High-power mid-infrared frequency comb source based on a femtosecond Er:fiber oscillator

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We report on a high-power mid-infrared (MIR) frequency comb source based on a femtosecond (fs) Er:fiber oscillator with a stabilized repetition rate of 250 MHz. The MIR frequency comb is produced through difference frequency generation in a periodically poled MgO-doped lithium niobate crystal. The output power is about 120 mW, with a pulse duration of about 80 fs and spectrum coverage from 2.9 to 3.6 μm, and the single comb mode power is larger than 0.3 μW over the range of 700 nm. The coherence properties of the produced high-power broadband MIR frequency comb are maintained, which was verified by heterodyne measurements. As the first application, the spectrum of a ~200 ppm methane–air mixture in a short 20 cm glass cell at ambient atmospheric pressure and temperature was measured.

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Currently, there is a large demand for gas detection systems in the mid-infrared (MIR) in many areas of science and technology. For these applications, high repetition rate femtosecond (fs) lasers and frequency combs are being developed actively due to fast data acquisition rates, high sensitivity, and multitarget detection properties inherent to broadband frequency comb spectroscopy [1,2]. The 3–4 μm MIR range is of particular interest since it contains the strong absorption features of the C-H stretching vibrational mode of methane (v_3 band) and many other more complex hydrocarbons. For a multitude of measuring tasks in the booming natural gas industry, agriculture, atmospheric and geosciences research, methane detection with real time monitoring, and quantifying different hydrocarbon isotopes are important [3,4].

Different versions of frequency comb spectroscopies have been developed since the invention of the frequency comb [2,5–9]. Broadband MIR frequency combs provide useful light sources for many spectroscopic applications. In particular, several MIR sources using single pass difference frequency generation (DFG) have been developed and are attractive because of their relative simplicity and the benefit of passive carrier-envelope offset frequency stabilization [10–15]. If the pump and signal fields are phase coherent and originated from the same source, the generated idler field is carrier-envelope phase slip free and requires only stabilization of the comb spacing, which is relatively easy to implement by stabilizing the source repetition rate. Hence, it was shown to provide a frequency synthesizer in the MIR and can be used in frequency standard applications [16]. Many DFG MIR sources exhibited a large tuning range with various nonlinear crystals, but some of them have a low power level of around milliwatt or less, and the coherence properties are not demonstrated [10–13] or only demonstrated over a few nanoseconds [14]. The DFG MIR sources based on Yb:fiber lasers provide high power of about 100 mW around 3 μm [11,12], but the coherence was reduced and even lost due to the Raman shifting [11].

In this Letter, we demonstrate a high-power MIR frequency comb source based on a fs Er:fiber oscillator with a stabilized repetition rate of 250 MHz. Applying self-phase modulation to up-convert a portion of the output from the Er:fiber oscillator, the coherence properties of the produced high-power broadband MIR frequency comb are maintained, which was verified by heterodyne measurements. In addition, we measured the spectrum of a ~200 ppm methane–air mixture in a short 20 cm glass cell at ambient atmospheric pressure and temperature as the first application.

A simplified diagram of the MIR fs laser is presented in Fig. 1. Intense ultrashort pulses centered at ~1.05 μm and ~1.55 μm are generated from a 250 MHz mode-locked Er:fiber oscillator. Half of the output from the Er:fiber oscillator is amplified by the first Er-doped fiber amplifier (EDFA) to about 450 mW. The amplified pulses are coupled into a highly nonlinear fiber (HNLF) to broaden the spectrum and generate the required ~1.05 μm seed pulses. Due to the process of self-phase modulation, the comb characteristics of the original radiation at ~1.55 μm are transferred to the seed at ~1.05 μm. After filtering, the ~1.05 μm seed pulse is amplified by a high-power Yb-doped fiber amplifier (YDFA) to about 1.2 W. After an isolator and a compressor, the pulse duration of pump pulses centered at ~1.05 μm is ~90 fs. The other half of the output from the Er:fiber oscillator is amplified by the second EDFA to about 450 mW to generate single pulses centered at ~1.55 μm; the pulse duration is ~60 fs. The DFG occurs in a periodically poled MgO-doped

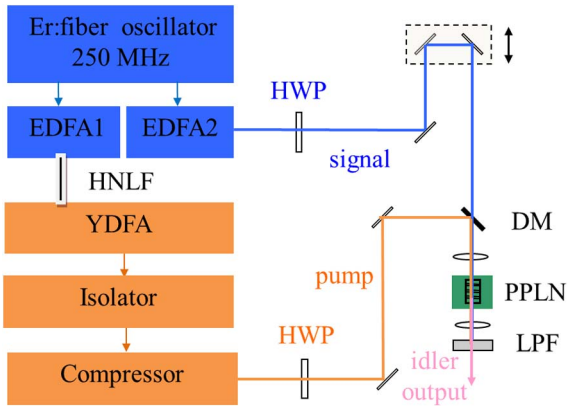

Fig. 1. Simplified diagram of MIR fs laser. EDFA, Er-doped fiber amplifier; YDFA, Yb-doped fiber amplifer; HNLF, highly nonlinear fiber; HWP, half-waveplate; DM, dichroic mirror; PPLN, periodically poled lithium niobate; LPF, longpass filter.

lithium niobate (MgO:PPLN) crystal, where the pump pulse and signal pulse overlap in time and space, resulting in an idler wave in the MIR ($\lambda_i^{-1} = \lambda_p^{-1} - \lambda_s^{-1}$). Pump and signal pulses are combined at a dichroic mirror before being collinearly focused into the MgO:PPLN crystal. Temporal overlap of the pulses is achieved by introducing a suitable optical delay in the path of the signal pulse. Since both pump and signal pulses are not completely converted, a Ge long-pass filter (>2.4 μm) is used to select the idler pulses in the MIR. The DFG system is placed on a water cooled breadboard to increase the long term stability.

To characterize the MIR fs laser we measured the spectrum, recorded the interferometric autocorrelation trace, and verified the coherence of the MIR comb with heterodyne beat experiments.

We measured the spectrum of the MIR comb presented in Fig. 2(b) with a 0.3 m McPherson 218 scanning monochromator and a sensitive thermal power meter (Ophir, 3A). The absorption features in the spectrum are due to water vapor in the laboratory environment (~20% relative humidity, ~0.6% concentration in air, ~6 m path

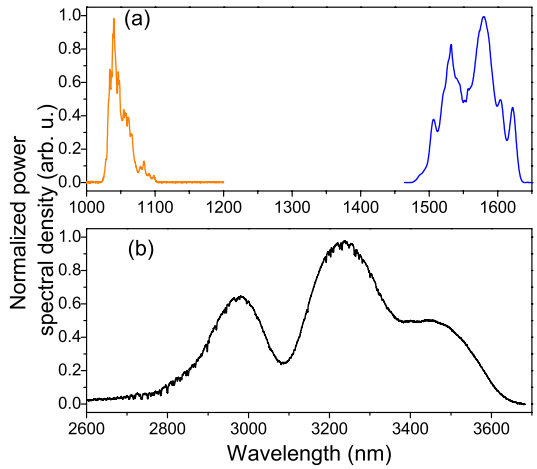

Fig. 2. Spectra of the DFG system. (a) Spectra of pump (orange, left) and signal (blue, right). (b) Spectrum of the MIR fs laser. The absorption features in the spectrum are due to water vapor in the laboratory environment.

length). By tuning the temperature of the MgO:PPLN crystal, the output spectrum and power can be slightly changed through different quasi-phase matching conditions. Experimentally, we determine the temperature of ~90°C for about 120 mW of maximum output power in the spectral range from 2.9 to 3.6 μm, showing a strong peak at the methane absorption band at ~3.3 μm, which corresponds to >0.3 μW/mode for a bandwidth of ~700 nm, and is higher than the value obtained for a similar DFG MIR source with a 100 MHz repetition rate [15].

The temporal characteristics of the MIR pulse were measured with a Michelson-type interferometric autocorrelator using two-photon absorption with an InGaAs photodiode (Hamamatsu, G8376-03). The interferometric autocorrelation trace is presented in Fig. 3 and corresponds to a pulse duration of about 80 fs, assuming a Gaussian pulse shape.

To verify the coherence we converted the MIR comb to a near-infrared (NIR) comb by second harmonic generation (SHG) with a $AgGaS_2$ crystal, because a fast MIR photodetector with the required bandwidth, an accessible narrow-linewidth MIR continuous wave (CW) laser source, as well as polarization optics for ~3.3 μm, are lacking. About 500 μW of SHG radiation centered at ~1.6 μm was generated by focusing the MIR comb into a 1 mm thick $AgGaS_2$ crystal with type I phase matching (Eksma Optics, AGS-401H). The heterodyne beat note between the SHG comb and a 1.602 μm CW distributed feedback diode laser (NEL, NLK1556STG) was detected with a fast InGaAs detector (Menlo Systems, FPD510-F) of DC-250 MHz bandwidth. The heterodyne beat note is presented in Fig. 4(a). The resolution bandwidth is 300 kHz, and the sweep time is 20 ms.

To investigate possible coherence degradation in the nonlinear broadening stage and the DFG process, the heterodyne beat note between the signal comb and the CW laser was detected under the same condition, and is presented in Fig. 4(b). Because the power per comb mode of the signal comb is much larger than the SHG comb, the corresponding beat note is stronger. The 3 dB linewidths of both beat notes are ~5 MHz, demonstrating at least a few hundred nanoseconds coherence time of the MIR source, which is largely limited by the nonstabilized frequency of the CW laser. More rigorous measurements can be performed to confirm low

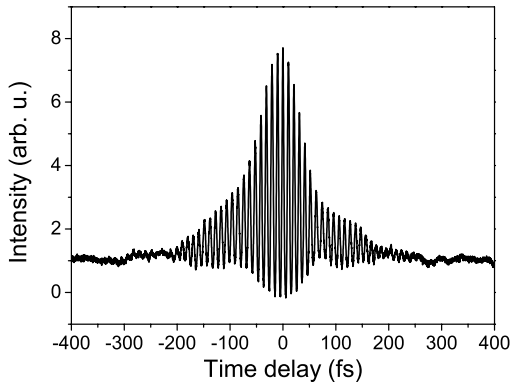

Fig. 3. Interferometric autocorrelation trace of the MIR fs laser. The autocorrelation trace corresponds to a pulse duration of about 80 fs assuming a Gaussian pulse shape.

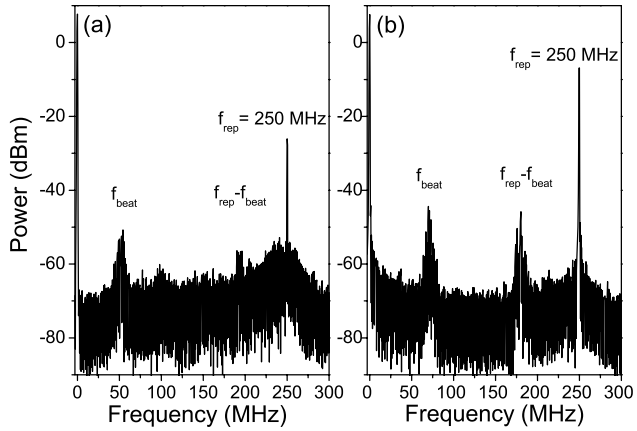

Fig. 4. Beat notes between the CW laser and the NIR frequency combs (a) SHG comb converted from the MIR comb and (b) signal comb. The resolution bandwidth is 300 kHz, and the sweep time is 20 ms.

phase-noise transfer from the NIR to the MIR with a frequency stabilized CW laser.

As the first application of the high-power MIR frequency comb, we measured the spectrum of ~200 ppm methane mixed with air in a short fused silica glass cell of 20 cm length at room temperature by inserting the sample gas cell in front of the scanning monochromator. The absorption spectrum is presented in Fig. 5(a). Because fused silica has strong absorption above 3.5 μm, the spectrum in Fig. 5(a) shows strong absorption in this region compared with Fig. 2(b). The experimentally determined transmittance is depicted in Fig. 5(b), and its comparison with calculations from the HITRAN data base [17] shows good agreement between measurement and literature values.

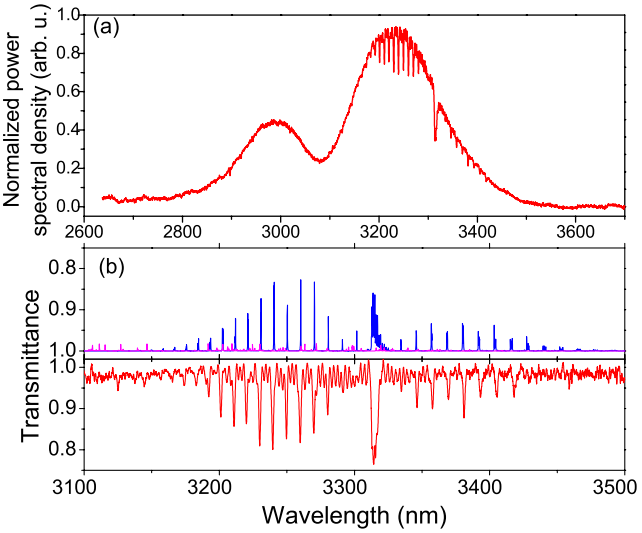

Fig. 5. (a) Methane and water vapor absorption spectrum and (b) experimentally determined transmittance (red, bottom) compared with calculations from HITRAN database. The HITRAN calculation is inverted for clarity. The calculations are done for the experimental conditions: blue (top, spiked curve), 200 ppm methane in a 20 cm glass cell; purple (horizontal), 0.6% water vapor in air, 6 m path length.

In summary, we demonstrated a high-power MIR frequency comb source based on a fs Er:fiber oscillator with a stabilized repetition rate of 250 MHz. We substantiated the coherence of the MIR frequency comb and verified its suitability for spectroscopic applications by a straightforward experiment of detecting methane in a short glass cell. The measured fundamental vibrational mode of methane at ~3.3 μm has a much larger absorption strength (~150:1) than the overtone in the NIR at ~1.65 μm [17], in which several parts per billion detection sensitivity has been demonstrated with a NIR frequency comb [6]. With the developed MIR frequency comb source, and signal improvement techniques, such as multipass [8] or cavity enhancement [6,7], as well as the development of a fast and sensitive MIR detector, several parts per trillion level of the detection sensitivity is possible. Many techniques can benefit from the developed MIR frequency comb source, such as frequency metrology and frequency standard [16], dual frequency comb spectroscopy [5,7,8], upconversion spectroscopy [9], and provide an ultrafast, highly sensitive platform for the gas detection and its applications.

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