Artemis Upgrade, Relocation and the New 100 kHz OPCPA Laser for ULTRA / ARTEMIS

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Introduction

The Ultra[1] and Artemis[2] laboratories provide ultrafast dynamics and spectroscopy facilities for UK and international scientists, addressing problems across physics, chemistry and biology. Synergies in the technology and experimental approaches of these two CLF facilities will be exploited in the coming years by relocation of Artemis to the Research Complex at Harwell, the home of Ultra. The acquisition of a new 100 kHz optical parametric chirped pulse amplifier (OPCPA) laser system, obtained from STFC and BEIS capital funding, will upgrade the facilities and underpin future laser technology for both facilities. Additionally the introduction of a third amplifier to the existing Artemis Ti:sapphire laser to boost the energy to 2x 10 mJ will be used to generate higher energy pulses in the ultraviolet (UV) and short-wave infrared (SWIR) spectral regions to enhance atomic and molecular optics (AMO) experimental capabilities.

We present an overview of the new laboratory and laser system to be installed in 2019, with some examples of the applications to be addressed over 2019/2020.

New Artemis Laboratories

The design of the new laboratory, consisting of separate laser and experiment rooms with services supplied from its own plant room is shown in Figure 1. The laser area will house the upgraded Ti:sapphire chirped pulse amplifier (CPA) and the new 100 kHz OPCPA (described in the next sections). The experiment room will consist of two independent experimental areas, each containing two optical tables. Interlocked wall shutters between the laser and experimental areas will control access to the lasers for each experiment.

Experimental area 1, primarily used with the 1 kHz CPA laser, will house the existing Artemis high harmonic generation (HHG) extreme ultraviolet (XUV) beamlines utilizing: (1) a grazing incident monochromator[3] and subsequent grazing incidence toroidal mirror coupled to either time-of-flight (ToF) or velocity map imaging (VMI) photoelectron spectrometers (PES); and (2) a flat field XUV spectrometer and imaging chamber containing XUV multilayer mirrors and an in-vacuum XUV charge coupled device (CCD).

Experimental area 2, primarily used with the 100 kHz OPCPA, will house a new HHG XUV beamline containing a grazing incidence monochromator and grazing incidence toroidal mirror (potentially upgraded to an ellipsoidal mirror), coupled to a hemispherical analyser for time- and angularly resolved photoelectron spectroscopy (TR-ARPES)[4] experiments, and a transient absorption spectroscopy beamline. Experimental area 2 will also house mid-infrared spectrometers and additional equipment for spectroscopy applications.

The external plant room will house vacuum pumping stations, cryogenic cooling and water cooling. This will help to isolate environmental noise from the laser and experimental areas, improve working conditions for users and allow access to services for maintenance with minimal disruption to operations.

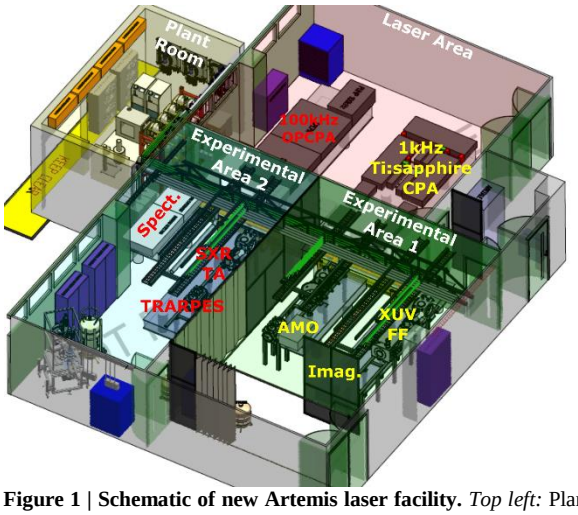

Figure 1 | Schematic of new Artemis laser facility. *Top left:* Plant room containing vacuum pumps and laser chillers. *Top:* Laser room containing upgraded 1 kHz Ti:sapphire CPA and new 100 kHz OPCPA. *Bottom right:* Experimental area 1 containing existing Artemis beamlines: atomic and molecular optics (AMO), XUV flat-field (FF) and imaging (Imag.). *Bottom left:* Experimental area 2 containing new Artemis beamlines: soft x-ray transient absorption (SXR TA) and time and angularly resolved photoelectron spectroscopy (TR-ARPES) and Ultra spectroscopy (Spect.) area

100 kHz OPCPA Laser System

With increasing application in areas such as ophthalmology and laser machining, ytterbium-based lasers have seen major advances in average powers over the last 10 years as a result of the commercial availability of kilowatt-scale short pulse laser systems. Using nonlinear optical processes, such as OPCPA, these industrial lasers can form a core light source in scientific laser laboratories. The OPCPA approach in particular allows efficient generation of wavelength tuneable outputs and shortening of the pump pulses to sub-10 fs.

The new laser system, shown in Figure 2, has been presented previously[5], so only a brief overview is given here. The system to be installed in the new laboratory is a 200 W ytterbium-based Trumpf Scientific laser system pumping a Fastlite BBO/LN-based OPCPA system. The laser will provide two tuneable outputs in the SWIR (1.45-1.85 μm) and mid-infrared (MIR: 2.3-3.9 μm) spectral regions, with a pulse duration of ~100fs and pulse energies of >50-100 μJ and >30-60 μJ across the full tuning ranges in the SWIR and MIR respectively. The system can be optimised to provide fixed outputs at ~1.7 μm and ~2.6 μm, delivering ~50 fs duration and >170/60 μJ (SWIR/MIR) energy pulses. Acousto-optic programmable dispersive filter (Dazzler, Fastlite) control of the seeding permits rapid and complex control of the laser's temporal and spectral characteristics, and stabilizes the carrier-envelope phase of the system to < 150 mrad[6]. Alternatively, one can obtain narrowband (~40 cm^{-1}) ~0.5 ps pulses across the full tuning ranges. Frequency conversion of these outputs will be developed to extend this capability to: the visible (<1 μm) and UV (200-400 nm) spectral regions through harmonic/sum-frequency generation; the XUV (20-100 eV) and soft x-ray

(SXR: >150 eV) via HHG; and further into the MIR (> 4 μm) through difference frequency generation / optical parametric amplification.

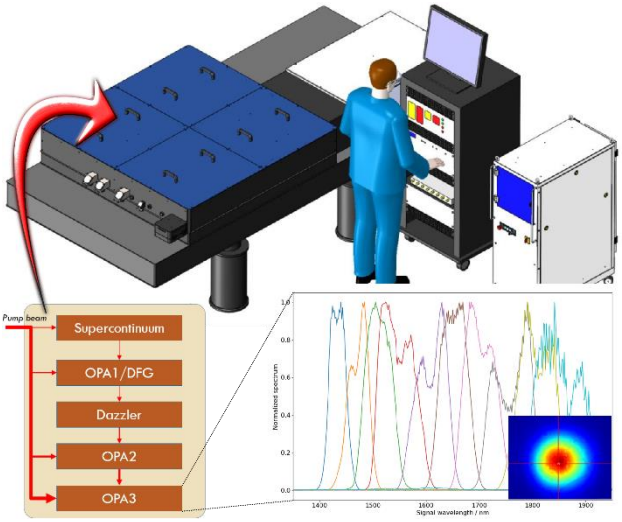

Figure 2 | Ytterbium pumped OPCPA system. *Top: 3D model of new laser. Bottom left:* Schematic of OPCPA architecture. *Bottom right:* Example of tuneable spectrum in the SWIR and the high quality spatial mode shown in the inset

Ti:sapphire Upgrade

The current two-stage Ti:sapphire CPA system delivers an output of ~12 mJ at a repetition rate of 1 kHz, central wavelength of ~780 nm and pulse duration of ~30 fs. Typically 8-10 mJ of this is used to pump a commercial OPA (Light Conversion TOPAS-HE) to deliver tuneable pulses across 1160-2600 nm with a maximum energy of ~1.5 mJ, with the remaining 2-4 mJ sent to a separate compressor and delay line. We will split the output of the first amplification stage to simultaneously pump the original second amplification stage and the newly introduced third amplification stage to generate two temporally locked beams of 10 mJ each, one of which can optionally be used to pump the OPA as in the old setup, as shown in Figure 3. Through various harmonic, sum- and difference-frequency generation processes, the wavelength output can be extended down to the UV (>200 nm) and into the MIR (<10 μm) spectral regions. A differentially pumped gas-filled hollow-core fibre (HCF) can be used to temporally compress the near-infrared (780 nm) or SWIR outputs to sub-10 fs durations[7,8], the latter of which is passively carrier-envelope phase-stable. HHG is used to generate XUV radiation (20-100 eV) for use in HHG spectroscopy, photoelectron spectroscopy and coherent diffraction imaging[9] experiments.

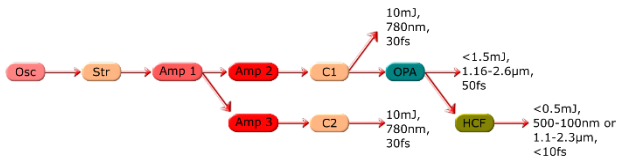

Figure 3 | Schematic of upgraded 1kHz Ti:sapphire CPA laser system. *Osc:* oscillator; *Str:* stretcher; *Amp:* amplifier; *C:* compressor; *OPA:* optical parametric amplifier; *HCF:* hollow-core fibre pulse compressor. The output from C1 can either be sent to the OPA or be used own its own for e.g. HHG or frequency-upconversion. The output from C2 can be used directly for HHG, frequency-upconversion and/or sum- and difference-frequency generation with the output from the OPA. Either output from C2 or the OPA can be compressed in the HCF to sub-10 fs duration pulses. Alternative geometries are also possible.

The few-cycle SWIR pulses can be used as a driver for HHG to generate SXR radiation for transient absorption spectroscopy. We will use the third amplifier upgrade to pump our few-cycle SWIR-OPA[8,10] to increase (up to ~1.5 mJ) the energy available to drive HHG and thus increase the SXR flux up to the oxygen K-edge (533 eV), to make it feasible to perform pump-probe experiments in this spectral region[11].

Vibrational Spectroscopies

The Ultra facility provides a wide range of short-pulse vibrational spectroscopy techniques. Key areas that will be addressed by the new system include time-resolved IR spectroscopies, such as 2DIR[12], Kerr-gated time-resolved resonance-Raman spectroscopy[13] and surface sum frequency generation (SSFG) spectroscopy[14].

The new laser will combine strengths of the current Ultra and LIFEtime laboratories, in terms of their broad bandwidth[1] and high repetition rate / stability,[12] to significantly enhance the experiments currently on offer.

As well as the broadband, ultrashort spectroscopies, the relatively narrowband, ~1 ps duration pump laser allows access to techniques requiring energy resolution with time-resolution, such as time-resolved Raman spectroscopy. In particular, the present OPCPA system provides a novel approach to techniques requiring narrow and broadband pulses (SSFG and EVV[15]).

XUV Capabilities

The existing Artemis beamlines will be enhanced as follows: the focal lengths of the toroidal mirrors inside the monochromator will be increased in order to allow higher energy pulses to be used to drive HHG for increased flux for AMO experiments[16]; the length of the imaging beamline will be extended to allow the use of a longer focal length optic for driving HHG, thus increasing the focal volume and HHG flux for use in coherent imaging experiments. The flat-field XUV spectrometer on the imaging beamline will be redesigned to enable the HHG spectrum to be recorded with optimum spectral resolution across the full HHG spectrum, up to and including the water window (~300-500 eV) via translation of the imaging detector across the image plane.

We will also introduce two new XUV beamlines for use with the new 100 kHz OPCPA system. The first will be similar in concept to the existing monochromator beamline, but specifically designed to accommodate HHG from a tighter focus due to the lower pulse energy. We will initially frequency double the SWIR output from the OPCPA to drive the HHG process with ~50-100 μJ energy, 50-100 fs duration pulses to generate high flux XUV radiation with photon energies in the range of 20-50 eV. Alternatively, we can use the SWIR output directly to reach photon energies up to 100 eV but with lower flux. A single harmonic can be selected using a purpose-built grazing incidence XUV monochromator, using a selection of gratings with 10-30% net transmission efficiency. The exit slit will then be imaged into the end station using a grazing incidence toroidal mirror to produce <20 μm size XUV spot (e^{-2} radius). A future upgrade to the imaging optics will enable demagnification of the source for even smaller spot sizes. A carefully designed telescope for the pump pulse will be used to ensure optimal spatial overlap with the XUV. A non-uniformly ruled cylindrical grating can be inserted into the beamline after the monochromator, but before the toroidal mirror, to be used as a flat-field XUV spectrometer to directly measure the HHG spectrum. Using the zeroth order diffraction of the monochromator grating, or a gold mirror, the full HHG spectrum can also be measured for rapid optimisation of the XUV source. The second beamline will be designed for transient absorption spectroscopy and consists of a differentially pumped high pressure gas jet/cell followed by grazing incidence toroidal mirror and newly designed flat-field XUV spectrometer.

Conclusions

The relocation of Artemis to the Research Complex at Harwell will enable the facility to capitalise on overlapping experimental areas and technologies with Ultra and provide a stronger platform for cross-discipline collaboration. The separation of the laser and plant room from the experimental areas, and the introduction of additional beamlines with more specific end stations, will provide a more efficient user experience whilst the laser upgrades will significantly enhance the experimental capabilities of both the Ultra and Artemis facilities. We are actively engaging with the user community through our user meetings, conferences and scientific collaborations and welcome suggestions from existing and potential new users to aid in shaping the future direction.

Acknowledgements

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References

1. Greetham, G. M. *et al.* Ultra: A Unique Instrument for Time-Resolved Spectroscopy. *Appl. Spectrosc.* **64,** 1311–1319 (2010).
2. Turcu, I. C. E. *et al.* Ultrafast science and development at the Artemis facility. in (ed. Vlad, V. I.) **7469,** 746902 (International Society for Optics and Photonics, 2009).
3. Frassetto, F. *et al.* Single-grating monochromator for extreme-ultraviolet ultrashort pulses. *Opt. Express* **19,** 19169 (2011).
4. Grønborg, S. S. *et al.* Observation of Ultrafast Free Carrier Dynamics in Single Layer MoS 2 . *Nano Lett.* **15,** 5883–5887 (2015).
5. Springate, E., Towrie, M., Greetham, G. & Wyatt, A. *ULTRA/ARTEMIS-100 kHz High Power OPCPA Progress Report*.
6. Maksimenka, R. *et al.* Highly stable, 15 W, few-cycle, 65 mrad CEP-noise mid-IR OPCPA for statistical physics. *Opt. Express* **26,** 26907 (2018).
7. Gierz, I. *et al.* Tracking Primary Thermalization Events in Graphene with Photoemission at Extreme Time Scales. *Phys. Rev. Lett.* **115,** 086803 (2015).
8. Wyatt, A. S. *et al. Short-Wave Infrared Few Cycle Pulse Generation and Characterization for High Harmonic Generation in the Water Window*.
9. Baksh, P. *et al.* High Resolution, Wide Field of View, Ptychographic Imaging of a Biological Sample using a High Harmonic Generation Source. in *Conference on Lasers and Electro-Optics* SF1I.4 (OSA, 2016). doi:10.1364/CLEO_SI.2016.SF1I.4
10. Wyatt, A. S. *et al.* Compression and amplification of SWIR single-cycle pulses for water window attosecond pulse generation. in *Optics InfoBase Conference Papers* CG_3_3 (Optical Society of America, 2014).
11. Johnson, A. S. *et al.* Apparatus for soft X-ray table-top high harmonic generation. *Rev. Sci. Instrum.* **89,** 083110 (2018).
12. Donaldson, P. M., Greetham, G. M., Shaw, D. J., Parker, A. W. & Towrie, M. A 100 kHz Pulse Shaping 2D-IR Spectrometer Based on Dual Yb:KGW Amplifiers. *J. Phys. Chem. A* **122,** 780–787 (2018).
13. Matousek, P. *et al.* Fluorescence suppression in resonance Raman spectroscopy using a high-performance picosecond Kerr gate. *J. Raman Spectrosc.* **32,** 983–988 (2001).
14. Neri, G., Walsh, J. J., Teobaldi, G., Donaldson, P. M. & Cowan, A. J. Detection of catalytic intermediates at an electrode surface during carbon dioxide reduction by an earth-abundant catalyst. *Nat. Catal.* **1,** 952–959 (2018).
15. Fournier, F. *et al.* Protein identification and quantification by two-dimensional infrared spectroscopy: Implications for an all-optical proteomic platform. *Proc. Natl. Acad. Sci.* **105,** 15352–15357 (2008).
16. Bellshaw, D. *et al.* Ab-initio surface hopping and multiphoton ionisation study of the photodissociation dynamics of CS2. *Chem. Phys. Lett.* **683,** 383–388 (2017).