From
Terawatts
to
Femtoseconds

Laser Research
at
The SERC Central Laser Facility
Rutherford Appleton Laboratory

Lasers – from terawatts to femtoseconds

by Professor M H Key, *Rutherford Appleton Laboratory*

Progress in laser technology has been rapid and sustained in the three decades since the Nobel prize-winning discovery of the first lasers. Developments have made lasers ubiquitous scientific tools as well as being widely incorporated in devices in the industrial, military, medical, communications, office equipment and consumer electronics sectors. In many cases they have become simple and cheap but, at the extremes of sophisticated scientific applications, the technology of lasers themselves has become a major challenge.

The SERC Central Laser Facility

It was the prospect of new scientific opportunities together with the escalating scale of the problems of developing and operating advanced high power laser systems which led to pressure on SERC from university and polytechnic researchers in the early 1970s for central provision of a high power laser. SERC responded by establishing, at the then Rutherford Laboratory, a Central Laser Facility (CLF) equipped with a 2-beam (108 millimetre) neodymium glass laser called Vulcan capable of 0.3 terawatts (3×10^{11} watts) of infrared (1.06 micron) power in subnanosecond (less than 10^{-9} second) pulses. Speed of construction was a paramount consideration and, built with the best available commercial components (then French and American), the laser and associated target irradiation facilities became operational in 1976 less than two years after the decision to build it.

The impact on UK contributions to research using high power lasers was immediate and significant.
The disparate activities of research groups previously working with comparatively poor equipment by international standards, were brought together into cooperative and highly effective work with Vulcan.
There was widespread international recognition of the contributions made to the study of high intensity interactions of laser radiation with matter, laser-driven implosions and compression of plasma, the physics of dense plasma and the development of methods of measurement applicable to plasmas of subnanosecond duration, submillimetre dimensions and temperature of hundreds of millions degrees Kelvin.

As the demands of its user community developed, Vulcan was modified continuously, so that today, more than ten years later, it is the world's most versatile high power laser facility. It has six 108 mm beams and two 150 mm beams (see figure 1) largely constructed with components designed and manufactured in the UK and capable of generating up to 5 TW (5×10^{12} W).
A wide range of pulse durations, wavelength and target irradiation geometries are available for two experiments in parallel by automated beam switching as summarised in table 1.

The versatility of Vulcan's operating parameters arises from the diversity of scientific research which it serves. Another facet of that diversity is the extensive network of international cooperation which has developed between the 80-strong academic community associated with Vulcan and their colleagues in some 16 institutions overseas.

Table 1: Vulcan laser output at 1 micron

Pulse duration	6-beam array	2-beam auxiliary
Long (0.7-25 nsec)	300 J/beam	500 J/beam
Short (70-500 psec)	80 J/beam	125 J/beam
Ultra short (20-30 psec)	15 J/beam	Unsynchronised

Long/short pulse synchronisation, better than 5 psec, between 6 beam array and auxiliary beams.

Repetition rate three full energy shots/hour.

Target irradiation facilities

- 50% efficient conversion to 0.5 micron
- 6 beam line focus and cluster geometry plus single beam backlighting
- 12 beam spherical geometry plus double beam backlighting
- ISI and random Phase Plate uniform target irradiation options
- Synchronised UV optical probe

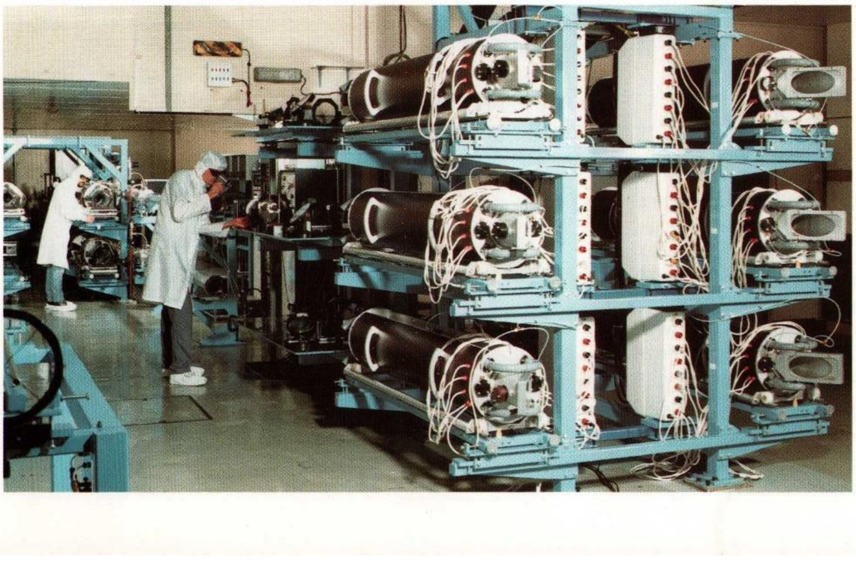

Figure 1: VULCAN laser output amplifiers. VULCAN output parameters are given in table 1.

The maximum power of Vulcan is modest compared to the 100 TW in the 1070 cm beams of the NOVA neodymium glass laser installation in the USA (Vulcan is fifth in the world league of high power capability) and, while occasional collaborative use of powerful lasers in the USA and Japan is helping UK groups to pursue their research programmes, local solutions are also being sought.

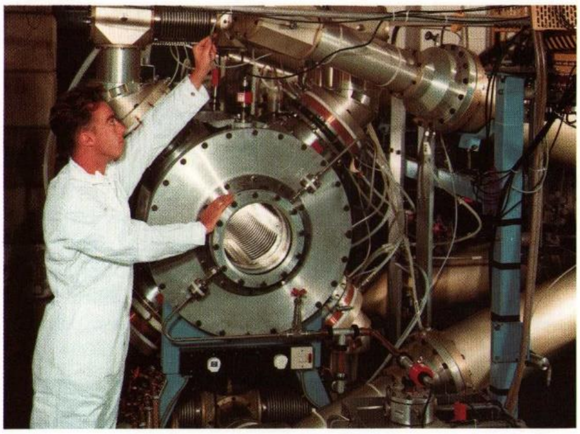

Figure 2: Output amplifier of the KrF laser Sprite. Operating parameters are given in table 2.

Development of high power KrF lasers

During the period of operation and development of Vulcan, staff at the CLF have played a leading role internationally in exploring the possibility of an alternative, less costly, high power laser technology using a novel laser architecture based on ultraviolet (248 mm) krypton fluoride excimer lasers.

Small discharge excited KrF gas lasers, operating at tens of Hertz pulse-rate and generating a fraction of a Joule in 10 to 20 nanoseconds, have in recent years become a commercial success (demonstrated by their use in the Laser Support Facility described below) but exploitation of the KrF laser for high power generation has required new ideas to overcome problems such as the small aperture limit of discharge excitation and the small upper state lifetime of the KrF excimer molecule (2 nanoseconds).

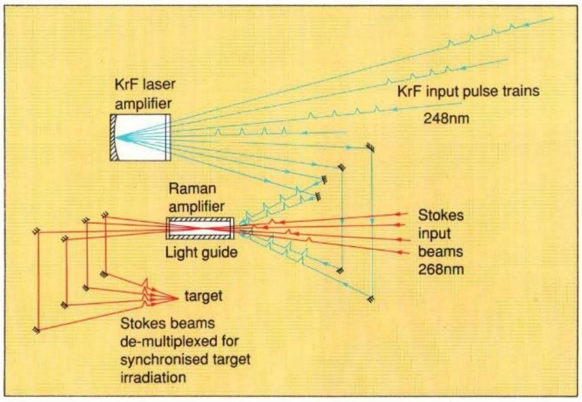

Figure 3: Schematic showing sixteen 248 nm pulses arriving at a KrF amplifier sequentially in four angularly separated beams each carrying four equally spaced pulses. Differential time delays synchronise the pulses in the four beams at a CH_4 gas cell which acts as a Raman amplifier. The energy of the 248 nm beams is transferred to four angularly separated beams at the 268 nm Raman Stokes wavelength. Each Stokes beam carries a single pulse synchronised with one of the groups of four 248 nm pulses in the pump beams, whose energy it extracts by stimulated Raman scattering in CH_4.

Research at the CLF has demonstrated a technologically successful solution to the first problem with modular high power electron beams exciting the KrF gas mixture for example, 500 keV, 500 kA in eight cold cathode diodes exciting a 27-cm diameter 1-metre long KrF laser amplifier, Sprite, pictured in figure 2, which has generated up to 200 J in 60 nsec. An elegant solution to the problem of the 2-nsec upperstate lifetime has also been developed by extracting the full laser energy of the KrF amplifier with a train of laser pulses at 2-nsec intervals spanning the whole period of electron beam excitation. Optical multiplexing and Raman beam combining is used to convert the train of pulses to a single pulse on target in the manner illustrated in figure 3. The single pulse can be of any duration less than 2 nanoseconds, giving the laser flexibility of pulse generation from picoseconds to nanoseconds. The capabilities of Sprite which have been made available to users, including a world record high energy picosecond pulse of 2.5 J, are summarised in table 2(a).

Table 2(a): Single beam output performance of SPRITE at 248 nm

Pulse duration	Energy (J)
50 nsec	150
3.5 psec	2.5

Repetition rate 12 per hour.

Lasers operating on this novel principle generate high power ultraviolet pulses considerably more efficiently and cost-effectively than neodymium glass lasers with harmonic conversion. Although the technology is relatively new, its promise is such that the recommendation of the CLF's review panel in 1988 was that Vulcan should be replaced in five to six years by a new KrF laser (Supersprite) with the specification shown in table 2(b). The advantage of the new technology is so significant that the major increase in performance of the proposed Supersprite laser compared with Vulcan, shown in table 2(b) relative to table 1, would be obtained with a cost, which is somewhat less than the investment in Vulcan.

Table 2(b): SUPERSPRITE specification

Laser	
No of beams	12
Maximum energy	3.5 kJ
Maximum power	300 TWatt
Pulse duration	1 psec-10 nsec
Wavelength	268-277 nm
Pulse rep rate	10 per hour
Target irradiation facilities	
a) 12 beam, long pulse, cluster or spherical	
b) 12 beam, short pulse, high power, cluster or line focus	

European possibilities

The rationale which caused UK universities and polytechnics to press for a UK Central Laser Facility is in a similiar way leading Europeans to consider the possibility of a European high performance laser facility which could have both the scientific diversity and technical versatility of the CLF with the significant additional feature of maximum power capability at world-leading level. Such a facility could open up important new scientific areas which are at present being considered by a scientific and technical working group composed of representatives from SERC and UK HEIs, CNRS (France), CNR and ENEA (Italy), MPG (West Germany) and CSIC (Spain), and due to report at the end of 1989. Meanwhile, development work on KrF lasers will continue at the CLF for the next two years pending a further review within SERC to assess the need for a national new laser (Supersprite) in the light of European developments.

The Laser Support Facility

Users of small lasers are also faced with a rapidly changing technology which can present both technical and financial problems for researchers requiring occasional access to advanced systems. Frequency tunable picosecond (10^{-12} sec) dye lasers have recently been adapted to generate pulses as short as tens of femtosecond (10^{-15} sec), and their use in analysing fast atomic and molecular processes has spread from physics to chemistry and biology. Nanosecond pulsed dye lasers have been engineered to produce intense narrow band light sources with sophisticated frequency scanning from the infrared to the vacuum ultraviolet.

Table 3: Laser support facility equipment

At Rutherford Appleton Laboratory

2 Excimer-pumped dye lasers
Triplemate Raman spectrometer with OMA/OSMA readout
Fluoride excimer laser
Laser generated X-ray source
Pico/femtosecond dye laser with amplification at 10 Hz and 6 kHz
Streak camera. Pump and probe system

For loan

4 Q-switched YAG-pumped dye lasers
1 Excimer-pumped dye laser
1 Fluoride excimer laser
1 Argon ion-pumped dye laser
Streak camera. Boxcar electronics

Full Raman shifting, harmonic conversion and mixing facilities are available on most dye lasers. Pump lasers may be used separately.

The availability at the CLF of a pool of laser expertise led to proposals from photochemists, supported by biologists and physicists, for some central provision for access to such small lasers and in 1985 the Laser Support Facility (LSF) was established to provide a loan pool of modern commercial lasers together with more complex systems located at the CLF for picosecond/femtosecond experiments and nanosecond spectroscopy. Table 3 summarises the facilities which are now available.

The LSF has proved popular and successful and, with its advent, the number of users of CLF has roughly doubled to a present level of some 150 academic staff and a similar number of research assistants and postgraduate and PhD students.

The future

The future course of the CLF has been charted in the recommendations of the Review Panel, chaired by Professor Challis, which reported to the Science Board in 1988 and by the response of the Science Board in accepting the Review Panel's recommendations and agreeing to fund the CLF at about its historical level of £3.2 million a year. About one-third of these resources will provide a slightly expanded Laser Support Facility and the remainder will be divided between operating Vulcan and developing krypton fluoride lasers, with the expanded LSF/KrF development work funded from savings on developing and operating Vulcan.

Science at the CLF

The diversity of scientific work at the CLF is such that a brief summary is difficult. Users of the high power lasers, Vulcan and Sprite, carry out typically 20 experiments a year usually involving collaboration between UK groups and often with participation from laboratories overseas. A few reciprocal experiments each year at laboratories overseas have participation from the UK. The more numerous small lasers provide for about 70 experiments a year usually by individual research groups.

Scientific applications of high power lasers

The extreme intensity (up to 10^{18} W cm^{-2}) and associated electromagnetic field strength (up to 2×10^{10} V cm^{-1}) which can be produced by focusing multi-terawatt laser beams, leads to the stripping of electrons from atoms and therefore to the production of plasmas which strongly absorb laser light and reach extremes of energy density in a thin surface layer of a solid target. Pressure can reach hundreds of megabars, temperature hundreds of millions K and density can approach 100 g cm^{-3}

The physics and applications of these plasmas provide a rich and diverse field of research including:

- Generation of intense single pulses of thermal X-rays and their application as time-resolved probes in other areas of science;
- XUV and X-ray lasers and their applications;
- Acceleration of charged particles in ultra high electric fields of laser driven plasma waves;
- New physics of laser interaction with matter at ultra high intensity;
- The physics of dense non-classical plasmas;
- Laboratory simulation of astrophysical plasmas;
- The physics of inertially confined fusion;
- Generation and applications of intense pulses of thermonuclear particles.

Thermally generated X-rays from laser produced plasmas are brighter than any other single pulse laboratory source for photon energies less than about 5 keV and sufficient for measurements using a variety of techniques. Subnanosecond time-resolved radiography of fast hydrodynamic phenomena, such as laser driven implosions, was pioneered at the CLF and is now widely used in other laboratories. Micro-radiography of living cells using 'water window' XUV radiation between the oxygen and carbon K edges has given about 50 nanometre resolution in contact images recorded in resists as illustrated in figure 4. Bragg reflection of soft X-rays from crystals is providing new information on the response of the lattice to both shock waves and thermal waves. Absorption spectroscopy with about 10 micron point sources of X-rays has yielded space-time maps of ionic ground state population density in laser produced plasmas. There is a valuable complementarity with synchrotron radiation sources which cater for high average power applications in that laser generated plasmas provide subnanosecond resolution of transient phenomena.

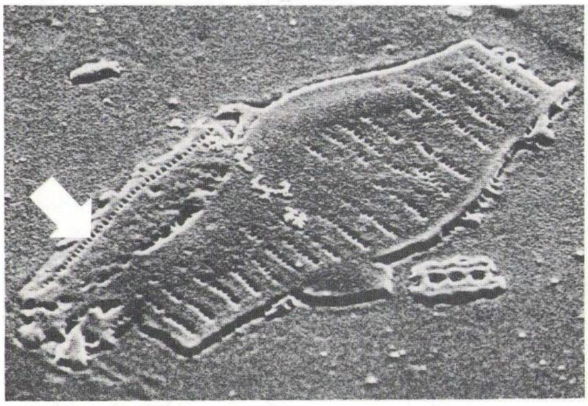

Figure 4: Contact X-ray micrograph of a diatom. The feature arrowed is 60 nm wide. The soft X-rays used to record this picture on a photoresist were generated by focusing a beam of the Vulcan laser on a gold target placed 10 centimetres from a cell containing the diatoms sandwiched between a thin Si_3N_4 window and a photoresist. The image is obtained by electron microscopy of the etched photoresist. (Royal Holloway Bedford New College – Rutherford Appleton Laboratory.)

Bright fluorescent sources of emission are needed for lasers, and laser-generated plasmas have enabled the development of XUV lasers. A new class of these lasers has been derived from early work in UK universities through the collaborative efforts of several teams working at the CLF. The new lasers operate by sudden cooling and recombination of a narrow column of plasmas produced by laser irradiation in a line focus. The shortest wavelength of laser amplification in 1986 (8.1 nm Balmer-α transition of hydrogenic F IX) was established at the CLF and, in a collaborative experiment using the more powerful Japanese GEKKO XII laser, a new limit of 4.5 nm has been established with the Balmer-α transition of Mg XII. At the shortest wavelengths, the length of amplifying plasma has been restricted to a few millimetres because of the high laser irradiation intensity required. The resulting amplification in a single transit has typically been less than 10 ×. Much larger amplification exceeding 10^4 × in a single transit and producing 10 kW output power in a beam of 10^{-2} radians divergence has recently been obtained at the CLF with 3p-3s transitions in neon-like Ge XXIII ions at 26.3 nm, as illustrated in figure 5. Work with high gain at longer wavelengths opens the way to study of the applications of XUV laser beams.

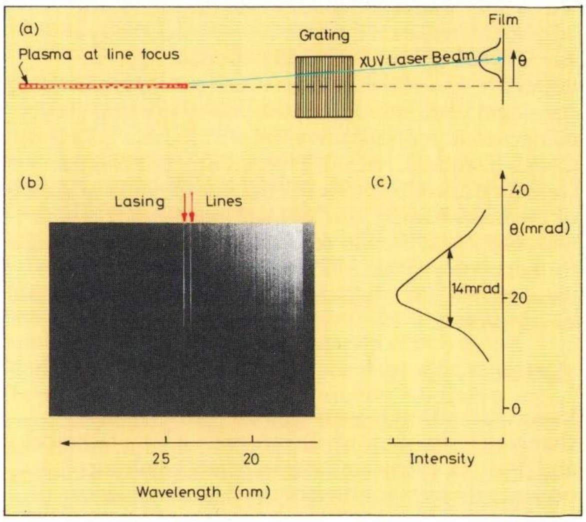

Figure 5: (a) Schematic of XUV laser experiment. (b) Spectrum of XUV laser lines produced by neon-like ions in a germanium plasma. (c) Variation of XUV laser intensity across the beam profile.

Operation of Vulcan at two frequencies with 1% beat frequency has been used to drive plasma waves with a relativistic gamma of 100, which is 10 × higher than in work elsewhere. Scattering of laser probe light from the resulting plasma waves has given important new evidence on their instability by turbulent break-up and, from their amplitude, electric field strengths of up to 3×10^6 V cm^{-1} applicable for charged particle acceleration have been inferred.

The physics of dense non-classical plasmas has been probed for the first time by absorption spectroscopy using measurements of the shift of the X-ray K edge to determine the state of the electrons and using EXAFS observations to show the correlations in positions of ions in the transition from a strongly correlated to more ideal plasma under varying levels of shock compression.

Intense black body emission from a gold laser generated plasma has created another plasma in thermodynamic equilibrium whose X-ray absorption spectrum, probed with an auxiliary laser produced plasma, has been measured for comparison with computer modelling. The results shown in figure 6 are important for the simulation of stellar atmospheres where validation of computer modelled absorption spectra is needed.

Many facets of the physics of inertially confined fusion have been studied at the CLF. Perhaps the most significant contribution by CLF users to international effort has been the experimental measurement of the rate of heat transport by plasma electrons and the development of a fundamentally new theoretical description valid in the limit where electron mean free paths are longer than the scale length of the strong temperature gradient. Current studies of energy transport are focusing on the unusual situation produced by intense picosecond pulses in which a transient heat conduction wave produced plasmas at solid density in a time too short for explosive expansion.

Laser driven implosions can generate thermonuclear reactions in deuterium/tritium mixtures, producing mono energetic alpha particles, protons and neutron emission at high intensity. Subnanosecond pulses of up to 10^9 fusion particles have been obtained in dodecahedral 12-beam irradiation with energy concentrated in 12 'hot spots' on a spherical target and the source has been applied to the study of charge particle energy loss in dense plasmas as well as to probing the growth of the Rayleigh-Taylor instability in laser accelerated targets.

Science with small lasers

Research at the CLF with small lasers covers many fields of physics, biology and chemistry, with chemistry accounting for more than two-thirds of the total.

An important effect of the LSF has been to enhance the spread of advanced techniques. This is well illustrated by ultraviolet resonance Raman scattering studies where methods developed at the CLF by groups from York University and the Royal Institution for an extensive study of anthroquinones were subsequently used by other groups for work on enzyme catalysis, measurement of organometallic complexes and for the analysis of hydrocarbon mixtures. Results have included the first observation of an acyl enzyme intermediate in naturally occurring enzyme catalysis and the first examples of resonance Raman scattering from metal-ligand-charge-transfer states of copper complexes and from transients in the photolysis of the Fischer complex. Picosecond Raman techniques are now being developed.

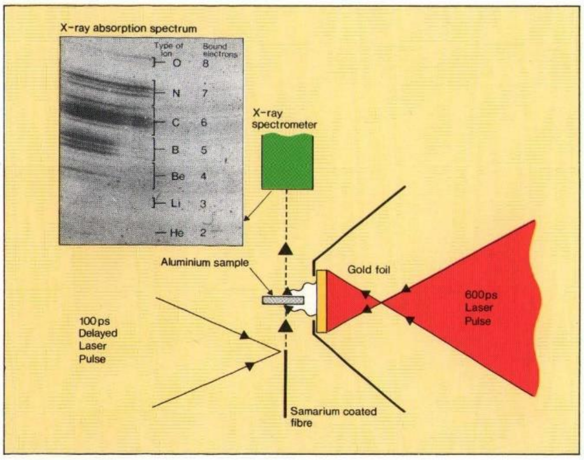

Figure 6: Measurement of the absorption spectrum of an aluminium plasma. The aluminium plasma is produced by thermal X-radiation from gold foil heated by the Vulcan laser. The X-rays used to measure the absorption spectrum of the aluminium plasma come from the tip of a samarium coated fibre heated by another beam from Vulcan. Clusters of absorption lines are due to the various stages of ionisation of the aluminium atoms in the plasma. (Queens University, Belfast.)

In biological research, good progress is being made in an ambitious study of *in vivo* DNA damage repair mechanisms by Birmingham University, Sussex University and Medical Research Council radiological unit collaboration using a radioactively labelled 'caged' repair inhibitor. Live mammalian cells permeated with this material in its inert, caged form are irradiated with a pulse of ultraviolet light of 248 nm wavelength which damages the cellular DNA but has no effect on the caged compound. Later a pulse of light of 351 nm wavelength breaks the cage to activate the repair inhibitor, and a subsequent radio-assay measures the uptake of the inhibitor and thus the amount of unrepaired damage. These are the first studies to be made of DNA repair with a time resolution better than 15 minutes. On a much shorter timescale, picosecond measurements of the first events in photosynthesis are in progress.

Research in physics has exploited the high peak (3×10^{15} W cm^{-2}) intensities produced by focused laser beams with sub-picosecond pulse lengths which result in electric field strengths comparable with the binding fields of atoms and molecules. A novel fast computing technique has been developed by physicists from Reading University to obtain the first ion-ion correlation data in multi photon physics.

Research with small-scale lasers also covers the fields of non-linear optics, materials processing and plasma X-ray generation. Figure 7 shows a polycrystalline film of high T_c superconductor prepared using excimer laser ablation of bulk material which has also given the first epitaxially grown thin film of the important non-linear optical material bariam titanate.

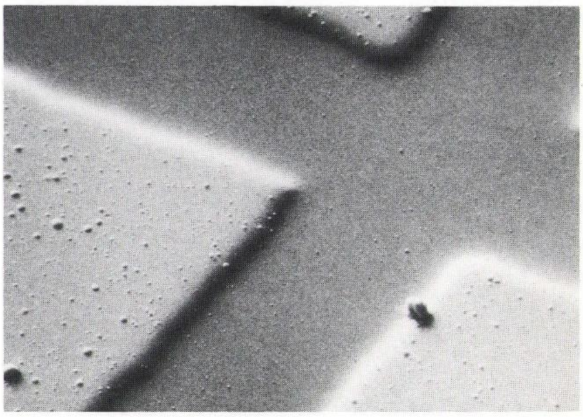

Figure 7: Patterned polycrystalline film of high Tc YBCO. These films were deposited by excimer laser ablation from bulk material on to heated subtrates.