A flexible mid-IR laser with high power for temperature-jump spectroscopy

Contact *paul.donaldson@stfc.ac.uk*

A.E. Edmeades

Central Laser Facility, STFC Rutherford Appleton Laboratory, Harwell Science and Innovation Campus, Didcot, Oxfordshire, OX11 0QX

M. Towrie

Central Laser Facility, STFC Rutherford Appleton Laboratory, Harwell Science and Innovation Campus, Didcot, Oxfordshire, OX11 0QX

P.M. Donaldson

Central Laser Facility, STFC Rutherford Appleton Laboratory, Harwell Science and Innovation Campus, Didcot, Oxfordshire, OX11 0QX

Introduction

Time resolved temperature-jump (T-jump) infrared (IR) spectroscopy using pulsed laser heating is a promising method for studying thermally activated processes in heterogeneous catalysis with high time resolution. In T-Jump spectroscopy, nanosecond heating can be achieved, followed by cooling times on millisecond time scales. A probe laser monitors the changes in the IR spectrum.[1,2,3] The microsecond to millisecond sample cooling times hinder the study of thermally activated chemistry on longer timescales. The Tokmakoff group introduced a 2 µm fixed wavelength 30W intensity modulated continuous wave (CW) Thulium fibre laser for creating T-jumps on the sub-ms timescale that can be sustained for seconds after initial heating, allowing longer timescales of observation of a heated sample.[4]

The current T-jump setup at CLF on Ultra B delivers 70 µJ pulses of 1 ns duration at up to 1 kHz.[2] Substantial T-jumps can be achieved with a single laser shot (50-100 ºC in ZSM-5 pellets [1]). By using multiple successive heating pulses it is possible to deliver up to 2 mJ of total laser energy over 30 ms (30 pulses), achieving higher, more sustained T-jumps through a stepped heating process that spans many milliseconds.[1] The T-jumps and their duration achieved by this laser are however substantially limited by the repetition rate of the pump laser. At 1 kHz, the time delay between subsequent pulses tends to be comparable to or longer than the cooling time of samples commonly examined.

We present a new mid infrared wavelength tunable T-jump laser based on a nanosecond pulsed fibre pumping a signal resonant periodically poled lithium niobate (PPLN) optical parametric oscillator (OPO). The fibre laser produces 4-2000 ns pulses at 1-4000 kHz with a maximum pulse energy of 1 mJ. Idler pulse energies up to 100 µJ (c. 84 kHz) and maximum average power of 8W at 10% duty cycle have been observed in the wavelength range of 2-5 µm. This new laser system is capable of causing greater sample heating on a µs timescale than the existing 1 kHz T-jump laser system. The flexibility in the fibre laser parameters provides the opportunity to design pulse sequences that control heating and cooling times. The IR wavelength tunability of the OPO allows the heating of a wide range of sample types to be optimised.

TRUMPF TruPulse Nano Fiber Laser

The pump laser used was a TRUMPF TruPulse Nano fibre laser. This 1064 nm commercial laser is a closed industrial module for use in scribing and marking applications based on a Neodymium gain material. It has 48 pre-programmed optical waveform shapes, controlled via varying electrical drive to the seed laser diodes. Each waveform has an optimum repetition rate for maximum power. The repetition rate can be varied up to an internally defined maximum for each waveform, with each pulse delivering up to 1 mJ of energy. Approximately linear control of the average power output is achieved using the active current set point. This defines the power-amplifier laser diode current while pulses are being emitted and can be varied by the user. Between pulse bursts, and between individual pulses for repetition rates below the optimum, the laser enters a simmer state. In this state, the pump laser diodes are supplied with a user defined low-level current, to pre-charge the amplifiers and avoid a slow ramp-up of pulse energies. The fibre output of the laser is unpolarized. The system can be externally controlled, allowing shot by shot selection of pulse duration, energy and repetition rate.

OPO

The TruPulse laser was used to pump a home-built signal resonant PPLN OPO. The PPLN crystal used was a 50mm long Covesion MOPO3-1.0 with seven channels. The OPO was a linear cavity design with two concave mirrors of radius = 100 mm from Layertec. The input mirror was high reflection, HR coated (>99.9%) for the signal and anti-reflection, AR coated for both the pump (<2%) and idler (<5%) on both sides. The output mirror was HR (>99.5%) coated for the pump and the signal and AR coated (<2%) for the idler on the curved side. The result was an 85mm long signal resonant cavity with two passes of the pump and idler light exiting from both ends. Select Non-Linear Optics(SNLO) modelling predicted a mode size of around 120 µm inside the cavity, not accounting for the presence of the PPLN crystal.

Setup

The T-jump laser was configured as shown in Figure 1. The output was characterised and compared in performance to the existing Ultra B T-jump setup.[2] A Faraday isolator was placed after the fibre laser output to polarise the pump beam and to protect the laser from the back reflected pump light. A wave plate and beam splitter cube were used to further attenuate the fibre laser output as required. The pump beam was focused into the OPO cavity using two 100 cm Thorlabs B-coated lenses paired up to give the equivalent of a 50 cm lens. Flat gold turning mirrors were used during alignment to allow clear visibility of the red alignment laser, with the final turning mirror being replaced with a flat dielectric 1064 nm HR mirror once alignment was complete.

A Thorlabs beam profiler with an ND4 filter was used to measure the spot size of the pump beam in the cavity and observed to be in agreement with the SNLO predictions (~120 µm).

A KTA crystal cut for 1064 nm second harmonic generation was used to generate an intercavity green beam for the first part of the alignment process. Once collinearity of the cavity with the incoming beam was achieved, this was then replaced with the PPLN in the cavity.

Initial detection of the OPO output was with a Kolmar Mercury Cadmium Telluride (MCT) detector with a germanium window used to block the signal and pump.

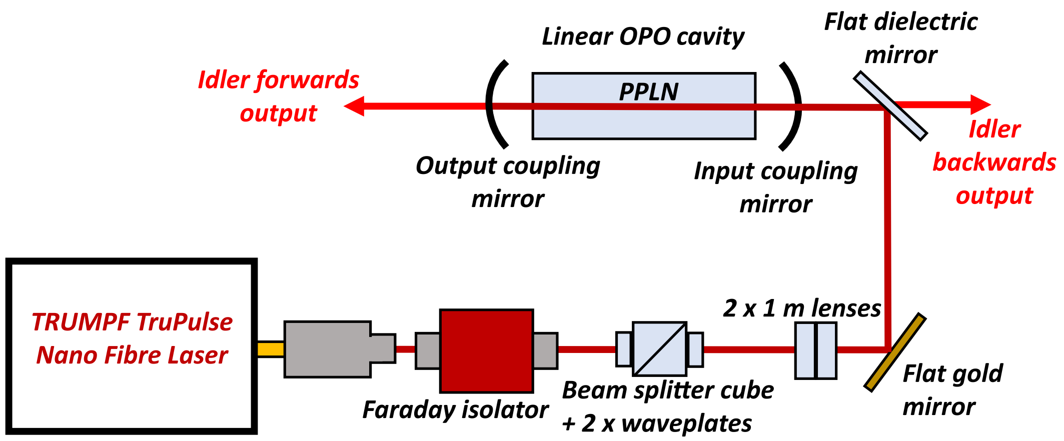

Figure 1: A T-jump laser setup based on a Nd fibre laser and PPLN OPO

Once the cavity was aligned, this sensitive detector was easily saturated, so a pyroelectric detector was then used for idler characterisation.

All seven PPLN channels were tested and assigned by using a Thorlabs fibre optic CCD spectrometer to measure the wavelengths of the weak visible beams emitted from the PPLN. These were assumed to be signal + pump sum frequency generated light, allowing calculation of the signal and idler wavelengths.

Most testing was carried out using a specific intensity profile optical waveform. This had an optimum repetition rate of 84kHz. The PPLN oven was set to 100 ºC, generating idler at around 3200 cm^{-1}. Unless otherwise stated, the active set point was set to maximum.

The pump laser was internally triggered. By replacing the constant 5V power supply to the laser emission gate with a modulated signal from a Stanford delay generator, bursts of pulses, each 0.2 ms long were produced. Using a Molectron pyroelectric energy head with a response time longer than 0.2 ms, the total energy for each burst was measured, then used to calculate an average pulse energy within the burst. For continuous pulse trains, without bursting, similar measurements were carried out with a thermopile power meter (Ophir).

Results

The beam profile of the idler beam coming out of the cavity in the forwards direction was measured using a Seek microbolometer array. When focused with a 20 cm CaF_2 lens, the FWHM was ~90 µm.

Initial investigations examined the idler output when the pump laser was pulsing continuously, not in a burst mode. For repetition rates from 1-6 kHz, average pulse energies were 66-89 µJ. Above 6 kHz, the OPO became unstable, which was most probably due to a thermal lensing effect as the PPLN crystal heated.

The active set point on the pump laser was used to control the pulse energy of the idler, resulting in almost linear control of both the pump and OPO output (Figure 2). Similar control was achieved through attenuation with the waveplate and polarizer, as displayed in Figure 3.

By using bursts, it was possible to increase the repetition rate further without the OPO becoming unstable. This is because the time between bursts allowed for cooling of the crystal to take place.

The highest pulse energies and highest total energy delivered in a burst were both observed at around 100kHz. Here, there was found to be an increase in intensity over the first 3 pulses, then constant intensity, as seen in Figure 4. This rise time varied with repetition rate, a very high energy first pulse being generated for 200 kHz and above. The rise time was altered by changing the simmer setting on the pump laser. After accounting for the relative delay between detectors, the rise of the OPO output at 100 kHz was 8 ns following the arrival of the pump, corresponding to around 12 round trips of the cavity.

The waveforms delivering the most energy in a burst were those that had optimum repetition rates of 80-100 kHz. Pulse energies for these waveforms were 80-100uJ, corresponding to generation of ~1.7mJ total energy in 0.2 ms.

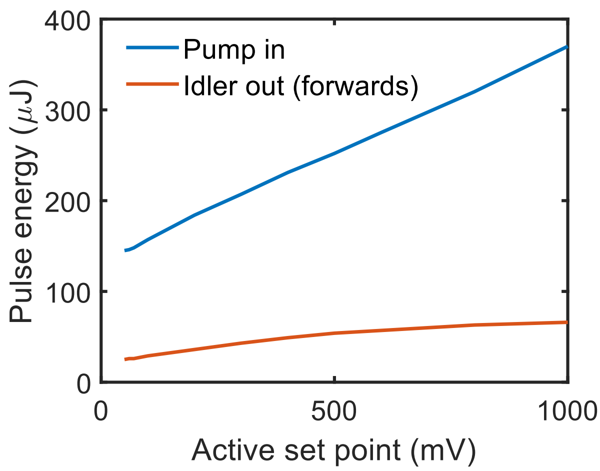

Figure 2: Average pulse energy of pump input to and idler output from the cavity, at 1 kHz in continuous pulsing mode, as a function of the active setpoint of the fiber laser

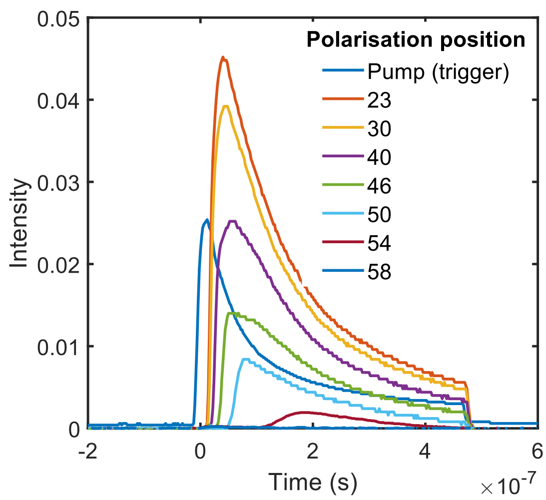

Figure 3: Temporal profiles of single idler output pulses, measured using a fast response MCT (Vigo), for different levels of pump attenuation, operating at 1 kHz in continuous pulsing mode

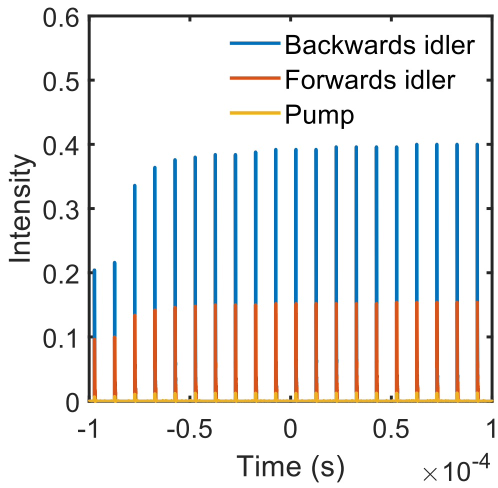

Figure 4: Temporal profile of a 0.2 ms burst of pump and idler pulses at 100kHz

Depending on the PPLN channel chosen, between 60 and 95% of the idler light was emitted in the forward direction. The reason for this forward bias is likely to be due to strong depletion of the pump beam in the first pass leaving less power for conversion to idler in the return pass. However, there may also be a contribution to forward bias if the mode matching between pump and signal is better in the first pass than for the return.

The highest energy pulses were recorded using the PPLN channel corresponding to an idler output of 3000 cm^{-1}, at 108 µJ. The lowest pulse energy recorded at 100 kHz was 25 µJ for the 2400 cm^{-1} channel.

Outlook and conclusions

A new T-jump laser based on a nanosecond pulsed fibre laser pumping a signal resonant PPLN OPO has been built. Pulse energies up to 100 µJ at up to 100 kHz have been achieved at 3200 cm^{-1} corresponding to a 100x increase in the amount of energy delivered to a sample in a given time, compared to the current Ultra B 1 kHz T-Jump system. This higher repetition rate will allow less time for sample cooling between subsequent pulses in a burst. Flexibility and parameter control of the pump laser present the opportunity to design pulse patterns that will achieve large T-jumps followed by T-holds and cooling on custom magnitudes and timescales.

Next steps will involve developing the capability to control pulse energy on a shot by shot basis, as well as gaining an understanding of the pulse patterns that are required to sustain the temperature of a sample after an initial T-jump. Delivering the energy in longer pulses reduces the peak intensity, allowing higher T-jumps to be achieved without reaching the threshold for plasma generation, known to occur for tightly focused ns IR beams in liquids.

With the TruPulse-pumped OPO developed, we observe some issues with thermal lensing causing instability at high powers. It is possible that this could be reduced by using longer focal length mirrors, or by replacing one of the curved mirrors with a flat mirror, resulting in a larger beam inside the cavity.

The TruPulse output is unpolarised, so half of its output is discarded. To utilise all of the energy from the pump laser, a second OPO can be implemented. This will enable pumping of a sample from both sides, as well as independent control of the parameters of the two lasers.

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