

Publishable summary: IMPROV – 1st reporting period (09/2010 – 08/2011)***Project context and objectives***

High power and in particular tunable mid-infrared short pulse laser systems operating in the wavelength range between 3 μm and 11 μm have a large potential in different applications, e.g. analytics, medicine and micromachining. Up to now, those systems are only be realised by multistage set-ups consisting of bulk four or five different units, a configuration which is of course very complex and expensive with low efficiency operation. This situation has not changed over the last years, but now with the project IMPROV a solution seems to be appearing.

IMPROV focuses first on the development, investigation and realisation and of such a highly integrated mid-infrared laser source. Its layout is based on a Master Oscillator Power Amplifier (MOPA) short pulse Thulium all-fibre laser operating around 2 μm . This laser source is used as front-end of an Optical Parametric Generator (OPG) enabling the generation of mid-infrared radiation at wavelengths between 3 and 10 μm . The latter one is based on highly efficient orientation-patterned quasi-phase matched GaAs crystals. Using this laser, a promising application, namely Resonant Infrared Ablation (RIA) for processing Organic Photovoltaic solar cells (OPV) is investigated within IMPROV.

The general structure of IMPROV can be described as follows:

- Development of a short-pulse mid-infrared MOPA laser source based on
 - Thulium fibre oscillator operating around 1.9 μm as seed source
 - High power Thulium fibre amplifier chain
 - OPG/OPA nonlinear wavelength conversion unit stage
- Evaluation of resonant infrared ablation (RIA) of OPV stacks
 - Preparation of a process evaluation set-up
 - Integration of the mid-infrared laser source
 - Ablation tests of single and multilayer elements
- Demonstration of the functionality of the mid-infrared tunable laser source regarding
 - Mechanical and optical parameters
 - Integration and compatibility with a practical industrial application

For the MOPA pump source, different integration aspects will be addressed in order to fully benefit of a waveguide device. This includes the development of fibre-coupled saturable absorbers, large mode area (LMA) photonic crystal fibres (PCF) with functionalities regarding wavelength tuning capability, mode-filtering and high power operation, pump/signal combiners based on LMA-PCFs and novel concepts for fibre amplifiers with integrated core-pumping schemes. The wavelength conversion unit will be realised with integrated wavelength tunability and structural design. This MIR-laser will operate in the wavelength region from 2.5 μm to 11 μm with a pulse energy of up to 30 μJ , a pulse duration between tens to hundreds of picoseconds and a repetition rate between 0.1 to 1 MHz. For validation of the developed laser source, tests concerning the processing of organic photovoltaic solar cells will be accomplished.

Work performed since the beginning of the project and the main results achieved so far

WP2: Saturable absorber and laser oscillator started with numerical simulations of the mode-locked 2 μm -fibre oscillator and corresponding saturable absorber mirrors (SAM) in order to make first estimations of cavity configurations and suitable optical parameters of the fibre oscillator and especially of the SAM. These investigations indicated, that the parameters of the SAM are more critical compared to those of the fibre sections and lay-out. The best laser performance could be achieved applying a SAM with a modulation depth of 30 % and a saturation fluence of 70 $\mu\text{J}/\text{cm}^2$.

Based on these design studies BATOP developed and grew first reflective absorbers with corresponding parameters by using low temperature molecular beam epitaxy. Assembled SAMs have been passed to LZH for mode-locking experiments. In a second step, two types of transmissive saturable absorbers were developed. Type 1 without a partial reflector with absorbtances of 25 % and 43 %, respectively, which can be used e.g. as mode-locker in fibre ring cavities and type 2 with a reflection coating, which can be applied as mode-locker and simultaneously as output coupler.

In addition to saturable absorbers, different fibre components like wavelength division multiplexers (WDM), output couplers and pump/signal combiners are needed for the development of 2 μm fibre lasers but are only commercial availability is limited. Therefore these components have been developed at LZH coupler facility. In particular, 1560 nm/1980 nm WDMs, 70/30 fibre couplers at 1980 nm based on SMF28 fibres and pump combiners for 793 nm clad pumping as well have been fabricated and successfully implemented in different short-pulse fibre oscillators.

At first, we build up a Thulium-fibre oscillator based on a ring cavity, mode-locked by non-linear-polarisation evolution and operating in a pure soliton regime. In order to tune the wavelength from the free running value of 1.90 μm to the long wavelength edge (planned wavelength was 1.98 μm), we used a corresponding interference filter placed inside the laser cavity. However, laser operation beyond 1.97 μm could not be achieved. In addition, the spectral bandwidth exceeded 8 nm yielding in a pulse duration below 500 fs, which doesn't fit with the required parameters. Therefore, a linear cavity set-up was realised, mode-locked by using a saturable absorber mirror provided by BATOP. The operation wavelength was fixed due to a linear Fibre Bragg Grating with a centre wavelength at 1978 nm and a bandwidth of 1 nm. This soliton laser system delivered 4.7 mW average output power at a repetition rate of 26 MHz, corresponding to a pulse energy of 180 pJ. In order to increase the pulse energy, the overall cavity dispersion of the oscillator was changed to net normal dispersion by applying a chirped Fibre Bragg Grating with a group velocity dispersion of 8 ps^2 (provided by MULTITEL). This system, which has been delivered to MULTITEL as a first fixed wavelength prototype, operated in the dissipative soliton regime with output pulse energies of 1.4 nJ and pulse durations below 90 ps.

In **WP3: Photonic Crystal fibre, combiners and amplifier integration** a comprehensive study on available 2 μm off-the-shelf components has been performed by MULTITEL, which are essential for building the whole laser system operating around a wavelength of 2 μm . This include active components, like Thulium-doped fibres, signal sources and pump sources and passive components, e.g. wavelength division multiplexers, splitters, isolators, circulators, polarizers, Fibre Bragg Gratings, etc. as well. In addition, these devices have been characterized and compared to the data sheets of the corresponding manufacturer. Additionally, preliminary amplification experiments were done with existing components and light sources at MULTITEL. As seed source a soliton shifted femtosecond Erbium fibre laser has been applied with a centre wavelength around 1.85 μm . Amplification up to 1.4 Watt at that wavelength could be achieved.

In parallel, NKTP developed pre-amplifier photonic crystal fibres with a core diameters $> 20 \mu\text{m}$ and NA < 0.1 , which can be used either for core or cladding pumping. As an optimum hole size is unknown, a coarse sweep of different hole sizes was performed. The fibres have been characterized

in respect of spectral attenuation in the core from 1200 up to 2400 nm and of pump signal absorption in the cladding from 600 to 2400 nm. Furthermore, NKT have tried scaling the design up to the 50 μm core envisioned in IMPROV. The resulting fiber has been tested in a laser set-up using cladding pumping, as reported in a scientific paper by Modsching *et. al.* in Optics Letters. (IMPROV acknowledged). Here single-mode performance was demonstrated (M^2 of 1.15) and an optical slope efficiency of 36% was found using pumping at 793 nm. This result is somewhat behind the state of the art on Tm doped silica fiber where >60% have been demonstrated by Nufern on smaller core fibers. A literature survey showed that the current Tm concentration of 2.5 wt% is insufficient, as the best performances have been achieved with > 4 wt% Tm concentration. Thus a second material batch with larger Tm concentration has been ordered, and will arrive at NKT in October 2011. In late 2011 NKT will produce a sweep of designs for a 50 μm core PCF based on the novel material. The fibers will be characterized and the optimum design reproduced to deliver D3.7 and D3.8 in February 2012 as planned in Annex I. If the novel material composition increases the slope efficiency then NKT expect to launch its first Tm doped CF product early 2012. Such a product would be IP protected as NKTs background IPR portfolio on double clad PCFs is also applicable at 2 μm .

It should be noticed that the larger concentration is not necessarily needed for core pumping, and this issue is currently investigated together with Multitel, and the first results are shown in deliverable report D3.4.

Looking ahead, NKT are considering the methods for realizing the pump/signal combiner given in D3.9. It was originally planned making a fused fiber bundle combiner, based on a foreground IPR from NKT. This concept has been tested since 2008 for similar combiners for Ytterbium-doped fiber operation at 1 μm . However, the development at 1 μm has faced unexpected problems and a solution is still not in place. In order to reduce the overall risk, NKT thus propose to partially outsource the development, as NKT have found a company claiming they can make such combiners at a cost of 50.000 Euro. Accordingly NKT have in September 2011 asked the coordinator for permission to redistribute our budget by moving 50.000 Euro from “Personnel” to “Consumables”.

After receiving the oscillator from LZH, amplification tests were done at MULTITEL. Pulses were pre-amplified and chirped to have 300 mW power and 800 ps pulse duration before the amplification. Two amplification methods were compared with four different pump wavelengths. The core pumping tests showed that the shortest necessary doped fiber length could be reached by pumping at 1560 nm with 38% slope efficiency.

At 1270 nm the slope efficiency was higher (44%) but a longer piece of doped fiber was required. The maximum total power achieved was 6.5 W that meant 1.8 μJ pulse energy at 3.6 MHz repetition rate.

Core pumping of large mode area photonic crystal fibres fabricated by NKTP were tested as well by butt coupling with passive LMA fibers. The maximum slope efficiency was measured to be 28%.

The highest output power was reached by clad pumping, however the doped fiber used for the experiment was more than 4 m long so the SPM became significant. The slope efficiency of the clad pumping was 43% and the maximum output power was 8 W that meant 2.22 μJ pulse energy.

WP4 Mid-IR Tunable laser source prototyping started with design studies of the frequency converter based on orientation-patterned GaAs crystals (OP-GaAs). It could be shown that a single grating with a period around 56 μm can accommodate the requirements of the planned polymer ablation tests due to a reasonable temperature tuning of the output wavelength. In addition, other opto-mechanical properties, like crystal length, width and thickness which are influencing the conversion efficiency have been reviewed. Damage threshold issues have been taken into account

and a corresponding design tool has been established. Applying this tool, slightly larger crystal dimensions than initially planned must be considered in order to avoid crystal damage and to handle pulse durations of 500 ps. The corresponding fabrication process has started thank to a dedicated photolithographic mask. In order to facilitate the optical design of the first fixed-wavelength prototype before the fiber pump from MULTITEL is available, a laboratory experiment has also been set up. It is based on an available 1064 nm source operating at low repetition rate and two PPLN crystals in an OPG/OPA configuration to obtain 1.98 μ m pulses suitable to pump OP-GaAs crystals and measure relevant parameters such as the beam quality and spectral widths of the mid-infrared signal and idler beams.

The objective of **WP5: Resonant infrared ablation for structuring OPV** is to validate the mid-infrared laser source for structuring organic photovoltaic material stacks. In the resonant infrared ablation process the wavelength of the mid-infrared laser will be tuned to one of the molecular vibrational transitions of the polymer to be ablated. For that reason, the IR absorption spectra of the prototypical materials used in an OPV were characterized in the wavelength regions that will be reached by the final laser setup. Focus was on OPV substrate materials – PET (Polyethylene Terephthalate), transparent conductive materials – PEDOT:PSS, as well as oligomeric hole transport materials – BPAPF. Attenuated total reflectance infrared spectroscopy (ATR FT-IR) was employed to characterise the substrate foils, since a measurement in transmission requires a sample thickness limited to a few tens of microns. The thin-film materials were applied on an IR transparent substrate material (e.g. NaCl or CaF₂), to avoid overlap in the spectra of thin-film coating versus substrate material. Reliable spectra could be obtained using measurements in transmission and in reflection. Using infrared spectroscopy the location and width of the absorption bands could be precisely determined, but it is difficult to obtain a one-to-one quantitative comparison of the strength of the absorption bands for different materials having different layer thicknesses. For that reason we explored the potential of Infrared Variable Angle Spectroscopic Ellipsometry (IVASE), which combines the chemical sensitivity of FTIR spectroscopy with thin film sensitivity of spectroscopic ellipsometry. As an outcome, a number of scenarios for selective patterning was proposed, based on the characteristic wavelengths for each material.

The expected final results and their potential impact and use

The overall aim of the project is the development, realisation and investigation of a highly integrated reliable, compact short-pulse Mid-infrared laser system based on a mode-locked Thulium fibre laser pumping a nonlinear wavelength conversion unit and its validation concerning Resonant-Infrared-Ablation of Organic-Photovoltaic-Devices.

The project IMPROV is driven by an industrial application that has a potential of large economical and societal impact. Indeed, a method for fabricating flexible organic solar cells can be an advance for European citizen through many every day life products. The potential field of micromachining applications based on the laser developed in IMPROV is not restricted to the OPVs market, but also addresses other optoelectronic polymer devices like Organic Light Emitting Diodes (OLED) or Organic Thin Film Transistors (OTFT) and other functional products like stents, particles filters etc.. Beyond micromachining other applications in the field of spectroscopy and medicine can also benefit from the advances in the project IMPROV.

The homepage of the project IMPROV can be found at:

<http://www.fp7project-improv.eu>