ULTRA laser system: a new dual-output 10 kHz Ti:Sapphire amplifier with UV–IR generation for time-resolved spectroscopy

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Introduction

ULTRA is a jointly funded STFC and BBSRC facility development which aims to advance time-resolved spectroscopy capabilities through the provision of flexible multiple-beam techniques, with high level of sensitivity required to probe complex molecules.

The ULTRA laser system provides synchronised 10 kHz outputs tunable across the UV – IR spectrum with femtosecond and picosecond pulse durations. This will allow user access to an array of linear and non-linear optical pump – probe spectroscopy techniques to investigate molecular dynamics across diverse applications on ultrafast to millisecond timescales. An overview of the recently commissioned ULTRA laser system and its benefits are described in this report.

Dual output Ti:Sapphire laser system

The central laser of the ULTRA system is a dual titanium sapphire chirped pulse amplifier (CPA) custom developed by Thales Laser. A single <20 fs femtosecond oscillator (Femtolaser) seeds two CPA arms. The first arm (A) is seeded with the oscillator to generate 10 kHz, 40-80 fs, 1 mJ pulses at 800 nm. The second amplifier (B) is seeded with a bandwidth filtered portion of the oscillator output to generate 10 kHz, 1-3 ps, 1 mJ pulses which are tunable across 775-825 nm. The seed pulses of both arms can be independently controlled in spectral phase and amplitude by two acousto-optic programmable dispersive filter devices (Dazzler, Fastlite). This enables computer controlled selection of pulse bandwidth / duration and the output wavelength (*A*: ± 5 nm, *B*: ± 1 nm).

For such high average power laser amplifiers, thermal lensing (caused by a variation of refractive index with temperature) in the amplifier medium can lead to serious aberrations and degradation of the beam quality. Cryogenic cooling of the titanium sapphire is used in this laser system to increase the thermal conductivity of the amplifier medium by several orders of magnitude[1]. This increase in thermal conductivity ensures efficient removal of heat from the titanium sapphire and consequently minimisation of thermal lens effects.

Wavelength tunability

A significant benefit of short pulsed lasers is their ability to use their high peak powers to efficiently drive non-linear optical processes which can provide tunable wavelength radiation. The two outputs from the Ti:Sapphire laser system are both split into three beams with 10, 45 and 45% of the total output powers. The 45% beams are used to pump optical parametric amplifiers (OPAs) (TOPAS, Light Conversion). These OPA devices convert the 800 nm Ti:Sapphire output to UV – IR wavelengths, with the wavelength tuned with computer control. The wavelength ranges of these outputs are described in table 1 and figure 1. The OPAs include a NOPA device, a non-collinear white light continuum amplifier, which generates shorter (<20 fs), tunable, broadband pulses (see table 1). The final 10 % of the output beam is used for harmonic generation in BBO crystals, generating 400, 267 and 200 nm outputs and/or a white light continuum.

OPA device	Tunability, nm
A. Femtosecond pumped (50 fs)	
OPA 1	235-2600
OPA 2 + DFG	1120-20000
NOPA	230-1000
B. Picosecond pumped (2.5 ps)	
OPA 3	235-2600
OPA 4 + DFG	235-20000

Table 1. Tunability of OPA outputs.

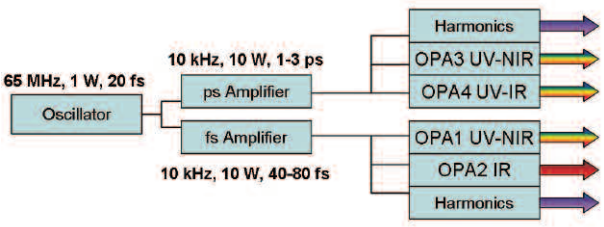

Figure 1. Amplification and wavelength conversion setup of ULTRA laser.

Synchronisation of outputs

Synchronisation of the two outputs A and B from the Ti: Sapphire laser system is essential for experiments requiring ps and fs output combinations. There are a wide variety of advanced multi-beam techniques which would utilise the capabilities of synchronised ps and fs outputs, such as 2D-IR [2] and stimulated Raman spectroscopy [3]. For example, 2D-IR could apply narrowband (ps) IR pumping with broadband (fs) IR probing, to observe vibrational coupling within a molecule. Typical spectra of the narrowband and broadband outputs are shown in figure 2.

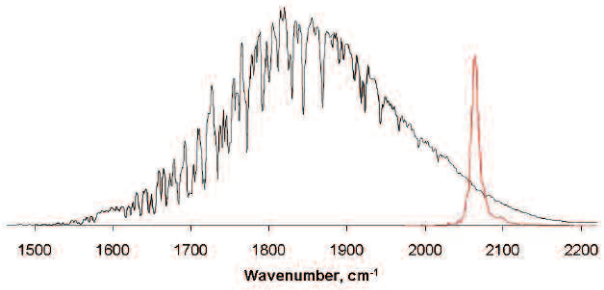

Figure 2. A Black: Broadband fs IR spectrum. Water vapour absorption is visible on the spectrum. The 75 lines/mm grating 0.25 m spectrograph provides ~400 cm^{-1} bandwidth, <5 cm^{-1} resolution in a single shot. Higher and lower numbers of lines gratings provide access to higher resolution or larger single shot bandwidth. B Red: Spectrum of the tunable narrowband ps IR output, ~11 cm^{-1} FWHM.

Outputs A and B are inherently synchronised as they are seeded by the same femtosecond oscillator laser. However, some thermal drift inside the two amplifiers may occur so the outputs could lose synchronisation to some degree over long periods. Some slow drift in relative timing (~300 fs, or 100 µm, per hour) has been measured. This is slow enough for experiments to begin with occasional optimisation of timing. A cross-correlation feedback correction is planned to be implemented to compensate for this slow drift in the future.

Stability and sensitivity

The shot-to-shot stability of the Ti: Sapphire laser system is ~1 % rms. The non-linear nature of the OPAs means that this becomes as large as 5% at the OPA outputs. By separating the probe laser pulse into two parts, reference and probe beams, much of this noise (spectrum and energy fluctuations) will be removed by normalisation to the reference beam spectrum shape and intensity. The aim is to bring the system to the sensitivity levels on the order of the detector shot noise, ~2000:1 rms.

Further sampling techniques such as reading a background with every second laser shot (effectively sampling at 5 kHz), significantly reduces contributions from 'slow' noise sources (e.g. vibrations from air-conditioning and cryo-cooling systems).

A final point to note is that the short pulses allow broad spectral windows to be probed in a single experiment, avoiding having to repeat the experiment several times with the probe laser set to different wavelengths to gain the complete spectrum. Customised 10 kHz, up to 512 pixel readout detectors spanning the UV- mid-IR have been developed by the STFC Technology Department. The mid IR spectra shown in figure 2 were taken using a 256 element mercury cadmium telluride sensor array.

We expect these techniques combined with the high repetition rate sampling should enable us to monitor ~10^{-6} changes in spectral intensities within 1 second.

References

1. S. Backus, C. G. Durfee III, G. Mourou, H. C. Kapteyn and M. M. Murnane, *Optics Letts*, **22**, 1256, (1997).
2. P. Hamm, M. H. Lim and R. M. Hochstrasser, *J. Phys. Chem. B*, **102**, 6123, (1998).
3. P. Kukura, S. Yoon and R. A. Mathies, *Analytical Chem.*, **78**, 5952, (2006).