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# Introduction

The introduction of the Time-Resolved Multiple Probe Spectroscopy (TRMPS)<sup>1</sup> capability at the ULTRA laser facility<sup>2</sup> in 2011 provided the ability to obtain information on long timescale chemical dynamics (ns - ms) in combination with ultrafast timescales (fs - ps) on a single instrument. An additional advantage of this multiple probe technique is the capacity to obtain more information per experiment on precious samples, which may quickly photo-degrade. These benefits have been successfully utilized in several protein dynamics studies<sup>3-5</sup>.

LIFEtime is a new BBSRC-funded instrument to be installed into the Research Complex at Harwell through 2014. The instrument builds on the facility's TRMPS capability and introduces higher (100 kHz compared to ULTRA's 10 kHz) repetition rate mid-IR laser systems for 10x more probe pulses per pump pulse.

Mid-infrared (IR) pulse shaping capability<sup>6</sup> will also be introduced to the laboratory to enhance two-dimensional IR (2D-IR) experiments, ongoing in the Central Laser Facility (CLF) since 2008<sup>7, 8</sup>.

## LIFEtime Laser System

The LIFEtime laser system, currently being commissioned, is a dual-output 100 kHz repetition-rate ytterbium-based laser (Pharos, Light Conversion, fig. 1). The custom dual-output provides individually configurable, synchronized sources for pump and probe lasers. The probe laser will run at 100 kHz, while the pump laser can run at variable repetition rate, single-shot to 100 kHz. This provides the possibility to easily configure the number of probe pulses (one every 10  $\mu$ s) per pump pulse, according to the timescales of interest in each experiment. The relative timing of the outputs will also be computer-controllable in steps of ~ 12 ns, according to the common oscillator roundtrips. The third aspect of the pulse timing control will be addressed by optical delay, for fs – ns timing.

Figure 1. Overview of LIFEtime's dual 100 kHz laser system.

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wavelength tunability from 210 - 16000 nm with < 300 and < 200 fs pulse durations for pump and probe outputs.

It should be noted that the longer pulse durations of the laser (typical of ytterbium-based systems) will reduce capability in terms of temporal response and bandwidth relative to ULTRA (50 fs). However, routes to shorter pulses and broader bandwidths are available through self-phase-modulation and non-colinear-OPA schemes, so this type of high average power ultrafast laser technology provides a potential route for next generations of molecular dynamics facilities. This potential is currently under review within the CLF.

### **Mid-Infrared Pulse Shaping**

The LIFEtime instrument will introduce novel mid-IR pulse shaping capability to the facility. The pulse shaper, based on a germanium acousto-optic modulator (PhaseTech Spectroscopy, see fig. 2)<sup>6</sup> will enable shot-to-shot pulse shaping, designed for rapid scanning of pump pulse interference in 2D-IR experiments with the ability to phase-cycle pulses, reducing background noise. Customized to work at 100 kHz, the device will allow several 2D-IR spectra to be acquired every second, dramatically improving current acquisition times of minutes. Although currently to be applied primarily to 2D-IR spectroscopy, applications of this technology are only just beginning to be explored.



Figure 2. Overview of LIFEtime's mid-IR pulse shaper application to 2D-IR spectroscopy.

# Time-Resolved IR Micro-Spectroscopy

LIFEtime's high repetition rate and pulse shaper ability to remove background scatter is suitable for 2D-IR, time-resolved IR and other ultrafast nonlinear methods, to be conducted in a spectro-microscopy mode with diffraction-limited resolution. A prototype time-resolved IR microscope for this purpose has been constructed, see fig. 3. This capability will open up new applications problems in protein crystallography, amyloidosis, tissue analysis and catalysis.

The laser system will drive multiple optical parametric amplifier systems (OPA, Orpheus, Light Conversion) to provide



Figure 3. Prototype time-resolved IR absorption microscope.

### Conclusions

The LIFEtime instrument will provide novel TRMPS and 2D-IR capability to Research Complex at Harwell for application to the biological sciences. The instrument will complement the current ULTRA facilities and introduce new and exciting technologies to the CLF.

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