Multi-amplifier laser system for time-resolved spectroscopy at HiLUX-Ultra

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Introduction

HiLUX is a UKRI Infrastructure Fund project, to deliver the next generation of ultrafast laser spectroscopy facilities. The facility will provide a range of capabilities in ultrafast XUV – IR spectroscopy, photoelectron and photoion spectroscopy, and non-linear spectroscopies. The facilities will be accessible to academia and industry in the UK and internationally.

To provide such capability, two laser systems were proposed (for HiLUX-Artemis and HiLUX-Ultra). Here we present an outline of the HiLUX-Ultra laser system, currently under construction by Light Conversion. It will supply a broad range of multi-kilohertz, femtosecond to picosecond pulsed laser outputs, in a multiamplifier format, enabling pump-probe spectroscopy measurements across femtoseconds to seconds timescales.

Overview

Figure 1 provides a schematic of the laser system. Five laser amplifiers will provide powers of 20 – 120 W each, at 10 – 1,000 kHz repetition rates, with pulse durations of < 350 fs.

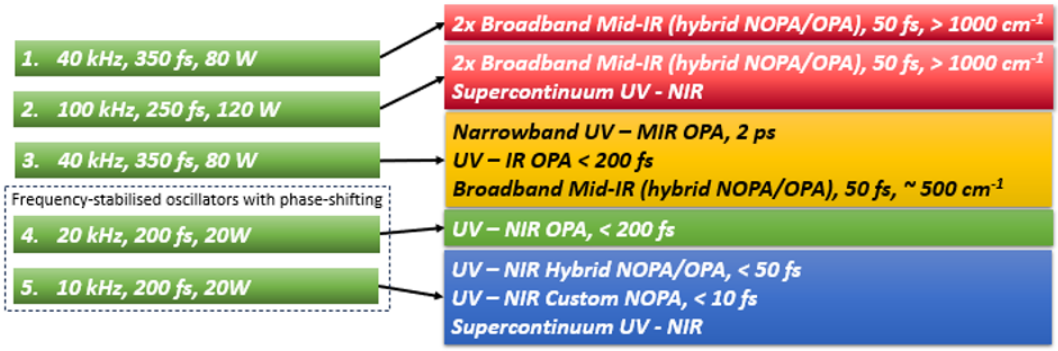

Figure 1. Schematic of laser system.

The five lasers will have options to be synchronised relative to each other, through frequency-stabilised oscillators, to < 200 fs. The synchronisation of multiple amplifiers, with different repetition rates, and control of timing phase relative to each other is a key requirement for time-resolved multiple probe spectroscopy (TRMPS) and the ability to measure dynamics over femtosecond to second timescales and beyond.[1,2]

The laser amplifiers will be based on ytterbium femtosecond technology due to its proven robustness, wall-plug efficiency, beam quality and stability performance. The average power of these systems (up to 120 W here) makes them ideal for driving optical parametric amplifier (OPA) systems at high repetition rates (up to 100 kHz) with a range of optical specifications matched to HiLUX's objective in delivering leading facilities.[3]

The laser amplifiers will pump commercial and/or custom OPAs based on requirements in terms of wavelength and pulse durations. The following sections outline the role of each amplifier and OPA capabilities.

The laser systems will be placed across one continuous optical table spanning three rooms, as light sources for five experiment end-stations, shown in Figure 2 and listed below:

- 2DIR Spectroscopy
- UV – IR TRMPS
- Short-pulse spectroscopy
- Surface sum-frequency generation spectroscopy
- Kerr-gated Raman spectroscopy.

1. Broadband 2DIR laser system

This laser system will be a pump and probe source for use in 2DIR spectroscopy.[4]

The chosen repetition rate of 40 kHz allows a balance of low noise through rapid IR pulse-shaper scanning of pump pulse interference patterns, while maintaining > 1 µJ pulse energies for high signal levels.

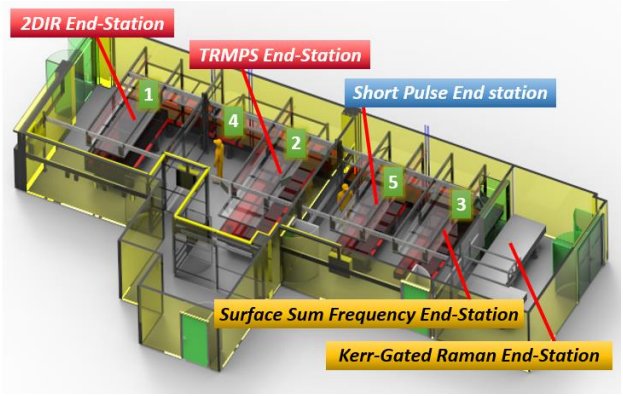

Figure 2. HiLUX-Ultra laser laboratory, under construction. End-stations labelled with associated laser systems.

Two OPAs will cover the mid-IR spectral range from 2 – 15 µm, based on a hybrid noncollinear OPA (NOPA)/OPA architecture.[5] This approach supports tunable, spectrally broad bandwidths (total of > 1,000 cm^{-1}) through a difference frequency generation between a 2 µm NOPA output mixed with a second, narrow but tunable, OPA. The outputs from these OPAs will be usable as 2DIR pump or probe pulses.

2. Broadband TRMPS probe laser system

This laser system will be a probe source for use in electronic and vibrational absorption spectroscopy[2]. High repetition rate is key to maximising information from samples in the TRMPS method. Here, we will work at 100 kHz, where shot-to-shot detection is feasible with array detectors and energies are suitable for the required nonlinear optical processes discussed below.

To enable a wide range of applications, the laser source must generate a large spectral bandwidth, across UV – IR, achievable through simultaneous non-linear optical frequency conversion methods, supercontinuum generation and optical parametric amplifiers.

Supercontinuum generation will supply 300 – 2,000 nm (to be tested) probe sources, compressible to < 100 fs by chirped mirrors. A single continuum source to cover this range has challenges in terms of stability, intensity, damage limits and detector types, so we anticipate three continua will be required (UV/blue, visible/red and near-IR).

Two OPAs will cover the mid-IR spectral range from 2 – 15 µm, with a useable probe spectrum of > 1,000 cm^{-1}, like that discussed in the previous section.

3. Picosecond pulse laser system

For nonlinear spectroscopy methods of time-resolved Raman and surface sum-frequency generation (SSFG), narrowband, picosecond pulses are typically used[6, 7] to support energy resolved spectroscopy. Here we use part of an 80 W, 40 kHz amplifier to pump a picosecond OPA with a second harmonic bandwidth compressor (SHBC) on the input beam[8]. The SHBC system can provide spectrally narrow pulses (< 10 cm^{-1}) at 515 nm, which can pump an OPA to generate tunable (320 – 2,600 nm) narrowband pulses for SSFG and Raman experiments. Additional difference frequency generation (DFG) stage on this OPA extends the wavelength of the narrowband pulses to 4,800 nm, provides opportunities in 2DIR-Raman spectroscopy[9].

For excitation of time-resolved Raman and SSFG experiments, a broadly tunable pump pulse will be generated by an OPA (210 – 2,600 nm). This can function as a pump source from femtosecond to nanosecond timescales. If timescales beyond a few nanoseconds are required in the Raman or SSFG experiments, pump light can be redirected from one of the frequency stabilised amplifiers, 4 or 5, discussed below.

This laser system will reserve a portion of the fundamental ytterbium laser amplifier for Kerr-gating of Raman and fluorescence. This pulse requires high energy, up to 1 mJ per pulse, to induce birefringence efficiently in polarisable media.[6] The Kerr-gating pulses will have an independent pulse compressor to tune the duration of the beam from 1 – 5 ps (typical 2 ps).

For SSFG[7], a spectrally broadband mid-IR source is required. When switching from Kerr-gated Raman experiments, the Kerr-gating beam will be compressed to < 350 fs and redirected to pump an OPA for the mid-IR spectral range, from 2 – 15 µm, like that discussed in Section 1, above.

4. Tunable pump laser system

For excitation of TRMPS or transient-2DIR[10] experiments, a broadly tunable pump pulse will be generated by an OPA (210 – 2600 nm). This can act as a pump source from femtosecond to second experiments. Automated pointing, timing and focusing control is being designed to enable seamless tuning of the pump across the wide spectral range for maximising applicability of this system. This system will run at 20 kHz, as electronic excitation of samples typically requires some recovery period or sample refresh, so 100 kHz pumping is not routinely workable.

5. Short pulse laser system

While the time-range of the TRMPS multi-amplifier approach can address many of the time-resolved spectroscopy experiments at the facility, < 100 fs synchronisation is a challenge to maintain across multiple amplifiers. This system will focus locally on the < 100 fs domain, but can also provide a long-timescale (femtoseconds to seconds) pump source for the other experiments.

As nonlinear compression of pulses is a key parameter here, and (as with system 4) electronic excitation typically requires some recovery, we select a lower repetition rate (10 kHz) with 2 mJ energy per pulse.

A < 50 fs hybrid NOPA/OPA system (like the MIR OPA in system 1 above, but with sum frequency generation rather than DFG final stage), will pump < 100 fs transient absorption experiments from 250 – 900 nm, using a chirped-mirror compressed supercontinuum as the probe.

For further capability to < 10 fs, a custom NOPA will be built[11] (with additional compression under consideration). These very short pulse systems naturally provide very broad spectral output, which will enable more advanced spectroscopies, such as impulsive Raman[12] and 2D electronic spectroscopy.[13]

Summary

We present here the diverse range of ultrafast laser sources which will be introduced to the Ultra facility by the HiLUX project. The systems enable a comprehensive, world-leading range of ultrafast spectroscopies while considering robust facility requirements. Installation is scheduled in multiple stages across 2025.

These laser systems provide access to such a range of powers, pulse durations and wavelengths that the facility will be able to adapt to new techniques as they are developed, well into the future. For example, as well as kHz repetition rates, these laser amplifiers can switch to MHz repetition rates, opening a route to short-pulsed imaging techniques in the future.

Acknowledgements

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