Principles, advances and applications of ultrafast mid-infrared lasers

Irina T. Sorokina[1] and Evgeni Sorokin[2]

[1] *Department of Physics, NTNU, Trondheim, Norway*

[2] *Photonics Institute, TU Vienna, Austria*
E-mail: sorokina @ntnu.no

Abstract: The talk reviews principles and advances in generation and applications of ultrashort pulses and frequency combs directly from the solid-state oscillator based on Cr^{2+}ZnSe laser, and highlights radical efficiency and sensitivity increase when applied to high-resolution molecular spectroscopy.

Keywords: Cr:ZnSe, ultrashort pulses, mid-infrared lasers

1. INTRODUCTION

The mid-infrared (mid-IR) wavelength range, which is often called a “molecular fingerprint” region, and in particular, the range between 2 and 5 µm is characterized by the presence of strong fundamental and overtone vibrational absorption lines of atmospheric constituents, vapours and gases. The ability of broadband solid-state lasers to spectrally cover in a single shot or by rapid laser tuning in the widest wavelength range, containing all the above molecular absorption lines, is the main advantage of these compact laser sources.

Detection of low concentrations of molecules, constituting air pollutants or green-house gases for the purpose of environmental diagnostics or even the human breath for the purpose of medical diagnostics is currently done using laser systems, which are based mainly on nonlinear optical conversion techniques and include optical parametric oscillators (OPO) and difference frequency generators. OPO is almost an ideal solution, but rather complex and costly. Another possibility would be to employ the semiconductor lasers, including heterojunction lasers, lead-salt, antimonite and quantum cascade lasers (QCL), which are probably the simplest and the most cost effective sources in the Mid-IR wavelength region. However, they provide narrow tuning ranges (compare ~140 cm^{-1} tuning range of a quantum cascade laser and ~ 1800 cm^{-1} tuning range of a Cr:ZnSe laser) and limited output power levels at room temperature. Besides, even at cryotemperatures the QCLs operate so far only above 3.4 µm, and this hurdle is not going to to be overtaken easily in near future [1].

The crystalline solid-state lasers [2], on the other hand, which operate at room-temperature and have the largest relative bandwidth of ~45% of the central wavelength, can provide very high power levels retaining good beam quality and narrow spectral linewidth at the wide tuning range. In combination with near-infrared diode lasers or Tm-fiber lasers as pump sources these lasers offer stability, efficiency and compactness as well as the broad spectral coverage and tuning ranges, which are generally inaccessible for semiconductor lasers.

In recent years a lot of attention of researchers was devoted towards the alternative compact and cost effective continuous-wave and ultrashort pulsed sources based on Cr^{2+}-doped crystals of the II-VI family [3,4]. For detailed reference list please see review articles [3-12]. In the last decade a number of such sources have been developed in the “molecular fingerprint” range between ~2 and 3.5 µm [11], which show not only mechanical stability and excellent laser properties, but operate now at 6 W power level in CW regime [10] and 20 W pulsed [8], tunable between 2 and 3.1 µm [11].

2. RECENT DEVELOPMENTS IN ULTRAFAST CR:ZNSE LASERS

The present paper reviews principles and the latest advances in generation and applications of mid-infrared (above 2 micron) ultrashort pulses and frequency combs directly from the solid-state oscillators based on Cr^{2+}ZnSe laser, and highlights the radical efficiency and sensitivity increase when applied to high-resolution molecular spectroscopy and gas sensing. The most recent breaking-through development in this field has been the first truly Kerr-lens modelocked operation of Cr^{2+}:ZnSe laser, which we have just recently obtained. At this point we can say that “Ti-sapphire of the infrared”, as one of the inventors of Cr^{2+}:ZnSe lasers named it, is really there and mature enough for commercial realizations and practical applications. Finally, we discuss the new perspectives, which are opening for these as well as for other ultrashort pulsed mid-IR fiber based sources like e.g. Tm^{3+} fiber lasers.

An ultrabroadband Cr^{2+}-doped ZnSe laser operates between 2 and 3 µm, and has been shown to operate as high-power tunable continuous-wave laser, tunable single-frequency source, in Q-switched and gain-switched

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regimes, as well as in actively and passively mode-locked regimes. The mode-locked Cr^{2+}:ZnSe laser is especially interesting, because the exceptionally broad gain band of the material allows few-cycle operation in an important for various applications mid-infrared range. The first ultrashort-pulse femtosecond operation has been reported already three years ago [13] and utilized a SESAM with a semiconductor Bragg mirror for achieving passive modelocking. Pulse duration could be shortened down to 80 fs using chirped mirrors, but output power stayed around 100 mW, limited mostly by absorption in the SESAM. Even more importantly, the pulse spectrum width was already at the limit of SESAM reflection band. In this configuration, the laser proved to be suitable for sensitive spectroscopic measurements [14]. Another novel and exciting observation has been made on the KLM regime in Cr^{2+}:ZnSe laser. Namely, the laser has operated in two distinct regimes: high-power soliton regime with ~300 mW of output power and 100 fs duration, and chirped pulse regime with 170 mW of output power and ~1 ps pulse duration. The mode-locked operation in the positive chirped regime was particularly long-term stable and self-starting, promising new ways of generation of frequency combs in this wavelength region.

We believe that the demonstration of a KLM Cr^{2+}:ZnSe laser opens a way to utilizing the 800-nm broad bandwidth of the material and reach ultrabroadband single-cycle operation. Most importantly, it was shown that it is possible to start and operate the modelocked laser in an open air. This simplifies setting up and adjustment of the laser significantly. At the same time, ultrabroadband operation should obviously require a purged or evacuated enclosure.

3. Conclusion

Summarizing, the recent availability of ultrabroadband coherent frequency comb sources, based on solid-state lasers, enables novel approaches which allow achieving simultaneously high resolution and record sensitivity at very short recording times, using multichannel and multiplex techniques. Most importantly, these sources are now available in the mid-infrared region around 2-3 microns, where molecular spectral lines are located, and are ready for introducing them into practice.

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