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4GLS—the UK's fourth generation light source at Daresbury: new prospects in biological surface science

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Abstract

4GLS is a suite of accelerator-based light sources planned to provide state-of-the-art radiation in the low energy photon regime. Superconducting energy recovery linac (ERL) technology will be utilized in combination with a variety of free electron lasers (IR to XUV), undulators and bending magnets. The 4GLS undulators will be optimized to generate spontaneous high flux, high brightness radiation, of variable polarization, from 3 to 100 eV. The ERL technology of 4GLS will allow shorter bunches and higher peak photon fluxes than possible on storage ring sources. It also provides pulse structure flexibility and an effectively infinite beam lifetime. The XUV and VUV free electron lasers will be used to generate short (femtosecond regime) pulses of extreme ultraviolet light that is broadly tunable and more than a million times more intense than the equivalent spontaneous undulator radiation. A strong feature of the scientific programme planned for 4GLS is dynamics experiments in a wide range of fields. Pump–probe experiments will allow the study of chemical reactions and short-lived intermediates on the timescale of bond breaking and bond making, even from very dilute species. The high intensity of the FEL radiation will allow very high resolution in imaging applications, using near-field approaches. For example, in the IR regime, resolution of the order of 30–50 nm should become possible, allowing the sub-cellular structure of live cells to be examined. The combination of high brilliance with short pulse lengths from multiple sources will allow development of techniques that probe the nature of biomolecule interactions with surfaces. These include methods for probing conformational changes on binding such as time-resolved sum-frequency spectroscopy and reflection anisotropy spectroscopy.

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1. The proposed 4GLS facility

1.1. Introduction

In current 'third generation' synchrotron sources radiation is extracted from insertion devices ('undulators' and 'multipole wigglers'), and from the bending magnets that ensure the circulation of electrons in storage rings. The electrons circulating in such rings typically orbit the ring around 10^{11} times, and the resultant equilibration of the electron beam with its surroundings results in fundamental limitations in source brightness and pulse length. In recent years, the energy recovery linac (ERL) concept [1–3] has been applied for the first time to the production of SR in two projects at the Jefferson Laboratory in Virginia. In an ERL device, a high density electron beam is produced by an RF superconducting linac array. This circulates the ring once (or only a few times), with its phase arranged so that when it returns to the linac structure it is decelerated, returning its energy to the RF field. The Jefferson team has already demonstrated recovery of in excess of 99.98% of the beam energy [4]. In parallel with these developments has been a related programme to demonstrate the potential of free electron lasers (FELs). Such sources extend the interaction between the electrons and the undulators into a regime where huge brightness increases can be obtained. So far a number of infrared user facilities, based on low energy electron linacs, have been established (e.g. FELIX [5] in the Netherlands, the Jefferson Laboratory [4], a number of university facilities in the USA, and CLIO in France) and there have also been a limited number of successful attempts to transfer the technology onto storage rings at higher electron energy and thus produce radiation into the VUV (Super-ACO (France), ELETTRA (Italy) [6], Duke University (USA), and NIJI-IV (Japan)). These FELs, like their laser counterparts, are dependent on mirrors to trap the emitted radiation in optical cavities. The SASE (self amplification of spontaneous emission) FEL, first developed at the University of California, Los Angeles, and also implemented at the DESY laboratory, is free of the constraint of mirrors and therefore has the potential for further extending output wavelengths from the XUV into the x-ray region [7, 8]. The properties of high intensity, extended wavelength range, and 'laser-like' beams produced by FELs make them the sources of choice when conventional lasers or SR fail to meet the peak flux or brightness criteria for an experiment.

1.2. The 4GLS concept

The 4GLS proposal is to site an IR-FEL, cavity VUV-FEL, and single-pass XUV-FEL at one experimental facility, which will also provide for undulator and bending magnet radiation (figure 1). At the heart of the facility is a photocathode injected energy recovery linac (ERL) and a single cycle electron returning ring, which allows the energy from the returning electron beam to be recovered in the ERL. The arrangement of ERL, bending magnets, undulators and FELs is designed to provide both single and bespoke combinations of sources at a multi-user facility. The machine configuration gives the advantage of a constant beam current and may be operated in either in high current (effectively CW) mode or in a 'temporal' mode offering a tailored train of sub-picosecond pulses. The frequency coverage provided by the 4GLS FELs (IR-THz, VUV and XUV) is designed to complement available table-top laser sources; it is envisaged that 4GLS may be powerfully combined with these sources in many experiments.

1.3. 4GLS photon sources

The 4GLS undulators and bending magnet sources. The 4GLS undulators will be optimized to generate spontaneous high flux, high brightness radiation, of variable

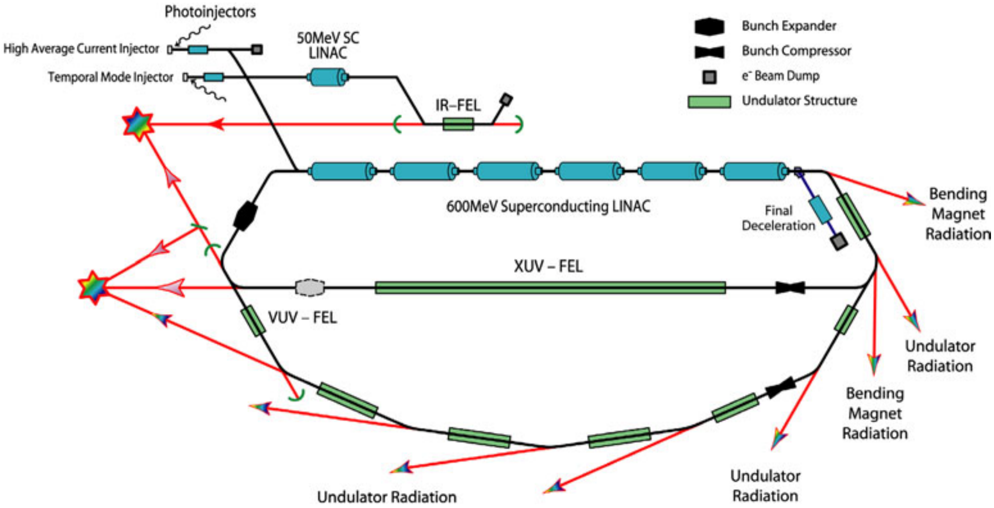

Figure 1. A schematic diagram of the 4GLS concept.
(This figure is in colour only in the electronic version)

polarization, over the photon energy range 3–100 eV. However, they will also generate usable radiation (in the higher harmonics) at energies up to and above 500 eV. Typical undulators will generate fluxes of 10^{16} photons/(s 0.1% bp 100 mA) and brightnesses up to 10^{19}photons/(s 0.1% bp 100 mA mm^2 mrad^2). Bending magnets on 4GLS will provide photons from the terahertz regime to over 1 keV. Due to the small physical size of the ring, it will be straightforward to obtain large vertical and horizontal apertures (tens of milliradians). This will be of particular advantage to users who need high intensity continuous flux. The bending magnet sources will also produce coherent radiation in the IR and terahertz regions, and because the electron bunch lengths on 4GLS will be of the order of the wavelength of the emitted light for energies below 0.02 eV (160 cm^{-1}), considerable flux enhancement due to multi-particle coherent emission will occur. This enhancement has recently been demonstrated at Jefferson Laboratory [9]. In the case of 4GLS, brightness of up to 10^{21} photons/(s 0.1% bp 100 mA mm^2 mrad^2) in the far-IR is anticipated.

The IR-FEL. The IR-FEL will generate sub-picosecond laser pulses of variable polarization in the range 3–75 μm (0.4–0.017 eV). Its main characteristics will depend on the detailed design, but pre-design goals are for average fluxes of around 10^{23} photons s^{-1} and peak fluxes of about 10^{28} photons s^{-1} at a repetition rate of around 10 MHz (with a micropulse energy of about 75 μJ). The use of photoinjectors together with superconducting linac technology means that the 4GLS IR-FEL will deliver short pulses (100 fs to a few ps), with good energy stability and high average power. The 4GLS IR-FEL will be based on a multi-pass optical cavity and its output will be diffraction and transform limited over its operating range. It will have the facility to operate either independently or in conjunction with the other 4GLS sources shown in figure 1.

The VUV-FEL. The proposed VUV-FEL is based on an optical klystron arrangement. Such systems have been successfully implemented in several storage ring facilities. The success of existing storage ring FELs has depended heavily on the development of very high reflectivity multilayer mirrors, with reflectivities of 95% or better [10], and this has limited the short wavelength performance of current devices to around 190 nm. However, it is expected that, by the use of pure Al mirrors, the limit will be extended to 120 nm in the near future [11]. Designs based on high gain harmonic generation (HGHG), which would eliminate the need for high reflectivity mirrors, are also under consideration for this FEL. The pre-design goals for the VUV-FEL are for an average photon flux output of around 10^{20} photons s^{-1}, a peak flux of about 10^{25} photons s^{-1} (10^{13} photons/pulse), and repetition rate of 6.25 MHz (with a pulse energy of about 15 μJ). The FEL will be designed to produce sub-picosecond VUV pulses that will be broadly tunable in the range from the visible to 10 eV. The output of the VUV-FEL will be 5–6 orders of magnitude more intense than the spontaneous VUV radiation from undulators on third generation sources.

The XUV-FEL. Without the constraints of a cavity and its associated mirrors, the XUV-FEL on 4GLS is designed to generate photons well into the extreme ultraviolet and even the soft x-ray region. Intense, short duration electron bunches fed into the long undulator lead to the SASE effect giving highly intense and coherent radiation. SASE FELs require long, high precision undulators for their operation. Again, the exact characteristics will depend on detailed design, but goals for average photon fluxes in the region of 10^{18} photons s^{-1}, and peak fluxes around 10^{26} photons s^{-1} (10^{14} photons/pulse), at 100 eV are envisaged (with a pulse energy of about 2 mJ/pulse). The output will be composed of macrobunches with a repetition rate around 60 Hz. Each macrobunch will contain 650 microbunches with a repetition rate of about 65 MHz within the macrobunch. Seeding schemes to improve the temporal coherence of this source are currently under development.

2. Applications of 4GLS in biological surface science

The 4GLS facility will uniquely combine undulator and bending magnet sources with free electron lasers of high power output and flexible timing structure over a broad wavelength range. This will enable experiments that have been hitherto impossible to undertake, because of the wavelength performance, and/or the time, or spatial resolution required. In particular, the source will be especially powerful in ‘pump–probe’ experiments. Applications where 4GLS is likely to have an impact in biological surface science include:

- macromolecular conformations at surfaces, and conformational dynamics;
- vibrational relaxation, energy transfer and redistribution at surfaces;
- intercellular signalling and studies of receptor systems on membrane rafts;
- studies of material–biological matrix interfaces and surface nanostructuring;
- functional imaging.

A small number of examples is discussed below.

2.1. Macromolecular reaction dynamics at surfaces

Biological processes occur over a wide range of timescales, from sub-picoseconds to hundreds of seconds and longer. Even those processes that are described as being slow, such as apoptosis and mitosis, can be broken down into component fast enzymic and diffusional events. This means that there is a requirement for fast measurements in order to study these processes fully. The fast time structure and extremely high power of FELs make them well suited to the investigation of processes on timescales from sub-picoseconds to milliseconds. One example where 4GLS offers substantial advantage is the study of intramolecular vibrational energy redistribution (IVR) in surface–adsorbate complexes. An understanding of this IVR is essential to improving our understanding of the adsorption, reaction and desorption of species at any catalyst or biointerface. The essence of the experiment is to use the IR-FEL radiation, tuned to a specific vibrational mode of an adsorbed molecule, to pump that mode and to use synchronized IR bending magnet radiation to probe the mid-IR and terahertz regimes at time intervals in the picosecond regime. The crucial surface–adsorbate bonds typically have very low frequencies in the far-IR, so the coverage of 4GLS IR-FEL and bending magnet radiation in the terahertz regime make it a uniquely powerful source for this experiment. The necessary temporal and spatial coherence in the RAIRS (reflection–absorption infrared spectroscopy) geometry is aided by the very small etendue of the 4GLS beams. Some demonstrator experiments of this type in the less challenging mid-IR have been carried out for molecules in solution. An example is a picosecond study of the population lifetime of the amide I vibration of myoglobin, using a pulse train from the Stanford IR FEL as both pump and probe [12].

2.2. Kinetics of DNA–DNA and DNA–protein interactions using reflection anisotropy spectroscopy (RAS)

RAS is an optical probe with very high surface sensitivity that is applicable to the study of surfaces in a wide range of environments. It was developed by Aspnes in the 1980s as a probe of semiconductor surfaces [13] and as a monitor of semiconductor growth [14, 15]. It relies on the difference in reflectance between orthogonal polarization states of light falling on the sample surface (and thus an isotropic bulk substance gives no contribution to the RAS signal). RAS has considerable potential for exploitation in a number of fields of research and has recently found application in the study of electrochemical interfaces [16] and in the determination of the geometry of adsorbed molecules on metal surfaces [17]. Its extension to biological interfaces (for example the study of interactions between DNA sequences at metal–liquid interfaces) is currently limited by the lack of a tunable source of intense linearly polarized radiation in the deep UV, as much of the identifiable spectral signal is expected to be found in this part of the spectrum. The 4GLS VUV-FEL will allow this range to be accessed, and, in addition, it should be possible to improve the speed of response of the technique from the millisecond to the nanosecond regime, allowing a much wider range of dynamic processes to be studied.

2.3. New horizons for sum frequency spectroscopy (SFS)

In recent years, vibrational sum frequency spectroscopy, SFS (or sum frequency generation, SFG) has begun to make a considerable impact in the analysis of conformation in large admolecules. It is specifically sensitive to interfaces, with the capability of probing buried interfaces involving gas, liquid or solid. The technique relies on the measurement of the (normally weak) second order nonlinear 'sum frequency' beam generated when two intense photon beams are coincident at a surface. The theory of these processes has been well developed in recent years [18–20]; a 'sum frequency' response is expected from an interface because of the lack of inversion symmetry at this boundary, whereas the bulk on both sides of the interface may be infrared transparent (and in fact, it is clearly important that the light incident on the interface is not strongly absorbed by the medium on at least one side of the interface). When one of the incident pulses is resonant with a vibrational or electronic excitation of the interfacial molecules or the substrate, an enhancement of the sum frequency response is observed (in some ways analogous to the surface-enhanced Raman effect). Typically, the technique uses IR and UV/visible photons from table-top laser sources (with the latter providing the 'resonant' excitation). As a result, the full potential of this technique has not been realized to date. Its application is severely limited by the lack of highly intense, tunable deep-UV sources, giving high sensitivity from only a few surfaces (e.g. Au) where the SFG response may be resonantly enhanced by available table-top UV/visible lasers. In addition, inadequate coverage in the IR, particularly to low wavenumber, can limit the types of vibration that can be probed. Due to the lack of suitable and sufficiently tunable IR laser sources, currently performed laboratory based SFG experiments are restricted to the measurement of vibrational sub-spectra in the C–H and O–H stretching regions [21]. In contrast, the wide spectral range available at the 4GLS IR-FEL would allow for the first time the recording of complete SFG spectra permitting a full vibrational characterization of active surface species. The range and tunability of the 4GLS VUV-FEL offers the opportunity to further enhance the sensitivity of SFG by utilizing resonance features at these higher photon energies, enormously widening the number of systems that can be studied.

4GLS will thus enable the full exploitation of SFG, in experiments from a range of surfaces (including insulators) never previously amenable to study in this way. The technique is potentially especially powerful in studies of biological interfaces, such as functionalized self-assembled monolayers, peptides and proteins, at the solid–fluid interface. Such measurements in the mid-IR region require access to FEL light sources because of the need for tunability of the light source and also because of intense adsorption from atmospheric water. The coverage of 4GLS into the terahertz regime in addition allows external vibrational modes of adsorbate/substrate complexes to be probed. The short pulses from the FEL sources will allow experiments to be conducted on picosecond and potentially sub-picosecond timescales, while the high flux lends itself to imaging applications using a scanning near-field optical microscopy (SNOM) probe. Important proof-of-principle experiments have been conducted which demonstrate the feasibility of this approach [22], although conventional laser sources yield very poor signal; clearly this is an area where 4GLS offers substantial advantages. Further opportunities in imaging are described briefly below.

2.4. Imaging of biological surfaces and interfaces

Biological imaging has benefited enormously from the rapid advances in laser technology over the past 30 years, and the special characteristics of the proposed 4GLS FELs would be expected to enable further developments and, potentially, entirely new imaging techniques. The power available from conventional lasers at visible wavelengths has resulted in techniques that break the ‘resolution limit’, including stimulated emission microscopy [23], 4pi-theta confocal microscopy [24], and scanning near-field microscopy [25]. These methods provide important information, but rely on incorporating fluorescence probes into the specimen. The future is expected to lie in the area of probeless functional imaging, where structures are imaged according to their chemical or structural properties, without the requirement for the introduction of probes that might perturb the system. The extended wavelengths of the 4GLS FELs would provide an excellent platform for the development of novel imaging techniques. For example, infrared microspectroscopy can probe chemical composition and protein secondary structure, but the resolution is limited because of the long wavelengths used. Near-field techniques are used to achieve higher spatial resolution, but require extremely high power levels. The high intensity of the infrared FEL would be well suited for scanning near-field infrared microscopy. Resolution of the order of 30–50 nm should become possible, in principle allowing the sub-cellular structure of live cells to be examined.

Circular dichroism and resonance Raman microscopies both have potential for functional imaging. They require bright radiation between 140 and 220 nm, a wavelength region that is not covered well by conventional lasers. Both methods will yield structural detail on proteins, carbohydrates, and nucleic acids, and time-resolved resonance Raman microscopy will allow the chemistry of sub-cellular domains to be investigated in real time. Sub-micron CD microscopy has already been demonstrated [26] using a variable polarization undulator, but the huge advantage in flux and the selectable polarization from the 4GLS VUV-FEL will allow greatly improved spatial resolution.

Terahertz imaging is a nascent but rapidly developing field. It requires wavelengths in the range from 10 μm to 3 mm (0.1–30 THz) which is differentially absorbed by tissue of different densities. The technique therefore has great potential in the field of medical imaging. The fast-pulsed table-top lasers [27] which are currently used do not provide the fluxes required to fully exploit the technique. The 4GLS IR-FEL, however would provide an excellent source for terahertz imaging.

3. Current status of the 4GLS project

The project is currently subject to assessment under the Office of Government Commerce 'Gateway' Process. The science case for 4GLS [21] ('Gateway 0') was approved in April 2002. The business case ('Gateway 1') was approved in November 2002. Funding for the first three years of the 4GLS project was announced by the UK Government in April 2003. This includes the research and development work necessary to produce a design study report, with the construction of an ERL-prototype. It is anticipated that, subject to successful passage through the remaining gateways, the full facility will be available to EU users in 2010.

4. Conclusions

The suite of sources that is encompassed by the 4GLS facility is unprecedented in its wavelength range, flux, brightness, and flexibility of pulse structure. In addition to the major advances that will be enabled by the individual components, the synergy between the separate sources will greatly enhance their value. In particular, the ability to operate at multiple wavelengths in pump–probe configurations with a high degree of pulse–pulse synchronization will allow researchers to obtain data at sub-picosecond time resolution and with higher information content than previously possible. There are also new opportunities for developing time-resolved probeless functional imaging to high resolution.

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