

Free-electron lasers

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Free-electron lasers (FELs) are lasers that use an electron beam from an accelerator to produce widely tunable, high power, ultrafast pulses of coherent radiation. FELs are today important sources of infrared and far-infrared radiation around the world. Quasi-CW high-power FELs are also operational. FELs that operate on the self-amplified spontaneous emission principle are leading candidates for X-ray lasers and fourth-generation light sources. We discuss the physics, technology, advantages and applications of FELs, and explore the frontiers of X-ray and high-power FELs. We also present details of plans for a compact, ultrafast, terahertz free-electron laser in India.

LASERS are ubiquitous sources of coherent radiation over a wide portion of the electromagnetic spectrum, from the infrared (around 10 μm) down to the ultraviolet (around 200 nm). However, in the far-infrared or terahertz part of the spectrum (around 30 μm to 1 mm), or at X-ray wavelengths (less than 10 nm), conventional lasers are not easily available. There is therefore considerable interest in alternate sources of intense, ultrafast, coherent radiation in these two portions of the electromagnetic spectrum.

The free-electron laser (FEL) is a new kind of laser in which the electrons are not bound in atomic or molecular systems. The source of these 'free' electrons is an electron accelerator, such as a linac or a synchrotron. It is therefore a classical rather than a quantum-mechanical laser. In essence, the free-electron laser converts part of the kinetic energy of the electrons into coherent electromagnetic radiation. This conversion takes place in the presence of a static magnetic field that is produced by a magnetic device called an undulator. FELs can produce continuously and widely tunable coherent radiation in any part of the electromagnetic spectrum, femtosecond pulses can be produced at any wavelength, and the laser intensity can be very high.

The history of FELs dates back to as early as 1951, when Hans Motz at Stanford¹ proposed the concept of the undulator, on which the device is based. The FEL in its present form was proposed by John Madey² at Stanford in 1970. The first experimental demonstration of an FEL was done by his group in 1976, when it was shown, using an electron beam from a linear accelerator (linac), that

the device could amplify radiation from a CO₂ laser³. A year later the same group successfully operated the FEL as an oscillator, at the same wavelength⁴. The first visible operation of an FEL, which was also the first storage-ring FEL, produced red light in 1982 at Orsay, France on the ACO storage ring⁵. FELs were now seen as a path towards short wavelength lasers, and many groups around the world started working on them. Meanwhile, the high-power potential of FELs was also appreciated, and FEL was identified as one component of the Strategic Defence Initiative in the US, during which programme a peak power of 2 GW in 15 ns bursts (30 J energy) was achieved. Presently, the shortest wavelength FEL lases in DESY⁶, Germany, at a wavelength of 80 nm. The highest average-power FEL operates at TJNAF, USA, with an average power of 10 kW in the IR – which is the highest average power in the world for any kind of picosecond laser.

These rapid advances over the last decade, enabled by important advances in accelerator technology, have evoked great interest in FELs, and established them as important sources of coherent radiation for the future – whether for X-ray FELs, or the next (fourth) generation of light sources, or high-power applications in industry and defence. Existing FEL facilities, mainly in the infrared and the terahertz, have already established themselves as excellent sources of coherent, widely tunable, short-pulse, high-power radiation.

Basic principle

Figure 1 shows a schematic of an FEL. The three major components of the device are: electron accelerator, undulator and resonator cavity. An electron accelerator can produce a beam of relativistic electrons with energy ranging from MeVs to GeVs, and peak currents ranging from few hundreds of milliamp to few tens of kiloamp. This electron beam passes through a magnetic device called an undulator, which produces a transverse magnetic field

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(i.e. perpendicular to the direction along which electron beam is injected) that is static in time, but varies sinusoidally in space. The amplitude of this magnetic field (B_u) is typically a few kilogauss, and the period (I_u) is typically a few centimetres.

If the FEL has to run as an oscillator, there has to be a resonator cavity as shown in Figure 1, which provides the positive feedback. One usually uses a Fabry–Perot resonator with mirrors at both ends. Radiation is typically coupled out through a hole in one of the mirrors or sometimes by putting a partially reflecting mirror in the cavity. When the FEL is to be operated as an amplifier, the resonator cavity is not there, but seed radiation has to be provided. One of the most exciting things about FELs is the ability to generate high-power coherent radiation with

neither a resonator cavity nor any seed radiation – the self amplified spontaneous emission (SASE) principle. SASE FELs are now seen as the most promising candidates for X-ray lasers.

Spontaneous emission

The heart of the FEL is the undulator (Figure 2) which produces a magnetic field that is static in time, but varies sinusoidally in space:

$$B_u = \hat{y}B_u \cos(k_u z),$$

where B_u is the peak value of the magnetic field, $k_u = 2\pi/I_u$, and I_u is the period of the undulator. When a high energy electron beam from an electron accelerator, moving along the z -axis, is injected into this undulator, the electrons experience the Lorentz force due to the transverse magnetic field, and start oscillating in the x - z plane. Like any oscillating charge moving with a relativistic speed, the electrons radiate in the forward direction in a narrow cone with a semi-angle of $\sim 1/g$, where g is the energy of the electron in units of its rest mass energy, which is typical of bremsstrahlung radiation.

The frequency of radiation can be easily calculated. In the rest frame of the electron, the undulator rushes towards it with a speed v_{z0} ($\sim c$), where v_{z0} is the z -component of the mean velocity of the electron beam. In this frame, the undulator has a period I_u/g_{z0} , where g_{z0} is the Lorentz factor corresponding to v_{z0} . The magnetic field of the undulator is transformed into an electromagnetic wave of wavelength I_u/g_{z0} . The electron oscillates in the electric field of this wave, and, classically, radiates at the same wavelength I_u/g_{z0} (assuming $b_{z0} \sim 1$). Transforming back to the laboratory frame, the radiation frequency is Doppler up-shifted by a factor of $2g_{z0}$, so that the wavelength of the emitted radiation is $I_R = I_u/2g_{z0}^2$. Here g_{z0} represents only the ‘longitudinal’ kinetic energy of the particle. One would prefer to write the expression in terms of g that represents the total kinetic energy of the particle. A simple Hamiltonian analysis shows that $g^2 = g_{z0}^2 (1 + K^2/2)$, where K , called the undulator parameter, is given by $K = eB_u I_u / 2\pi mc$ – it is simply the dimensionless vector potential of the undulator field. Then the wavelength of the emitted radiation can finally be written as,

$$I_R = \frac{I_u}{2g^2} \left(1 + \frac{K^2}{2} \right),$$

which is called the resonance equation. Typically, the longitudinal distribution of the electrons is random within a distance I_R . The radiation from individual electrons therefore adds incoherently and is termed as spontaneous emission. The electrons radiate only for a finite time, since the length of the undulator is finite, and so the

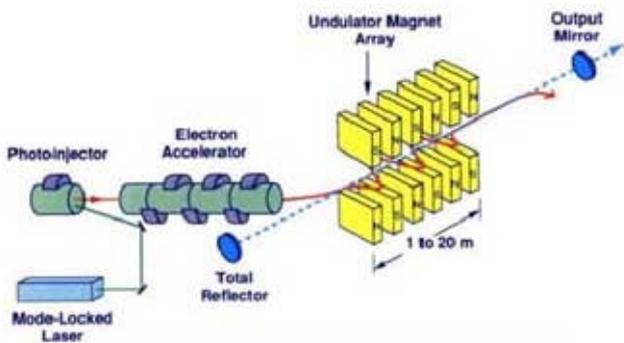


Figure 1. Schematic of a free-electron laser (FEL), showing the three main components: the electron accelerator that produces the electrons; the undulator that produces a static, spatially sinusoidal magnetic field, and the mirrors that provide positive feedback. Not all FELs require mirrors.



Figure 2. Photograph of a typical pure permanent magnet undulator. This one was developed at the Beam Physics and FEL Laboratory, for the CUTE-FEL. Portions in blue and red are support structure for the undulator. The magnets themselves are mounted in the steel ‘jaw’ at the centre in two arrays. The top surface of the magnets is clearly visible.

radiation spectrum develops a Fourier-transformed linewidth, given by $1/N_u$ (where N_u is the number of undulator periods).

It is interesting to note that an electron radiates even at higher harmonics in the undulator. This means that an electron beam in an undulator is essentially a nonlinear medium, and one can make use of this property of the medium for producing shorter wavelengths – as one does in conventional lasers.

Stimulated emission

As the electron beam travels down the undulator, it gets bunched as a result of interaction with the undulator magnetic field and the co-propagating radiation beam of frequency ω_L (which could be the spontaneous emission discussed above, or external seed radiation). This bunching can be explained in terms of the longitudinal Lorentz force arising due to the coupling between the transverse velocity v_x of the electron, arising from the interaction with the undulator magnetic field, and the oscillating magnetic field B_L of the radiation beam. This force is known as the ponderomotive force. For a plane electromagnetic wave, $B_L = \hat{y}B_L \cos(k_L z - \omega_L t + \phi_L)$, and v_x can be shown to vary as $\sin(k_u z)$. Then the magnitude of the ponderomotive force $F_z (= e v_x B_L)$ is given by

$$F_z \propto \begin{bmatrix} -\sin(k_L z + k_u z - \omega_L t + \phi_L) \\ +\sin(k_L z - k_u z - \omega_L t + \phi_L) \end{bmatrix}$$

The term $(k_L z + k_u z - \omega_L t)$ plays an important role in the study of the interaction between the electron and the radiation, and is called the ponderomotive phase, ψ . For $\omega_L \sim \omega_R$, i.e. near resonance, the z -component of the electron velocity is $\omega_L/(k_L + k_u)$, as can be derived from the resonance condition. Consequently, the argument of the first sine term is slowly-varying as the electron moves. On the other hand, the argument of the second sine term oscillates rapidly between +1 and -1, and its contribution averages to zero. Different electrons see different phases ψ and therefore experience the ponderomotive force with different magnitudes and directions, and this leads to bunching of the electrons (Figure 3). The distance between successive bunchlets is $2\pi/(k_L + k_u)$, which is approxima-

tely λ_L . All the electrons in a bunchlet radiate at the same phase, and successive bunchlets radiate with a phase difference of 2π . Hence this radiation develops coherence, and is termed as stimulated emission.

To summarize, the FEL mechanism involves three steps:

- (i) the electrons oscillate inside the undulator and emit spontaneous emission;
- (ii) this radiation acts back on the undulating electrons and bunches them;
- (iii) the bunched electrons emit coherent stimulated radiation and amplify the co-propagating electromagnetic wave.

Technology issues

The FEL is a laser driven by an accelerator, and the technology of an FEL is dominated by the technology of the accelerator driving it. The other important component is the undulator. While it would not be possible for us to delve deep into the technology of accelerators and undulators here, a few points need to be emphasized.

It is important to realize that successful operation of an FEL imposes several challenges on the quality of the electron beam and the undulator magnetic field. The electron beam should have an rms energy spread typically less than $1/2N_u$ (say 1% for a typical value of $N_u = 50$) in order to avoid gain degradation of the FEL due to inhomogeneous broadening. The rms emittance of the electron beam should be less than the radiation wavelength for maintaining good overlap between the radiation beam and the electron beam throughout the length of the undulator. The gain in an FEL depends on the peak electron beam current, which should be sufficiently high (typically in amperes) for the FEL to lase. The rms error in the peak undulator magnetic field should be typically less than at least 1% to avoid deterioration in the gain. The undulator field quality should be such that it does not produce significant wander in the electron beam trajectory, resulting in poor overlap between the radiation and the electron beam. All these tolerances become more demanding at shorter wavelengths.

Details of the physics of FELs and these technological issues can be found in a number of textbooks on the subject, including those by Marshall⁷ and Brau⁸.

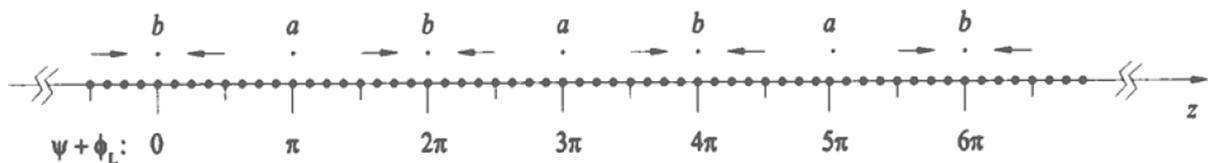


Figure 3. Schematic of bunching in an FEL. Electrons shown by solid dots and arrows show the direction of the force acting on the electrons having a particular phase. The electrons get bunched at location ‘b’ and anti-bunched at location ‘a’.

Advantages of FELs

FELs have a number of important advantages over conventional lasers and synchrotron-based light sources.

Wide tunability

As shown by the resonance equation, the wavelength of the FEL radiation can be continuously tuned by even an order of magnitude by varying either the magnetic field B_u , or the electron energy g . There are many operating FELs around the world that provide this kind of very broadly tunable radiation in the infrared and ultraviolet. For example, the FOM FELIX FEL in the Netherlands provides radiation tunable between 5 and 30 μm . FEL facilities that use a single accelerator and multiple undulators can provide radiation over a truly staggering spectrum. For example, the FELI facility in Japan, using multiple undulators, each tuned to a different range, delivers radiation from 230 nm to 100 μm , covering continuously the entire spectrum from UV to FIR!

Short pulses

In conventional lasers, it is typically not possible to produce ultra-short pulses at all wavelengths, because mode-locking can be achieved only in lasers with a broad emission spectrum – for example, picosecond pulses cannot be achieved in He–Ne lasers at 632 nm. Similarly, synchrotron light sources do not produce very short pulses. With FELs, one can obtain ultra-short pulses at all wavelengths because the structure of the radiation pulse mimics that of the electron pulse. Picosecond electron pulses are routinely produced in S-band linacs, and femtosecond pulses are not difficult to produce.

High brightness

FELs can produce, both in the infrared and in X-ray regimes, radiation whose brightness (both average and peak) is several orders of magnitude higher than the best that can be achieved with synchrotron-based light sources. The best numbers, in the X-ray regime, for present non-FEL sources are: average brightness of 10^{20} photons/s/ $\text{mm}^2/\text{mrad}^2/0.1\%$ bandwidth, and peak brightness of 10^{23} . SASE FEL-based fourth-generation light sources are expected to produce an average brightness of 10^{26} , and peak brightness of 10^{34} ! In part, this is because of the high-brightness electron beams that can be generated in linacs, using the modern technology of photocathode guns, which enable a more efficient conversion of the kinetic energy of the electrons to radiation. Similarly, in the infrared, FELs can provide brightness that is 3–4 orders of magnitude higher than synchrotron radiation sources or conventional laboratory sources⁹.

Short wavelength

With the new principle of SASE in FELs, which requires neither mirrors nor coherent seeds, there is the possibility of generating high-power, short-pulse, tunable, X-ray lasers. Such SASE FELs would produce radiation well beyond the capabilities of existing radiation sources, and have therefore been identified as fourth-generation light-sources.

High power

High-power operation of conventional lasers has drawbacks related to heating and breakdown of the laser medium, and single-shot operation. With FELs there is in principle no fundamental limit to the operating power, because the lasing medium (the electron beam) exits the interaction volume at close to the speed of light, and electrons, being point particles, cannot breakdown. In addition, long-duration, quasi-CW operation is possible – unlike, for example, chemical oxygen iodine lasers (COIL), that can deliver a high-power pulse only around once in ten minutes.

Applications of FELs

FELs have applications in a wide variety of fields due to their special properties of wide tunability, high power, ultra-short pulses and high brightness. The wide tunability of FELs enables them to fill the gaps in the electromagnetic spectrum left by conventional lasers and other conventional sources – these are in the far-infrared and X-ray. The capability of ultra-short pulses and high peak power enables applications even in the IR and visible, wherever short-pulse lasers are not always available. The high brightness enables a wide variety of applications in spectroscopy and related fields. There are around fifty operating FELs around the world, and seventeen more are being built. Over ten are operating as user facilities, mainly in the infrared (Table 1). More details of applications can be found on the websites of FEL facilities listed in Table 1. Details of the compact, ultrafast, terahertz free-electron laser (CUTE-FEL) being planned in India, are given in the Appendix.

We discuss here in detail applications in condensed-matter physics, chemistry, biology and medicine. Applications to fundamental physics are discussed in the section on X-ray FELs, and defence applications in the section on high-power FELs.

Condensed-matter physics

There are a variety of applications in condensed-matter physics, occurring in the infrared part of the spectrum,

Table 1. Some of the major FEL facilities around the world

Location	Wavelength (μm)	Peak/average power	Application
Stanford University, USA	3–60	1.2 MW 1 W	Condensed matter physics (CMP); biology; medicine
University of California, Santa Barbara, USA	30–2500	– 10 kW (CW)	CMP; semiconductors
Duke University, USA	2–10	2 MW 3W	CMP
Vanderbilt University, USA	2–8	3 MW 6 W	Medicine; biology; CMP
Los Alamos National Laboratory, USA	4–6	10 MW 1.5 W	Medicine
FOM Institute of Plasma Physics, The Netherlands	5–110	5 MW 0.5 W	Nuclear/molecular physics; biology; medicine
Laboratoire pour l'utilisation du Rayonnement Electromagnetique, France	2–18	10 MW 9 W	CMP
Free-Electron Laser Institute, Japan	0.2–100	10 MW 2 W	CMP; medicine; isotope separation; biology; materials

that use the wide tunability of FELs¹⁰. One can perform the spectroscopy of various materials, particularly semiconductors and superconductors, with far-infrared radiation. Owing to the high-peak power of FELs¹¹, one can also perform nonlinear spectroscopy, which is not easily possible at these wavelengths using conventional lasers. The short-pulse nature allows transient spectroscopy of electron–phonon and electron–electron coupling in semiconductors.

For example, the FEL at Duke University makes use of the high peak power and broad tunability of FELs, to perform nonlinear spectroscopic studies of semiconductors, quantum-wells, polymers, etc. These studies are useful in understanding the properties of matter and also in developing models for predicting the different properties. At the CLIO FEL and also at the Stanford FEL, the short-pulse (picosecond) nature of the radiation from an IR/FEL is used in the study of electron relaxation time and intraband carrier dynamics in semiconductor samples. At the Vanderbilt University, the tunable radiation from the IR/FEL is used to perform studies on band discontinuities at semiconductor interfaces using the internal photoemission technique. In this technique, photons from the FEL are used to pump electrons across the energy barrier caused by a heterogeneous band discontinuity and the barrier height is derived directly from the spectral position of the threshold. Another interesting application of the Vanderbilt FEL has been in performing two-photon spectroscopy of direct as well as indirect band-gap semiconductors using photoconductivity, photoluminescence and absorption measurements.

Applications to materials: Interfaces of semiconductor crystal hetero-junctions, viz. ZnSe/GaAs, ZnMgSse/GaAs, AlGaAs/GaAs, CdS/CdTe, InAs/AlSb are of great interest from the point of view of applications in optoelectronics. There are type-I and type-II quantum wells, which

have different types of band discontinuities. The band discontinuities of the band structure are measured using the FEL internal photo-emissions technique. One end of the crystal is grounded and the energy of the excitation photon is varied. The generated photocurrent goes as the square of the energy difference between the photon energy and the conduction band excess. Band discontinuities at hetero-junctions can be determined using the appropriate wavelength radiation, like the one generated by the FEL in the far-infrared (FIR). THz-FELs become important for closely spaced band-discontinuities. These problems are not yet attempted due to lack of FELs in the appropriate range.

Additionally, internal structure of the magneto-excitons in GaAs/AlGaAs quantum wells can be probed using the intense THz radiation. The idea is to excite the carriers within the sub-bands with the help of highly intense THz radiation. An internal excitation leads to changes in the photo-luminescence, which is monitored under the influence of a magnetic field. The nonlinear interaction between the exciton and the exciting visible/infrared photon leads to the absorption and emission of the THz radiation, creating optical sidebands. In closely spaced energy levels, one can study the dynamics and new types of nonlinear phenomena using intense THz radiation. The closely spaced energy levels are useful if light modulation is required at THz frequency. Interband light modulation is carried out using the inter-subband transitions within the quantum well, stimulated by the use of FEL radiation of the appropriate photon energy. The interband transition is carried out between conduction and valence bands of the quantum well, with the appropriate energy photon. The quantum well width is such that the two levels are tuned to the FEL radiation frequency (or vice versa) to make an inter-subband transition within the subband levels. The light causing interband transition can be modulated using this procedure. Using the coupled wells, the FEL-THz

radiation is used to excite the carriers in the subbands of the first quantum well and if these upper levels are resonant with the level of the adjacent quantum well, then the THz photon absorption/emission induced tunnelling can occur. This is useful in the study of the various coupled quantum well devices.

Future applications: Another important use of FELs is in the annealing process of materials. Direct excitation of the lattice vibration by FELs is called FEL-annealing. FEL radiation tuned exactly to the vibrational level can induce resonant vibration, leading to the reconstruction or re-crystallization of the solid. This is used in the case of disordered atoms that can bind the free atoms at room temperature. Diamond-like structures or ion-implanted materials can be annealed or re-structured using intense FEL radiation. Fabrication of silicon nano-structures has been done using IR/FEL. The THz-FEL offers novel applications, where specific molecule vibrational/rotational modes of the bonds can be excited so as to restructure a particular design.

Photocurrent dynamics on single quantum well infrared photo detector (SQWIP) has been studied using mid-IR FELs. High intensity FEL leads to the inter-subband transitions in SQWIP, which are used for the investigation of its photocurrent. To make these devices in the THz region, it would be important to study them under the THz-FEL radiation. Carrying out the ionization of the excitons and studying their formation and dissociation dynamics, is currently a topic of intense study. Further topics of interest include the study of low frequency protein motions, phonons and superconducting band-gaps. With the advent of intense femtosecond THz radiation, new problems involving the electronic (electron–electron, electron–hole, etc.) scattering and collective electronic excitations like plasmons, polaritons, excitons, charge density waves, etc. can be studied. Also, the study of intense THz radiation on spintronic devices is not yet done. This also offers a new area of research. Initial studies with intense THz radiation have shown many interesting and unexplained phenomena not seen before, e.g. luminescence from ZnSe excited by ps-FEL pulses in the mid-infrared shows blue-band edge emission, etc.

Chemistry

Applications of FELs in chemistry are envisioned for both scientific research and industrial production. The status and prospects for scientific use were evaluated by the Solid State Sciences Committee of the National Research Council quite some time back¹², where it was concluded that chemistry and surface chemistry would be areas that would particularly benefit from FEL advances. The most important spectral region for chemical applications is the vibrational infrared region (3–30 μm), although many in-

teresting applications involve the near infrared (1–3 μm) as well.

Radiation from an IR/FEL can be used to manipulate molecular vibrations and chemical reactions in both the condensed and the gas phase. The infrared pulses will make possible new studies on intramolecular vibrational energy transfer by means of infrared multiple photon excitation and dissociation (IRMPE and IRMPD). With broadly tunable IR/FELs, the technique can be extended to practically all molecular species. The pump-probe type experiments involving IR/FEL and a second light source (which can be another FEL, a conventional laser or a synchrotron light source), will allow quantitative studies of many surface phenomena, such as time-dependent redistribution of surface molecules, diffusion, desorption and so on.

In the following, we discuss some of the application areas of FEL in IRMPE/IRMPD and isotope separation processes. We also examine infrared spectroscopy and dynamics of some interesting systems in condensed/gas phase and on surfaces as well. Finally, we deal with various aspects of the industrial applications of FELs, which have been advocated for this promising laser source¹³.

Photochemistry: A large body of experimental and theoretical work on IRMPD has been accomplished with CO₂ lasers. A substantial portion of the work is concerned with isotope-specific multi-photon dissociation leading to isotopic enrichment in the gas phase of small molecules for S, O, C, B, T, D, etc. The most extensive work has been done on carbon isotopes using single and multiple-wavelengths, and also in single- and two-component mixtures^{14–16}.

While the CO₂ laser frequencies are suitable for many multi-photon dissociation reactions, there are important fundamental vibrational frequencies such as the O–H and C–H stretch in the 2.5–3.5 μm regions and C=O and N=O stretch in the 5.5–6.5 μm region, which can be excited directly at these wavelengths. The spectral dependence of yield and selectivity by exciting different vibrational modes of the same molecule, can throw new insight into the multi-photon dissociation process. Excitation of weakly anharmonically coupled modes with the other normal vibrations of the molecule can possibly lead to mode-or bond-selective chemistry.

The FEL is now well established as a continuously tunable source of radiation throughout the infrared, permitting investigation over a broad range of photon energies using a single source. The pulse energy and power are adequate to secure reasonable yields. The FEL also offers a unique opportunity for delivering two-colour photons in the required time synchronization, by supplying electrons from the same accelerator to two appropriately designed undulators in sequence. FELs can also be used for research, including unusually high instantaneous peak power for the exploration of the Stark effect and

power broadening, and to produce coherent, phase-locked trains of transform-limited or chirped pico-second pulses.

Spectroscopy and dynamics: Traditionally, molecular vibrational spectroscopy is studied by analysing the amount of light absorbed as a function of infrared frequency after passing through either a solid, liquid or gas-phase sample. A serious limitation of this approach is its inherent low sensitivity and its lack of selectivity. In the ultraviolet and visible regions, well-established techniques exist, such as resonance enhanced multi-photon ionization and laser induced fluorescence, which combine a high sensitivity with species selectivity. In a complementary experiment, mass-selective gas-phase infrared spectroscopy was demonstrated using pulsed high-power FEL radiation (19.2 μm macropulse of 5 μs duration having 50 mJ energy) from the FELIX FEL in the Netherlands. Gas-phase C_{60} molecules are resonantly excited by FELIX to very high internal energies by absorption of several hundred infrared photons per molecule, where they undergo auto-ionization¹⁷. The surprising observation of C_{60}^+ formation (35–40 eV) meant 600 photons were absorbed at 19.2 μm . This observation could be explained on the basis of intramolecular vibrational energy redistribution. Therefore, scanning the FEL and recording the mass-selected ion yield provided the absorption spectrum, and it resembled the infrared spectrum of C_{60} thin-film. The observed four peaks correspond to the four well-known infrared allowed F_{1u} fundamental modes of icosahedral C_{60} . The sensitivity of the present technique is many orders of magnitude better than conventional absorption spectroscopy and this procedure can comfortably be adopted for many more systems.

The excellent properties of the FEL can be exploited to induce some biological level interaction between photons and living tissues. The average power of the FEL is sufficient for most thermal coagulation therapy and photodynamic therapy, which is a light-activated chemotherapy for cancer and other indications. In advanced atherom atherosclerosis, a large amount of lipids, particularly cholesterol esters, accumulates on the arterial wall. Recently, an IR/FEL was employed, tuned to 5.75 μm , corresponding to the stretching vibrations of ester bonds, to remove them by direct dissociation of cholesteryl oleate. Interesting results were obtained for wavelength-sensitive removal process and such photodynamic processes are of great importance for biomedical applications¹⁸.

The use of IR/FEL allows adsorbate vibrational spectroscopy by infrared-visible sum-frequency generation (SFG) beyond the frequency range accessible by bench-top infrared lasers. SFG relies on the second-order nonlinear surface polarizability. It depends directly on the overlapping infrared and visible laser pulse energy and inversely on the pulse duration and irradiated area. It is resonantly enhanced when the infrared laser is tuned in resonance with an adsorbate vibration that is both infrared and

Raman-active. The feasibility of this technique¹⁹ is demonstrated by recording the SFG vibrational spectrum near 5 μm of the CO adsorbate on a Pt surface, resulting either from direct CO adsorption from an electrolyte saturated with CO or from electrochemical decomposition of methanol on Pt/HClO₄. This system is of particular interest to the prospect of fuel cell development.

The apparatus consists of a synchronized and frequency-doubled mode-locked YAG laser (green 532 nm, 200 ps, 100 mW) with the CLIO FEL, which is tunable from 2 to 17.5 μm . The SFG spectrum of CO/Pt interface following the adsorption of CO from the CO-saturated electrolyte showed a sharp resonance at about 2030 cm^{-1} , which corresponds to linearly bound CO. However, the corresponding spectrum of CO from the methanol-dissociative adsorption on the Pt electrode clearly showed two resonances at 2053 and 1977 cm^{-1} , indicating that linearly and doubly-bounded species coexisted in the system.

Future industrial applications: The high power, widely tunable spectral range and quasi-CW operation of free-electron lasers makes them intrinsically well suited to large-scale material/chemical processing¹⁶. Two important processes have been identified: (i) to purify chemical compounds by removing a few impurity molecules from a large number of desired molecules, and (ii) photochemical processes having high quantum yield like laser-initiated chain reactions. One of the most promising opportunities for the FEL is micro-fabrication of semiconductor circuits. By tuning the laser wavelength and/or varying the composition of the gas or liquid, it should be possible to construct various features of the microcircuit. Another interesting result is the observation of significantly enhanced material removal from polymers when certain vibrational modes within the polymer were excited with tuned infrared radiation. Other material studies that have been investigated include ablation of carbon materials, vaporization of fused silica, texturing and micromachining polymers and metals and the production of single-wall carbon nanotubes^{20–22} (A. Loper *et al.*, unpublished).

Isotope separation: Nuclear reactor technology is another promising field for the application of FELs²³. FEL features are well suited for obtaining enriched fuel, reprocessing of spent nuclear fuel, and synthesizing new strategic materials. Such materials are required not only to improve the neutron economy, but also to suppress the activation and disintegration of materials due to α -particle decay. The interesting materials with small neutron absorption cross-sections are: 90-Zr, 50-Ti, 53-Cr, 56, 57, 58-Fe and 96, 97-Mo. On the other hand, the isotopes 157, 155-Gd, which are added as a burnable poison, have unusually large absorption cross-sections for thermal neutrons. Depending on the isotope shift of the elements, either atomic vapour laser isotope separation or molecular laser isotope separation process could be employed.

Therefore, from this brief account of FEL applications in chemistry, it is apparent that significant advances in our fundamental knowledge of photon–molecule interaction and also in the technology of lasers have been made. The application of this new knowledge and technology to more general areas in chemical processing like selective chemical synthesis, purification, and dynamics has a great future. Opportunities in this field of study are enormous and lots of exciting questions are at hand. If they can be answered, selective chemistry with FELs might revolutionize the world of science.

Biology

A coherent, high-brightness light source that can operate from the far infrared to X-ray wavelengths, and can provide picosecond pulses at all these wavelengths, appears to be the embodiment of a combined wish-list of a whole generation of biophysicists. Light at the shorter wavelengths promises to elucidate the structures of biomolecules. It should help structural genomics leapfrog the yawning gap between what we know of the human genome, thanks to the human genome project, and what we know about the protein structures that these genes code. Light at longer wavelengths promises to improve our even more abysmal knowledge of the dynamics of these structures. Light in the FIR region probes the realm of the collective motion of the whole, or of substantial domains of proteins. All biology finally stems from the dynamic interaction of the structures that the cells make, and any machine that boasts of the power to elucidate both structure and dynamics of biomolecules will surely grab the attention of the whole of the biological community. We have already described that an FEL, in principle, can be such an ideal light source. Here we discuss some of the promises that FELs (especially the FIR FELs) hold for biology.

Protein dynamics: FELs in the FIR – Information contained in the vibrational spectrum: Protein sequence, and even the crystal structure obtained from X-ray diffraction, give a static depiction of the protein structure. In a living cell, the action of the protein depends largely on its ability to be flexible. Stability of a globular protein structure is only a few kcal/mol, and this limited stability engenders a structural flexibility that has been optimized over billions of years of evolution. In key functions such as ligand binding, catalysis of a substrate from one form to another, or in signal transduction, protein conformational changes are key factors in achieving the function. These conformational changes typically involve a large fraction of the atoms in a protein. The coordinates that change (the reaction coordinates) are a combination of the vibrational degrees of freedom available to these atoms. These vibrational modes are typically of rather low frequency,

as they involve a large mass movement. Thus the low-frequency vibrational modes of the proteins and the time-scale of their relaxation as the protein goes from one state to another, are of paramount interest.

What it is possible to see at FIR frequencies: What is the appropriate frequency range for investigating protein motion? The answer to this question depends on the types of motion we are interested in. Protein vibrational spectrum spans from the OH stretches at $\sim 3500\text{ cm}^{-1}$ down to the domain motions that occur at a few wavenumbers. The local changes involving the deformation of the secondary structures are in the mid infrared. The larger-scale motions are more important in understanding the phenomena that involve the movements of large portions of the protein. For example, signal transduction may involve dimerization, enzymatic activity may involve an induced fit, allosteric effects may be mediated by conformational changes of specific domains, charge transport may involve coordinated changes in the conformation, ion channelling may involve the large-scale twist of trans-membrane helices, protein translocation may involve partial folding and unfolding, chaperoning may involve the assembly and dis-assembly of sub-units and transcription may involve the conformational changes involved with DNA binding. Thus a large fraction of proteins of interest undergo large-scale motions in order to carry out their functions. In fact, the list should not stop with proteins. The DNA molecule has a sequence-dependent flexibility, which may be the key to controlling which gene is expressed at which stage. This flexibility can be measured by recording the FIR spectra of DNA fragments with different sequences. In fact, predicting the sequence features from the infrared spectrum is also a possibility that is being investigated with FELs. It is perhaps fair to say that there is no set of processes in a living cell whose core mechanisms cannot be investigated fruitfully with an IR/FEL.

Time-resolved spectra and the conformational energy landscape: While the steady-state FIR spectra are informative and easier to collect, it is the time-resolved FIR spectra that carry information about the dynamics. The relaxation of the vibrational excitations tells us about energy flow through the protein. If two conformations were separated by an energy barrier, then the timescale of transformation between these two states would have its signature on the relaxation times of the vibrational modes that are coupled to the reaction coordinate. This not only helps identify the relevant reaction coordinates, but it also puts an upper bound on the fastest possible conformational transition.

Light-driven proteins – pump probe studies: In favourable cases, protein transformation can be driven by a pulse of visible light. Then the evolution can be followed with a pulsed FIR probe (which is time-synchronized to this

visible 'pump' pulse) that measures the time-dependent changes in the FIR spectrum. Calculations of the normal modes of proteins will provide a backdrop for understanding these changes. For many proteins, light-driven conformational changes are the essence of their function. For example, the atomic-level structures of eight different states of the light-driven proton pumping molecule, bacteriorhodopsin, have been elucidated but the final understanding of how the protein conformation changes from any one of these states to the next, must come from time-resolved spectroscopy. Probing the dynamics in the visible domain has provided information primarily about the changes taking place in the chromophore. How the protein itself is involved in this process has to be answered at the infrared wavelengths that are sensitive to the dynamics of the protein structure itself.

Proteins without a light trigger: Bacteriorhodopsin is but one example, and many other extensively investigated proteins that exhibit light-driven changes, such as myoglobin and haemoglobin, the photosynthetic reaction centre, and the family of fluorescent proteins, are all waiting to be understood with this tool. However, most proteins are not light-driven yet their conformational dynamics is just as interesting and just as important from a functional point of view. One needs to impart an appropriately short impulse, and the time-resolved dynamics could then be studied with the FIR pulses provided by an FEL. There are various ways to achieve this impulse. For example, the first steps towards the folding of a protein can be studied if a light pulse is used to initiate the unfolding process, say by breaking a protein–ligand bond or by changing the pH of the solution. The low-frequency vibrational modes that evolve first may point towards the domains, hinges and other structures that form initially during folding. The conformational changes required for the binding or unbinding of a substrate can be investigated likewise. The vibrational signatures of the intermediate states of a functioning protein, and the rates of transitions between them, should provide an understanding of the conformational free energy landscape that governs protein motion.

FELs at other wavelengths

FELs in other spectral ranges are also useful devices for biological investigations. Ultimately, we would like to know the structural changes at the atomic level, with a time resolution that can resolve the motion. This can be achieved by starting the conformational transition with a femtosecond light pulse, and subsequently probing it with a coherent X-ray pulse at the appropriate time to obtain a single-shot diffraction pattern that is good enough to elucidate the structure at an atomic level. Resolving the structural fluctuations at an atomic level in the picosecond

timescale promises to finally bring the experimental results to the time regime accessible to the Newtonian molecular dynamics simulations.

An interesting aspect of FELs in the soft X-ray frequencies is their potential to perform X-ray microscopy of biological specimens in their native aqueous environment. Water has a transmission window of around 4 nm, and a collimated X-ray source at this wavelength can potentially form images with resolution of the order of a few nanometers.

Medicine

The Vanderbilt FEL facility dedicated to medical applications, has been operating for over a decade now. The tunability of the FEL has been used to perform surgery; initially on animals, but more recently on humans. Photodynamic therapy for cancer treatment can be performed using high peak power in an infrared FEL. In one case, the FEL was successfully used to excise a malignant brain tumour. The advantage of the FEL over conventional lasers was that the wavelength could be tuned precisely to the one at which the malignant cells have the highest absorption cross-section, and focused to a tight spot. In this way the collateral damage to the surrounding healthy cells was minimized and the surgical procedure could be successfully performed – which it could not have been with conventional lasers or other surgical methods. At the FELI FEL in Japan, polishing and surface-hardening of teeth is done using tunable IR radiation.

Frontiers of FELs

The two major frontiers of research on FELs are at short wavelengths and high powers. Here we discuss these two frontiers in some detail.

X-ray FELs

Since photocathode guns are capable of producing bright femtosecond electron pulses, one can generate femtosecond X-ray pulses using an FEL, which, so far, has not been possible using synchrotron radiation sources. One of the greatest challenges, on which the FEL community the world over is working, is to have the FEL deliver high-power coherent radiation in femtosecond pulses at X-ray wavelengths. From the resonance condition, one can calculate that for a typical undulator period of around 25 mm, one can generate coherent X-rays using an electron beam energy of a few GeV. The availability of highly reflecting mirrors at normal incidence is generally a problem at X-ray wavelengths, which means one cannot have an FEL oscillator operating at such wavelengths.

However, using the SASE principle, one can have mirror-less lasing of an FEL, in a single pass, at X-ray wave-

lengths²⁴. Given a sufficiently bright electron beam, one can produce GWs of coherent X-rays using this scheme. Having such a powerful source of coherent X-rays is expected to lead to revolutionary breakthroughs in many areas of science, such as atomic physics, warm dense matter, femtosecond chemistry, imaging/holography of biomolecular system, X-ray fluctuation spectroscopy, etc. (unpublished report, 2000).

X-ray FELs also have potential applications in fundamental physics, such as spontaneous pair creation from vacuum in the presence of an external electric field (the Schwinger mechanism). The advantage of X-ray FELs over high-power optical lasers is that, because of the much shorter wavelength, the diffraction limit is three orders of magnitude lower, allowing the laser to be focused to a much smaller spot-size (of the order of 1 nm). This allows high electric fields (E) and large accelerations. Also, since the adiabaticity parameter ($h = m_e c \omega / eE$) is ~ 1 (for optical lasers, even exawatt lasers, $h \ll 1$), X-ray lasers would probe the adiabatic, non-perturbative, strong-field regime. To have a reasonable rate of pair-production, one would require an electric field $E \sim 10^{17}$ V/m, which is achievable with a TW X-ray FEL with a diffraction-limited spot-size. Other possible applications include the Unruh effect, axion production, etc.²⁵.

The SASE process (Figure 4) is one in which the broad-band incoherent shot-noise from undulating electrons is amplified to high-power coherent radiation, due to the collective instability excited in the electron beam–undulator–radiation system. Successful operation of an X-ray FEL however poses several challenges. In order to achieve such high gain in a single pass, one typically requires the undulator length to be more than 20–30 gain lengths (where one gain length is the length along the undulator over which the radiation power gets e-folded). Typically, the undulator for an X-ray FEL based on the SASE principle is many tens of metres long, and the number of periods (N_u) could be as high as 3000. Building an undulator that produces a sinusoidal magnetic field of high quality (having rms field errors of around 0.1%) over such a long length, is a challenge in itself.

For the electron beam to maintain strong interaction with the co-propagating radiation beam over such a large distance, it needs to be of good quality: small energy-spread ($< 0.1\%$), low emittance (< 0.1 p nm-rad), and high current (\sim kA). The technology to generate such high-brightness electron beams has been available only since the mid-90s, through the development of photocathode rf guns (Figure 5). There are now ambitious projects at the Linac Coherent Light Source at SLAC²⁶, designed to lase at 0.15 nm, and at the TESLA X-ray FEL at DESY, designed to lase⁶ at 0.1 nm. Much preliminary research, prototyping and testing, at UV and longer wavelengths, has been, and continues to be done at these and other laboratories. SASE lasing and saturation have been established at wavelengths down to 80 nm.

High-power FELs

The impetus for the development of FELs came in the 1980s, as a consequence of the Strategic Defence Initiative, more commonly known as the ‘Star Wars’ programme. Recognizing the potential of FELs for high-power operation, there were plans to build FELs that could deliver average power in MWs. These plans were probably premature because the requisite accelerator technology simply did not exist then. Consequently, for a long time, the highest average powers were in watts! Recent technological developments, e.g. photocathode gun technology, superconducting CW RF technology, the technique of energy recovery in the superconducting structures, etc. have removed these bottlenecks.

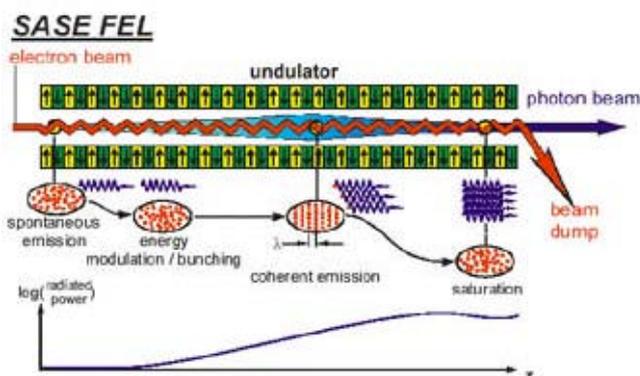


Figure 4. Schematic of a SASE FEL. In the first part of the undulator the electrons emit incoherent shot-noise radiation, which increases in magnitude and starts bunching the electron beam. The bunched beam then emits coherent stimulated radiation, whose intensity increases exponentially and finally saturates. All this happens in a single pass, and therefore requires a very long undulator.



Figure 5. Photograph of a prototype photocathode rf gun structure developed at the Beam Physics and FEL Laboratory, CAT for short-wavelength FEL applications. The final gun will produce 100 A, 8 ps electron bunches and can be upgraded to higher currents and shorter pulse-lengths.

Presently, the highest average power of an operating FEL has been pushed up to around 10 kW at 3.1 μm at the Thomas Jefferson National Accelerator Facility in USA. This is achieved using a photocathode gun source, and a 48 MeV electron beam, with an average current of 5 mA. The key ingredient in the efficient operation of this facility is the use of superconducting RF cavities, along with an energy recovery technique that recovers over 90% of the electron beam power! In the near future upgrades are planned that will extend operation beyond 10 kW average power in the near IR, and kilowatts of power at wavelengths from 0.3 to 30 μm . A similar kW IR/FEL is also being developed at the Japanese Atomic Energy Research Institute.

FELs are also being used for generating high-power long wavelength (mm wave) coherent radiation, for applications like plasma heating in fusion research. At the FOM Plasma Physics Institute in the Netherlands, the FEL has produced 730 kW at 1.5 mm, with an electron beam of 1.8 MeV, for a duration of 10 ms. Efforts are underway to implement energy recovery and produce kilojoules energies in millisecond pulses. Other high-power millimetre (mm) wave projects are on at the University of California at Santa Barbara, the University of Central Florida, Tel Aviv University, and KAERI in Korea.

Defence applications: High-power FELs find an important use in defence, for directed-energy applications. Also, the high power mm-wave FELs mentioned above, could be used for generating high-power microwaves which have important defence applications.

Presently, chemical lasers are used for missile defence, and they can give very high average powers of around a MW. But they have two major disadvantages. First, the wavelengths are fixed: 1.3 μm with COIL, or around 3 μm with DF. These wavelengths may be very slightly tunable, but it is not possible to widely tune them or to get high power at very different wavelengths. The latter could especially be an issue for transmitting high-power laser beams into space, either for missile defence or satellite powering. Second, they are only capable of single-shot operation: lasing for a few tens of seconds, with a down time of over an hour. For example, the MIRACL laser (a 3.8 μm , MW class DF laser operated by the US Navy), has a maximum lase duration of 70 s, and in a decade has operated for only 50 min over 150 lasing tests (an average of 20 s per test). The long recovery time of chemical lasers is a disadvantage for missile defence applications, where it is unlikely that missiles will come at a convenient rate of one an hour! This problem can be addressed by building multiple laser systems, but this would multiply the costs and increase command and control complexity.

FELs can overcome both these disadvantages. FELs can produce high average-powers not in single-shot, but in quasi-CW mode, for long periods of time. The wave-

length at which the power can be delivered by the FEL can be widely tuned with ease (say from 1 to 10 μm). In addition, FELs provide the only source of high-power radiation at wavelengths very different from around 1 μm , and particularly in the UV. Efforts are already underway at the Jefferson Lab, USA, to lase in the UV with an average-power of over a kW, and in the IR with over 10 kW, within the next year.

With the recent results at Jefferson Lab., the US Navy is seriously reconsidering proposals for a MW-level, ship-based FEL for defence against sea-skimming and cruise missiles. The Navy also is concerned about asymmetric threats such as multiple incoming devices, where the danger is not in the sophistication of the threat, but in the depletion of a defensive system's magazine. Here, the CW operation of the FEL 'infinite magazine' would be invaluable.

Conclusion

Internationally, terahertz free-electron lasers have already established themselves as competitive, and relatively inexpensive light sources. Linac-based X-ray FELs have been identified as radiation sources for the future. Their properties of wide tunability, high power and short pulses make them unique as fourth-generation light sources, which are likely, in twenty years, to completely replace synchrotron-based light sources. Their flexibility in terms of wavelength, power and pulse structure exceeds that of conventional lasers. Their capability for long-duration, high average-power operation also makes them important for strategic applications. Internationally, this technology is still maturing. This is therefore the right time for us to leapfrog, through a focused and sustained thrust in FEL technology.

The Beam Physics and FEL Laboratory is working on a CUTE-FEL for wide-ranging applications in physics, chemistry and biology. The successful lasing of this FEL (targeted for 2006) could be a first step towards a more ambitious femtosecond X-ray FEL in India.

Appendix. A CUTE-FEL in India

With the wide spectrum of FEL applications described above, particularly in the far-infrared or terahertz region (30 μm –1 mm), it is of interest to consider the utility of building such FELs in India. Such a FEL would not only serve as a unique tool for experiments in the terahertz region with ultrafast high power pulses, but could also be the first step in a roadmap towards X-ray and high-power FELs for wide-ranging applications in basic and applied science, industry and defence.

In this context we looked into the possibility of building a compact, ultrafast, terahertz FEL (CUTE-FEL) in India. After detailed discussions with potential users

around the country, it seemed that, as a first step, a terahertz FEL that can provide radiation from 50 to 100 μm , with a peak power of the order of 1 MW and pulse-width in picoseconds, would be an interesting and internationally competitive facility. A schematic of such an FEL is shown in Figure 6. Preliminary design studies showed that with an electron accelerator of 10 MeV energy and 20 A peak current, it should be possible to provide radiation of the specifications given above. Detailed design studies are on.

Present status

Most of the sub-systems shown in Figure 6 have already been developed, or are in an advanced stage of development.

For terahertz lasing we have designed, built and field-mapped a 5 cm period, 2.5 m long undulator, in two equal sections of length 25 periods each (Figure 2). The quality of the undulator is within specifications. For example, we

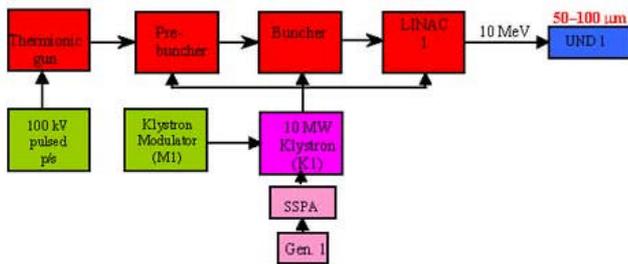


Figure 6. Block diagram of the layout of a potential terahertz FEL that could provide intense (1 MW peak power) tunable radiation from 50 to 100 μm , with a pulse length of a few picoseconds over the entire spectrum. The low electron energy (10 MeV) makes the FEL compact (80 m^2) and relatively inexpensive (Rs 5 crores).

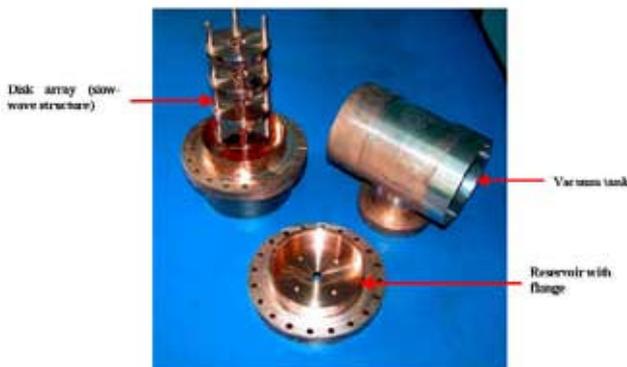


Figure 7. Components of a Plane Wave Transformer (PWT) linac structure developed at the Beam Physics FEL Laboratory. The disk array, which is responsible for producing the accelerating field, fits into the vacuum tank that has ports for RF feed and vacuum pump. The electron beam to be accelerated passes through the holes at the centre of the disks. The total length of the linac is just 21 cm, and it has successfully accelerated electrons up to 3.5 MeV.

had calculated a requirement of a field quality of better than 1% in $\Delta B/B$ – we obtained 0.9%. We have also measured the optical phase error (less than 2 degrees with steering compensation and gap tuning), and the beam wander (less than 1.5 wiggles amplitudes). The rms error in the undulator period was measured to be less than 100 μm .

A major challenge has been the indigenous development of the linear accelerator technology. We also chose to build a rather unconventional structure – the plane wave transformer (PWT) linac²⁷. This is a much more open structure, with strong coupling between the cells – and consequently with reduced fabrication tolerances. The only PWT linac working in the world is at UCLA²⁸, where it is routinely used, mainly for FEL applications. After building a number of prototypes and ascending a steep learning curve, we now have a four-cell, 21 cm long structure, PWT3 (Figure 7) that has been fabricated to the required tolerances (30 μm) and surface finish (0.2 μm CLA), which can hold UHV (1×10^{-8} torr), resonates at the desired frequency of 2856 MHz, has a loaded Q of 8000, and which has been conditioned with high-power RF.

We have also developed a 69 kV, 330 A, 25 MW, 10 μs , line-type pulse modulator for powering the 10 MW, 10 μs , 2856 MHz klystron that we have bought from TOIRY, Russia. We have successfully extracted up to around 7 MW of power from the klystron, and are only limited from going to higher power by the fact that our waveguides are presently pressurized with N_2 ; higher power will require pressurization with SF_6 .

We have also successfully obtained a 500 mA beam from a 40 kV thermionic gun, built by another group in our centre²⁹. We have built a small beamline connecting the gun to the linac, and obtained transmission of the electron beam through the linac when it was not powered. After sorting out synchronization issues, at the time of going to press we have accelerated electrons up to 3.5 MeV. This is only the second PWT linac in the world, and the first to accelerate beam from a thermionic gun. It is also perhaps the first electron linac in the country to deliver an external electron beam.

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