Report of 4GLS & DIAMOND Working Party

Remit: To consider the relative roles of 4GLS and DIAMOND in the provision of low energy radiation for the UK community

Membership:

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Background:

DIAMOND is a 3 GeV electron storage ring for synchrotron radiation to be built at the Rutherford Appleton Laboratory. 4GLS is a collection of sources proposed to be constructed at Daresbury Laboratory. The main component of this is now a 600 MeV energy recovery linac (ERL) which will incorporate conventional undulators and bending magnets together with both cavity-based and SASE (self-amplified spontaneous emission) FELs (free-electron lasers) for operation in the ultra-violet and beyond. In addition, the facility will house an infra-red (IR) FEL which can be used alone or in conjunction with the ERL source(s). The panel met on three occasions, 12 June, 27 June, 17 July, to consider the way in which these two facilities might complement each other. Not all members were present at all meetings, but the final report was circulated to all members for comment. Additional information was provided by Daresbury Laboratory staff. A report entitled *"Low Energy Provision on Diamond"* (http://www.diamond.ac.uk/Publications/32/DM-WG-TN-001.doc), which examined the demand and the possibility of providing IR and VUV (for circular dichroism experiments) specifically on DIAMOND, was presented earlier this year by the DIAMOND scientific directors. The present report concentrates on the core question of the complementarity of DIAMOND and 4GLS as radiation sources for the UK community, but does touch on some of the fascinating scientific opportunities which are emerging from the latest design of 4GLS.

The regions of the spectrum, scientific applications and source characteristics were divided into four sub-topics:

1. VUV/XUV undulator and bending magnet radiation
2. UV FEL radiation
3. Sources of visible/UV CD experiments
4. IR sources – synchrotron radiation and FEL

The report deals with each of these in turn. For each topic an introduction and brief overview of the conclusions is given below, while fuller descriptions are given in 4 appendices.

1. VUV/XUV undulator and bending magnet radiation

The vacuum ultraviolet (VUV) and extreme ultraviolet to soft X-ray (XUV) photon energy ranges, typically defined as from a few eV to a few hundred eV in photon energy, underpin a substantial amount of current synchrotron radiation science mainly in photoemission, photoionisation and photoabsorption studies of gas-phase atoms and molecules, solid surfaces and the solid state. There are several beamlines devoted to this work on the SRS, and many such beamlines world-wide. The fundamental wavelength emitted by undulator sources on third-generation synchrotron radiation machines is defined principally by the physical period of the magnet structure and the (square of the) energy of the electrons in the storage ring. Thus, an undulator on a 3 GeV storage ring has a fundamental energy which is 25 times larger than that of the same undulator on a 600 MeV storage ring or ERL. Undulators can be tuned to lower energy by increasing the strength of the periodic magnetic field, defined by the tuning parameter K, but at very large values of K the device becomes a wiggler, emitting a very high background of higher-energy synchrotron radiation. Operating undulators at very high K-values to achieve lower energy fundamental radiation is therefore undesirable, being inefficient and creating unacceptably heavy heat loadings of the first optical element. The useful tuning range of individual undulators is thus limited. In our evaluation we have assumed the maximum value of K for operation would be 5, as described in Appendix 1.

Based on this limitation, fig. 1 in Appendix 1 shows the locus of the first harmonic output flux of undulators optimally designed for each separate photon energy on the DIAMOND and 4GLS sources; in the case of DIAMOND, curves are shown for both 5 m and 8 m straights, and these are compared with the most favourable 15 m straight on 4GLS. Clearly 4GLS offers excellent performance in the 5-100 eV energy range not accessible, under these criteria, at DIAMOND. The output of the long 4GLS undulator is actually also higher than that of the DIAMOND 8 m undulator in the 100 eV to 200 eV range, but in this range the difference is dominated by the greater length of the straight on 4GLS while not all insertion devices on this machine are likely to be so long. Evidently both sources can deliver good undulator radiation in this intermediate energy range with comparable flux. This graph provides a useful quantification of the complementarity of the two sources at low and high energies. It shows that experiments mainly exploiting 5-100 eV radiation should be located at 4GLS and those aimed mainly at higher energies should be installed at DIAMOND. Of course, one can expect that some low energy beamlines at 4GLS will deliver intense radiation to higher energies of more than 200 eV, and that higher energy beamlines at DIAMOND may offer some (non-optimal) operation down to perhaps 50 eV, to ensure reasonable flexibility for users. Further information on some possible specific undulators is given in Fig. 2 of Appendix 1, while Fig. 3 defines the ultimate useable energy limits of all plausible undulators on the two sources.

Of course, DIAMOND can also provide low energy conventional synchrotron radiation from bending magnets, so it is important to include these sources in the comparison. For this purpose, it is especially important to consider the whole beamline to compare photon fluxes delivered at the sample, because while standard optics will accept essentially all the radiation from an undulator, the usable flux from a bending magnet depends on the horizontal acceptance angle of the optics. The comparisons in Table 1 (Appendix 1) show that even with this allowance these bending magnet beamlines are not competitive, delivering photon fluxes two orders of magnitude less than the undulators, even at relatively low resolution operation.

The provision of low energy undulator radiation on 4GLS will only be matched by the MAX-III facility in Sweden (although the MAX-III source will be a conventional storage ring lacking the extra features of the latest ERL-based 4GLS proposal); all other new synchrotron radiation sources in Europe and the USA operating at too high an electron energy to be optimised in this photon energy range. We should also note that the ERL design of 4GLS offers a far more flexible time structure to the source than any conventional storage ring design, and provides another aspect of complementarity to DIAMOND. While very few synchrotron radiation experiments in the VUV/XUV energy range in the past have exploited the time domain, this situation may change in the future as a result of the more flexible source characteristics and the growing interest in two-colour experiments mentioned elsewhere in this report, especially in the context of the free electron lasers.

2. UV FEL radiation

4GLS offers two types of FEL in the VUV/XUV energy range, a cavity-based FEL with a potential range of 3-10 eV (although the upper end of this range depends on mirror materials as yet unproven for this purpose) and a SASE FEL with a potential energy range of 10-100 eV. Both of these devices offer predicted average and peak fluxes very significantly in excess of conventional undulators, and also have the potential for novel time structures, the ERL providing much shorter pulses than those of a conventional storage ring. Clearly, these have no parallel in DIAMOND, nor is there any realistic potential for DIAMOND to operate in these modes as a 3 GeV storage ring.

One can envisage a variety of novel new science opportunities from such sources, such as pump-probe experiments (coupled, for example, to VUV undulator radiation) in molecules and solids to probe excited states and short-lived species, as well as ultra photon-hungry experiments such as spin-polarised photoemission using time-of-flight detection. Further information on the anticipated performance of these devices, and illustrative examples of new scientific opportunities, are given in Appendix 2.

3. Sources of UV/VUV for CD experiments

One of the two areas of exploitation of synchrotron radiation at low energies which has already been considered for installation on DIAMOND is circular dichroism (CD) experiments in the near UV to VUV (around 3-10 eV). These experiments have gained considerable support in recent years on the SRS and a new dedicated BBSRC-supported SRS beamline is currently being established for this purpose. This facility is designed to operate in the 160-240 nm wavelength (7.7-5.2 eV) appropriate to the biological work based on aqueous solutions for which the absorption of water defines the shortest useful wavelength. The main application is in protein folding and characterisation, with the presence of broad CD bands in this spectral range providing a valuable spectral fingerprint. The CD may only be approximately 10^{-5} of the total absorption, so the experiments are only viable with high-flux stable radiation sources. This feature of synchrotron radiation, together with its broad band character allowing spectra to be measured further into the UV, offers huge advantages over conventional (Xenon arc lamp) light sources. For some users or potential users there is significant interest in extending the spectral range somewhat further into the VUV for chemical studies of large molecules (including those of biological interest); in most systems the short wavelength limit is defined by absorption in the solvent (as in the case of water mentioned above), but the ultimate limitation of the current methodology will be absorption in the photoelastic modulators used for switching the circular polarisation. Current experiments are based on bending magnet radiation, and this same type of source is envisaged for these experiments on DIAMOND. In this spectral energy range the flux of synchrotron radiation delivered by bending magnets is almost independent of the source energy and there is no significant difference between the expected characteristics of DIAMOND and 4GLS. Wavelength scanning (or white beam dispersive) experiments, therefore, could be equally-well accommodated at either source. At DIAMOND one important advantage seen as of great importance by the biological community is the extensive support facilities for experiments in biology and protein handling, associated with the large protein crystallography programme. On 4 GLS there may be some simplification at the technical level in matching the extraction optics to the source.

Of course, the fact that 4GLS offers undulator and FEL operation in the spectral range of interest raises the question of whether these sources could offer extensions of the CD technique beyond those currently envisaged. Such experiments would be complementary additions to, rather than replacements for, the mainstream experiments on bending magnets. There are two key areas in which such developments might be considered: (i) removing the need for photoelastic modulators by arranging for appropriate light polarisation from the source itself and (ii) exploiting the enhanced flux and time structure of output from either an undulator or the cavity-based UV FEL to perform single-wavelength time-resolved CD experiments on much shorter time scales than are currently possible.

The working group did explore some ideas of this type, although they would need to be evaluated in far more detail before being pursued seriously (some further details are discussed in Appendix 2). In particular, the need for the photoelastic modulator (which ultimately limits both time-resolved and short wavelength experiments) might be circumvented by using a double-undulator system delivering both forms of circular polarisation in non-parallel beams which could pass through the sample and be detected simultaneously in two detectors. The ultimate goal of a time-resolved experiment might be based on a single-pulse measurement, but the extremely demanding signal-to-noise requirement of CD experiments does make this very difficult. If the UV FEL can be operated successfully over the full spectral range of interest (see Appendix 2), the estimated delivery of 10^{14} photons in a single pulse would be sufficient to measure CD signals of around 10^{-4} of the total absorption. More demanding CD measurements on the pulse-length time-scale might be achieved if the timed event can be repeated many times. Despite these detailed issues, however, it is clear that in the longer term there is potential to explore new types of CD experiment using the more advanced facilities of 4GLS. In particular, the FEL should certainly open up new opportunities in the fast time regime, including those important for biological processes (microsecond), that will not be accessible by any other source.

4. IR sources – synchrotron radiation and FEL

In considering the infra-red region of the spectrum it is helpful to distinguish between experiments requiring broad-band radiation and those which involve single wavelength scanning. In the former case, the only source is bending magnet radiation, and there is very little difference in the flux emitted from bending magnets on DIAMOND and 4GLS. Current use of this broad-band IR synchrotron radiation falls into two areas: spectroscopy, especially of surfaces, in the far IR for which the SR advantage is greatest, and infrared microspectroscopy (IMS) of a wide range of materials, especially biological systems, for which the need for finely-focussed beams gives SR a large advantage even in the mid-IR spectral range. The DIAMOND working party on “Low Energy Provision on DIAMOND” suggested that future synchrotron IR needs of the UK community could be met by one dedicated far-IR spectroscopy station and three stations for IMS. The IMS community in the UK is growing fast and can be accommodated on DIAMOND given minor modifications in three dipole areas. RAIRS and far IR spectroscopy would require somewhat more significant modification to one dipole vessel. Bending magnets on 4GLS could be optimised for IR work, and might offer some technical advantages for the far-IR spectroscopy, but for the IMS work, in particular, there seems to be no good reason to switch plans for these stations from DIAMOND. In the IR range generally it would appear to be more sensible to concentrate efforts at 4GLS on time-resolved and two-source experiments which exploit the special features of this source described below.

For narrow band studies, the IR FEL component of the 4GLS proposal offers some fascinating possibilities, especially in combination with broad-band IR radiation or UV radiation from the ERL, for two-colour experiments and time-resolved studies. Of course, the IR FEL is more directly in competition with relatively conventional table-top lasers, but the wide range tunability and scanability, particularly in the far IR, favours the FEL. Even more significant, however, are the opportunities offered by operating the IR FEL in tandem with conventional and (UV) FEL sources on the ERL, including the possibility of operating to the two colour sources in a highly synchronised fashion in the time domain. In this regard, the 4GLS facility will provide entirely new opportunities for the UK community and has no counterpart at DIAMOND, or elsewhere. The capabilities for novel non-linear and time-resolved spectroscopic experiments will attract new users from across Physics, Chemistry, Biology and Materials Science as well as many within the current IMS and RAIRS communities using synchrotron radiation. The added value arising from the combinations of UV FEL/IR FEL/Synchrotron IR cannot be overemphasised.

5 Conclusions

i. For mainstream applications of undulator radiation 4GLS and DIAMOND are complementary. 4GLS is substantially superior for photon energies in the 5-100 eV energy range, DIAMOND is superior for all photon energies above 200 eV. An intermediate overlapping range should be provided at both facilities to aid researchers working at the boundaries.

ii. The two UV FEL devices proposed for 4GLS, covering the 3-100 eV photon energy range, will offer unique facilities in terms of both flux and time structure for new and demanding experiments which cannot be matched at DIAMOND or elsewhere.

iii. Current plans for UV/VUV biological CD spectral measurements can be met equally well in terms of flux provision by bending magnet sources on either DIAMOND or 4GLS. DIAMOND probably has significant advantages in terms of infra-structure support for protein and nucleic acid studies, 4GLS may have some slight technical advantages in optical design. In the longer term, however, insertion devices on 4GLS offer the potential for a new generation of CD experiments, especially in the time domain.

iv. The output flux of IR radiation from bending magnets is essentially identical on DIAMOND and 4GLS, and for the mid-IR in particular, and spectro-microscopy experiments, either source could be used. In the far IR, especially relevant to some spectroscopic experiments such as the investigation of surfaces, the need for a very large aperture of acceptance probably favours 4GLS. The IR-FEL of 4GLS provides a unique facility in the UK which, especially in conjunction with broad-band radiation from the ERL devices, may open up wholly new areas of science.

In summary it is clear that 4GLS will provide radiation sources which are entirely complementary to, rather than in competition with, those of DIAMOND, and that the two sources together provide the suite of UK national facilities envisaged by the Woolfson and Blundell Committees.

Appendix 1: Comparison of VUV/XUV undulator and bending magnet radiation from 4GLS and DIAMOND

Peak Flux Output

The peak undulator output curve gives an overview of the optimum capabilities of each machine. For a specific photon energy, the output from the undulator period that gives the maximum flux is plotted; each point on the graph represents a different undulator. The curves are therefore effectively the upper envelope of the output curves of all possible undulators.

The curves are truncated where *K*=5 to indicate where power loading will start to make devices totally unworkable. Undulators can be designed to give output below this energy, but the period chosen would be longer than that for maximum flux in order to reduce the power to realistic values, resulting in lower flux. Figure 1 shows the comparison for first harmonic output from DIAMOND and 4GLS. DIAMOND can optimally produce photon energies above 100 eV (total power 5kW), 4GLS above 5 eV (total power 1kW).

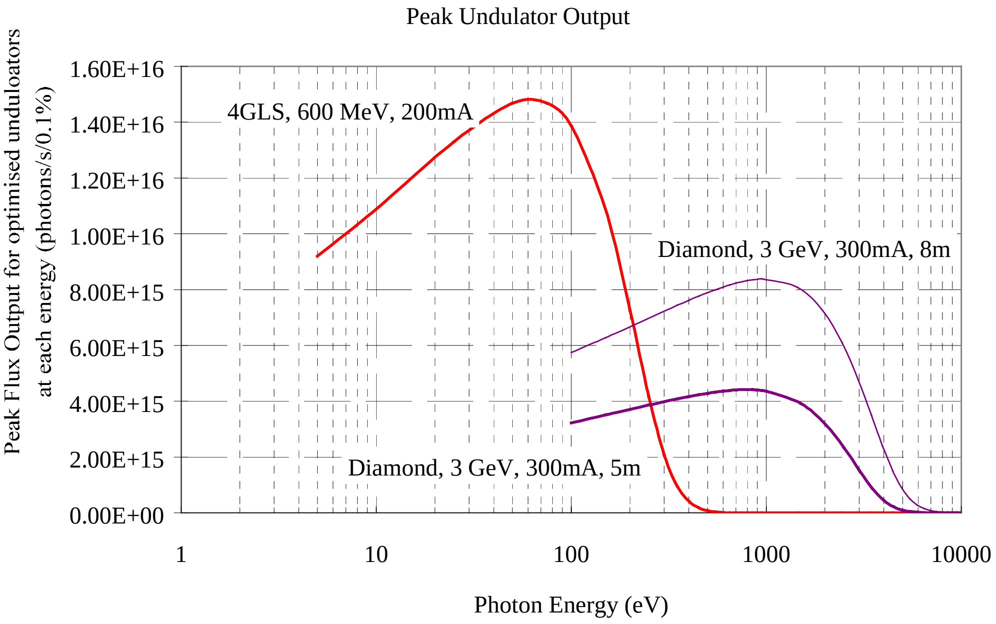


Figure 1. *Output of optimised undulators at each photon energy; for Diamond 5m and 8m straights are shown, for 4GLS a 15m a straight is shown.*

Figure 2 shows calculated flux and brightness performance of typical specific undulators for both machines. For DIAMOND, the comparison includes a 200mm period undulator; this period is longer than optimal and so does not reach the flux output of the envelope curve shown in Figure 1, but it can operate at lower photon energies. The highest photon energy provided on 4GLS is achieved using U28 which reaches 500eV using the 5th harmonic.

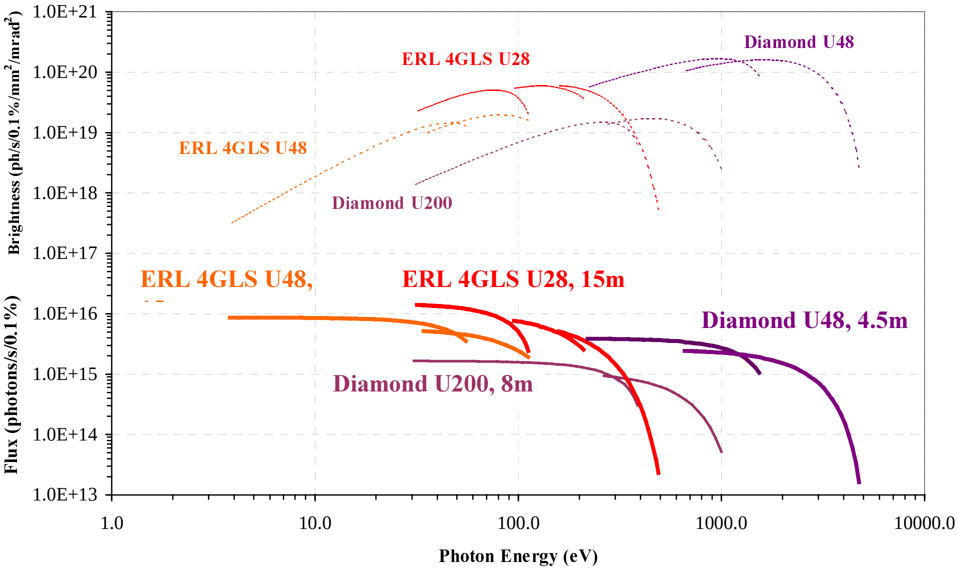


Figure 2. *Comparison of flux and brightness into the central cone for specific examples of undulators on 4GLS and DIAMOND. Note that a 200 mm period undulator (U200) is shown for DIAMOND; this long period would be required to give 30 eV photons at acceptable power levels, but is sub-optimal in its output flux.*

To see more simply the effect of machine energy on the photon energy range covered by undulators, realistic upper and lower limits for the period and *K* value can be chosen and the photon energy plotted for a fixed ID length, typically 5m. The lowest energies are provided by the highest *K* values and longest periods; the highest energies by the shortest period and lowest *K*. Undulator periods can range from values approaching the minimum gap to values where the number of periods is too small for good undulator output; this gives 10mm<period<200mm. The *K* value can range up to 5 (limited by power) and down to 0.1 (flux is a factor of 100 less than peak output). These upper and lower limits are shown in Figure 3 along with the ranges for 4GLS and DIAMOND.

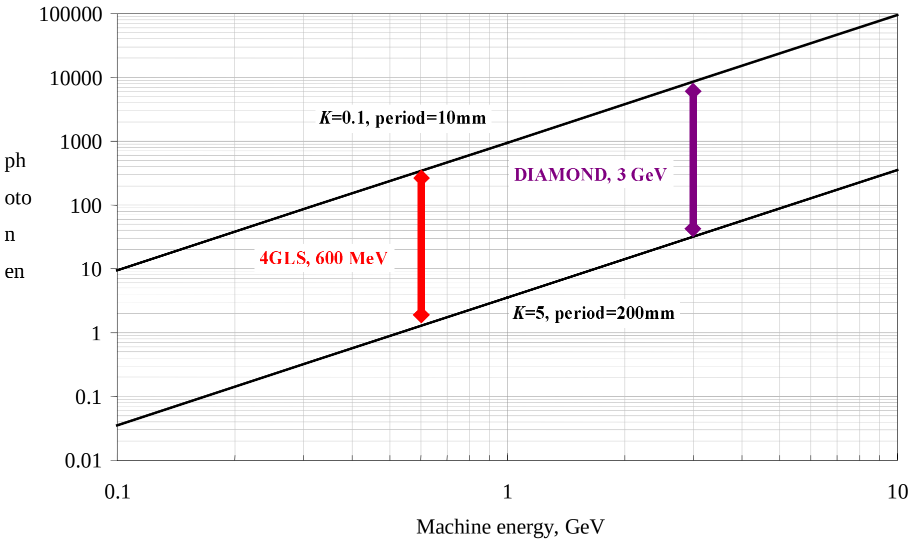


Figure 3. *The photon energy range covered by feasible undulators as a function of machine energy. The upper and lower limits are shown by black lines. The red and purple lines show the range of photon energies covered by undulator first harmonics in 4GLS and DIAMOND.*

Estimate of realisable flux at sample

All of these figures so far are in terms of the output flux of the undulators. A more relevant comparison is to calculate the flux delivered to the sample by a typical beamline and a specific insertion device. The region 5-100 eV divides into normal incidence (5-30 eV) and grazing incidence (20-100 eV) monochromators. Schemes for both have been modelled and the results presented in Table 1.

Table 1. *Estimated flux at sample for a spherical grating monochromator (30-100eV) and a normal incidence monochromator (5-10 eV) for sources on DIAMOND and 4GLS).*

source	**eV**	**source flux, 0.1% bp**	**sample flux (1) 1e4 RP**	**sample flux (2) 1e5 RP**	**power, kW**
DIAMOND U200 (8m)	100	1.58E+15	2.36E+13	1.42E+12	0.255
	50	1.65E+15	2.47E+13	1.16E+12	0.567
	30	1.66E+15	2.44E+13	9.46E+11	1.057
4GLS-erl U28 (15m)	100	7.41E+15	1.09E+14	4.67E+12	0.037
	50	1.25E+16	1.85E+14	7.50E+12	0.295
	30	1.40E+16	2.08E+14	7.98E+12	0.651
4GLS-ERL U48 (15m)	10	8.50E+15	6.80E+13	2.40E+12	0.410
	5	8.60E+15	6.30E+13	2.10E+12	0.890
Diamond BM	100	8.56E+13	3.47E+11	4.62E+09	0.240
	50	6.40E+13	2.40E+11	2.88E+09	0.240
	30	5.55E+13	1.92E+11	1.92E+09	0.240
	10	2.50E+13	5.90E+10	4.80E+09	0.240
	5	1.90E+13	8.60E+10	3.30E+09	0.240

It can be seen that the lower energy 4GLS delivers more flux over the 5-100 eV region than DIAMOND with significantly lower power, also making the design and operation of high-resolution optics more viable. The calculations have been performed for two different (but both high) values of the resolving power.

10^4 resolving power

For 10^4 resolving power, the calculations confirm that only reflectivity and diffraction efficiency needs to be taken into account as virtually all the light can be transmitted through the slit sizes required. Hence performance depends directly on source flux.

10^5 resolving power

In this region, significant losses of > 67% occur at slits; this figure depends strongly on the detailed beamline design and on optics quality, requiring demanding slope errors of < 0.1 arc sec. (achieved at BESSY II). Non-ideal performance will reduce the beamline efficiency, reducing the accuracy of calculated estimates. However, the flux at the sample will still depend strongly on the source flux.

Bending magnets on DIAMOND

For beamlines on bending magnets, the much larger beam divergence results in very much poorer transmission through slits, while an important limitation is how close the optics can be mounted to the source and thus what divergence can be accepted. The flux from bending magnets is three orders of magnitude lower that from undulators on either DIAMOND or 4GLS.

Appendix 2: UV FEL radiation

1. Background and Physical principles

4GLS offers two types of FEL in the VUV/XUV range (a cavity-based FEL and a SASE FEL), which have no direct parallel on the DIAMOND machine. These sources are not only planned to operate in the photon energy range (below about 100 eV) identified as inappropriate for DIAMOND, but are quite unique in character and thus have no parallels in conventional synchrotron radiation sources. For completeness, however, we summarise here the main characteristics of the proposed devices, and outline some of the novel scientific opportunities.

A FEL is a laser in which the lasing medium is a relativistic electron beam, and its physical structure is very similar to that of an undulator, comprising a periodic array of dipole magnets. In a cavity-based FEL, mirrors are used at either end to reflect the emitted light back into the device, as in a ‘conventional’ laser. One advantageous characteristic of a FEL is that they are very easily tuned, for example by simply changing the undulator field strength (usually by magnet gap variation), over an energy range which, by the standards of conventional lasers, is extraordinarily wide. At shorter wavelengths, into the XUV spectral range, increasing problems are found in designing sufficiently reflective mirror materials which do not degrade rapidly during laser operation and which do not have a restrictive tuning range. This has led to the design of SASE (Self Amplified Spontaneous Emission) XUV and X-ray FELs, such as the TESLA Test Facility (TTF), TESLA XFEL and the BESSY FEL. In a SASE FEL, the electrons make a single pass down a very long undulator and amplification of the spontaneous emission occurs through interaction of the electron bunch and the electromagnetic field.

2. Proposed 4GLS FELs

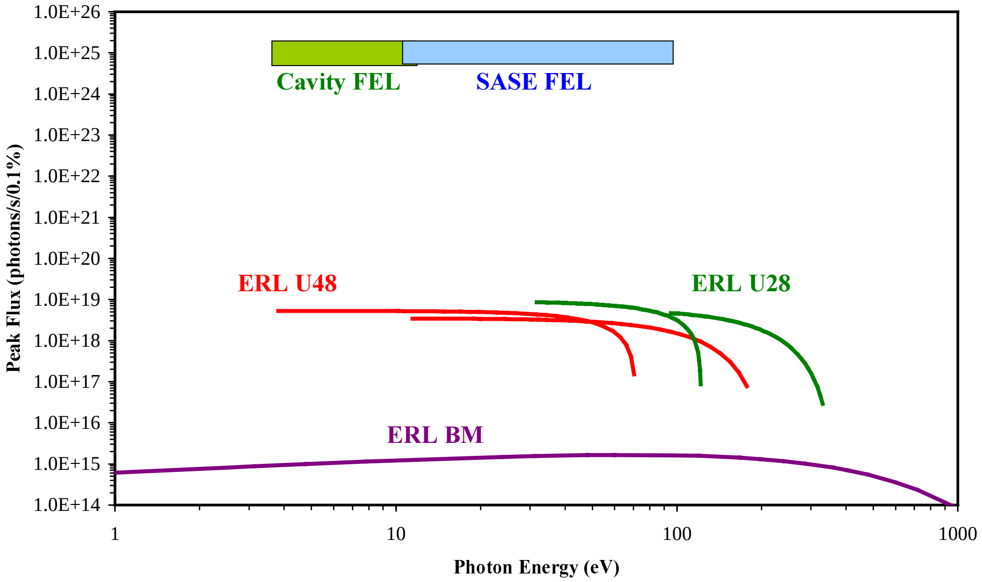
The cavity-based UV-FEL

This is an integral part of the ring and will operate from the start-up of 4GLS. It is intended to generate pulses of VUV radiation in the range 3-10 eV with broad tunability, and at selected higher energies with reduced tunability. In the sub-10 eV regime it is anticipated that it will have an average brightness of around 5 x 10^{22}

photons/s/mm^2/mrad^2/0.1%BW, and a peak brightness of around 10^{27} photons/s/mm^2/mrad^2/0.1%BW (see Table). This is around 10^6 times more intense than the VUV radiation from undulators either on 4GLS or on DIAMOND. The pulse length will be of order a few hundred fs, with the aim of reducing this in future.

The cavity-FEL offers very good pulse-to-pulse stability compared with a SASE FEL, and importantly may be used with variable polarisation (i.e. linearly- through to circularly-polarised pulses may be produced), not yet proven using the SASE design. It also offers some flexibility in pulse tailoring (e.g. changing the structure within pulses, or the gaps between them). However, the operational range is limited at the high photon energy end by the mirror material. In 2000 lasing at 6.4 eV was demonstrated at the Duke University FEL, and this record was broken in February 2001, when the EUFELE FEL at ELETTRA lased at 6.5 eV. This project has recently received EU funding to transform the ELETTRA FEL into a user facility operating at photon energies up to 8 eV (155 nm) within the next 3 years. Thus the goal for the 4GLS cavity FEL of lasing at 10 eV when 4GLS is commissioned is probably realistic. However, while the design of stable mirrors, particularly multilayer materials is improving rapidly, it remains the case that the use of these materials restricts the tuneability of the laser increasingly as the lasing energy is increased. Thus for lasing at energies of above around 10 eV, a SASE FEL appears currently to be the source of choice.

Parameters for cavity and SASE FELs on 4GLS	Cavity VUV-FEL 3-10eV	SASE XUV-FEL @58eV	SASE XUV-FEL @100eV
Average photon flux (ph/s)	10^{21}	10^{18}	10^{18}
Peak photon flux (ph/s)	10^{25}	10^{25}	10^{25}
Number of photons per pulse	10^{14}	10^{14}	10^{14}
Energy per pulse (mJ)	0.16	1.5	1.1
Average brightness (ph/(s mm^2 mrad^2 laser bandwidth))	10^{22}-10^{23}	10^{22}	10^{22}
Peak brightness (ph/(s mm^2 mrad^2 laser bandwidth))	10^{27}	10^{29}	10^{29}



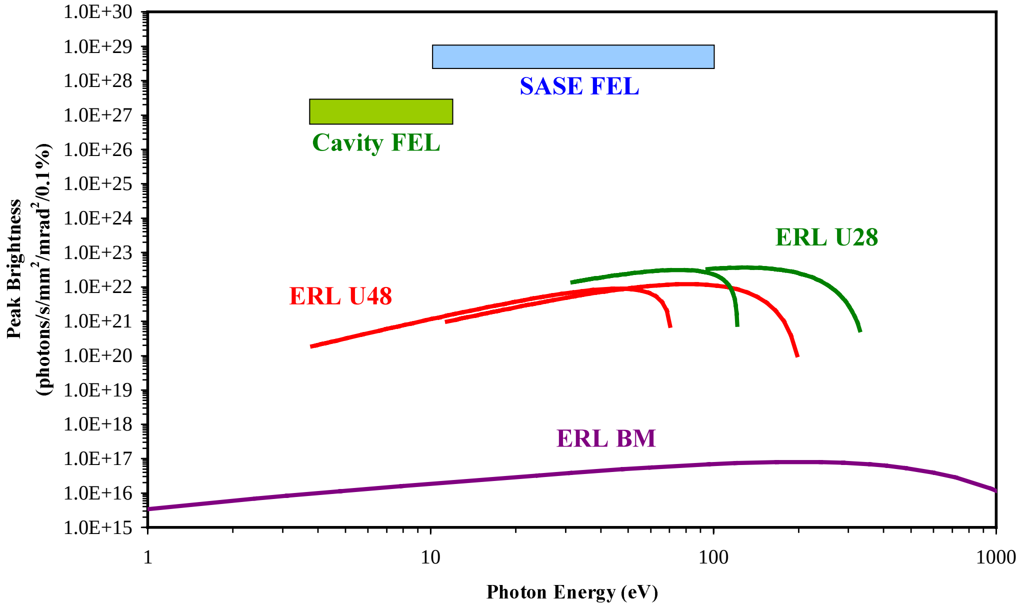


Figure 4. Comparison of ***peak*** *flux and brightness of the VUV and XUV FELs with those of conventional undulators on 4GLS*

The SASE XUV-FEL

The SASE FEL on 4GLS is intended as a development project to be built after the initial commissioning of the source. This is because the SASE technology is not currently as established as that for cavity-FELs. The XUV-FEL is aimed at generating photons up to 100 eV in energy (wavelength 124 Å) in the fundamental with a calculated peak brightness of 10^{29} ph/(s mm^2 mrad^2 laser bandwidth), again with pulses of the order of 100 fs duration initially. The brightness figure for the 4GLS XUV-FEL is similar in magnitude to the figures quoted for the TESLA Test Facility (TTF) and the BESSY FEL project. The 4GLS XUV-FEL is aimed at extending the short wavelength laser capability of 4GLS beyond the 10 eV that can potentially be reached by the cavity VUV-FEL. It is not planned to extend into the regimes covered by the proposed X-ray FELs such as the TESLA XFEL project, the BESSY FEL project and LCLS (Linac Coherent Light Source, Stanford) which are designed to produce X-rays down to 1 Å in wavelength. The 4GLS FEL capabilities thus complement those of the proposed X-ray FELs.

The aim of the SASE XUV-FEL is to obtain intense pulses of coherent, short wavelength radiation, but the physics of the SASE process means that extremely high electron current densities are needed and this implies very short electron bunches. The electron injection systems that do this have repetition rates that fall in the 100s of Hz to kHz regimes. They are pulsed sources and cannot operate in a manner that would produce a quasi-continuous photon beam. All of the 4GLS FELs necessarily would operate in the equivalent of single bunch i.e. a pulsed mode.

2. Scientific Opportunities provided by the FELs

Both FELs make possible entirely new science in experiments requiring pump-probe techniques (which profit, in the case of the cavity VUV-FEL, from the natural synchronisation of the different elements of the source), time resolution or coincidence techniques. The very high peak brightness makes experiments on very dilute samples using flux hungry techniques possible, in some cases making ‘single-shot’ experiments feasible. Pulse lengths into the 100 fs range make possible ‘real-time’ monitoring of chemical reactions, carrier transport in porous solids and biological processes such as protein folding. A few examples for both FELs are given below.

Scientific opportunities using the cavity-based VUV-FEL

The cavity VUV-FEL radiation will be sufficiently energetic to excite photoelectrons from close to the Fermi energy. The very high intensity means that these electrons may be used to determine the spin-polarised density of states function close to E_F in clusters or other dilute magnetic materials. This may be achieved with either very high energy or very high lateral resolution allowing experiments such as SPLEEM (spin-polarised low energy microscopy); such a facility could have a major impact on the design of new permanent magnets and magneto-optic recording materials.

Particularly in the range 160 nm – 190 nm (approx 8 eV-6.5 eV), the VUV-FEL offers possibilities for fast UV-CD in the ns and ps regime, with some 'one-shot' experiments probably being feasible (see appendix 3). This has important applications in time-resolved studies of protein folding and the study of conformational changes in other macromolecules. This energy range is appropriate to the cavity-based FEL for which the possibility of variable polarisation offers a substantial advantage. This may also be exploited in low energy photoelectron CD from chiral molecules – here the chiral effect may be as large as 10 %, even for randomly oriented molecules, and is greatly enhanced by using pump-probe techniques to align or state-select the molecules in the gas phase. Thus in this case, single-shot experiments become feasible using the UV-FEL. The FEL also offers a good source for confocal microscopy, with spot sizes smaller than 50 nm over a wide range of wavelengths.

In combination with other radiation, in pump-probe experiments, the UV-FEL has multiple applications. For example in solid state physics, the UV-FEL may be used to study electron-hole pair dynamics in semiconductors – relevant for example to the study of transients in Shottky diodes, of the design of photovoltaic cells (in fact the LURE super-ACO FEL, operating at up to around 1 eV is being used for the study of surface photovoltage effects in silicon and Si/SiO_2 junctions).

The UV-FEL also offers the possibility of using pump-probe experiments to study excited states in molecules and clusters and transient molecule spectroscopy, where the thresholds for excitation lie below 10 eV, and intramolecular vibrational relaxation dynamics for some large molecules. This is described in more detail below, as the full exploitation of these techniques will in some cases require the higher energy photons provided by the SASE FEL.

Scientific opportunities using the XUV SASE FEL

An important application of the XUV FEL is to pump a reaction process, which is subsequently monitored as a function of time. Examples include the use of the high flux from the FEL to create transient molecules in sufficient concentration to allow their subsequent study by coincidence techniques – this has relevance to the study of short-lived intermediates involved in, for example, the creation or breakdown of pollutants in the atmosphere.

The timescales accessed by the FEL (a few 100 fs initially) are sufficiently fast to allow the study of the internal vibrational relaxation of large molecules such as biomolecules. Electronic relaxation plays an important role in photochemical reactions (for example studies of radiation damage of biomolecules), and the initial substrate-complex interactions of enzymatic reactions. The timescales accessed are also sufficiently fast to study bond-breaking and making (around 100 fs), and thus to allow the ‘real-time’ monitoring of chemical reactions and their transition states.

The very high brilliance of the FEL will allow the study of single clusters in the gas phase, and the Coulomb explosions which may be induced by a focussed XFEL beam. Spectroscopic studies on other low density systems such as radicals, chiral molecules, and radioactive or exotic nuclei (prepared using the radioactive beams facility) will also be possible. The last two examples have clear applications in astrophysics, since almost nothing is known about the spectroscopy of atoms or ions based on isotopes which do not occur naturally on earth but are to be found in stellar atmospheres. Other areas accessed by the high brilliance of the SASE FEL are multiphoton experiments and the generation of plasmas in which ensembles of atoms are excited or ionised coherently.

Appendix 3: Source Provision for Circular Dichroism

1. Introduction

Synchrotron Radiation Circular Dichroism (SRCD) is an emerging technique in the Biological Sciences with potential applications in protein folding, the study of macromolecular interactions, conformational changes and enzyme mechanisms, and secondary structure determination, with specific applications in fold recognition for structural genomics.

The UK is now considered the world leader in this technique and with the advent of the new station 12 on the SRS this dominance should continue. Much effort in the past 2 years has been on proof of principle, spearheaded by the BBSRC's establishment of the Centre for Protein and Membrane Structure and Dynamics at Daresbury. The developments on the old 3.1 station at the SRS have shown the advantages of SR sources over conventional lab-based CD. The additional flux available on station 12 relative to conventional instruments, approximately a factor of 10^2 at a wavelength of 240 nm, and more importantly, a factor of more than 10^4-10^5 at 180 nm, means that smaller amounts of precious biological materials can be used, and that proteins can be examined in solutions with higher concentrations of salts and buffers (resulting in conditions which are more "physiological-like" and that better mimic those appropriate to crystallisation). An additional direct result of the higher flux at VUV wavelengths is that aqueous spectra can be measured to lower wavelengths (the practical limit is now ~160 nm due to the water solvent, as opposed to ~190 nm due to limitations of the beam in conventional instrument). The superior beamline characteristics of the new station 12 on the SRS should lead to rapid advances in biological studies.

The goals for future development of CD beyond the lifetime of the SRS are to maintain and improve on the quality of data obtainable on station 12 and to enhance CD capabilities generally in order to promote new science. The instrumental advantages that may be offered by the new sources DIAMOND and 4GLS include: 1) higher flux, 2) an improved potential to exploit shorter wavelengths for non-aqueous samples and 3) better time and better spatial resolution (smaller spot sizes). It is important to recognise, however, that the new beamline on the SRS far outstretches any other facility world wide for this work, so even maintenance of this level of performance will be vitally important to this scientific community. Nevertheless, further improvement in these capabilities will mean that the information content will increase and permit the examination of 3D folds (important for future Structural Genomics studies) as well as discrimination into more finely graded secondary structural types. Finally, the smaller spot size and higher beam intensity mean that much smaller amounts and concentrations of samples will be necessary (which will enable high-throughput screening for Structural and Functional Genomics projects). All of these will be possible as consequence of the higher flux available on DIAMOND or 4GLS.

In addition to this mainstream CD activity, 4GLS does offer some advantages in time and spatial resolution which could open up new areas of application of CD, enabling, in principle, studies of rapid and tissue-localised biological processes such as: protein folding, real-time conformational changes associated with catalysis, vision, and for the first time, ion conductance across membranes (potentially monitoring optically single channel formation across membranes or in patch pipettes). Spatial resolution could permit examination of compartmentalised functioning in cells. Thus, the availability of both types of SR sources will allow continuation of the recently achieved success at the SRS and open up new opportunities in biological sciences heretofore not possible on existing conventional sources or SR machines.

2. Technical assessment

2.1 Flux

High flux is important for CD because the dicroism signal may be only 10^{-5} of the absorption. This is why the SRS is preferable to laboratory sources since the SRS offers a higher flux in the 160 nm to 190 nm range. The flux available from bending magnets on DIAMOND and 4GLS will be essentially the same so other considerations will determine the preferred location of such beamlines. The absence of hard x-ray radiation and resultant lower heat load on the optics, together with the flexibility engendered by the ERL design favours 4GLS, but the close proximity of a large-scale effort on protein crystallography and associated back-up laboratories favours DIAMOND. However, the flux from either a wiggler or an undulator on 4GLS will be an order magnitude higher than these bending magnet sources and it should also be possible to focus the output from an undulator onto a smaller specimen, so for some specific experiments this may be the preferred option. Calculations of the output around 5 eV from an insertion device on 4GLS ERL indicate a yield of up to 8x10^{15} photons/sec on the specimen compared to 10^{14} photons/sec from a bending magnet on either source (assuming a 35 mrad horizontal acceptance angle). The exact performance of a bending magnet on DIAMOND for low energy photons does depend on the vertical angular aperture which can be extracted, and the possibility of modified dipole vessels on DIAMOND has been discussed by the SAC and TAC, but in the energy range appropriate to CD this issue is of marginal importance.

2.2 Polarisation modulation and time-resolution

Measurement of the weak CD signal requires the use of periodic switching between left- and right-circular polarisations and the use of lock-in techniques, and this switching is currently achieved through the use of a photoelastic modulator (PEM). The PEM introduces a limitation to the time resolution and also ultimately imposes a short wavelength limit on the measurement due to absorption (although for most samples the solvent, such as water, is the source of this limit). In the present work on the SRS the PEM is modulated at 50 kHz giving a 20 μsec limit on the speed of data collection but current experiments need only a 1 msec response and so are not limited. These experiments are carried out in two modes:

a) time-resolved single wavelength monitoring

b) spectral scanning

A major improvement being implemented at station 12 on the SRS is the use of a white beam and multi-detection which will offer sub-msec response. Notice that white beam studies have the potential to compensate for the lower flux at a single energy relative to an undulator. In addition to these intrinsic time limitations, however, the useable time resolution of an experiment is also limited by flux, and to push this aspect in the future there may be an advantage in using an insertion device on a low energy source such as 4GLS. One might also envisage an experiment in which the insertion device delivers the polarisation switching directly, allowing one to dispose of the PEM, although current methods of undulator switching are much too slow (see below).

3. Long term development of CD

Clearly both DIAMOND and 4GLS are able to provide bending magnet radiation for the current aspirations of the biological CD community, and there may be logistical considerations favouring DIAMOND. In the longer term one might envisage a new generation of CD experiments which exploit the higher flux, special insertion device properties and time structure of 4GLS and which could only be performed on such a source. For example, two slightly out-of-line helical undulators could shine the two opposite light polarisations simultaneously through the specimen and onto two different detectors, obviating the need for any polarisation switching. One might also consider exploiting the special time structure of the output of an insertion device on a laser-excited ERL since in principle this would offer experiments on the μsec, nsec and psec time scale. The ultimate time resolution would be defined by the time scale of one pulse of such a source. For the most demanding experiments, the limitation on time-resolved experiments is defined by the need for approximately 10^{15} photons per measurement point to achieve a 1% signal-to-noise ratio in a 10% absorbing sample with a CD signal of 1 part in 10^{5}. Unfortunately, even the expected output of the cavity-based FEL (for which existing devices are predicted to reach the full spectral range of interest in the next 3 years) corresponds to only about 10^{14} photons per pulse. Single-pulse CD may not be achievable in this worst-case scenario, but experiments at lower signal-to-noise ratio or measuring stronger CD signals should be viable. Moreover, the UV-FEL could allow the exploitation of ellipsometric techniques [1] in combination with stroboscopic and lock-in detection to obtain ps time resolution.

Reference

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Appendix 4: Infrared Radiation Sources

1. Synchrotron radiation (bending magnet) IR studies

There are more than 30 IR beamlines at synchrotron light sources throughout the world based on storage ring bending magnets. They are used for a wide range of fundamental and technological problems including: heterogeneous catalysis, solid-state photochemistry, high-pressure geochemistry, cancer diagnosis, food science and forensic analysis.

Reflection-Absorption Infrared Spectroscopy (RAIRS) operates in the far–IR (600-20cm^{-1}) where the flux advantage over a conventional IR source for small samples, such as a single crystal surface oriented at grazing angle, approaches 2 orders of magnitude. The technique provides information on the chemical bonding between molecules and surfaces and cannot be matched for its combination of sensitivity and spectral resolution. The SRS is a world-leading source for this work. Recent activity by UK groups includes studies of molecular bonding on surfaces, photon decomposition of phosphorus trifluoride on nickel and adsorbed MOCVD precursors. Future work will include the bonding of fullerenes and self-assembled organics, problems that are relevant to molecular electronics, biosensors and other areas of applied science.

Infrared Microspectroscopy (IMS), in which microscopic dots of material are analysed by infrared spectroscopy, benefits from the higher brightness of SR sources throughout the whole mid-IR range (4000 cm^{-1} – 600 cm^{-1}). Conventional IR microscopes will resolve to about 30 - 100 microns, whereas the SRS can resolve to below 10 microns, and for DIAMOND this could be reduced to 3 microns. The great versatility of synchrotron IMS as a means of micro-chemical analysis has led to a rapid increase in the range of applications by UK groups. It has attracted users in the biomedical sphere, where IR analysis at the sub-cellular level is being actively exploited. Other work includes studies of high T_c superconductors, single crystals of zeolite catalysts, earth sciences and archaeology. IMS remains non-destructive even when applied to the most sensitive molecular and biomolecular materials. In France and the USA there has been growing interest in the technique from the cosmetics industry and the forensics services.

Projected demand and future provision

The existing UK RAIRS community is relatively small (5 user groups), but the experiments are difficult and demanding in time. RAIRS is used mainly for surface science but is expected to develop with the growth of the nanoscience in the UK. The IMS community is larger and more diverse. Experience worldwide points to further growth, *eg* a further five IMS beamlines are planned at the ALS in Berkeley which will bring the total in the USA to eighteen. Currently there is one station at the SRS (beamline 13), which is used for both RAIRS and IMS. Another (beamline 11) has been funded specifically for IMS. Evidence collated by the “Low Energy Provision on DIAMOND” working party suggested that future synchrotron IR needs of the UK community could be met by *one dedicated RAIRS station* (which could also provide for more general spectroscopic experiments in the far-IR such as geology and materials studies at high pressures) and *three stations for IMS*.

The flux output of synchrotron radiation in the IR is almost independent of electron energy and bending magnets on DIAMOND and 4GLS are thus essentially equivalent sources for this purpose. There is a technical issue associated with the fact that the natural opening angle of IR synchrotron radiation is very large, especially in the far IR, and this large aperture needs to be allowed for in the design of the dipole magnet areas and associated vacuum vessels. The DIAMOND Low Energy Working Party has established that relatively minor modifications in the dipole area to the current design for DIAMOND (changes to the shield wall and positioning a plane mirror in the crotch area) would allow 11 mrad vertical and 35 mrad horizontal collection angles to an IR station, compared to the optimum for IMS of 20 mrad x 20 mrad. Coupled with the small source size of DIAMOND this will ensure excellent performance at 3 micron spot size with flux up to 5x that achievable on the new beamline 11 at the SRS. Provision for at least three such modified dipole areas in the initial DIAMOND design is a minimum requirement.

Accommodation of RAIRS and far-IR spectroscopy stations on DIAMOND would require modification to the dipole vessel itself to open up the vertical collection angle to at least 30 mrad. Such a modification is feasible and inexpensive and would meet envisaged future needs if carried out at just one dipole area on DIAMOND. These far IR studies could also be accommodated (but much more readily) on a suitably-designed dipole magnet on 4GLS.

2. IR FEL sources

In contrast to synchrotron IR sources (10s of milliwatts of average power in nanosecond pulses in a broad band across the infrared range) with IR FELs (up to 10 watts of average power in a narrow band, in picosecond pulses with 10 megawatts peak power) we are making an enormous leap. In general terms, accelerator-based laser sources are expected to play a key role in leading developments in biophysics, medical physics, nanomaterials, condensed phase dynamics and atomic and molecular science [1]. In the case of the IR FEL, non-linear and time resolved spectroscopic experiments become available which are not possible with synchrotron sources. The FEL source also has capabilities beyond those of current table-top IR lasers, particularly in terms of its long wavelength capability (> 10 μm), its ease of scanning and its high power (the current micropulse power envisaged for the 4GLS IR FEL are 10 x higher than achievable with a table top source).

The 4GLS Facility

The 4GLS facility will offer additional advantages stemming from the combination of VUV FEL, IR FEL and synchrotron IR sources in both pump-probe and non-linear two photon experiments *eg*

- UV FEL pump – Synchrotron IR probe experiments investigating photochemical dynamics in biological systems, surfaces and interfaces.
- IR FEL – Synchrotron IR probe experiments investigating folding dynamics in biological systems or vibrational relaxation dynamics of large adsorbates.
- UV FEL – IR FEL sum frequency spectroscopy of interfaces.

Unique experiments in the above categories arise from the long wavelength capability and tuneability of the IR FEL and the short wavelength capability and tuneability of the UV FEL.

Apart from the synergistic advantages of having a synchrotron IR source coupled with laser sources, the advanced design of the energy recovery linac (ERL) provides flexibility to construct a dipole magnet area, and to define beam parameters, which are totally optimised for IR emission. It follows that the 4GLS facility could provide an excellent source for IMS and RAIRS experiments in general.

It should be emphasized that the two-source experiments mentioned above require both spatial and temporal synchronisation of the two light sources. It is important that the evolving design of 4GLS aims to provide synchronisation to the sub-picosecond time scale between the IR and UV FEL sources.

Near Field Microscopy

There are a number of drivers towards IR microscopy at sub-micron spatial resolution, particularly in probing biological material at the sub-cellular level. The synchrotron IMS technique probes at down to 3 microns within cells of 10-30 microns in diameter. Biologists are more interested in the length scale of organelles within the cell, say 100 nm. This can be achieved with apertureless near field techniques which require laser sources [2]. The high flux and long wavelength tuneability of the IR FEL suggests applications in this area.

References

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