METROCOMB

Femtosecond comb optical parametric oscillators for high-resolution spectroscopy in the mid-infrared

Final Publishable Summary Report

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The research leading to these results has received funding from the European Union Seventh Framework Program (*FP7/2007-2013*) under grant agreement no. 605057

1 EXECUTIVE SUMMARY

A frequency comb is a light spectrum consisting of a series of discrete, equidistant elements. Frequency combs may be generated by a number of techniques, including frequency modulation of a continuous wave laser or stabilisation of the pulse train generated by a mode-locked laser. If the wavelength range of the comb sources can be extended to cover the mid-IR region then such a source would be ideal for coherent Fourier-transform spectroscopy in the absorption-rich mid-IR 'molecular fingerprint' region delivering real-time acquisition of molecular spectra and real-time imaging with chemical identification for applications in large fast-growing global markets including environmental monitoring, real-time analysis of chemical /bio threats and explosives, trace molecular detection, quantum technologies and medical breath analysis.

The main objectives we aimed to solve in developing this technology to the point of being able to produce reliable products were: Define the stabilisation mechanism to enable highly stable combs to be generated in the 1 - 4.5 µm region; Validate the concept of a compact VECSEL pumped OPO for stable comb generation; Extend the accessible spectral range to longer wavelengths in the 5 -12 µm region.

The research necessary to extend the application areas of femtosecond frequency combs through the development of compact, robust, low-cost, commercially-exploitable sources is now possible; taking advantage of the fact that ultrafast laser pulses of femtosecond widths, separated by nanoseconds, manifest themselves as a phase-coherent comb of frequencies spread over a wide spectral band. Furthermore, the development of femtosecond frequency combs in the infrared region of the electromagnetic spectrum and beyond offers enormous opportunities for exploitation in broad spectrum detection and metrology. Robust industrial laser sources such as those produced by the SME supply chain grouping brought together in this project have be used by the leading research groups in this consortium to develop frequency comb based spectroscopy systems offering unprecedented detection sensitivity and measurement accuracy.

METROCOMB is a €2.01M project (EU contribution just under €1.5M) coordinated by M Squared Lasers Limited, with a project consortium consisting of 8 of Europe’s leading photonics research groups and small to medium sized companies from 5 different countries. The project has produced results which can be exploited across the supply chain covering optics, crystals, lasers and OPOs.

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2 Project Context and Objectives

2.1 Context

A frequency comb is a light spectrum consisting of a series of discrete, equidistant elements. Frequency combs may be generated by a number of techniques, including frequency modulation of a continuous wave laser or stabilisation of the pulse train generated by a mode-locked laser. The latter mechanism has been the subject of great interest since its discovery in 1999 which lead the Nobel Prize in Physics being shared by John L. Hall and Theodor W. Hänsch in 2005. Cavity modes in an ultrafast laser form a frequency-domain "comb" whose teeth are spaced at the pulse repetition rate. The modes do not lie on a scale intercepting 0 Hz, but have a “DC-offset” (delta) physically describing the phase-slip between the pulse envelope and carrier in each cavity roundtrip. By controlling delta to a constant value, the comb is precisely defined in frequency, and can be used for spectroscopy and metrology.

Mode-locking is a technique in optics by which a laser can be made to produce pulses of light of extremely short duration, on the order of picoseconds (10^{-12} s) down to femtoseconds (10^{-15} s). The basis of the technique is to induce a fixed phase relationship between the modes of the laser's resonant cavity. The laser is then said to be phase-locked or mode-locked. Interference between these modes causes the laser light to be produced as a train of pulses. Depending on the properties of the laser, these pulses may be of extremely brief duration, as short as a few femtoseconds.

Femtosecond combs provide a revolutionary new spectroscopic technique. The multitude of frequencies present in the comb, when incident upon a spectroscopic sample, are absorbed on many molecular lines. In this way the comb simultaneously probes the whole of the molecular spectrum; in comparison to conventional approach where a single frequency is scanned to interrogate each line sequentially. The comb, therefore, provides a powerful new way of conducting high-resolution molecular spectroscopy. One example of an application is breath analysis, in which the comb can, in a short space of time, generate a “fingerprint” of the breath and reveal trace quantities of gas, leading to a health diagnosis.

This technology is rapidly developing beyond the original high-cost and high-maintenance laboratory based lasers used to realise the meter in terms of the SI second and to measure optical frequency standards and fundamental physical constants. A multitude of everyday spectroscopic-based monitoring and measuring applications are now within reach if frequency combs can be realised from compact portable laser sources.

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2.2 Project Objectives

The research necessary to extend the production of femtosecond frequency combs into the infrared region of the electromagnetic spectrum and beyond is now possible, taking advantage of the fact that ultrafast laser pulses of femtosecond widths, separated by nanoseconds, manifest themselves as a superposition of light at different frequencies over a wide spectral band. Robust industrial laser sources such as those produced by the SME grouping brought together in this proposal can be used to develop frequency comb based spectroscopy systems offering unprecedented detection sensitivity and measurement accuracy. Moreover, if the wavelength range of the comb sources can be extended to cover the near IR region and into the far IR then such a source would be ideal for coherent Fourier-transform spectroscopy in the absorption-rich mid-IR 'molecular fingerprint' region delivering real-time acquisition of molecular spectra and real-time imaging with chemical identification for applications including environmental monitoring, real-time analysis of chemical / bio threats and explosives, trace molecular detection, and medical breath analysis. The main objectives we aimed to solve in developing this technology to the point of being able to produce reliable products are:

• Define the stabilisation mechanism to enable highly stable combs to be generated in the 1 - 4.5 µm region.

• Validate the concept of a compact VECSEL pumped OPO for stable comb generation.

• Extend the accessible spectral range to longer wavelengths in the 5 -12 µm region.

2.3 Project Team

A highly experience team of European academic and industrial organisations was assembled for the METROCOMB project. The project team is introduced below:

M Squared Lasers Limited (M2) M2 manufactures next-generation compact lasers and related systems. The company expertise spans the entire laser performance spectrum, from ultra-narrow, highly stabilized continuous wave to broadband femtosecond sources, and from deep ultraviolet to terahertz wavelengths. M Squared has longstanding experience and demonstrated success in delivering innovative solid-state laser products, meeting customer application requirements, and delivering the highest levels of customer service and support.

In this project, M Squared sought to expand the applicability of its core product lines with the object of increasing its customer base and sales

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revenue. In this context, M Squared was particularly interested in the development of novel vertical external cavity surface operating lasers and their use to pump frequency combs that can be used in quantum technology and metrological/sensing applications.

The principle responsibilities of M Squared were the project management and demonstration activities of the work.

LASEROPTIK GmbH (LO) was founded in 1984 and since the company lives and constantly grows based on its reputation and customer´s recommendations in the laser business. From the beginning, the primary goal of LASEROPTIK was to support customers to produce better lasers with improved optical components. This goal still remains as a business philosophy and was an important factor for the company to become a fully integrated producer of UV-, VIS- and NIR laser optics. As an owner-managed high-tech company we attach great importance to our social and environmental responsibility.

The principal responsibilities of LO were the supply of specialised optical components to the consortium.

Laser Quantum GmbH (LQ) manufacture compact, reliable, low-noise solid-state laser sources with long operational lifetime operating at GHz repetition rates. Laser Quantum UK supply pump lasers to both Laser Quantum GmbH and M Squared. In both cases, the pump laser is mounted on the same base plate and incorporated into the complete system.

The principal responsibilities of LQ were the interaction with the Heriot Watt to guide the developmental outputs of the frequency combs. LQ was also involved in the demonstration activities.

Raicol Crystals Ltd. (RAI) specialise in manufacturing high quality Periodically Poled nonlinear optical crystals and electro-optic devices. In particular, Raicol produce Periodically Poled Magnesium doped Lithium Niobate (PPMgLN), an efficient nonlinear optical material for frequency conversion applications in the visible and mid-IR wavelength range. The high nonlinear coefficient of PPMgLN makes it suitable for compact low power solid state laser systems.

As a supply chain SME, Raicol Crystals has provided nonlinear crystals to the consortium.

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Radiant Light SL (RAD) are specialist suppliers of advanced solid state instruments for laser tuning. They design, manufacture and market state of the art frequency conversion systems that expand the wavelength coverage of lasers and laser-based systems. They offer the latest technology in broadband laser tuning; their optical frequency conversion instruments include Optical Parametric Oscillators and Harmonic Generators which extend laser wavelengths from the UV to the IR, with high performance and ease of use.

The principal responsibilities of RAD were the interaction and guidance for the RTD performers as well as the demonstration activities at the end of the project.

Heriot-Watt University (HWU) a leading institution in science, technology and business and excels as Scotland's most international university. We have the structures to support and enhance research within a stimulating environment in key topical areas. They are continually investing in new research leadership posts and facilities, and have introduced ambitious talent development programmes for academic staff and research students.

The Institute of Photonics and Quantum Sciences (IPaQS) at HWU carries out broad range of world leading research in photonic physics, engineering photonics and quantum sciences. IPaQS builds on HWU 40+ years of history in world-leading research in photonics and spans a broad range of research – from lasers and optical sensing approaches to future manufacturing methods to the fundamentals of quantum information.

The principal responsibilities of HWU in the project were to apply the research expertise to the SMEs lasers and optics technologies to design and validate the necessary stabilisation and comb generation techniques for integration with and development of the SMEs product range.

Universite de Neuchatel (UNINE) The Academy of Neuchatel was founded in 1838. Now the University of Neuchatel (UNINE) is an internationally recognised institution with 4380 students from Switzerland and beyond (nearly 20% of students from abroad). The Faculty of Science has high-level research laboratories including various fields such as atomic clocks, plant survival and geothermics. The Faculty of

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Science has a notable reputation for its involvement in several national and international projects. While research is supported by the Swiss National Science Foundation, the European Union and other funding sources, the University interacts with both academic and private entities. The Institute of Physics includes the time-frequency group which studies frequency standards and atomic time-frequency metrology.

The principal responsibilities of UNINE were the development of improved vertical external cavity surface emitting lasers in the near infrared for use to pump optical frequency combs based on OPO technology.

Fraunhofer UK Research Limited (FHI-CAP) The Fraunhofer Centre for Applied Photonics is the first Fraunhofer Centre to be established in the UK and is based at the University of Strathclyde incorporating what was previously the Institute of Photonics. Fraunhofer UK Research Limited is a Research and Technology Organisation (RTO) which is incorporated as a not-for-profit, limited by guarantee company which provides applied research and development services to industry.

Fraunhofer Gesellschaft is Europe's largest organisation for applied research and the recently established Fraunhofer UK Research Ltd. (FHI-CAP) will be a hub for industry-driven laser research and technology for a variety of sectors including healthcare, security, energy and transport. The Fraunhofer Centre is based in the University's world-class Technology and Innovation Centre, which is transforming the way universities, business and industry collaborate to find solutions to global challenges, create jobs and support the economy.

The principal responsibilities of FHI-CAP were the development of mid-infrared, ultrafast vertical cavity surface emitting lasers based on the GaSb material system with the intention to use them to pump mid-infrared frequency combs based on novel nonlinear crystal materials such as ZGP and OP-GaAs.

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3 Main Scientific and Technological Results

3.1 Project Overview

The METROCOMB work programme builds on a strong existing foundation of knowledge in the fields of ultrafast photonics, tunable lasers, semiconductor and solid state lasers and frequency combs. The project research activities are structured into three logical research related work packages, a diagram of which can be seen in Figure 1.

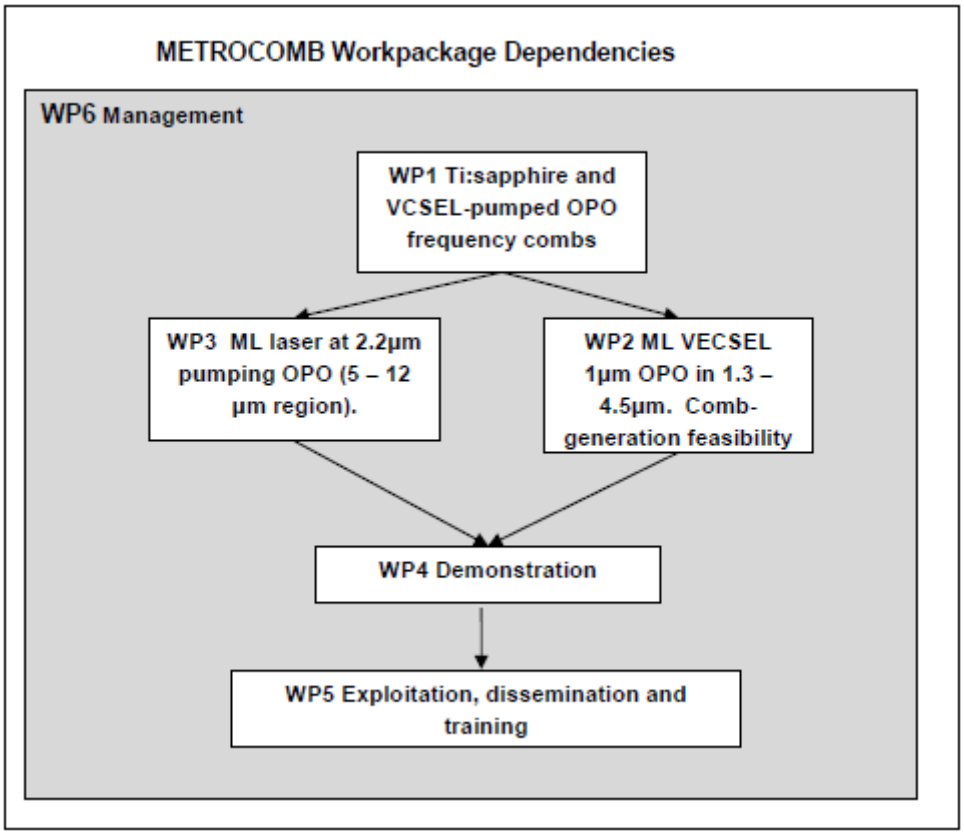

Figure 1 Workpackage overview and dependancies.

The 3 research workpackages and their activities are described below:

Workpackage 1: 'CEO- and Rb-cell-locked fs OPO-comb based on Ti:Sapphire-pumped OPO operating in 1.0 – 4.5 µm region.' The main aims of this workpacakge were to develop the initial frequency comb setup using Ti:S and VECSEL pumping through computational modelling, and experimental physical modelling and testing, advancing the existing comb generation technique knowledge. A detailed noise and performance assessment will be conducted which will develop a manufacturing strategy based on existing manufacturing technologies.

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Workpackage 2: ‘Mode-locked VECSEL operating at 1 µm pumping OPO operating in 1.3 – 4.5 µm region. Feasibility of comb-generation’ The aim of this workpackage was to focus on the development of an optimised mode locking VECSEL for comb generation. Experimental and modelling techniques were to be applied to establish efficient frequency conversion and comb stability techniques. The developed highly compact frequency comb was planned to be prototyped in the laboratory and tested to correlate the earlier experimental and computational work.

Workpackage 3: ‘Mode-locked laser operating at 2.2 µm pumping long-wavelength OPO (5 – 12 µm region).’ The aim of this workpackage was to develop a highly innovative compact femtosecond 2-3 µm pump sources then investigate suitable mode locking regimes for a VECSEL operating at 2.2 µm, finally a ML laser-pumped OPO in 5 – 12 µm region was planned to be developed to offer the broad wavelength coverage.

3.2 WP1: CEO- and Rb-cell-locked fs OPO-comb

In the first period of the project a CEO-locked PPKTP comb was demonstrated. The OPO operated at a signal wavelength of around 1060 nm. In addition to this, two approaches to harmonic pumping were evaluated, with one ("n = 1 Harmonic Pumping") being identified as providing a cleaner frequency comb structure than the other. Fabry-Pérot filtering of a 333-MHz laser to 10 GHz was implemented and locking investigated in two alternative approaches, namely dither locking to the transmitted comb signal or dither locking to an auxiliary Rb-stabilised ECDL laser at 780 nm.

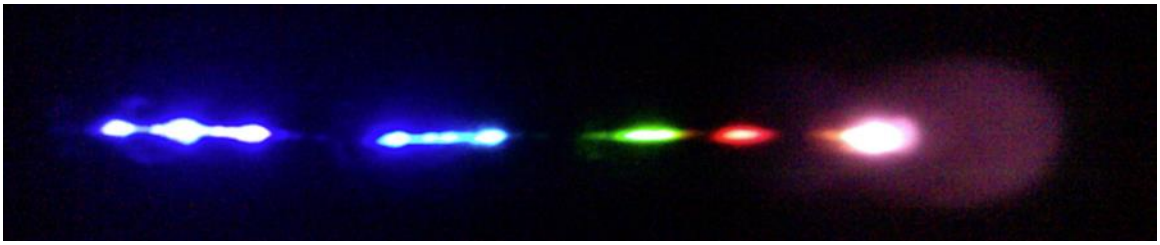

Figure 2 OPO visible outputs. Left to right: SHG pump; pump+signal SFM; SHG signal; pump+idler SFM; pump.

In the second period of the project a broadly tunable, fully stabilized, 1.95 – 4.0 µm frequency comb was demonstrated, and a full characterisation completed by the WP1 partners. This result was reported at CLEO 2015 [1] and CLEO/Europe 2015 [2], and has

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also been published in Optics Letters [3]. An atomically referenced 1 GHz femtosecond OPO comb was also demonstrated, whose repetition and offset frequencies were referenced to Rb-stabilised microwave and laser oscillators respectively. This result was reported in Optics Express 23, 16466 (2015) [4].

Following on from the results generated in the 1st period a stabilized 10-GHz frequency comb generated by filtering a 333.3-MHz OPO frequency comb with a Fabry-Pérot (FP) cavity was developed, which was directly stabilized to the incident fundamental comb and whose modes were clearly resolved by a Fourier transform spectrometer with a spectral resolution of 830 MHz. This result was reported in Opt. Lett. 40, 2692 (2015) [5] and at CLEO/Europe 2015 as a postdeadline paper [6].

The noise characterisation of such a system was carried out for harmonic pumping of a 1.33 GHz OPO by a 333 MHz Ti:sapphire laser, resulting in certain specific problems being identified which are relevant to the adoption of the approach in a commercial system (Figure 3).

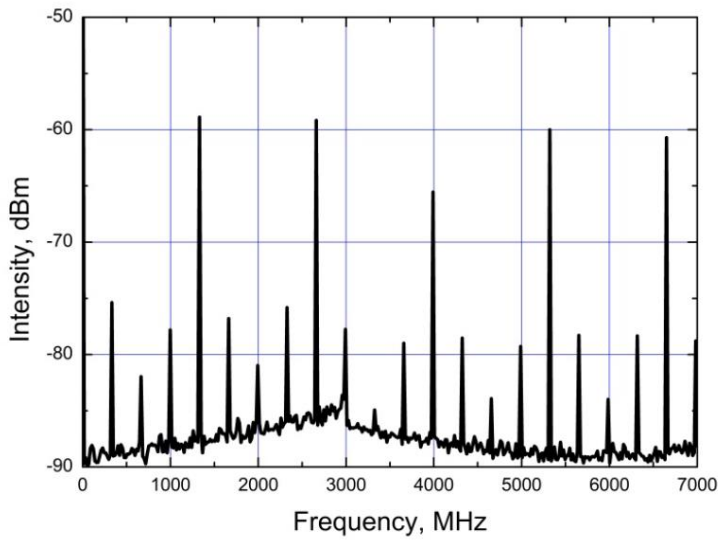

Figure 3 1.33-GHz repetition rate frequency in time domain

To reach GHz frequencies harmonically pumping of an optical parametric oscillator can be utilized, which can offer broad wavelength coverage, short pulse durations and can be locked to produce low-noise frequency combs. Synchronously pumping an OPO limits its repetition rate to that of its pump laser, but it is possible to operate the OPO at a harmonic of this when the OPO cavity length is an integer or integer fraction of the pump cavity length. Here, we demonstrated the first example of a fully stabilized frequency comb from a harmonically pumped 1-GHz OPO (see Figure 4).

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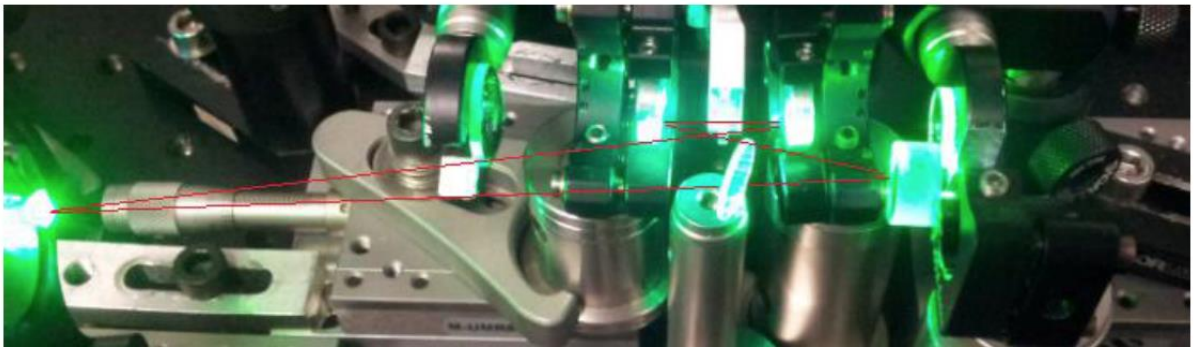

Figure 4 Harmonically pumped 1-GHz OPO cavity.

The experimental configuration and the locking of fREP and fCEO is shown in (Figure 5). A Ti:sapphire pump laser (Gigajet, Laser Quantum) produced 30-fs pulses with 1.45-W average power centered at 800 nm with a full-width half-maximum (FWHM) bandwidth of 32 nm and a repetition rate of 333 MHz. A 90% reflector was used to steer 1.3 W of pump power into the OPO, with the remaining 10% coupled into a photonic crystal fiber (PCF) for supercontinuum generation.

The locking scheme of the fCEO frequency was the same for fundamentally and harmonically pumped OPOs.

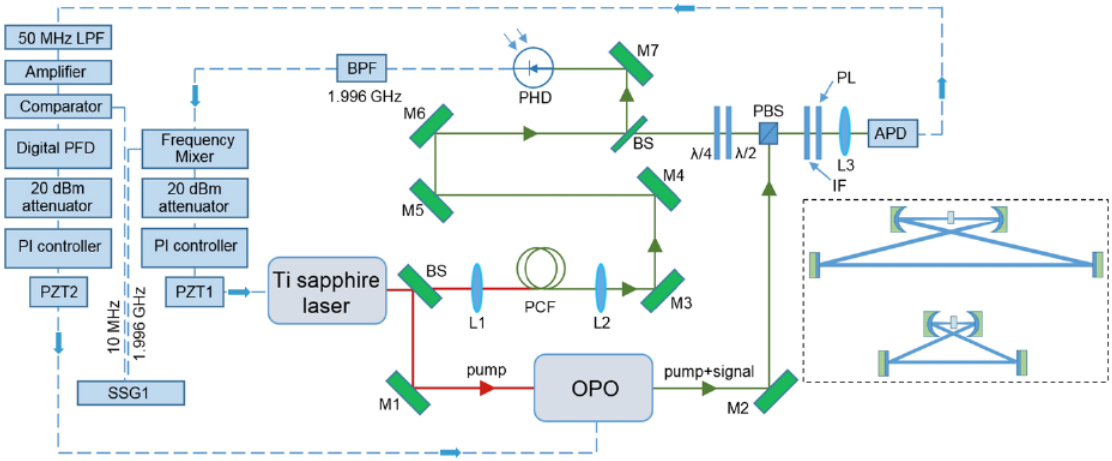

Figure 5 Stabilization layout and (inset) cavities of the fundamentally / harmonically-pumped OPO combs.

3.3 WP2: Mode-locked VECSEL operating at 1 µm pumping OPO operating in 1.3 – 4.5 µm region

In WP2 a characterisation and optimisation of ultrafast VECSELs lasers was achieved and performance limits were evaluated. A testbed setup was developed and self-modelocking

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was simulated and studied. Optimized cavity designs for efficient frequency converters based on OPOs have been discussed and developed with the requirements on the VECSEL performance for comb stabilisation being discussed and initial plans for joint experiments developed. A cavity diagram can be seen in Figure 6.

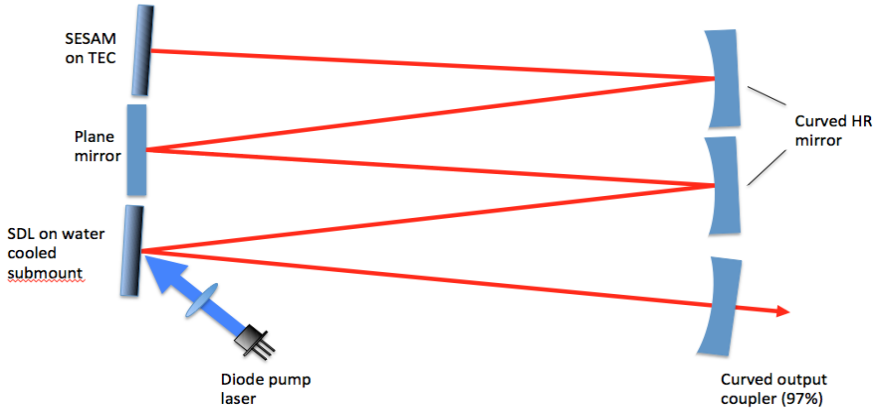

Figure 6 Cavity diagram of a modelocked vertical external cavity surface emitting laser.

The theoretical and experimental assessments of various solid-state laser types and architectures for production of ultrashort (<1 ps) pulses with a high average output power level (up to 1 W) at around 2 µm spectral region were performed in WP3. In particular, optimal cavity configurations for both soft- and hard-aperture Kerr Lens Modelocking (KLM) effects in the ultrafast VECSEL set-ups were proposed and studied Figure 7). The numerical analysis revealed that KLM efficiency in the semiconductor gain chip could be high enough for generation of ultra-short pulses from a self-mode-locked 2 µm VECSEL.

The WP2 partners evaluated all relevant parameters of M2's commercially available Dragonfly laser including its noise properties. The Dragonfly has a pulse energy, which is unique for semiconductor lasers and which is highly attractive. However, the peak power is typically only calculated from the autocorrelation trace and the average power. The realization of the first ultrafast VECSEL pumped OPO confirms for the first time the high peak power of the source in a direct way (the parametric gain is determined by the peak power and corresponds well to the simulations).

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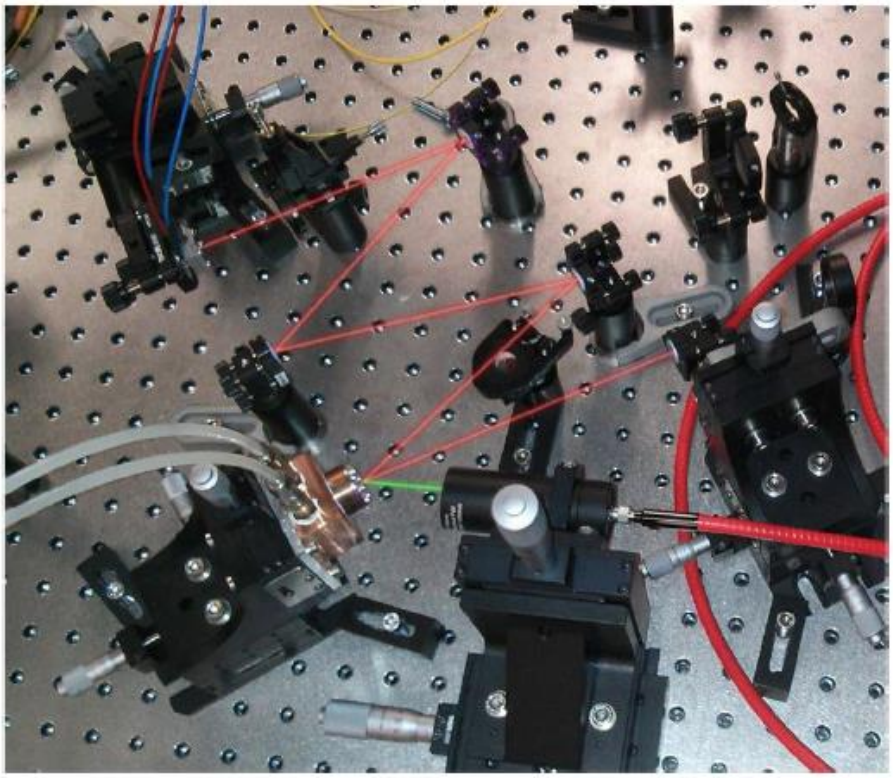

Figure 7 Picture of the experimental setup of a modelocked VECSEL.

The WP2 partners demonstrated the first OPO that is synchronously-pumped by an ultrafast VECSEL. A schematic for the setup can be seen in Figure 8.

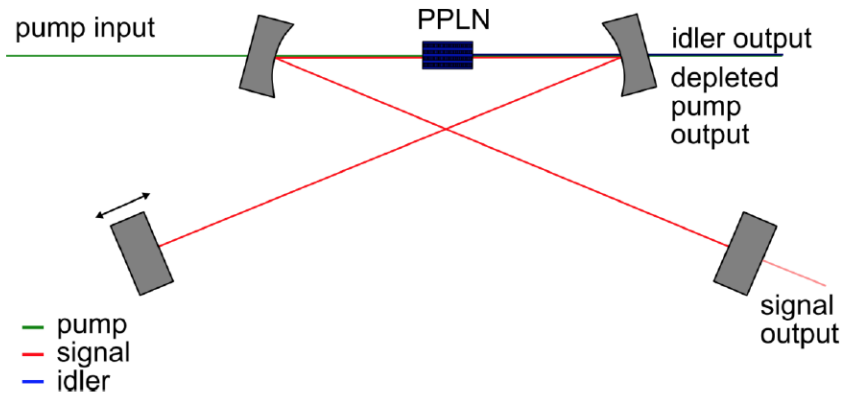

Figure 8 Schematic of a linear cavity OPO to be pumped by the VECSEL ultrafast laser.

The overall signal and idler efficiencies are good, and both pulse duration and tuning behaviour are in excellent agreement with numerical simulations. The compact, cost-efficient, and air-cooled system is an excellent commercial alternative to significantly more complex laser systems for mid-IR applications. A photograph of such a system can be seen in Figure 9.

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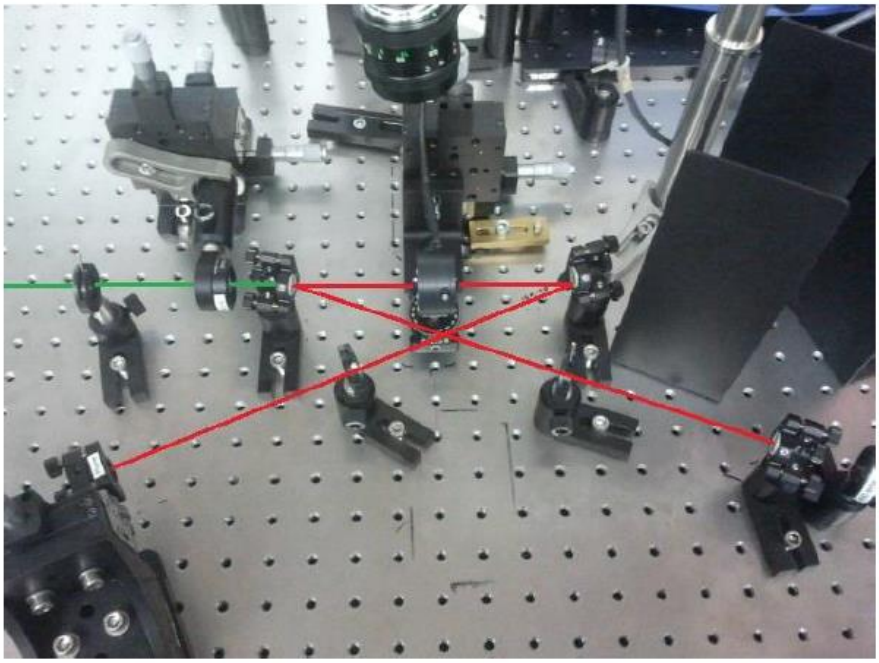

Figure 9 Picture of the OPO setup with the pump beam drawn in grean and the oscillating signal beam drawn in red.

UNINE demonstrated first pulse compression results for an unamplified VECSEL, compressing the ~2 ps long Dragonfly pulses down to 420 fs with the available power of 330mW from the Dragonfly. A schematic of the setup is shown in Figure 10). These results are highly important for future products of the partner M2.

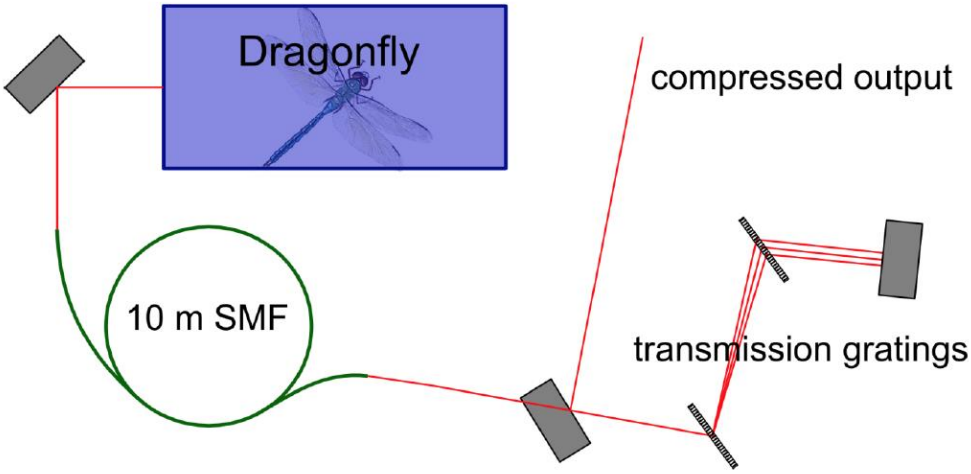

Figure 10 External compressor setup for the Dragonfly laser.

UNINE also studied the progress from a standing-wave OPO cavity towards a ring cavity. It has been realized both with a Covesion and the RAI crystal. The RAI crystal gave better performance and higher power levels. Optimized mirrors from LO helped to further optimize its performance. The tuning range and operation are according to the expectations.

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3.4 WP3 Modelocked laser operating at 2.2 µm pumping long-wavelength OPO (5 – 12 µm region)

This work package has investigated GaSb-based VECSELs for mode-locked operation. These VECSEL structures were developed during the course of the previously EU funded VERTIGO program under Grant 034692 and made available to this project (Courtesy of J. Wagner and M. Rattunde from the Fraunhofer Institute for Applied Solid State Physics (IAF) in Freiburg, Germany). Samples were characterised and good CW performance achieved (Figure 11).

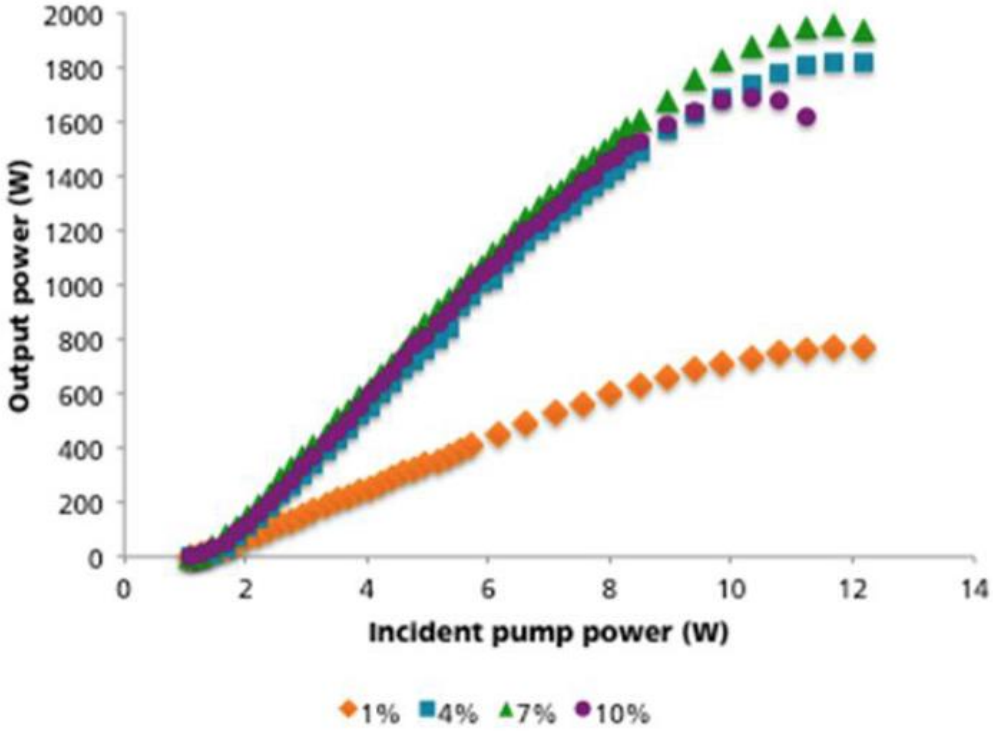

Figure 11 CW performance of 2 µm GaSb VECSEL ship.

Absorber-free mode-locking of the devices was investigated. In particular, optimal cavity configurations were proposed for both soft- and hard-aperture KLM effects in the ultrafast VECSEL set-ups. The numerical analysis also revealed that KLM efficiency in the semiconductor gain chip could be as high as in a typical Ti:sapphire laser system that demonstrates the possibility of generation of ultra-short pulses from a self-mode-locked 2 µm VECSEL. Despite all efforts, no sensible performance was obtained of the absorber-free system.Semiconductor saturable absorber mirror (SESAM) mode locking is currently the best-suited technology for the development of high-power, high-pulse energy and reliable ultrashort pulse laser oscillators. However, to date, most of the SESAM devices have been fabricated using III-V compound semiconductors (AlAs/AlxGa1-xAs, GaAs/InxGa1-xAs)

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which have been used intensively for femtosecond pulse generation in a range of near-IR lasers only (~ 0.8-1.5 µm). The saturable absorber devices for mode locking of lasers at around 2 µm or longer wavelengths have not been well developed to a mature level so far, except for some initial laboratory demonstrations restricted to a low-power regimes and/or picosecond pulse generation in some fibre, solid-state or semiconductor disk lasers. In WP3 pulsed operation of a 2 µm VECSEL system was successfully achieved, using a semiconductor saturable absorber mirror and a Silicon etalon for dispersion control. A total output power of up to 25 mW was measured in this operation mode. An autocorrelation trace to ultimately confirm stable mode-locking remains to be detected as the available output pulse power is thought to be below the detection limit of the autocorrelation unit. These results illustrate, that considerable further design and growth effort will be required to improve the performance to a level sufficient to pump a mid IR OPO system, as proposed in the METROCOMB project.

Alternatively to the semiconductor laser system approach we also developed a Tm-doped laser systems for femtosecond pulse generation as a better-established and a lower-risk option. Namely, the gain media from crystalline classes of double tungstates (Tm:KYW) and sesquioxides (Tm:Lu2O3) have been chosen for further mode-locking experiments under direct laser diode pumping. The schematic for this system can be seen in Figure 12).

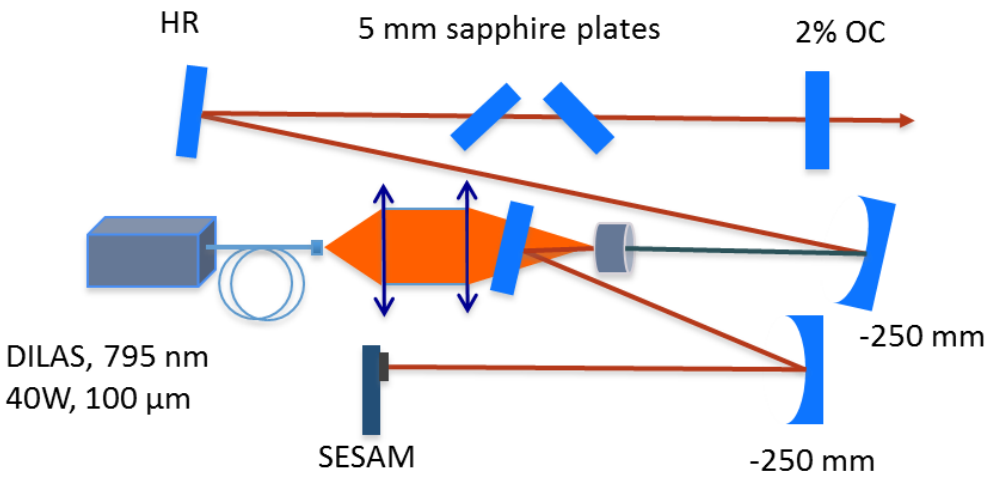

Figure 121 Experimental set-up for passive mode-locking of the Tm:Lu2O3 ceramic laser at around 2060 nm.

The output performance with respect to pulse stability, pulse duration (sub-ps), pulse power (> 10 kW), repetition rate (100 MHz) is sufficient to pump the proposed mid-IR OPO. A photograph of the experimental setup is shown in Figure 13.

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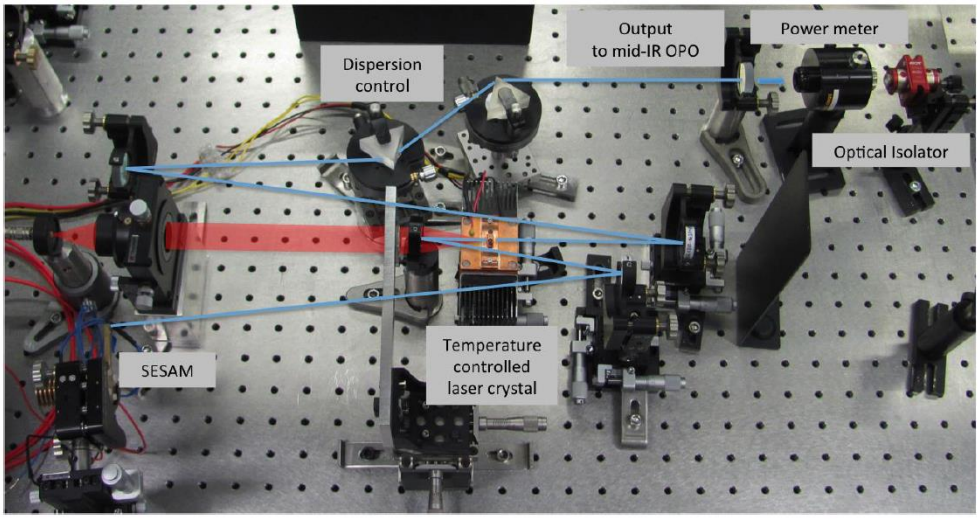

Figure 13 Experimental setup of the ultrafast thulium laser.

For the long-wave light generation, a ZGP OPO was configured a diagram and photograph of which can be seen in Figure 14.

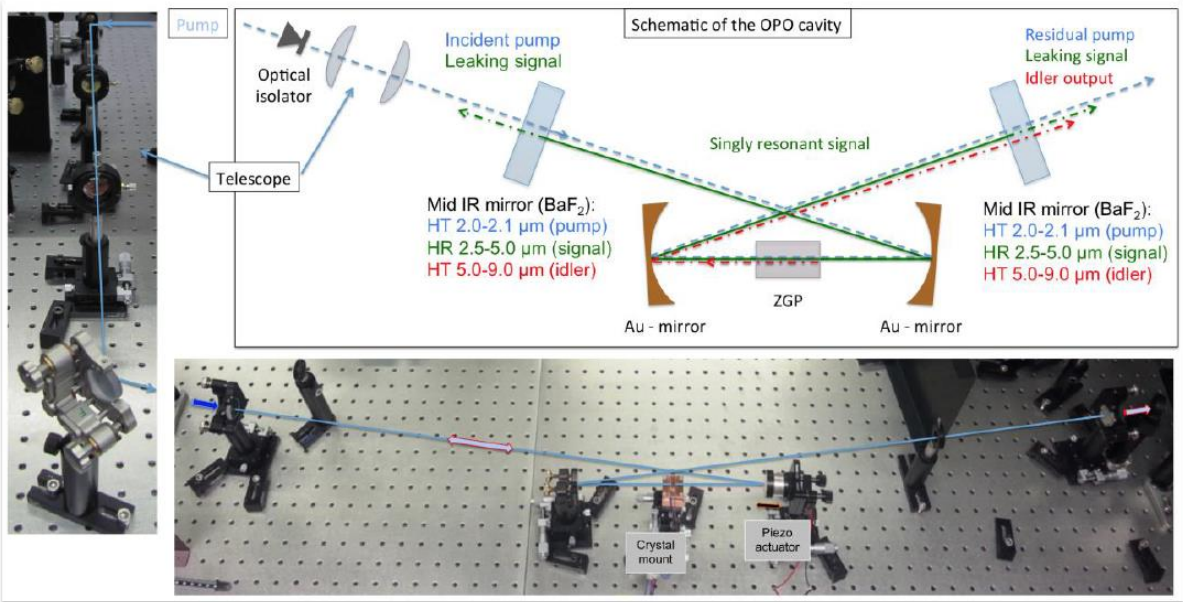

Figure 14 Photograph and schematic of the OPO resonator.

After initial failing to achieve OPO threshold conditions at the maximum available pump power, a more in depth characterisation of the OPO losses was undertaken and the found parameters were used to feed the simulation programme previously developed. The results of this simulation show that the calculated threshold for the current pumping condition and cavity arrangement is below the available pump power only for certain crystal lengths. Thus the found threshold power does not leave a large error margin, as the simulation is not taking mitigating effects, such as birefringence related polarisation, non-optimised mode overlap or the presence of multiple pulsing instabilities of the mode-locked pump into

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consideration. A further theoretical study on a possible ring resonator setup reveals, that the threshold can be reduced under certain conditions and that this approach could be more likely to allow OPO operation in the mid-IR range.

3.5 WP4: Demonstration

The results from UNINE on pulse compressing VECSELs to obtain the shortest pulses possible from the Dragonfly architecture have been demonstrated and improved on at M2 as part of the demonstration activities. In the VECSEL configurations adopted in METROCOMB, pulse compression is achieved in two stages. First, an extracavity pulse compressor using two transmissive diffraction gratings is used to reduce the pulse duration of the primary laser output beam. A schematic of this first-stage pulse compressor is shown in Figure 15. The pulse output from the standard M2 Dragonfly laser cavity was compressed resulting in a 2.5 ps pulse duration with an average power of 1.3W (see Figure 15).

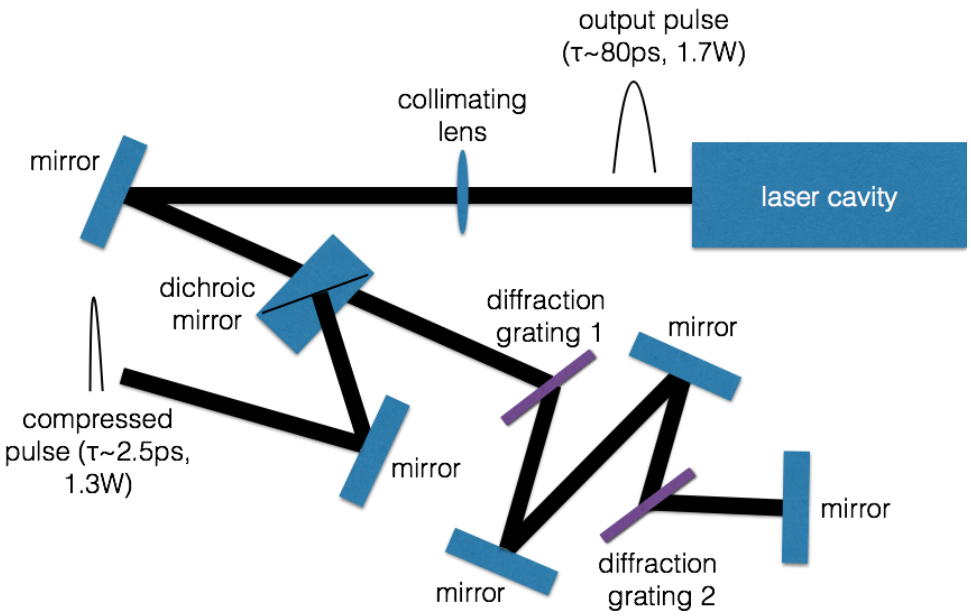

Figure 15 ***Schematic diagram of the first compressor stage.***

In a second stage, a 5 meter long polarisation maintaining fibre was used to impose self phase modulation on the output which broadened the spectrum. A block diagram of the approach is given in Figure 16.

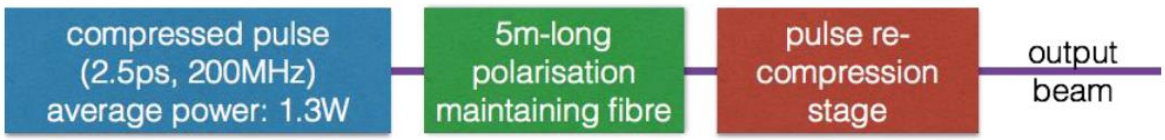

Figure 16 ***Schematic diagram of the proof-of-concept SPM pulse compression experiment.***

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After this, the spectrally broadened pulse was compressed again, leading to <300 fs pulse durations (see Figure 17).

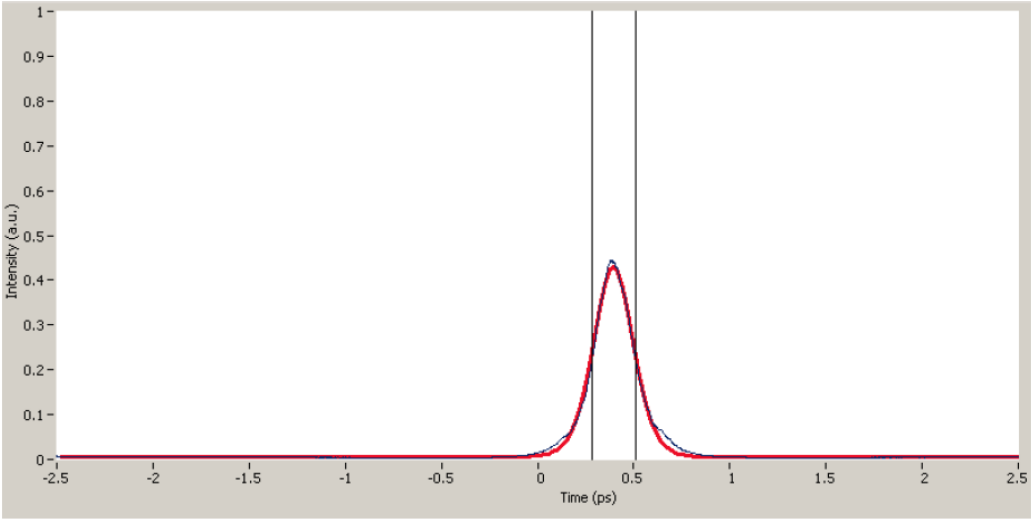

Figure 17 ***Resulting pulse shape after re-compression in blue, sech2 fit in red.***

.For the sensing aspect of the demonstration activities, we present results using an OPO based laser frequency comb. The OPO laser frequency comb can be tuned to cover 2 - 4 µm mid-infrared region, representing an ideal source for sensing applications. A Fourier transform infrared (FTIR) spectrometer is used for conducting spectroscopy. A schematic of implementing standoff sensing is shown in Figure 18.

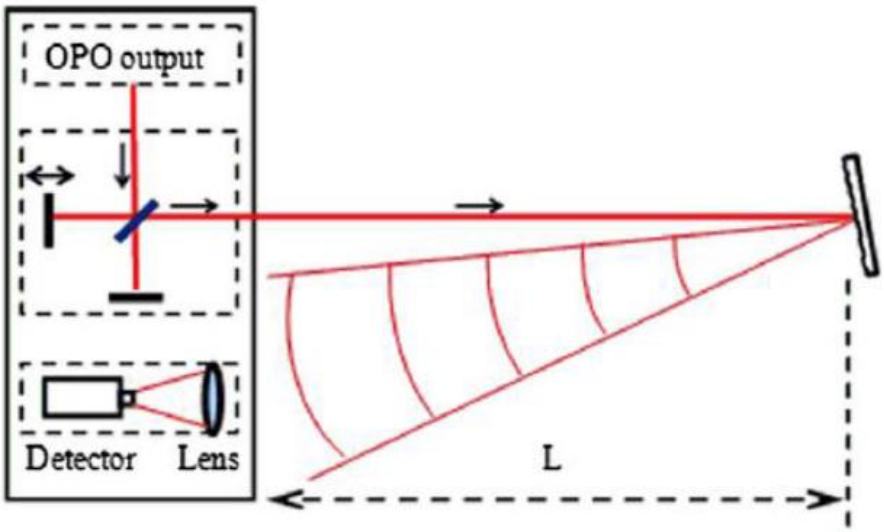

Figure 18 Layout of the stand-off detection system, showing the definition of the stand-off distance, *L*.

Mid-IR idler light with a collimated beam diameter of 5 mm was directed to the Michelson interferometer of the FTIR spectrometer. After the interferometer, the beam was directed to

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the surface of interest. Light diffusely reflected (or scattered) from the surface was collected by a CaF2 lens. The OPO was tuned by altering the OPO cavity length or by translating to crystals with slightly different grating periods, and the measured idler comb spectra are shown in Figure 19.

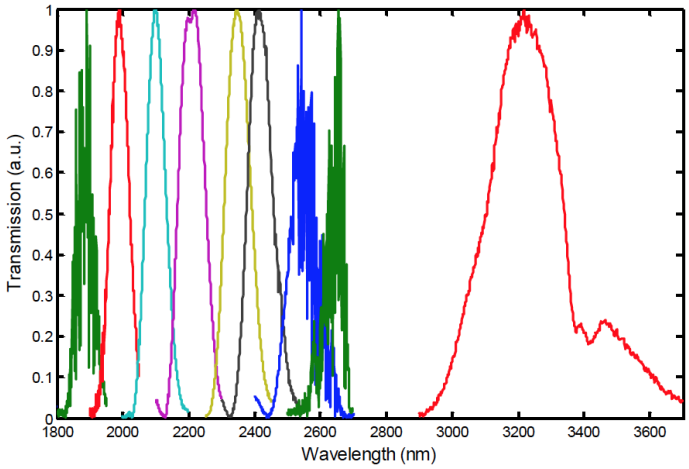

Figure 19 The measured OPO idler spectrum [Res = 1 cm-1 (30 GHz)].

Using this setup, we have shown simulated transmission spectrum of water vapour (assuming a concentration of 1% and at the pressure of one atmosphere, resolution = 0.1 cm-1) with a path length of 2.5 m at around 1.9 μm and 2.6 μm respectively, and compared them with the measured OPO idler spectra. Wavelength of the measured absorption lines agree well with the simulated one. An example of the experiments are shown in Figure 20.

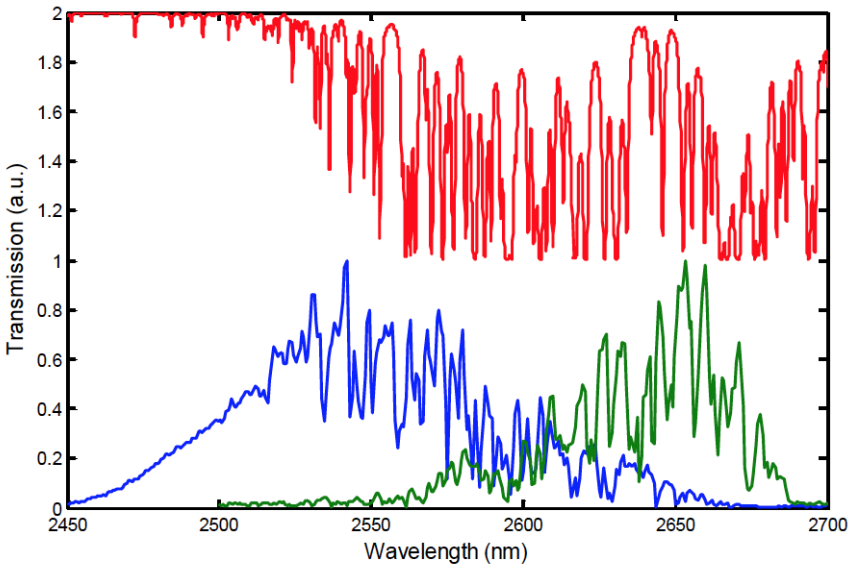

Figure 20 Red line: simulated transmission spectrum of water vapour (concentration = 1%, path length = 2.5 m, at a pressure of one atmosphere, resolution = 0.1 cm-1). An offset of 1 is added on the y axis. Blue and green liens: measured OPO idler spectrum [resolution = 1 cm-1 (30 GHz)].

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4 Potential Impact, Main Dissemination Activities and Exploitation of Results

4.1 Potential impact

The results and products developed in METROCOMB have many potential OEM applications for a femtosecond comb source. The global market size for laser technologies related to METROCOMB was in excess of $509million in 2011, and is forecast to grow tenfold to $5billion by 2018.41 Based on predicted growth and routes to market, the following sectors are considered to be the most likely initial target markets.

Key OEM markets for femtosecond comb

- **Remote gas and chemical leak detection – oil & gas and petrochemical industries**
- **Chemical warfare agent & explosives detection – military and homeland security**
- **Industrial process monitoring and control**
- **Atmospheric, pollution and environmental monitoring**
- **Medical diagnostics**

The unique selling points of a femtosecond comb will make significant sales into these markets based upon:

- **High power (250mW; 100kW peak)**
- **Broad wavelength coverage (1.0-4.5µm)**
- **Compact / portable configuration**
- **Robust & reliable**

The first key market application targeted by METROCOMB is Mid-IR sensing Mid-IR lasers are a specific segment of the global laser market, which reached almost $7.5billion in 2011, a 14% growth. The mid-infrared wavelength range is ideal for spectroscopy and thermal imaging applications because it matches well with molecular vibrational frequencies. In addition, mid-IR lasers are known for being eye-safe and covert.

Sensing applications are expected to drive rapid growth of the mid infrared (~1.8 - 15µm) laser market with a predicted annual growth rate of 30%, compounded annually through to 2014. The global market for mid-infrared sensing technologies is growing rapidly; at $509million in 2011, it is forecast to grow tenfold to $5billion by 2018. This strong growth is anticipated to come as technology develops from bench-top to portable units for in-field applications.

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come as technology develops from bench-top to portable units for in-field applications. Key technological advances that will drive uptake within all of these applications are high sensitivity, high speed, flexible detection (range of target species) in a compact and robust package, enabling mobile platforms and integration into commercial and industrial processes.

Gas and chemical leak detection within the oil & gas and petrochemical industries offers a number of potential applications for a femtosecond comb source including prospecting; pipeline leak detection; and safety monitoring of production platforms, refineries and petrochemical plants. The high peak power and broad wavelength span of the femtosecond comb will enable long-range (>100m), multi-species, hyperspectral detection and imaging at sensitivities over 1000x greater than existing market leading imaging technologies. Significant cost benefit to the industry is anticipated as the detection and location of very small leaks in real-time will actively reduce product loss, prevent hazardous incidents and avoid costly shut-downs, increasing operational efficiencies and reducing risk. Platform and pipeline leaks can cost producers $100m's in lost production and fines; $1.5b of natural gas is lost from pipelines in the US alone each year.

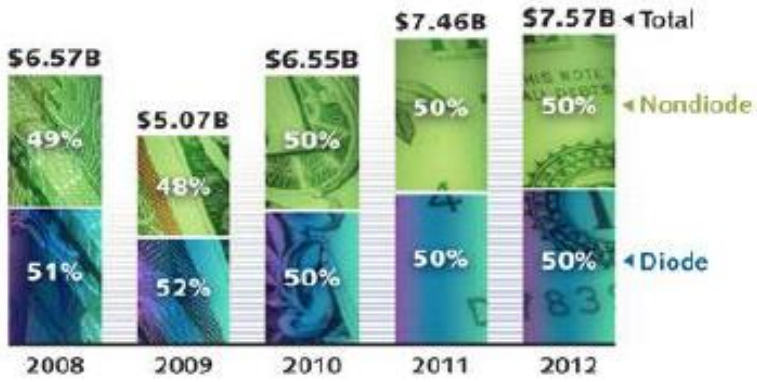

Figure 21 Worldwide commercial laser revenues

The explosives detection market is forecast to grow by ~10% p.a. from $1.5billion in 2013 to $2.2billion by 2019.46 There are currently no true stand-off detection technologies that can detect trace amounts of explosives or constituent chemicals at more than a few metres. The high peak power and broad span of a femtosecond comb offers substantial technical advantages over existing technologies and will enable capabilities not currently available to this market. This sector is dominated by large, multi-national companies (e.g. Smiths Detection, L- 3 Communications, Safran) and hence OEM supply is likely to be the best route to market. Industrial applications of infrared molecular spectroscopy are numerous and can be found in almost every manufacturing and production sector including agriculture,

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chemicals, food, life sciences, process control, pharmaceuticals, textiles, polymers, wood, soil analysis, etc. The global spectrometer market is forecast to exceed $10.3billion by 2015. Japan, Europe and the US currently represent 80% of this market but expanding markets in India and China are expected to present significant opportunities for spectrometry. Specifically, the molecular spectroscopy market is the largest product segment within this. The diagram in Figure 22 shows the segmentation of the molecular spectroscopy market by technique predicted to reach $3.3billion by 2012. These reports all confirm the major trend in spectroscopy development is towards robust, portable instruments for in-field use.

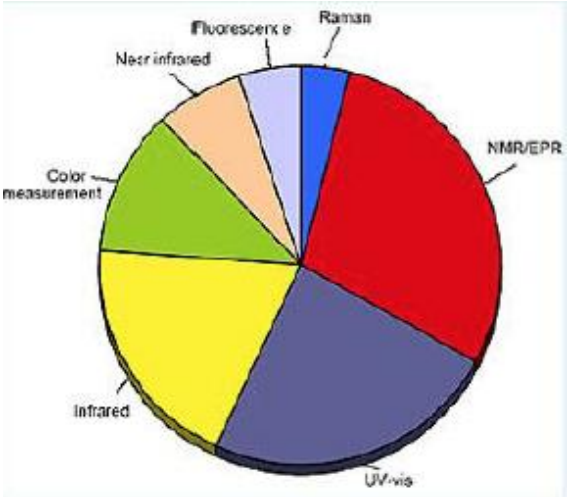

Figure 22 Breakdown of the commercial laser market.

Dispersive spectrometers currently dominate the near infrared range with FT-IR being the more common technique in the mid and far IR. Existing dispersive spectrometers typically use either a single, narrow-band laser source, or a broadband source that requires calibration and narrow-band filtering, resulting in significant attenuation of the illumination and low power within any given wavelength band, limiting the sensitivity. A compact, robust femtosecond comb OEM source with high power, high spectral resolution and broad wavelength coverage would significantly enhance dispersive instrument capabilities. In addition, a femtosecond comb would extend their useful wavelength range from < 2.5μm into the mid-IR. The key players in IR spectroscopy are FOSS, Thermo Scientific, Perkin Elmer, Bruker & JASCO. The applications are diverse with the pharmaceutical and chemical industries accounting for around 20% each and a range of others (oil & gas, food & agriculture, medical, semiconductor, biotechnology and research) each contributing between 5% and 10% market share.

The returns to the SMEs and the EU of investing in this project are expected to be several times higher than the co-financing provided; the business case behind this for the METROCOMB SME participants over the 5 years following the project is conservatively

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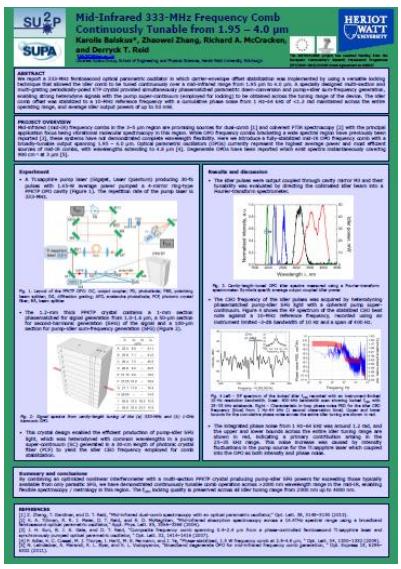
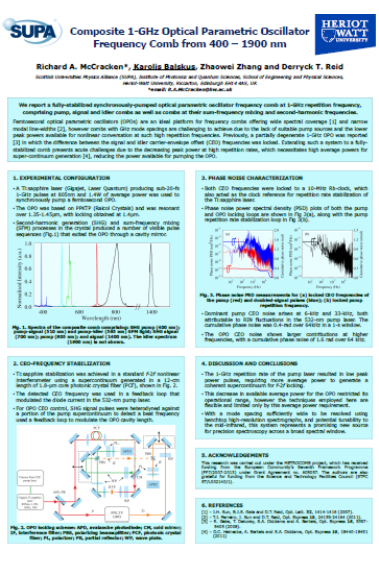
estimated and detailed below, producing additional sales revenues of more than €30M. The rational and mechanism behind these increased profitability figures are detailed by the SME participants. The overall economic return to the SMEs, increase in pre-tax profits, is forecast to be in the region of €16M. The SME partners believe that these figures could easily be twice as high and, 10 years from the end of the project, up to a factor of ten higher than the projected year 5. The company tax payable on the additional profits generated over a period of 5 years from the end of the project would be of the order of €3M, assuming a tax rate of 20%, thereby fully repaying twice the REA funding requested.

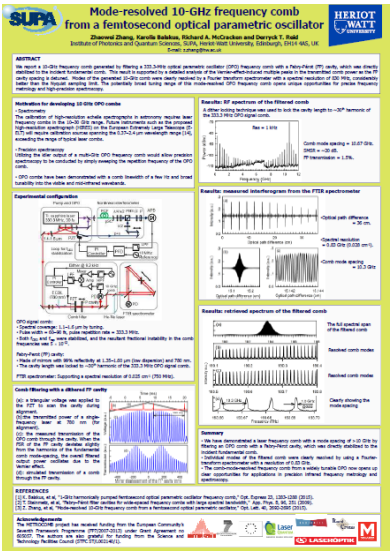
As the SME partners will form a supply chain, M2 and RAD will be the main drivers of the increased sales revenues and profitability through sales of new products in the markets identified. The gross profit margin on sales reflects the SMEs current levels of profitability and the net profit before tax for these new unique products is expected to be in the order of 50%.

4.2 Main dissemination activities

Over the course of the project METROCOMB has been promoted at 12 scientific conferences and meetings. This has involved a combination of presentations and posters communicating the objectives of the project and results achieved.







In addition, the METROCOMB partners have disseminated the project results via 12 papers in scientific journals, many of which have been open access.

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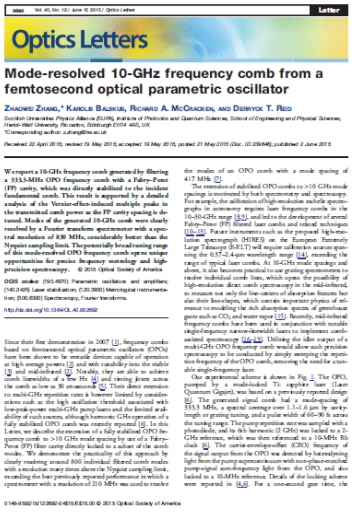
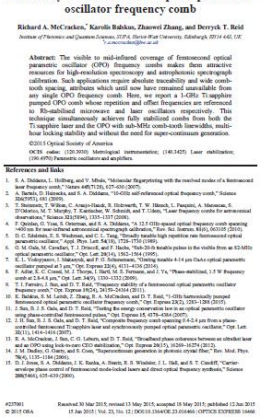
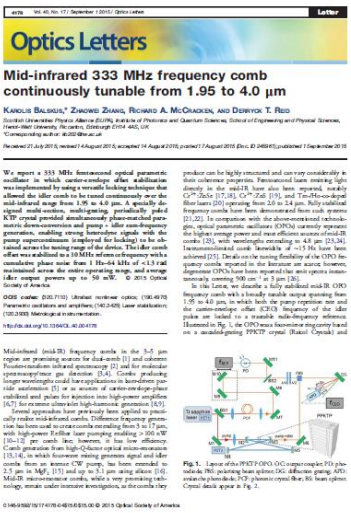

Optics Letters

Mode-resolved 10-GHz frequency comb from a femtosecond optical parametric oscillator


Atomically referenced 1-GHz optical parametric oscillator frequency comb


Optics Letters

Mid-infrared 333 MHz frequency comb continuously tunable from 1.95 to 4.0 μm

The project website, press release, YouTube video and flyers have all been used to disseminate the project to a wider audience.

4.3 Exploitation of Results

In addition to the direct new product sales anticipated to impact the SME supply chain, there will be the potential to grant sub-licences or enter into technology agreements for the use of the technology developed in METROCOMB with organisations who are not partners in METROCOMB. The broad appeal of the technology being developed and numerous potential application areas beyond the current areas of activity of the SME partners makes this likely. Revenues generated from sub-licensing the technologies will be shared by the SME partners in line with the level of financing of the results and the detailed agreements for exploitation reached during the project.

The METROCOMB project and successful commercialisation would allow the SMEs to expand their teams of world-class technologists and to build on its strong competency in the miniaturisation of efficient laser sources.

The results and products developed in METROCOMB have many potential OEM applications for a femtosecond comb source. The global market size for laser technologies related to METROCOMB was in excess of $509M in 2011, and is forecast to grow tenfold to $5billion by 2018. Based on predicted growth and routes to market, the following sectors are considered to be the most likely initial target markets:

- Remote gas and chemical leak detection – oil & gas and petrochemical industries
- Chemical warfare agent & explosives detection – military and homeland security

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- Industrial process monitoring and control
- Atmospheric, pollution and environmental monitoring
- Medical diagnostics
- Mid-IR molecular spectroscopy and sensors
- Quantum technologies

The unique selling points of a femtosecond comb will make significant sales into these markets based upon:

- High power (250 mW; 100 kW peak)
- Broad wavelength coverage (1.0 - 4.5 μm)
- Compact / portable configuration
- Robust & reliable

The key technical differentiation that this project facilitates and the innovations it contains establishes a clear technological advance for the SME's markets. No other companies have yet developed FT spectrometers in the mid-IR illuminated by femtosecond comb sources. In addition to this, the pioneering VECSEL technology and its introduction as a source of mid-IR radiation will offer a low-cost alternative to diode-pumped solid-state lasers. These key advantages will confer significant commercial opportunity and would underpin the success of the supply chain and future growth of the companies involved.

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5 Project Details

Title	**METROCOMB: Femtosecond comb optical parametric oscillators for high-resolution spectroscopy in the mid-infrared** (GA no. 605057)
Coordinator	**M Squared Lasers Limited**, United Kingdom
Consortium	**LaserOptik GmbH** Germany **Laser Quantum GmbH** Germany **Raicol Crystals Limited** Israel **Radiant Light SL** Spain **Heriot-Watt University** United Kingdom **University of Neuchatel** Switzerland **Fraunhofer UK Research Limited** United Kingdom
Duration	1st August, 2013 – 31st July, 2015 (24 months)
Funding Scheme	SME-2013-1: Research for the benefit of SMEs
Budget	EU contribution: €1,499,000.00
Website	http://www.metrocomb.eu/
For more information	metrocomb@m2lasers.com

The research leading to these results has received funding from the European Union Seventh Framework Program (*FP7/2007-2013*) under grant agreement no. 605057

6 References

[1] Karolis Balskus, Zhaowei Zhang, Richard A. McCracken, Derryck T. Reid, "Mid-Infrared 333-MHz Frequency Comb Continuously Tunable from 1.95 µm to 4.0 µm," CLEO 2015, STh1N.7

[2] Karolis Balskus, Zhaowei Zhang, Richard A. McCracken, and Derryck T. Reid, "Mid-Infrared 333-MHz Frequency Comb Continuously Tunable from 1.95 – 4.0 µm," 2015 European Conference on Lasers and Electro-Optics - European Quantum Electronics Conference, ED-P.6

[3] Karolis Balskus, Zhaowei Zhang, Richard A. McCracken, Derryck T. Reid, “Mid-infrared 333 MHz frequency comb continuously tunable from 1.95 to 4.0 µm,” *Optics Letters*, vol. 40, no. 17, pp. 4178-4181, 2015.

[4] Richard A. McCracken, Karolis Balskus, Zhaowei Zhang, Derryck T. Reid, “Atomically referenced 1-GHz optical para metric oscillator frequency comb,” *Optics Express*, vol. 23, no.12, pp. 16466, 2015.

[5] Zhaowei Zhang, Karolis Balskus, Richard A. McCracken, Derryck T. Reid, “Mode-resolved 10-GHz frequency comb from a femtosecond optical parametric oscillator,” *Optics Letters,* vol. 40, no. 12, p. 2692, 2015.

[6] Zhaowei Zhang, Karolis Balskus, Richard A. McCracken, Derryck T. Reid, “Mode-resolved 10-GHz frequency comb from a femtosecond optical parametric oscillator,” The European Conference on Lasers and Electro-Optics, PD_A_6

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