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ESTABLISHING A RELATIVISTIC ULTRAFAST ELECTRON DIFFRACTION & IMAGING (RUEDI) UK NATIONAL FACILITY*

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Abstract

RUIDI (Relativistic Ultrafast Electron Diffraction & Imaging) is a proposed facility which will deliver single-shot, time-resolved, imaging with MeV electrons, and ultrafast electron diffraction down to 10 fs timescales. RUEDI is being designed to enable the following science themes: dynamics of chemical change; materials in extreme conditions; quantum materials; energy generation, storage, and conversion; and in vivo biosciences. RUEDI is proposed to be built at STFC's Daresbury Laboratory in UK. The Conceptual Design Review and Outline Instrument Design reports were published in November 2022 and summarized in this paper, with a Technical Design Review report to follow in November 2023.

INTRODUCTION

Ultrafast electron diffraction (UED) at MeV beam energies has been developed at many facilities around the world. UCLA in California, USA was the first to demonstrate pump probe experiments where a laser pulse was used to initiate chemical change, with a MeV electron beam used to probe the crystal structure through electron diffraction with a varying time delay [1]. Many other instruments have since been developed including at SLAC [2] and BNL in the USA [3], DESY in Germany [4], Shanghai, China [5], and Daejeon, Korea [6].

Ultrafast electron microscopy (UEM) at the MeV-scale would improve the temporal resolution limits of current keV Transmission Electron Microscopy (TEM) instruments, at a compromise with reduction in spatial resolution, and also allow thicker, more realistic samples to be imaged. Demonstrators have been carried out, for example at Osaka University [7] and UCLA [8], and design studies ongoing at various laboratories worldwide [9-12], although none yet operational as a user facility.

The RUEDI (Relativistic Ultrafast Electron Diffraction & Imaging) project represents a desire to establish a relativistic ultra-fast electron diffraction and imaging capability in the UK, as a mid-range facility funded by EPSRC as part of the UKRI infrastructure fund. Currently a two-year design study has been funded, of which this paper provides a brief overview of the current status, and funding for the full construction of the project has been applied for, with the outcome to be known late in 2023.

The facility is planned to be located at the STFC Daresbury Laboratory in the UK, where it will be co-located with facilities such as CLARA [13] and SuperSTEM [14]. Preliminary electron diffraction studies using the VELA accelerator at Daresbury were carried out in 2014 to 2015, although for that work only static measurements were made due to the absence of a pump laser source [15].

Five science themes have been identified for RUEDI, and have held user consultation meetings in 2022 [16]: dynamics of chemical change; materials in extreme conditions; quantum materials; energy generation, storage, and conversion; and in vivo biosciences.

MACHINE OVERVIEW

RUEDI will use relativistic electrons to probe the structural dynamics of matter. The operating modes of RUEDI can be split into diffraction and imaging categories and for each category we define two modes of operation. Single-shot requires a large number of electrons in a single pulse to directly capture the full image to study irreversible processes. In stroboscopic mode, the images can be built up over many, potentially thousands, of shots. This mode can only be used to fully study reversible processes or those in which the samples is being continually renewed (for example a microfluidic cell). In stroboscopic mode, fewer electrons are needed for each shot, and thus the spatial-temporal resolution can be improved due to lower space-charge effects in the bunch. All modes will be capable of operating in a pump-probe regime with the pump normally being a laser pulse (although other sources may be available) and the electron beam acting as the probe. A wide range of pump laser wavelengths/durations/intensities will be provided. RUEDI will initially be limited to a repetition rate of 100 Hz, however, technology will be specified in order to operate at repetition rates of up to 1 kHz in the future. Table 1 lists the desired electron beam parameters at the sample for the different proposed operational modes.

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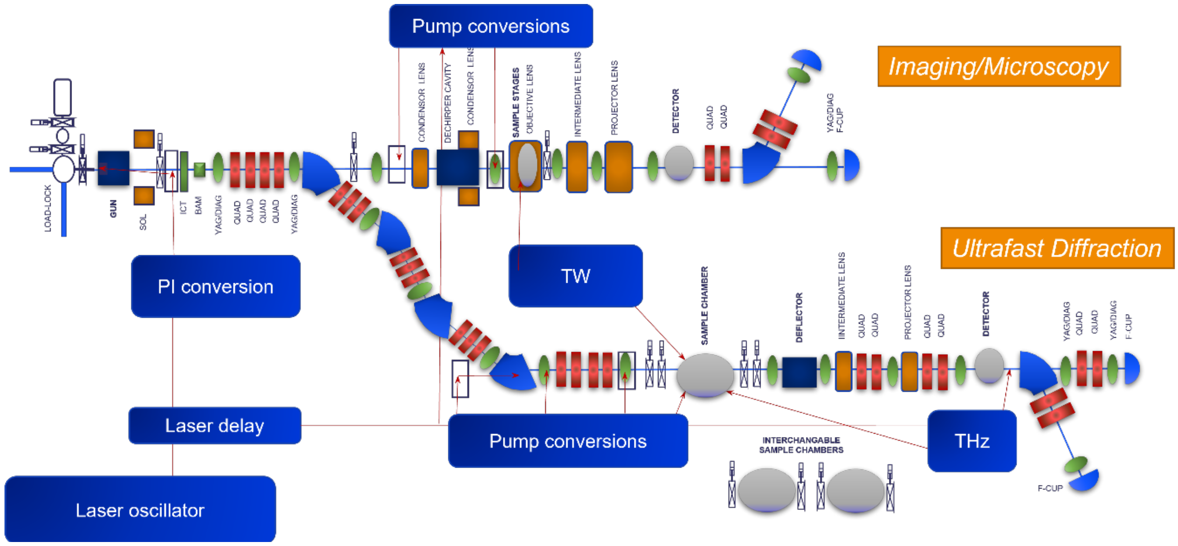

Figure 1: RUEDI schematic layout. RF cavities are shown in dark blue, dipole magnets in blue, quadrupoles in red, solenoids in orange, and diagnostic stations in green. The sample stations and detector are shown in grey. The components of the laser system are shown in blue boxes which roughly correspond to the location of laser tables.

Table 1: Operating Modes and Desired Design Parameters of RUEDI

Mode	**Kinetic Energy**	**Electrons per bunch**	**Temporal resolution**	**Spatial resolution**	**Momentum resolution**	**Spot size**
Imaging *(single-shot)*	2 MeV	$10^6 – 10^8$	1 – 10 ps	1 – 100 nm	-	10 µm
Imaging *(stroboscopic)*	2 MeV	10^4 - 10^6	< 800 fs	< 1 nm	-	10 µm
Diffraction *(single-shot)*	4 MeV	10^6	50 – 20 fs	-	0.1 Å^{-1}	200 - 400 µm
Diffraction *(stroboscopic)*	4 MeV	$10^4 – 10^5$	50 - 10 fs	-	0.1 Å^{-1}	50 - 100 µm
Diffraction *(streaking)*	4 MeV	10^7	10 fs	-	0.1 Å^{-1}	100 µm

Imaging will be offered at ps-scale time resolution, but not the ultrafast fs-scale of the diffraction operating modes. Diffraction will be offered at as fast a temporal resolution as can be managed, aiming for 10 fs. The limits on this resolution are a convolution of the electron bunch length, laser pulse length, and the relative jitter between the two. A further streaking diffraction mode will be offered. This mode is single-shot, time-resolved, where the time information is implanted onto a transverse plane via a deflector/streaker [17]. The time resolution of this mode is limited by that of the streaker and will be ~10 fs. In this mode, the full diffraction image cannot be observed, making it most suitable for polycrystalline materials, or gas or liquid jets.

The overall schematic layout of the proposed RUEDI facility is shown in Fig. 1. A normal-conducting S-band (2998.5 MHz) RF photocathode gun is used to generate the electrons and accelerate them to 4 MeV. Initial design studies of this gun are presented in [18]. This electron gun feeds the two beamlines for imaging, and ultrafast diffraction. A single laser oscillator and amplifier drives the facility, which is then split into the feed for photoinjector gun and also used for pump conversions for the two beamlines. Having all the laser beamlines driven by the same oscillator means they are inherently synchronised – important for the desired 10 fs time resolution of the instrument. Scanning the delay time between the pump laser and probe photoinjector allows time-resolved experiments to be carried out. Further laser conversion to TW pump, and THz diagnostics is shown.

IMAGING BEAMLINE

The first quadrupoles and dipole are switched off and the beam transported from the gun to the sample via solenoids only, to provide axial symmetry. The series of solenoid magnets then act as the traditional lenses in a TEM. Two condenser lenses are used to focus the electron beam onto the sample, which is inserted into the objective lens. This lens and the following intermediate and projector lenses are used to magnify the image of the sample onto the detector. Magnification factors up to 6500 will be made available. This will allow a 1 nm feature to be magnified to the 6.5 µm pixel size of the detector, which is planned to be a 16-64 megapixel direct electron detector with single electron sensitivity. Pump laser conversions to a wide variety of wavelengths, are located as close to the sample as possible, with multiple laser insertion points to enable co-linear propagation with the electron beam or oblique incidence illumination if preferred. A keV-scale ion source is also proposed to feed into the sample location to allow for ion implantation and tracking [19].

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Using solenoidal lenses (instead of quadrupoles for example), limits the beam energy capable of being focused by the objective lens to ~2 MeV, due to magnetic field saturation limits. To minimise the deleterious effects of space-charge on the electron beam quality, the gun is operated at 4 MeV, as in the diffraction beam modes, and transported to the sample at high energy. A second RF cavity, is located just before the sample location is then used to decelerate the beam from the 4 MeV to 2 MeV to minimise the amount of low energy beam transport. This is a 3-cell normal-conducting standing wave S-band cavity. This cavity is also used to remove the energy-time chirp the electron bunch receives from the gun, which is vital to lower the energy spread sufficiently so that the chromatic aberration of the microscope lenses do not degrade the image resolution. Beam dynamics studies of this beamline are described in [20]. A comparison of using solenoids or quadrupoles as the magnetic lenses is given in [21].

ULTRAFAST DIFFRACTION BEAMLINE

The diffraction beamline has been designed to offer as good time resolution as possible. The electron beam produced from the gun is transported through a 4-dipole magnetic arc to the diffraction line. This separates the diffraction beamline from the imaging beamline and keeps them parallel. The magnetic arc is tuned so that it both supresses the time-of-arrival jitter of the electron bunch at the sample location, and temporally compresses the bunch from the gun to fs-scale at the sample. The beam dynamics of this scheme are described in [22]. Quadrupoles are used before and after the arc to provide matching and the final focus onto the sample.

Unlike the imaging beamline, the sample does not have to be located inside a magnetic lens. Thus, larger sample chambers can be used to house the sample. This allows more flexibility of sample environments, stages, and both electron beam and laser beam diagnostics, as well as laser injection and extraction. To maximise the number and type of experiments which can be carried out, it is proposed for the sample chamber to be interchangeable. This allows for maximum flexibility, whilst only having one interaction point between pump lasers and probe electron beam to optimise beam transport for both.

Initially, three types of sample chambers are planned: a cryogenic sample chamber, with capabilities to cool down to 5 mK, a sample chamber which allows gas and liquid jets to be used, and a chamber which only takes solid sample stages, but has the best achievable sample control and diagnostics. The sample chambers can all be kept under vacuum and wheeled in and out of the beamline position when not in use. Vacuum valves on the beamline both upstream and downstream of the sample chamber location will enable the sample chamber to be changed without venting the machine vacuum.

After the sample chambers, the beamline is designed for two purposes – diagnostics, and transport of the diffraction pattern to a detector. Solenoidal magnetic lenses are used to magnify the diffraction pattern onto a detector, and quadrupoles used to correct any asymmetry that is brought about by the magnetic arc and use of quadrupoles in the final focus system.

An S-band RF transverse deflecting cavity (TDC) is located just after the sample chamber. This can be used for two purposes: electron bunch length measurement, and the single-shot streaking mode. An electron spectrometer is located at the end of the beamline after the TDC to measure the longitudinal phase-space of the electron bunch.

Since the TDC is not located at the sample position, it cannot be used to directly measure the bunch length seen by the sample. To measure this, THz streaking can be used at the sample location. The THz will be generated from the same laser as all the other systems for accurate synchronisation. As the THz will thus be synchronised to the pump laser, it can also be used to measure the time-of-arrival of the electron bunch. A second THz interaction point is located after the detector. Since not all experiments require the zero-order spot to be measured, a detector with a hole in the centre can be used to pass the undiffracted beam through to the diagnostics. Thus the THz (and electron spectrometer) can be used for online shot-by-shot diagnostics of the electron beam.

LASER SYSTEMS

The laser system is responsible for generating the electrons and delivering a comprehensive suite of pump-probe pulses for scientific applications and achieving the best temporal resolution for studying ultrafast processes in matter. The common Ti:Sa laser source is split to feed the photoinjector and both beamlines. A wide range of pump sources is created by: second, third, and fourth harmonic generation from the 800 nm fundamental; optical parametric amplification (OPA) and conversion for almost continuous coverage of far-IR to UV wavelengths; few-cycle THz pulses, which will be produced by optical rectification in nonlinear crystals from Ti:Sa and OPA mid-IR drivers; hollow-core fibre post-compression for delivering ultrashort pulses in the mid-IR, visible and DUV/VUV wavelengths.

In addition to providing a comprehensive set of pump sources, two additional unique capabilities are currently planned. The first involves combining the UED beamline with an XUV beamline. This would enable experiments that use the electron beam as a broadband excitation pump, which is then probed by transient XUV absorption spectroscopy. The second unique capability is the provision of a TW class laser delivering high-intensity laser pulses capable of reaching relativistic intensities of ~10^{19} W/cm^2 and high-energy nanosecond pulses. This will allow the facility to provide a new platform for studying materials in high-energy density conditions in both real and reciprocal space.

To achieve the required pump-probe timing resolution, laser and RF systems, will be synchronised to an ultra-stable master oscillator clock. Synchronisation systems based on RF and all optical architecture have been considered. All lasers, and the THz diagnostics are derived from a common seed to reduce relative jitter. Low charge cavity beam arrival monitors are under consideration.

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