Ti:sapphire laser intracavity difference-frequency generation of 30 mW cw radiation around 4.5 μm

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A cw mid-IR coherent source based on difference-frequency generation is designed and characterized. For mid-IR generation, a periodically poled MgO:LiNbO₃ crystal is placed inside a compact Ti:sapphire laser cavity. This provides high-power pump radiation for the nonlinear process. Optical injection by an external-cavity diode laser ensures single-frequency operation of the Ti:sapphire laser, while signal radiation is provided by a fiber-amplified Nd:YAG laser. Mid-IR radiation can be generated with 3850–4540 nm tuning range, narrow linewidth, Cs-standard traceability, and TEM₀₀ spatial mode. 30 mW power is obtained at 4510 nm. © 2010 Optical Society of America

Mid-IR coherent sources are ideal tools for spectroscopic interrogation and manipulation of molecules, that possess strong and rich ro-vibrational absorption bands in this spectral window. To date, several kinds of mid-IR sources based on nonlinear frequency generation have been developed [1–7]. In the last few years, quantum-cascade lasers (QCLs) [8] have been demonstrated to be compact, stand-alone mid-IR sources that can be operated in cw mode, at room temperature, with watt-level power [9], and intrinsically narrow emission linewidth [10]. However, for accurate frequency measurements in high-resolution and high-sensitivity experiments (e.g., sub-Doppler spectroscopy of weakly absorbing molecular transitions), Cs-standard traceability, frequency stability, and relatively high power are needed all together. Actually, none of the mid-IR sources identified has all the desired features.

Recently, we demonstrated operation of a comb-referenced cw IR tunable source emitting less than 80 μW at 4.5 μm wavelength [11]. In this work, we demonstrate that the IR power can be increased by more than 2 orders of magnitude, up to 30 mW at 4510 nm, while preserving the very peculiar features of our single-pass difference-frequency generation (DFG) source. In order to increase the generated IR power, we chose to exploit the Ti:sapphire laser intracavity circulating power by putting the nonlinear crystal inside the cavity itself, following a theoretically proposed scheme [12]. To the best of our knowledge, this is the first experimental work reporting on a DFG process inside a cw laser cavity. Placing a nonlinear crystal in a laser cavity may increase the challenges due to the increased optical losses. Moreover, such losses prevent addition of other optical components (e.g., Lyot filters, thin etalons, etc.) that are especially needed for single-frequency operation of broadband-gain Ti:sapphire lasers. In this work, we show that these nontrivial issues can be addressed.

Following an original design reported in 2002 by Cummings et al. [13], in the last few years we have fabricated a cw ring Ti:sapphire laser, which is optically injected by an external-cavity diode laser (ECDL), and characterized its intensity-noise properties [14]. At the secondary waist position of the laser cavity, we placed a periodically poled (PP) MgO:LiNbO₃ crystal, which is not affected by photorefractive damage due to its 5% MgO doping level. The experimental layout is shown in Fig. 1. The dashed box represents the baseplate of the Ti:sapphire laser cavity, which is thermoelectrically stabilized within 1 mK around room temperature. The Ti:sapphire gain crystal is water-cooled to efficiently remove the excess absorbed power from the 532 nm pump. The transmission value T of the output coupler (about 1.2% at 861 nm) was chosen as the best trade-off between a large power enhancement factor and a good coupling efficiency of the injecting ECDL. The former is maximum for T = 0, while the latter is maximum when T equals all other round trip powers.
losses, including the pump depletion in the DFG process. Indeed, these losses experimentally amount to about 2.4% at 10 W signal power. The PP crystal (dimensions, \( l \times w \times t = 20 \text{ mm} \times 9.8 \text{ mm} \times 2 \text{ mm} \); poling periods, 23.0, 23.1, 23.2, 23.3, 23.4, 23.5, 23.6 \( \mu \text{m} \)) is Brewster-cut for \( \lambda_p \). The quasi-phase-matched DFG process is achieved by using the 23.3 \( \mu \text{m} \) period and thermoelectrically stabilizing the crystal at 28.4 \( ^\circ \text{C} \). We measured the sagittal pump, signal, and idler beam waist inside the crystal as \( w_{p,s,i} = 110, 65, \) and 70 \( \mu \text{m} \), respectively. An idler beam waist larger than the signal waist is determined by the pump and signal beam sizes along \( x \) and \( y \) and by diffraction at each crystal propagation \( z \) position during the nonlinear generation process [15]. The pump, signal, and idler beams are angularly separated, due to dispersion at the output facet of the crystal. The idler beam is reflected out of the Ti:sapphire cavity by a gold mirror and is then collimated by lens L3 (\( f = 100 \text{ mm} \)).

The Ti:sapphire laser is injection-locked by a fiber-coupled ECDL with 834–862 nm tuning range, thus permitting tuning of the idler within the 3850–4540 nm range. However, the ECDL was operated constantly at the 861 nm wavelength (corresponding to an idler wavelength of 4510 nm) and 60 mW power throughout all measurements. We chose this wavelength value primarily to test the achievable DFG efficiency in the least favorable wavelength bound of PP-MgO:LiNbO\(_3\), due to the combined effects of the 1/\( \lambda_i^2 \) dependence of the DFG process and strong crystal absorption. An external 40 dB optical isolator is required to avoid feedback effects from the Ti:sapphire cavity, in addition to the internal one, having the same isolation level. A monolithic-cavity Nd:YAG laser seeds a Yb-doped fiber amplifier that can deliver a nominal power of 10 W, providing the signal radiation for the DFG process. The pump laser is phase-locked to the signal laser by use of an optical frequency comb as a transfer oscillator (see [11] and references therein for details). Lenses L1 (\( f = 300 \text{ mm} \)) and L2 (\( f = 250 \text{ mm} \)) perform an optimal matching of the Nd:YAG and ECDL beams to the Ti:sapphire cavity mode. The cavity length is actively stabilized to keep it resonant with the injecting ECDL, by the polarization-based Hänisch–Couillaud technique [16]. Proportional-integral processing electronics feeds the correction signal back to the PZT-mounted dichroic mirror, which is highly transmissive for the signal and highly reflective for the pump. A locking bandwidth as high as 8 kHz was achieved.

Figure 2 shows the characterization of the DFG source in terms of both generated power and beam profile. The intracavity power reported in the \( x \)-scale of Fig. 2 has been inferred from the measured Ti:sapphire power and the output-coupler transmission value. Here we would like to state the key point of the present work, i.e., that placing the nonlinear crystal inside an active laser cavity boosts the pump power (and that of the idler as well) considerably in comparison to the single-pass scheme. With the waist dimensions reported above, the confocal parameters are much longer than the crystal and the plane-wave approximation can be adapted to calculate the DFG efficiency. Taking into account the absorption from the PP crystal (assuming it to be the same as non-doped LiNbO\(_3\) [17]) and assuming a mean confocal parameter, with \( P_p = 40 \text{ W} \) and \( P_s = 9.8 \text{ W} \) in the theoretical

![Image](image-url)

**Fig. 2.** (Color online) Characterization of the DFG source. Generated idler power curves at 4.5 \( \mu \text{m} \) with 23.3 \( \mu \text{m} \) poling period for different signal powers. Data points in each curve correspond to \( P_{32} \) ranging from 1 to 5 W in 0.5 W steps. The dashed line shows the decreasing \( P_p \) with increasing \( P_s \) due to the DFG-induced intracavity losses at wavelength \( \lambda_p \). Inset, far-field beam profile.

The idler beam profile in Fig. 2 was recorded with a pyroelectric camera looking at the far-field IR radiation scattered by a semitransparent screen placed across the beam. With the knife-edge method, we measured the beam shape after the collimating lens L3 along both \( x \) and \( y \), at different \( z \) propagation distances. The measurements of the beam profile yield a quasi-\( \text{TEM}_{00} \) mode, with a slight astigmatism. Moreover, we have coupled the idler beam to a high-finesse mid-IR Fabry–Perot cavity, achieving a transmission value of about 40%, in agreement with the 41% value based on the 270 ppm mirror losses (100 ppm absorption, and 170 ppm transmission). Comparison of this measured transmission with that measured (35%) for our previous single-pass DFG through the same cavity [11], shows that the Ti:sapphire cavity guarantees an idler beam with a near-\( \text{TEM}_{00} \) mode, differently from the single-pass DFG source, where the pump diode laser was not fiber coupled and had a non-Gaussian profile. In addition, transverse modes were not detected to a barely visible level.

To compare the stability of the present intracavity source with the single-pass one, we measured the frequency-noise power spectral density (PSD) with the same procedure as before. The resonance peak of the Fabry–Perot cavity is used as a frequency-to-amplitude converter. Figure 3 shows the measured frequency noise. Since a side-of-fringe discriminator cannot discriminate between pure frequency and amplitude noises, we checked that the contribution of amplitude noise was negligible. A direct comparison with the corresponding spectrum measured for the single-pass DFG source (see Fig. 2 in [11]) shows that a discrepancy is found at Fourier frequencies in the 1–10 kHz range, where the present source PSD is almost an order of magnitude larger. We did an investigation to

formula [18], \( P_i = 44 \text{ mW} \), in reasonable agreement with the measured value of 30 mW. The calculated IR power that can be generated at shorter wavelengths is higher (e.g., at 3850 nm it is more than double that at 4510 nm), due to the 1/\( \lambda_i^2 \) dependence of the DFG efficiency and to the weaker absorption of the MgO:LiNbO\(_3\) material.
search for possible sources of such excess frequency noise. First, we quantitatively checked the injection purity level of the fiber amplifier and the Ti:sapphire laser from the Nd:YAG laser and the ECDL, respectively. The measurements were performed with a heterodyne scheme: a portion of the injecting laser frequency was shifted by 200 MHz with an acousto-optic modulator; the beat note with the amplifier/laser injected by the unshifted laser was detected by a fast avalanche photo-diode and recorded by a Fourier-transform spectrum analyzer. In the former case, 98% of the fiber-amplifier emitted power was at the Nd:YAG laser frequency (20 kHz span around the carrier). In the latter case (see inset in Fig. 3), 98.6% of the Ti:sapphire laser emitted power was at the ECDL frequency (10 MHz span around the carrier).

In conclusion, we have reported on a tunable mid-IR coherent source based on intracavity DFG, with narrow linewidth, Cs-standard traceability, and up to 30 mW power at 4.5 μm. Possible applications range from manipulation of cold molecules 19–21 to detection of rare isotopologues of molecular species absorbing in that spectral region. Indeed, our recently developed high-sensitivity spectroscopic technique 22 is already benefiting from the present mid-IR source.

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