HNLF). In this embodiment, broadening block 110 generates pulses having a bandwidth of ~600 nm. The positive chirp accumulated within ND-HNLF 110 is compensated for using a pair of anomalous dispersion fused silica wedges for temporal compression 114, resulting in a few-cycle pump pulses 116. Frequency resolved optical gating may be used to measure the duration of the pulses. Pulses 116 have a temporal duration of 10.6 fs, corresponding to a two-cycle pulse.

Intrapulse difference frequency generation 120 is accomplished via OP-GaP in this embodiment. After compression 114, the few-cycle pulse 116 is focused into an OP-GaP crystal with an orientation patterning period of Λ=61.1 μm and a thickness of 1 mm. The OP-GaP crystal exhibits a high nonlinear coefficient (d_{eff}=27 μm/V at 1.3 μm) and broad transparency across the pump and mid-IR wavelength regime, making it ideally suited for intra-pulse down-conversion 120.

Few-cycle pulse 116 is focused into the nonlinear crystal 120, resulting in a nonlinear polarization and forward emission of mid-IR light 122 having an optical period on the order of the temporal duration of the pump pulse. In the frequency domain this corresponds to DFG between the spectral components within the few-cycle, near-infrared pump spectrum. Since the few-cycle pulses occur as a coherent pulse train, the pump spectrum consists of a comb of frequency modes given by v_n=f_0+n×f_{rep}, where f_0 is the carrier-envelope offset frequency and f_{rep} is the repetition rate of the pulse train. The pairwise difference between the nth and mth pump modes yields a mid-IR comb with frequencies v_i=(n−m)×f_{rep}. A critical and advantageous aspect of the intra-pulse difference frequency is that f_0 is subtracted out from the pump field, providing a phase-stable, offset-free mid-IR comb consisting of exact harmonics of f_{rep}.

FIG. 4 shows experiment results using several $\chi^{(2)}$ materials 120 for intrapulse difference frequency generation in a system such as the system shown in FIG. 3. The generated mid-IR light 122 is collected after the $\chi^{(2)}$ crystal 120 and long-pass filtered at 3.6 μm (not shown). This setup generated mid-IR spectra 122 spanning across both infrared atmospheric transmission bands (3-5 μm and 8-12 μm) and containing up to 0.25 mW of power.

A typical spectrum optimized for bandwidth exhibits over 1.5 octaves of bandwidth from 4 to 12 μm. Results are shown for three types of $\chi^{(2)}$ crystal 120: PPLN, GaSe, and OP-GaP. Similar results are obtained with cadmium silicon phosphide (CSP), and silver gallium sulfide (AgS).

FIG. 5 is a schematic block diagram illustrating a dual comb spectroscopy system based on two comb sources 502, 504 similar to the one shown in FIG. 3. Comb source blocks 502 and 504 each include a bandpass filter 530 before amplifier 106, for example a 10 nm bandpass filter to filter the oscillator 102 spectrum bandwidth of $\Delta\lambda$ 40 nm centered

around 1570 nm to a bandwidth of $\Delta\lambda$≈10 nm. The filtered spectra 108 are then stretched temporally in 1 m of PANDA 1550 fiber (not shown) and amplified using a quad-pumped erbium doped fiber amplifier 106 (EDFA). A normal dispersion Er gain fiber is used to enable non-linear amplification and pulse compression. The output of amplifier 106 yields pulses 108 having powers ranging between 300-400 mW depending on the quality of the fiber components and splices. The output of amplifier 106 is spliced to nonlinear spectral broadening block 110, here a normal dispersion, highly non-linear fiber (ND-HNLF) for controlled spectral broadening. The parameters of the HNLF in this embodiment are as follows: the dispersion is D=−1.0 ps/nm km, the dispersion slope is D_s=0.006 ps/nm² km, and the non-linearity is γ≈10.8 (W km)$^{-1}$. The fiber 106 length that gave the best compression (typically 30-50 fs) was spliced directly to the ND-HNLF.

The ND-HNLF 110 was connectorized with a FC/APC connector (not shown) and connected to an off-axis parabolic collimator (not shown) to output-couple the light into free space without imparting unwanted dispersion onto the broadened pulses 112.

The pulses 112 were then compressed 114 with either a bulk fused silica window (5 mm) or a pair of fused silica wedges. The anomalous dispersion from the fused silica material compensated for the normal dispersion and self-phase modulation in the ND-HNLF 110. The compressed pulses 116 were characterized using second harmonic generation FROG. Experimentally, the duration of compressed pulses 116 was 10.6 fs.

After pulse compression, the few-cycle pulses 116 were focused into an orientation patterned GaP (OP-GaP) crystal 120 with a length of 1 mm and an orientation patterning period of Λ=61.1 μm. The pulses were focused using an off-axis parabolic mirror to avoid imparting unwanted dispersion onto the few-cycle pulses 116. The generated mid-IR light 122 was collected with a ZnSe lens (not shown) and filtered using either a 3600 nm or 4500 nm long-pass filter (not shown) to remove the pump spectrum.

Block 502 generates first mid-IR frequency comb 122A and block 504 generates second mid-IR frequency comb 122B. For dual-comb spectroscopy (DCS), the two combs 122A and 122B were combined together using a mid-IR, 50-50 beamsplitter. One optical path from the beamsplitter travelled through a 15 cm absorption cell 506 containing some quantity of a sample gas. After absorption cell 506, the light was collected onto a liquid nitrogen cooled HgCdTe (MCT) detector 512. This is used as the main photo-detector for the DCS experiment. The second path from the beamsplitter travels to either a Fourier transform spectrometer (FTS) 508, where a low-resolution spectrum of the mid-IR light can be measured, or to a second MCT photo-detector 510, where a DCS reference spectrum can be acquired for the absorbance measurement. Note that the two mid-IR frequency combs 122A, 122B are mutually phase coherent, for example by locking their seed Er: fiber mode-locked lasers to a common 1550 nm continuous wave laser. Due to the nature of intra-pulse difference frequency generation according to the present invention, the mid-IR combs are offset free. However, the pump combs still exhibit an offset frequency. Therefore, the carrier-envelope offset frequencies of both combs were stabilized, for example with two inline f-2f interferometers (not shown).

The embodiment of FIG. 5 was used to perform high-resolution spectroscopy in ethanol. In addition to simultaneously measuring the four broadband absorption features in ethanol at atmospheric pressure, it also recorded a highly

resolved ethanol spectrum at a lower backing pressure of 10 torr in order to discern the individual ro-vibrational lines. This is a better result than was found in the literature.

FIG. **6** is a detailed block diagram of the portion of the invention which generates higher-power (1-10 W) few-cycle conditioned pulses **116** which are configured to be ready for the intrapulse DFG process **120** (see FIG. **2**). In this embodiment, pulse generating laser **102** is an Er: Fiber Mode-locked laser generating a near-IR pulse train. An optical isolator **602** is shown to prevent light from leaking back into laser **102**. Light amplifier **106** includes both a lower-power pre-amplifier **604** that employs core-pumped Er: fiber, as well as a higher-power amplifier **606** that employs cladding-pumped Er/Yb: fiber. Nonlinear spectral broadening block **110** comprises temporal compression of the amplified pulses using gratings followed by nonlinear spectral broadening in normal dispersion highly nonlinear fiber (ND-HNLF) **608**. Final temporal compression to few-cycle duration occurs in block **114** which comprises chirped dielectric mirrors **610** and fused-silica wedges **612**.

FIG. **7**A is a schematic block diagram of a mid-IR infrared comb source **800** implementing a beat note **808** as a lock parameter for comb offset frequency. While frequency stabilization using beat note **808** is not required for MIR light generation, it is useful for applications such as dual-comb spectroscopy. A long-pass filter (LPF) **122** allows the mid-IR light **122** to pass and focus onto a MCT detector **804**. Additionally, the offset frequency (f$_0$) is detected here. The light **803** reflected from LPF **122** is coupled into optical fiber **814** to heterodyne with a cavity-stabilized 1.55 μm laser output **816**, which is used to fully stabilize the comb.

FIG. **7**B is a plot showing results obtained with the implementation of FIG. **7**A. FIG. **7**B shows the near infrared spectrum of the few-cycle pulse extending between 1 and 2 microns, as well as a mid-IR spectrum between 3 and 4 microns and visible spectrum in the region of 0.5-1 microns.

FIG. **7**C shows three replicas of the carrier envelope offset frequency of the mode-locked laser **102** which are detected directly in the output spectra **122** at 600 nm, 900 nm and 3500 nm.

Comb source **800** generates a mid-IR frequency comb **122** by driving few-cycle pulses **116** within a $\chi^{(2)}$ material **120** (see FIG. **2**). Frequency combs **122** span 3 μm to 5.5 μm. In addition to generating mid-IR frequency combs, the system also provides the carrier-envelope offset frequency of the driving pulse train **116** directly, which is used to fully stabilize the mode-locked laser **102**. Elements **102-114** generate near-IR pulses **116** as shown in FIGS. **1-6**. The offset frequency beat **808**, measured directly from the MCT detector **804** (along with dual comb spectroscopy **806**), as a consequence of cascaded non-linear processes, allows stabilization of the combs to an in-loop timing jitter <100 as.

In one embodiment, driving pulses **116** in the 1.55 μm region pump periodically poled lithium niobate (PPLN). The system amplifies **106** and spectrally broadens **110** the output **104** of a 100 MHz Er: fiber oscillator **102** to generate 3 nJ few-cycle pulses **116** (10.6 fs, 2.1 cycles) centered at 1.55 μm. The few-cycle pulses **116** are focused with a 25 mm focal length silver-coated off-axis parabolic (OAP) mirror **117** into 1 mm-long PPLN crystal **120**, achieving a minimum beam diameter of 16 μm. The broadband output **120** of the PPLN crystal is collected with another OAP mirror **122**, optically filtered **812**, and measured on a Fourier transform spectrometer (see FIG. **5**) or sent to a liquid nitrogen-cooled mercury cadmium telluride (MCT) detector **804**.

The modes of a frequency comb are defined through two radio frequency parameters, the repetition rate (f$_{rep}$) and the

offset frequency (f$_0$), such that a given mode, n, has a well-defined optical frequency, $v_n = nf_{rep} + f_0$. For DFG that occurs strictly within the original input pulse, the resulting mid-IR light will be "offset-free", meaning the offset frequency of the comb will subtract out in the DFG process. However, due to the high peak intensities within the crystal, cascaded $\chi^{(2)}$ nonlinear processes give rise to additional comb modes throughout the spectrum with which we observe f$_0$ in heterodyne beats at multiple wavelengths, as shown in FIG. **7**C. For example, at 600 nm, we observe the 2f-3f interference, from cascaded QPM 1.8 μm tripling and 1.2 μm (third-order phase-matched) doubling at a beat frequency, fb,

$$f_b = 3 \times (nf_{rep} + f_0) - 2 \times (3/2 nf_{rep} + f_0) = 3f_0 - 2f_0 = f_0 \tag{1}$$

At 900 nm, a f-2f interference results from QPM 1.8 μm frequency doubling beating with slightly broadened comb light. At 3500 nm, we observe an "f-0" beat note between the "offset-free" DFG comb, and DFG between doubled light from 2 μm (with a factor of 2f$_0$) and the original 1.5 μm comb (containing a single f$_0$). The two mid-IR combs heterodyne together on the MCT photo detector to generate an offset frequency tone in the RF spectrum.

System **850** implements optical frequency control at 1.55 μm, which ensures a high degree of mutual coherence necessary for applications like DCS. A near-IR comb tooth **814** heterodyned against a stable 1.55 μm laser **816** signal using photodetector **818** provides one lock parameter f$_b$ **808** as feedback **812**, and an offset frequency beat **808** provides the second feedback, **810**. An additional external f-2f interferometer branch is usually required to detect an offset frequency beat note; however, in this configuration, the need for such a branch is bypassed by utilizing the offset frequencies directly detected throughout the spectrum. The beat note **808** from MCT detector **804** is used for mid-IR spectroscopy to lock the offset frequency of combs **502, 504**, as shown in FIG. **7**D. The integrated phase noise on the offset frequency is 115 mrad, integrated from 50 Hz to 2 MHz, and comparable to other methods of offset frequency stabilization. This demonstrates that using the f$_0$ in the mid-IR results in similar performance to a traditional f-2f lock, allowing for simultaneous comb stabilization and mid-IR spectroscopy from the same detector.

The PPLN crystal **120** contains a set of discrete poling periods, ranging from $\wedge$=24.1-35.6 μm in steps of approximately 0.6 μm. The longer (shorter) poling periods provide QPM for DFG into shorter (longer) wave-lengths in the mid-IR. By tuning across these poling periods, mid-IR light spans 3-5.5 μm (FIG. **2**(*a*)). The maximum power measured in the mid-IR spectra is found to be 1.3 mW. However, on average these spectra yield approximately 500 μW, which is sufficient to saturate high-speed, low-noise MCT detectors in lab-scale linear spectroscopy measurements.

FIG. **7**B shows the full optical spectrum after propagation through one PPLN poling period. The spectrum of the input few-cycle pulse spans 1-2 μm (dashed curve). Below 1 μm, the sharp spectral features come from higher-order quasi phase-matched (QPM) harmonic generation from the input spectrum. The mid-IR light arises from difference-frequency mixing within the input spectrum.

The bandwidth generated (300-500 nm) in the mid-IR is limited by group velocity dispersion elongating the pulse (112 fs²/mm at 1.55 μm) and the phasematching bandwidth for a single poling period. Instead, by using crystals with a chirped poling period (aperiodic poling), we significantly increase the QPM bandwidth. To implement a chirped poling period, we use a fanout PPLN tilted at approximately

45 degrees, such that the pulse experiences different poling periods as it passes through the crystal. Through these chirped grating periods, bandwidths >1000 nm are generated. The chirp, and thus the generated bandwidth, is limited by the crystal angle in the current implementation. A chirp of the opposite sign results in a significantly more modulated spectrum.

While the exemplary preferred embodiments of the present invention are described herein with particularity, those skilled in the art will appreciate various changes, additions, and applications other than those specifically mentioned, which are within the spirit of this invention. For example, the potential for achieving still broader spectral coverage and higher powers should be enabling for a wide range of diagnostics of chemical and biological species, including applications in astronomical heterodyne spectroscopy and nonlinear spectroscopy.

What is claimed is:

1. An apparatus for generating a mid-IR frequency comb comprising:
   an oscillator configured to generate near-IR mode-locked light pulses, the pulses having a duration under 500 fsec;
   a normal dispersion broadening element for receiving the near-IR pulses and generating nonlinearly spectral broadened pulses;
   an anomalous dispersion compression element for receiving the nonlinearly spectral broadened pulses and forming few-cycle, temporally compressed, conditioned pulses;
   a comb generating element for receiving the conditioned pulses and generating a first mid-IR frequency comb by intrapulse difference frequency generation;
   a reference near-IR oscillator for generating a reference near-IR signal;
   a filter for selecting a near-IR comb tooth from the first mid-IR frequency comb;
   a heterodyne element for heterodyning the reference near-IR signal and the near-IR comb tooth and generating a heterodyned signal;
   a detector for detecting the first mid-IR frequency comb;
   wherein the detector further detects a beat note comprising a carrier envelope offset frequency of the mode-locked laser; and
   a feedback mechanism for locking the first mid-IR frequency comb using the beat note and the heterodyned signal.

2. The apparatus of claim 1 wherein the oscillator is a Er:fiber laser.

3. The apparatus of claim 1 wherein the broadening element comprises a normal dispersion highly nonlinear fiber.

4. The apparatus of claim 1 wherein the compression element comprises anomalous dispersion fused silica.

5. The apparatus of claim 1 wherein the compression element comprises anomalous dispersion fused silica wedges.

6. The apparatus of claim 1 wherein the compression element comprises anomalous dispersion dielectric chirped mirrors.

7. The apparatus of claim 1 wherein the comb generating element comprises a poled lithium niobate crystal.

8. The apparatus of claim 1 wherein the comb generating element comprises an orientation patterned GaP crystal.

9. The apparatus of claim 1 wherein the comb generating element comprises one of the following nonlinear crystals: a $CdSiP_2$ crystal; an AgS crystal; or a GaSe crystal.

10. The apparatus of claim 1 further comprising:
   a second oscillator, a second broadening element, a second compression element, and a second comb generating element for generating a second mid-IR frequency comb;
   an absorption cell containing a sample gas;
   optics for directing the first mid-IR frequency comb and the second mid-IR frequency comb into the absorption cell in such a manner as to cause dual frequency comb spectroscopy in the absorption cell; and
   a mercury cadmium telluride (MCT) detector for detecting light from the absorption cell.

11. A method of generating a mid-IR frequency comb comprising the steps of:
   (a) generating near-IR mode-locked light pulses, the pulses having a duration under 500 fsec;
   (b) receiving the near-IR pulses and generating nonlinearly spectral broadened pulses having normal dispersion;
   (c) receiving the nonlinearly spectral broadened pulses and forming few-cycle, temporally compressed, conditioned pulses while applying anomalous dispersion to account for the normal dispersion of step (b);
   (d) receiving the conditioned pulses and generating a first mid-IR frequency comb by intrapulse difference frequency generation;
   (e) generating a reference near-IR signal;
   (f) selecting a near-IR comb tooth from the first mid-IR frequency comb;
   (g) heterodyning the reference near-IR signal and the near-IR comb tooth and generating a heterodyned signal;
   (h) detecting the first mid-IR frequency comb, and detecting a beat note comprising a carrier envelope offset frequency of the mode-locked laser; and
   (i) locking the first mid-IR frequency comb using the beat note and the heterodyned signal.

12. The method of claim 11 wherein step (a) is performed by an Er:fiber laser.

13. The method of claim 11 wherein the step (b) is performed with a normal dispersion highly nonlinear fiber.

14. The method of claim 11 wherein step (c) is performed with either anomalous dispersion fused silica or dielectric chirped mirrors.

15. The method of claim 11 wherein step (d) is performed with one of the following: a poled lithium niobate crystal; an orientation patterned GaP crystal; a $CdSiP_2$ crystal; or an AgS crystal.

16. The method of claim 11, further including the steps of
   (i) generating a second mid-IR frequency comb according to steps (a)-(d);
   (k) providing a sample gas in an absorption cell;
   (l) directing the first mid-IR frequency comb and the second mid-IR frequency comb into the absorption cell in such a manner as to cause dual frequency comb spectroscopy in the absorption cell; and
   (m) detecting light from the absorption cell.

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