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### The Free Electron Laser (1982)

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Continuing advances in the science and technology of new sources of tunable radiation will be essential to obtaining increased and deeper understanding of gaseous and condensed state materials. The demonstration of a new source or radiation, the Free Electron Laser, promises to be a unique source in the region of wavelength shorter than 200 nm and longer than 25 micrometers. The characteristics that make the FEL worth special attention include tunability, high average power, potentially stable output power and frequency, very short pulses with high peak powers, coherence, bandwidth. The combination of these characteristics makes possible exciting studies in the area of condensed matter physics, spectroscopy of atoms, molecules and ions, and surface studies in presence of absorbed molecular species. Many other scientific investigations of interest to materials characterization and technology will become possible as the awareness of the capabilities of the FEL radiation increases.

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# **↑ The Free Electron Laser**

Report of the
Free Electron Laser Subcommittee
of the
Solid State Sciences Committee
(A Resource Paper)

Commission on Physical Sciences, Mathematics, and Resources National Research Council

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### **PREFACE**

This report constitutes a resource paper prepared for the Solid State Sciences Committee (SSSC) of the Assembly of Mathematical and Physical Sciences (AMPS) of the National Research Council (NRC) by an SSSC Subcommittee. This study was undertaken by the SSSC (D. E. Eastman, Chairman) as a result of recognition that Free Electron Lasers (FEL's) are versatile sources of radiation. These sources are of potential value in providing high power, tunable radiation at frequencies in the electromagnetic spectrum where such sources do not now exist.

Accordingly, an SSSC Subcommittee was appointed in the Fall of 1980 to provide an information base on the scientific opportunities made possible by development of FEL's for the SSSC and the community it serves. The subcommittee comprised a number of experts in FEL design and construction and in areas in which the FEL might be used. The group met in Washington during December 1980, and this report presents its findings.

In summary, the group concluded that the FEL indeed was a promising source in the wavelength regions  $\lambda > 25 \,\mu \text{m}$  and  $\lambda < 200 \,\text{nm}$  and recommended that the SSSC monitor this area closely over the next two years. If the promise identified now is realized, a deliberate, yet rapid assessment of funding priorities in this area would be necessary.

Finally, on behalf of the SSSC and the community, I would like to express our appreciation to all the participants in this study for their hard and conscientious work.

# J. Silcox, SSSC Liaison.

### 1. CONCLUSIONS AND RECOMMENDATIONS

Continuing advances in the science and technology of new sources of tunable radiation will be essential to obtaining increased and deeper understanding of gaseous and condensed state materials. Historically, every new source of radiation has led to a significant increase in the types of interaction that can be investigated. The demonstration of a new source of radiation, the Free Electron Laser, promises to be one such significant advance. At present there are at least four on-going and funded projects for construction of the sources, with differing objectives and hence of differing capabilities, which are expected to come "on line" by late 1981 or early 1982.

• We conclude that the Free Electron Laser promises to be a unique source in the region of wavelength shorter than 200 nm and longer than 25  $\mu$ m.

The characteristics that make the FEL worth special attention include tunability, high average power, potentially stable output power and frequency, very short pulses with high peak powers, coherence, narrow bandwidth.

The combination of these characteristics makes possible exciting studies in the area of condensed matter physics, spectroscopy of atoms, molecules and ions, and surface studies in presence of adsorbed molecular species (as in catalysts) to name just a few. Particular promises of unique applications are seen in the study of nonlinear phenomena and of temporal transient effects. It is also clear to the committee that many other scientific investigations of interest to materials characterization and technology will become possible as the awareness of the capabilities of the FEL radiation increases.

 We, therefore, recommend that the SSSC actively and closely monitor the program of the already planned FEL's and their initial impact on physics and chemistry over the next two years through a standing subcommittee.

If the promise of the current projected capabilities is realized, extensive novel scientific opportunities will become available. It will then be important to provide careful evaluation of these opportunities in order to arrive at a deliberate yet rapid assessment of the priorities. Such an assessment leading to conclusions regarding further support of new facilities and experimental capabilities should be carried out by an appropriate ad hoc panel of the Academy whose deliberations would be greatly aided by the existing subcommittee. We regard it as important that the current status in funding and auxiliary support (e.g., accelerator time) be sustained in order that the machine physics be adequately established and that access to the lasers for at least a few exploratory applications experiments be made available.

### 2. SUMMARY

Optical spectroscopy and the study of optical interactions with materials have been important tools for the elucidation of the structure and spectra of elementary excitations in gaseous and condensed state materials. wavelength region covered by these tools spans from about 100 nm in the ultraviolet to about 1000 µm in the far infrared. The optical excitations in the 100 nm to 1 µm region are generally characteristic of electronic excitations of the materials, which for example in solids will include excitations across direct or indirect bandgaps (and the associated excitons, etc.) and in gases and liquids, the excitation of a molecule from one electronic state to a higher one, having the same or different vibrational and rotational quantum numbers. Optical excitations in the 1  $\mu$ m to 25  $\mu$ m region are characteristic of vibrational excitations in molecules (gases or liquids) and of phonons in solids. The longer wavelength (i.e., lower frequency) excitations (from  $\sim 25 \mu m$  to  $\sim 1000 \mu m$ ) characterize phonons, plasmons, superconducting gaps, etc., in solids and rotational transitions in gases. Linear as well as nonlinear optical interaction studies have been seen to be important. Further, steady state spectra and transient spectra following a perturbation from an external source, provide complementary information regarding the ground and excited state, respectively. Historically, such studies in the  $\sim 25 \mu m$  to  $\sim 1000 \mu m$  region have yielded good but by no means complete pictures of materials of interest to science and technology.

Spectroscopic investigations in this entire range of wavelengths require sources of radiation

- whose frequency can be tuned continuously to match that of the absorbing species,
- whose power output is sufficiently high to carry out the needed measurements in reasonable lengths of time,
- whose output level is sufficiently stable to obtain meaningful information regarding small changes in reflection or transmission,
- whose bandwidth is sufficiently narrow to resolve the narrowest structure in the spectra of interest,
- whose temporal structure is such that transient measurements of the shortest lived excited states can be carried out,
- and finally whose coherence properties are such that nonlinear interactions such as harmonic generation, multiphoton dissociation, etc., can be carried out.

It is at once clear that while a single source that possesses all the above characteristics and covers the entire region of interest from 100 nm to 1000  $\mu$ m would be ideal but also that in practice, different wavelength regions may be best served by a variety of sources. Recently, a new source of laser radiation,

the Free Electron Laser (FEL), has been demonstrated and may satisfy many or all of the above needs. The present report evaluates and compares the capabilities of the FEL with other sources of tunable radiation currently available for optical interaction studies.

Tunable coherent sources have been developed to varying degrees over a broad wavelength range. Below 100 nm, there are essentially no good tunable coherent sources. Between 100 and 200 nm useful sources exist but have limited power presently. For the range between 200 nm and 25  $\mu$ m, a large number of well developed sources exist which could satisfy most of the imaginable scientific needs. From 25  $\mu$ m to 1000  $\mu$ m, tunable sources are not as well developed and could represent a unique opportunity for Free Electron Lasers.

A wide variety of tunable laser and more conventional techniques 1s used for photon spectroscopy from the X-ray to the infrared. As a general rule broad band synchrotron or thermal sources are used with dispersion or interferometric spectrometers for linear spectroscopy at moderate resolution over wide spectra bands. Tunable (and fixed frequency) lasers are generally used to measure coherent effects, high resolution spectra, nonlinear effects, or short-time duration phenomena. The noise properties of laser sources and the dynamic range limits of sensitive detectors are sufficiently comparable that both approaches are used when a large ratio of signal-to-noise is required.

### 2.1 The Free Electron Laser

A free electron laser consists of a high energy electron beam and a periodically alternating, static transverse magnetic field called a "wiggler." The period of the alternations is typically a few centimeters, the length of the wiggler a few meters, and the field strength typically a few kilogauss. When a relativistic electron beam is injected into this magnetic field, the electrons execute forced transverse oscillations. If a laser beam is now propagated parallel to the electron beam, the oscillating transverse electric field of the laser can interact with the transversely oscillating electrons. Laser amplification will occur if the following resonance condition is satisfied:

$$\lambda_{L} = \frac{\lambda_{w}}{2\gamma^{2}} \left[ 1 + \left( \frac{eB\lambda_{W}}{2\pi mc} \right)^{2} \right]$$
 (1)

where  $\lambda_L$  is the laser wavelength,  $\lambda_W$  the wiggler wavelength,  $\gamma mc^2$  the electron energy, B the rms magnetic field of the wiggler, e the electron charge, m the electron mass and c the velocity of light. From Eq. (1) it is evident that the laser wavelength can be tuned by varying the wiggler wavelength, the electron energy, or the wiggler magnetic field. Experiments conducted at Stanford, Columbia, and NRL have demonstrated laser action according to these principles.

Free electron lasers have the potential capability of operating as powerful sources of continuously tunable, coherent radiation in the wavelength region from millimeters to the VUV. The following table summarizes the range of potential values of the most important operating characteristics of FEL's.

<ul> <li>Wavelength</li> </ul>	$50~\text{nm} \lesssim \lambda \lesssim 1~\text{mm}$
<ul> <li>Average power</li> </ul>	$P_{av} > 1 \text{ kW}$
<ul> <li>Peak power</li> </ul>	$P_p > 1 MW$
• Linewidth	$\frac{\Delta\lambda}{\lambda} < 10^{-6}$
<ul> <li>Overall efficiency</li> </ul>	> 10%

Pulse length

However, not all of the above characteristics can be achieved with a single FEL. Different electron beam sources will be needed to cover the range of wavelengths and power.

 $10^{-12} \lesssim \tau_{\rm p} \lesssim \infty$ 

There are now several experimental FEL programs under way in the U.S., Europe and the Soviet Union. Experimental results obtained to date have demonstrated amplification at 10.6  $\mu$ m (Elias et al., 1977), and laser oscillation at 3.4  $\mu$ m with a peak power of 7 kW (Deacon et al., 1977) and at 400  $\mu$ m with 1 MW peak power (McDermott et al., 1978). Approximately ten new experiments are under construction at the present time in the U.S. alone, mostly with DOD funding. These experiments will span wavelengths from 200 nm to the millimeter region, and are expected to demonstrate improved power and efficiency with pulse lengths from  $10^{-11}$ s to CW. Of these facilities, only the 20  $\mu$ m-1000  $\mu$ m facility at Bell Laboratories will be dedicated to solid state research. The other facilities are dedicated to the development of devices, although limited amounts of time may be available for interesting scientific experiments.

### 2.2 Solid State Studies

Solid state studies where the unique combination of tunability, high power, coherence, and temporal structure of the FEL laser power output can play a singular role include the understanding of dynamics of phonons, bulk and surface plasmons and superconducting gaps. For example, the characteristic excitations of a two-dimensional electron gas in a MOSFET depend on both electric and magnetic fields. These can only be studied using tunable radiation (rather than using a fixed frequency laser together with tuning of the properties of the electron gas by changing the electric or magnetic field) as shown by the recent measurements of cyclotron resonance in the extreme quantum limit (Wilson, et al., 1980). Transient spectroscopic studies utilizing the  $\sim 10^{-10} - 10^{-11}$  second time duration of the FEL output radiation in the far IR

include the dynamics of charge carriers in semiconductors, identification of vibrational decay channels in liquid and phonons in solids following a higher power transient excitation provided either by another synchronized (short wavelength) excitation source (e.g., a laser) or even the electron beam bunches emerging from the FEL's, quasiparticle scattering times in superconductors, etc. Such transient studies, so important in the exploration of dynamic effects in solids are beyond the reach of any of the currently available sources in the 25  $\mu$ m to 1000  $\mu$ m region.

### 2.3 Research in Chemistry

Molecular and chemical studies would profit from a pulsed tunable infrared source with energies comparable to that of laboratory  $CO_2$  lasers, tunable picosecond pulses of substantial energy especially for  $\lambda < 200$  nm and a strong new laser for  $\lambda < 150$  nm. Vibrational relaxation studies, multiphoton absorption, fast chemical kinetics, and photochemistry would be substantially strengthened by these new tools.

### 2.4 Surface Chemistry

A FEL that operates in the infrared or far infrared has several potential applications in surface chemistry. Four specific areas of applications which should be considered are: i) vibrational spectroscopy of adsorbed molecules on single crystals (high area samples may be studied using conventional infrared techniques) with high resolution and at high ambient pressures, ii) time resolved spectroscopy that follows the transient response of surface species, iii) far infrared spectroscopy of the metal-oxide interface between small particles or clusters of metal atoms and oxide surfaces which stablize them, and iv) vibrational (rotational) excitation of molecules reacting on a surface. Except possibly, for the last, none of these applications require the very high power available from a FEL. However, adequate power will simplify, and may add convenience to conventional linear spectroscopy of surfaces. The area of vibrational spectroscopy of adsorbed species on single crystal oxide surfaces is an example of an important surface problem where this may be the case. At the present time most applications to surfaces of FEL operating at short wavelengths ( $\lambda < 100 \text{ nm}$ ) can be met with existing synchrotron sources. However, FEL may be useful for photodesorption experiments in the region covering 50 nm where the absorption cross-sections are generally very small.

### 3. BACKGROUND

### 3.1 Introduction

Optical spectroscopy of gaseous and condensed state materials has always experienced a significant expansion to new phenomena and new materials when the technology of either tunable sources or detectors has experienced a breakthrough. There have been laser and nonlaser sources which have served the field of spectroscopy well in the 250 nm  $-25 \mu m$  region. At shorter or at longer wavelengths the existing sources lack the versatility and flexibility to explore many of potentially exciting phenomena. A recent development in the field of lasers - the demonstration of a Free Election Laser (FEL) holds the promise for alleviating some of the restrictions on currently feasible experiments. In what follows we review first the status of existing laser technology and current spectroscopic techniques, discuss current experiments in free electron lasers, and then outline some areas in which the FEL is likely to be valuable.

### 3.2 Status of Existing Laser Technology

A large number of tunable laser devices exist both as commercially available units or as specifically developed sources in various laboratories. An extensive review of the operating principles of tunable coherent sources has been given by Colles and Pidgeon, 1975, and by Mooradian, 1979.

The wavelength range of operation may be divided roughly into three regions: less than about 200 nm; between 200 mm and 25  $\mu$ m; and between 25  $\mu$ m and 1 mm. Tunable coherent sources below about 150 nm become increasingly difficult to produce as the wavelength decreases. Most present sources rely on nonlinear mixing techniques such as 4-wave processes in atomic vapors. The short wavelength transmission limit for nonlinear crystals is usually reached around 200 nm. Wavelengths as short as 30 nm have been produced by frequency multiplication techniques but with little tunability and very little power output. Several laboratories have used nonlinear mixing techniques with commercially available excimer lasers (KrF, ArF, etc.) to carry out linear and nonlinear spectroscopic measurements down to 120  $\mu$ m. With present technology, several kilowatts of peak power can be generated with continuous tunability down to 80 nm using frequency mixing of excimer laser and dye laser radiation in atomic vapors. This technique may be extended to 30 nm with tens of watts of peak power.

In the region between 150 nm and 25  $\mu$ m, tunable coherent sources have been rather well developed so far, and are continuing to be developed at a rapid pace. While no single source exists which can, as a primary laser, tune continuously over this entire region, there are several different types of laser systems that have been demonstrated to operate over parts of this region. These include

- a. Dye lasers which generally cover the region from 350 nm to 1100 nm.
- b. Parametric oscillators which are tunable over regions from  $\sim$ 600 nm to  $\sim$ 25  $\mu$ m.
- c. F-center lasers covering the region from  $\sim 800$  nm to  $\sim 4 \mu \text{m}$ ;
- d. Semiconductor diode lasers tunable over the region from  $\sim$ 650 nm to  $\sim$ 35  $\mu$ m.
- e. Transition metal-ion doped crystal lasers covering the  $\sim 1 \,\mu$ m to  $\sim 2.5 \,\mu$ m region.
- f. Alexandrite lasers with a tunability from ~700 nm to ~800 nm.
- g. High pressure molecular gas lasers which provide primary tunable radiation from  $\sim 9 \mu m$  to  $\sim 12 \mu m$ .

Detailed operating characteristics of these sources are given in Appendix I.

In the region between 25  $\mu$ m and 1000  $\mu$ m, much less success has been achieved in developing tunable sources. Optically pumped molecular gas lasers offer convenient sources of CW (hundreds of mW's) and pulsed (up to MW's) radiation at a large number of discrete frequencies in this region with very limited (tens of MHz) tuning around each line (Chang, 1979). Present microwave semiconductor technology has produced pulsed solid state oscillators at frequencies beyond 300 GHz. Usable sources for spectroscopic applications between 100  $\mu$ m and 1000  $\mu$ m will come available in the near future. Conventional electron tube devices such as carcinotrons and extended interaction oscillators are limited to wavelengths longer than ~300  $\mu$ m. Efficient frequency multiplication using these sources may extend tuning to shorter wavelengths. Nonlinear mixing processes in the far IR have been demonstrated to be efficient in such devices as GaAs Schottky diodes.

### 3.3 Current Spectroscopic Techniques

When linear spectral information of moderate precision is required over a broad range of wavelengths, it is best acquired using broad band continuous sources together with a diffraction grating or other dispersion spectrometers from X-ray to the visible region, and using a Fourier transform interferometer in the infrared. The continuous radiation sources used include synchrotron radiation at short wavelengths and black body radiation out to mm wavelengths. Much useful spectroscopy has been done in this way including, for example, photoemission and extended fine structure absorption spectroscopy (EXAFS) at X-ray wavelengths, and the measurement of molecular vibrations in the infrared. Tunable laser and submillimeter microwave sources can conveniently do very high resolution measurement such as those required for the investigation of Doppler broadened molecular spectra.

Nonlinear phenomena and time dependent effects can be investigated with flash lamp sources where the band to be pumped is relatively broad. Their

limited spectral brightness and the limited sensitivity of fast detectors (especially at infrared wavelengths) limit the general usefulness of this technique. Tunable laser sources are proving very useful in these applications. Fixed frequency laser sources are also used in special cases where the available wavelength is appropriate.

Many spectroscopic phenomena such as magnetic resonances can be tuned with electric or magnetic fields or with temperature. Significant tuning ranges are often available at infrared wavelengths which permit the use of fixed frequency laser sources for spectroscopy.

A wide variety of special techniques is used to recover weak spectroscopic effects, or effects which appear in the presence of strong background signals. Great demands are often placed on source stability as well as the detector sensitivity and dynamic range. Techniques such as photo-acoustic spectroscopy and thermal detection can directly measure small amounts of power absorbed in a sample. Many such experiments take advantage of the power available from laser sources.

At visible and shorter wavelengths photon detectors are available which can detect single photons with good quantum efficiency. Wide dynamic ranges are also available. Noise limits are often set by the statistics of the signal photon stream. Infrared photon detectors with high quantum efficiency are available for wavelengths out to 240  $\mu$ m. These are typically amplifier noise limited to power sensitivities of NEP  $\approx 10^{-16}$  to  $10^{-17}$  W/ $\sqrt{\rm Hz}$ .

At longer infrared wavelengths thermal detectors must be used. Room temperature pyroelectric detectors have NEP  $\gtrsim 10^{-10}$  W/ $\sqrt{\rm Hz}$  while cooled bolometers can have NEP  $\sim 10^{-15}$  W/ $\sqrt{\rm Hz}$ . Thermal detectors are used over the whole infrared region. The noise and dynamic range of infrared experiments is often limited by ambient temperature black body radiation so that the values of NEP obtained in practice can be orders of magnitude worse than those quoted above. Low detector noise levels are obtained only with cooled narrow-band instruments. The signal-to-noise ratio can be limited by detector dynamic range even with weak thermal sources. The more sensitive infrared detectors have dynamic range of  $10^4$  to  $10^5$ .

When laser sources are used to observe weak spectral features, the source power is usually large enough so that room temperature detectors can be used and background photon effects are not important. The signal-to-noise ratio is then typically limited by fluctuations in laser power which are often troublesome at the level of 10<sup>4</sup> to 10<sup>5</sup> in a 1 Hz bandwidth. For these reasons, tunable laser sources do not hold a general advantage over thermal sources for measurements of weak spectral features at moderate resolution.

### 3.4 Free Electron Lasers

The first experimental demonstration of an FEL operating in the IR was conducted by Madey and coworkers in 1976 (Elias, et al., 1976) followed by a

number of other exploratory experiments (Deacon et al., 1977; Granatstein et al., 1977; Marshall et al., 1977). Theoretical developments have proceeded rapidly and at the present time innovations (e.g., tapered wigglers) are being considered for incorporation into the second round of experiments. One the of the main thrusts at present is to verify the theoretical understanding of the machine physics, a necessary step if FEL's are to be fully exploited. We believe this is likely to be achieved over the next two to three years as the newer experiments come on line and are carried to fruition. It is also important that as the sources become operational, time should be made available not only to demonstrate machine principles but also for demonstration user applications. In our judgment, in the current situation, this is likely to be achieved.

Current machine physics thrusts are devoted towards improvements in the power extraction and efficiency of extraction and towards FEL operation at shorter wavelengths. Other parameters may well also be significant to users in the long term. For example, improvements in stability of output (better than  $10^{-5}$ ) would improve significantly the prospects for novel applications in linear spectroscopy. However, this is not a priority item at the present time and is unlikely to be attacked until user interest builds to an appropriate level.

In Appendix II, we provide an account of the ideas underlying the FEL and the possible improvements so far identified. In the remainder of this section we outline the experiments currently under way in this country.

The experimental arrangement of the initial Stanford experiment (Elias et al., 1976) is illustrated in Fig. 1. The wiggler magnet consisted of a superconducting double helical winding 5.3 m long with a uniform 3.2 cm period, producing a transverse field of 0.24 T on axis. In the first series of experiments, the electron beam was operated at 24 MeV, with a peak current of 70 mA. No mirrors were used, and the gain at 10.6  $\mu$ m was measured using a CO<sub>2</sub> laser with a peak power of 100 kW. The maximum gain observed at 10.6 µm was 7% per pass; absorption was observed when the electron energy was tuned below resonance. The maximum energy extraction was 0.25%, in agreement with Eq. (2) in Appendix II. In a second series of experiments, the electron energy was increased to 43 MeV, the peak current was increased to 0.6 A, and mirrors were installed (Deacon, et al., 1977). Laser oscillation was observed at 3.4  $\mu$ m with a peak power of 30 kW, corresponding to 0.1% energy extraction. Although these results are not astonishing in themselves, they verify the theoretical predictions, at least for uniform wigglers, and provide evidence beyond any doubt that free electron lasers do work.

At the present time, several experiments are in progress to improve the extraction efficiency observed in Madey's experiments by using tapered wigglers (see Appendix II). In an experiment being undertaken at Los Alamos, a small electron linac is being modified to provide 20 MeV electrons with a peak current of 25 A. In the first series of measurements, gain and energy extraction will be measured using a  $CO_2$  laser at  $10.6 \mu m$ . To trap and

decelerate the electrons in a tapered wiggler, the CO<sub>2</sub> laser will be operated for short pulses, no more than a few nanoseconds long, to produce high peak powers in excess of 1 GW, as discussed in Appendix II. The gain is expected to be quite small (1-2%) from a tapered wiggler.

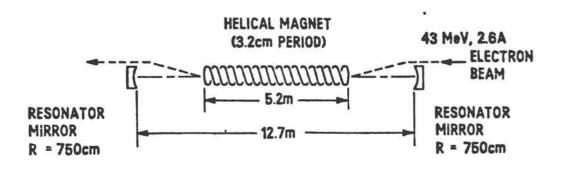


Fig. 1. Experimental arrangement used by Madey and coworkers [Phys. Rev. Lett. 38, 892, (1977)] to demonstrate free-electron laser.

However, at an input laser power of about 1 GW, the predicted energy extraction efficiency is about 3%. This will represent an order-of-magnitude improvement over the results predicted and obtained from uniform wigglers. Following this series of experiments, this device will be converted to an oscillator. The nominal wavelength will still be  $10.6 \mu m$ , but it should be possible to tune the device from  $8 \mu m$  to  $15 \mu m$ , or even longer wavelengths. The peak power will be of the order of megawatts, and the average power of the order of tens of watts. This facility should be operating in 1982, and limited time may be available on a noninterference basis for interesting scientific experiments.

The free electron laser experimental program at the University of California, Santa Barbara, was initiated in January 1980 with funds from ONR and AFOSR. The UCSB FEL device is based on the use of a 3 MeV electrostatic accelerator as the source of electrons. Initially (1981) this device will operate in the FIR region (100  $\mu$ m - 1 mm). However, using a two-stage FEL technique (Elias, 1979) it will be possible to operate the device in the visible-UV region (1982). The important operating characteristics of the UCSB FEL are listed below:

Wavelength (1981)  $100 \mu m - 1 mm$ 

Peak power 20 kW

Average Power 100 W - 12 kW

Pulse length (1980)  $100 \mu s - \infty$ 

(1981)  $< 100 \text{ ns} - \infty$ 

Resolution  $\Delta \lambda/\lambda < 10^{-4}$ 

Expected overall

efficiency  $\eta \gtrsim 10\%$ 

Running time for scientific applications will be available on a limited basis beginning January 1982.

The Brookhaven FEL experiment is intended to study the operation of FEL's in an electron storage ring. A storage ring allows one to build a laser operating in the wavelength region below 150 nm and perhaps down to a few tens of nm. A tunable laser in this spectral region is clearly a unique instrument. The experiment will be done on the 700 MeV storage ring of the National Synchrotron Light Source. Initial data will be taken at 300 nm to check the present theoretical understanding of the system. Subsequently, one will go to shorter wavelengths. Using the 700 MeV storage ring the minimum lasing wavelength will be limited to around 100 nm because the maximum gain per pass that can be obtained is near 10%. In this wavelength region the expected laser power is of the order of 10W, the line width 10<sup>-6</sup>; the laser pulses will be about one nanosecond long and separated by 50 ns. If this experiment confirms the present understanding of the FEL storage ring system, one can, with a specially designed storage ring, increase the gain by up to two orders of magnitude extending the system capabilities to the region of a few tens of nm.

The Bell Labs FEL project is specifically aimed towards obtaining tunable laser radiation in the 10  $\mu$ m to 1000  $\mu$ m region with the objective of using the far infrared radiation for solid state spectroscopy of a variety of types. The electron accelerator will be a Microtron (Kapitza and Melekhin, 1978) which will provide electron bunches of duration  $\sim 100$  ps in a train  $\sim 10$   $\mu$ s long and with peak current of > 5 A. The output energy of the electrons will be tunable from  $\sim 8$  MeV to  $\sim 10$  Mev. Initially an electromagnet wiggler is planned which will allow the FEL tunability from 100  $\mu$ m to 400  $\mu$ m. Expected peak powers and average powers are 100 kW and 10 W, respectively. The next stage of its development will include addition of wigglers with different periodicity to extend the tunability of the FEL.

Several experiments have been conducted to investigate free-electron lasers operating in the Raman regime (Granatstein, et al., 1977; Marshall, et al., 1977; McDermott, et al., 1978; Caponi, et al., 1979). The best results were

achieved in a joint effort by Columbia University and the Naval Research Laboratory (McDermott, et al., 1978). The experimental arrangement is shown in Fig. 2. A pulsed electrostatic accelerator was used to produce a high-intensity electron beam with an electron energy of 1.2 MeV and a peak current of 25 kA. An annular beam formed with a cold plasma cathode was expanded to fill the region between the annular mirrors. The longitudinal magnetic field used to guide the electrons was given a ripple component to act as the wiggler. The experiment demonstrated very high gain at a laser wavelength of 400  $\mu$ m, in good agreement with theoretical predictions.

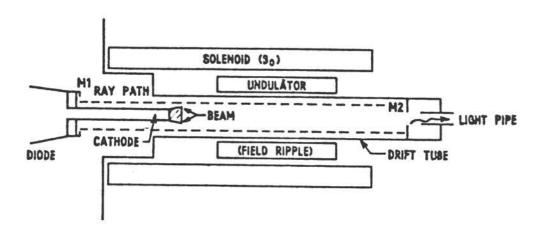


Fig. 2. Experimental arrangement used by McDermott, et al. [Phys. Rev. Lett. 41, 1368, (1978)] for free electron laser operating in the Raman regime.

A peak power of 1 MW was obtained, corresponding to an extraction efficiency of the order of  $3\times10^{-5}$ . This rather low extraction efficiency is attributed to the fact that the 50 ns electron beam pulse was too short for the laser oscillation to build up from noise to saturation. In addition, the electron-beam voltage was not steady during this period, so that the resonant laser wavelength fluctuated. An improved experiment is now under construction.

Comparison of these results with those obtained by Madey and coworkers illustrates the differences between free electron lasers operating in the Raman and Compton regimes. To operate in the Raman regime and take advantage of plasma effects, it is necessary to have a very high intensity electron beam. In addition, it is necessary to operate at low electron energy. As shown by Eq. (3) in Appendix II, the threshold current density for plasma effects, which is proportional to  $\omega_p^2$ , increases with the third power of the electron energy. Since low electron energies correspond, in general, to long wavelengths [see Eq. (1)], lasers operating in the Raman regime are more likely to be found in the

infrared to microwave region. With regard to the technology, such low-energy, high-current beams are best formed with pulsed, electrostatic generators. Low current, high duty factor electrostatic accelerators will be useful for high average power at this and somewhat shorter wavelengths. For even shorter wavelengths, higher electron energies are required. These are best provided by rf accelerators, (microtrons, linear accelerators and storage rings) which produce low-intensity beams. Because of the low intensity and high energy of electron beams used for short wavelengths, lasers in this regime will necessarily operate in the Compton (single-particle) regime. These ideas are summarized in Fig. 3. It should be realized that the operational regions indicated there are not sharply defined, especially in the long wavelength region.

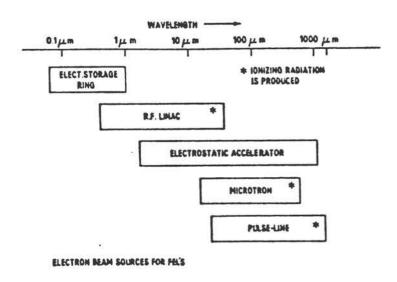


Fig. 3 Electron accelerators for FEL's in different wavelength regions.

### 3.5 Applications

3.5.1 Solid State Studies The far infrared region of the spectrum covers most of the important elementary excitations of the solids as shown in Fig. 4. (Patel and Shaw, 1981) These are bulk and surface phonons, bulk and surface plasmons, quasiparticle bandgap in superconductors to name just a few. All of these have been subject to extensive spectroscopic studies using conventional far IR sources and Raman scattering (in the case of phonons and plasmons), tunneling spectroscopy in the case of superconductors, etc. There are, however, specific situations which can either be handled only with great difficulty or not at all using currently available sources. The far IR FEL with its tunable high power, pulsed operation characteristics should provide unique opportunities for linear as well as nonlinear, and steady state as well as transient studies. It can be, in general, argued that the availability of very sensitive detectors removes some of the advantages of the far IR FEL for linear optical absorption studies. However, if the FEL source is available anyway, the

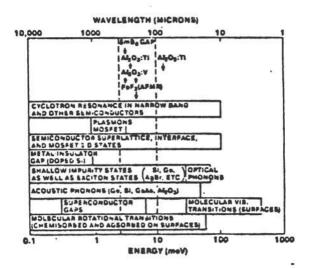


Fig. 4 Important elementary excitations in solids in the far-infrared region.

detector constraints are much relaxed, and linear spectroscopy may still be carried out using the FEL because of its high average power capability. Examples of linear far IR spectroscopy where fixed frequency lasers have been used in conjunction with tuning the absorption characteristics of materials being investigated include the studies of ferromagnetic systems (where magnetic field provides the tuning), two dimensional electron gas in MOSFETs (where either an electric or a magnetic field can be used for tuning the characteristic absorption of the electron system), etc. However, often it is seen that such fixed frequency laser - absorption tuning spectroscopy leads to erroneous conclusions when the resonance system, spins, electrons, etc., themselves change their interactions as a function of the tuning parameter. A characteristic example of such an inadvertent mistake is in the study of cyclotron resonance of the two-dimensional electron gas in the MOSFETs in the extreme quantum limit when the average separation between the electrons is greater than the cyclotron radius (Wilson, et al, 1980). In such a situation, the availability of a tunable far IR FEL for linear spectroscopy would be a tremendous advantage.

Optical properties of superconductors have been investigated extensively for the purpose of correlating superconducting properties with phonon properties of interesting systems. Often these studies need to be carried out on interesting materials such as Nb<sub>3</sub>Sn, V<sub>3</sub>Si, ErRh<sub>4</sub>B<sub>4</sub>, etc. which can not be grown in the form of large single crystals appropriate for measurements using far IR Fourier spectrometer. The far IR FEL laser whose output can be focused down to a small spot size will simplify acquisition of data that would complement the information gathered from neutron scattering experiments.

It is, however, the area of nonlinear spectroscopy and that of transient studies that stand to be the principal beneficiaries of the capabilities of the far

IR pulsed FEL source. The types of nonlinear studies in the long wavelength region that promise to be interesting and useful are stimulated emission phenomena (where the gain drops only as  $\lambda^{-1}$ ), inelastic scattering from a variety electronic excitations, optical pumping of  $V^{++}$  and  $Ti^{+++}$  ions in  $AQ_2O_3$  to generate stimulated emission of phonons, etc. Yet another example of optical pumping is the utilization of a scheme that allows energy storage in a medium for high peak power Q-switched operation of conventional lasers. A particularly interesting example is the liquid  $N_2$ :CO system. The 4.3  $\mu$ m band of liquid  $N_2$  has a long energy storage time and an efficient collisional transfer to CO vibrational level. This scheme would make possible a high peak and high average power  $\sim$ 4.8  $\mu$ m laser source.

Time resolved spectroscopy has found a well deserved place in understanding electronic energy transfer mechanisms in atomic, molecular, solid and liquid systems. The development of picosecond pulse dye lasers in the visible region has allowed one to probe on a  $10^{-11} - 10^{-12}$  second time scale the relaxation of the hot electron-hole gas in semiconductors, vibrational relaxation in solids and liquids through the use of Raman scattering techniques, and the excitation/de-excitation mechanisms in biologically important systems such as rhodopsin, etc. In solids the far infrared excitations are phonons, plasmons, and quasiparticles in superconductors, whose transient study is going to provide an even greater understanding of materials and their associated relaxation phenomena than what has been possible to date. Fig. 5 (Patel and Shaw, 1981) shows some typical lifetimes of interest.

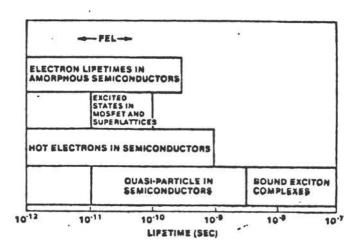


Fig. 5 Characteristic lifetime of a variety of solid state excitations in the farinfrared region.

The short pulse, high power far infrared free electron laser could be advantageously exploited for transient spectroscopy by synchronizing the free

electron laser to a tunable source of picosecond visible pulses such as a dye laser pumped by a mode-locked CW ion laser (Heritage and Jain, 1978). Since both the free electron laser and the mode-locked ion laser require stabilized rf driving oscillators, this could be readily achieved by using a common rf reference frequency and equalizing the cavity lengths of the free electron and visible mode-locked lasers. The result would be the generation of synchronous trains of intense tunable optical and far infrared pulses of picosecond duration whose relative timing would be varied for time-resolved spectroscopy. Since both trains would have high peak intensity, either the FIR or visible pulses could be used for excitation of materials and the other for time-resolved probing.

An example of the use of this capability is the measurement of the dynamics of charge carriers in semiconductors. The FIR pulses are an ideal probe for the measurement of local electrical conductivity without the need for electrodes. The visible pulses, on the other hand, are ideally suited for the generation of free carriers by band-to-band transitions. By the selective choice of polarization and photon energy of the visible laser, hot carriers could be preferentially excited with a particular excess kinetic energy and their subsequent relaxation to minimum energy states could be accurately probed with the FIR pulses, providing important information about electron-phonon and electron-electron coupling. Similar measurements could be made of rapid radiative and nonradiative recombination, trapping, and plasmon generation and decay. Also, it would be possible to modulate the FIR pulses by generating overdense plasmas with the optical pulses, enabling even shorter FIR pulses to be produced (Nurmikko, 1978).

Another use of this measurement capability would be the identification and study of vibrational and rotational states in liquids and phonons, plasmons, defect states, and other low energy excitation in solids by time-resolved FIRvisible double-resonance spectroscopy. Recently, Kaiser, et al., 1978, have used synchronized trains of tunable near-infrared and optical picosecond pulses to make high resolution spectral measurements of vibrational states in liquids. Particular vibrational states were excited by the near IR pulses and then probed with the visible pulses by inducing selective electronic transitions to states from which visible fluorescence could be observed. The excellent selectivity of the transient double resonance approach enabled them to make extremely high resolution spectra which previously had been possible only in the gaseous phase. In addition, time-resolved measurements of vibrational dephasing times give information about molecular collisional dynamics. The extension of this approach to the far infrared would provide an important capability for the study of low energy excitations of materials in the condensed phase. In particular, the study of bulk and interface defect states in crystalline solids could be greatly enhanced by this technique. Other possible studies include 1) study of quasiparticle scattering times in highly excited superconductors where one uses the far infrared free electron laser radiation both for pumping as well as probing at and near the superconducting band gap, 2) studies of phonon propagation and interaction in materials, and 3) studies of nonequilibration excitation and relaxation in the two dimensional electron gas system in MOSFETs, etc. Such studies in the  $10-1000 \, \mathrm{cm}^{-1}$  are just not possible at the present time, but without any doubt they hold the key to essentially a new window on the interplay of a variety of different interactive systems in solids. A far infrared free electron laser under mode locked operation will provide high power tunable laser pulses with duration of  $\approx 10^{-10} \, \mathrm{sec}$  to  $\approx 10^{-12} \, \mathrm{sec}$  and make the above studies possible.

The study of fast phenomena in the far IR represents a genuinely new frontier in science where the far IR FEL will play a unique role. A comparison of the wavelength region and power levels achieved with both typical phenomena and with conventional laser sources is given in Fig. 6. (Patel, 1977; Patel and Shaw, 1981)

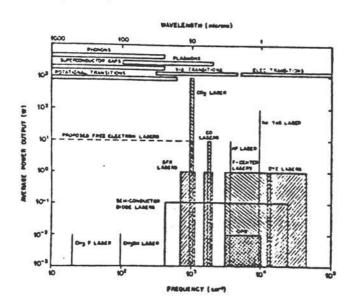


Fig. 6 Wavelengths of operation and power output for a variety of laser sources.

3.5.2 Gas and Liquid Phase Chemistry IR and FIR can be used to study spectroscopy and dynamics, i.e., the positions of molecular vibrational levels and the rates of transitions between them. For both types of measurements a monochromatic source is essential. Vibrational energy level measurements can usually be carried out satisfactorily with conventional equipment and in any case are not in the scientific forefront. There is need, however, for precise far infrared spectroscopy mainly of pure rotational transitions in astrophysically interesting molecules.

Chemical dynamics measurements in the  $2 \mu m - 25 \mu m$  wavelength region determine the rates of vibrational relaxation either by intermolecular energy transfer or by intramolecular vibrational redistribution. The "hot" intermediate

is detected either by the difference in its electronic absorption or by infrared emission. Moreover, the rates of relaxation are usually very rapid-subnanosecond in the liquid phase and typically in 10-1000 collisions in the gas phase. The need to have a high concentration of a vibrationally excited molecule which rapidly relaxes places a premium on a pulse of short duration and high energy in the  $3-25 \mu m$  wavelength region.

The most outstanding contribution of laser physics to chemistry is the CO<sub>2</sub> laser (Patel, 1964a,b, 1965). Through its high peak power it has created a new phenomenon - the multiphoton infrared absorption which reveals characteristics of previously inaccessible upper molecular vibrational levels and has produced isotope selective ground electronic state dissociation. A broad array of studies has been carried out but systems that can be investigated are severely limited to molecules which happen to absorb CO<sub>2</sub> laser radiation. The remarkable experiments already done with CO<sub>2</sub> lasers could be extended to all infrared absorbing molecules if tunable infrared sources of comparable strength were available. Such a source might result from new developments in infrared laser mixing or from a FEL.

Research in photochemistry has become much more rewarding since the development of visible and UV lasers. Moreover, from 157 nm ( $F_2$  excimer) to 1064 nm (YAG) there is now an array of excellent sources. What is still lacking? The region below 157 nm has no strong laser. Below about 250 nm one has discrete lines but no strong tunable laser. Finally, below 266 nm picosecond pulses have not yet been generated. The FEL could make a unique contribution to chemistry if it could fill even one of these needs.

3.5.3 Surface Chemistry and Catalysts The characteristics of a FEL which may be particularly applicable to surface studies are very short pulses for the investigation of the transient response of surface concentrations, high average power for vibrational excitation of adsorbed molecules and tunable, high intensity source for the far infrared. A tunable source throughout the infrared finger print region, 200-4000 cm<sup>-1</sup> will also make vibrational spectroscopy of adsorbed molecules more convenient than using presently available sources. The advantages of the FEL for studying surfaces at UV and X-ray frequencies are not so apparent because of the potential of conventional synchrotron sources. Photodesorption is an example of an experiment where the very high intensity of the FEL at short wavelengths may be superior to synchrotron sources because of the small cross-section for the processes Because filters to avoid second order effects from monochromators are not readily available at short wavelengths, the spectral purity of the FEL may be a second advantage for photoelectron spectroscopy. There may well be fluorescence and coincidence surface experiments at < 100 nm that will require the intensity and time resolution of a FEL but these have not been clearly identified to date.

Electron energy loss spectroscopy (EELS) is now being widely used (Ibach, et al., 1977) to investigate adsorption on single crystal metals. It has the

advantage of high sensitivity and is compatible with other ultra-high vacuum spectroscopies, e.g., LEED, Auger, etc., which provide structural information about the surface. However, EELS allows neither high resolution nor in-situ investigation of adsorbed molecules in equilibrium with a gas at high pressure. The latter situation may be handled using absorption-reflection spectroscopy where the FEL may offer an improvement in signal/noise, e.g., by making multiple reflections practical. A coherent source of radiation has an advantage of a well defined angle of incidence and more power per mode (compared to nonlaser source) which will be be especially useful for internal reflection spectroscopy, e.g., one could operate near the critical angle without energy loss.

Most of the vibrational spectroscopic studies of adsorbed molecules probably involve stable species that exist at high coverage but are not involved in catalyzed reactions. In order to identify kinetically significant adsorbed species it is essential to obtain time resolved spectra. If the response of surface species can be compared to the rate of product formation in the gas phase and if surface and gas phase concentrations respond in parallel, this provides good evidence that the surface species is kinetically significant. Time resolved vibrational spectroscopy of adsorbed species is now possible on a time scale >0.1s. This needs to be extended down to the  $\mu$ s scale or below to investigate kinetically significant species on good catalysts. This work must necessarily be performed on single crystals or flat surfaces in order that transport to and from the surface is not rate limiting.

The nature of the chemical interaction between metals and oxides is an important problem in practical catalysis. Oxide supports affect the rate of catalyst sintering, activity, selectivity, etc. In some cases the metal-oxide interaction must involve strong chemical bonding, e.g., when the metal spreads out into monolayers instead of clustering or crystallizing. Presumably the nature of these metal-oxide bonds can be analyzed if the vibrational spectrum of the interface can be obtained. The vibrational frequencies of these bonds will occur in the far infrared, a region where a FEL may provide a strong source.

The vibrational excitation of adsorbed molecules could involve multiphoton processes in some cases. Perhaps a more important application would be single quantum excitation where intramolecular energy redistribution does not occur. On surfaces, activation barriers can be low and a single quantum of vibrational energy may be sufficient to effect selectivity between two reaction paths or induce surface transport of a molecule. For molecules physically adsorbed or in a precursor state to chemisorption, photo-desorption may be possible.

3.5.4 Other Studies Other studies which do not necessarily derive from the high power, pulsed, tunable output characteristic of the far IR FEL include high resolution spectroscopy of gaseous molecules at pressures low enough so that collision broadening is negligible (a situation of considerable importance in the rotational spectra of molecules of interest for astrophysical investigations). For

these studies, the FEL will have to be stabilized well enough to have long term frequency stability approaching  $1:10^6$ , which is not expected to be difficult. The availability of high average power ( $\sim 1W$ ) combined with the possibility of continuous tuning and the narrow linewidth clearly point towards a unique capability of using the opto-acoustic detection techniques which should allow measurement of far IR spectra of gaseous molecules having an absorption coefficient  $\sim 10^{-10}$  cm<sup>-1</sup> without having to use path lengths longer than 10 cm. Molecular fragment lifetimes are very short in the laboratory environment but are quite long in the interstellar environment. Rotational spectra of such species can be studied only using the pulsed high power output from the far IR FEL.

Further, the high average power capability of the FEL in the far IR points to applications in the area of diagnostics and heating of the fusion plasmas. The areas of remote sensing in the atmosphere out to 500 kM altitude using resonance fluorescence above 100 kM (using the 200 nm - 100 nm region of the spectrum) for atomic and ionic species and differential absorption LIDAR (DIAL) for molecular species (using the 1  $\mu$ m - 25  $\mu$ m spectral region) are of interest. Lunar ranging using the short pulse capability of the FEL is also an attractive possibility.

### APPENDIX I

# Summary of Existing Sources

Dye Lasers

Wavelength range 350 nm - 1100 nm

(with several dyes)

CW power A few watts

mode purity Single frequency

Mode locked Pulse width ≤ 1 psec

operation

Pulsed energy 400 joules/pulse reported

typical 100 joules

Extended wavelength To 110 nm by nonlinear mixing

coverage and stimulated scattering and

out to 25 nm with

reduced power but more than adequate for high resolution

linear and nonlinear

spectroscopy.

Parametric Oscillators\*

Wavelength Visible to 4.5 μm

using Li NbO<sub>3</sub>;

out to 20 µm with CdSe.

Power output

Mostly pulsed with a

few watts of average power

in near IR.

Mode purely

Single frequency demonstrated.

Mode-locked

Short pulses with with mode-locked pump.

F-Center Lasers

Wavelength

 $2.2 \mu m - 3.2 \mu m$  (commercial device)

 $0.8 \ \mu m - 3.4 \ \mu m$ 

demonstrated using different F-center and various crystals.

Power

Hundred of mW typical; pulsed output

only with pulsed pump.

· Mode purity

Single frequency

Mode-locked

Synchronouly pumped with pulses as short

as 0.3 psec.

Semiconductor Diode Lasers

Wavelength

 $0.65 \mu m$  to  $35 \mu m$  with many different diodes.

mW's; CW not generally

Power output

scalable upward in

power but 1 W CW possible at

low temperature.

Mode purity

Single frequency

Mode-locked

A few picoseconds.

Transition-Metal Ion Doped

Crystal Lasers†

Wavelength

 $1 \mu m - 2.5 \mu m$  (about

1000 - 2000 cm<sup>-1</sup> per

impurity ion)

Power

10 W CW demonstrated

hundreds of watts possible with

several percent efficiency

in modest laser. Several joules/cm<sup>3</sup>

extraction.

Mode purity

Single frequency

Mode locked

Demonstrated in CW laser with subpicosecond possible.

Extended tuning

To ultra-violet and out

to 25 µm with nonlinear techniques

(both CW and pulsed).

Alexandrite:

 $(BeA(_2O_4:Cr^{3+})$ 

Wavelength

701 - 818 nm

Mode purity

Single frequency possible

Mode-locked

Possible but not yed demonstrated.

Power

Hundred of watts average with several pulses per pulse possible. Also CW operation.

70 W avg. power in long pulsed mode demonstrated.

High Pressure Molecular Gas Lasers

Wavelength

Between  $9-12 \mu m$ 

(12-20 µm by Raman shifting

3-4 µm by tripling

Power output

Joules per pulse with

several hundred Watt average

powers possible.

Mode purity

Single frequency

Mode locked

100 psec demonstrated but shorter possible.

<sup>\*</sup> No longer commercially available but can be made to order.

<sup>†</sup> Under development and may be commercially available soon.

### APPENDIX II

Free Electron Laser: Operating Principles, Limitations and Proposed Improvements.

The original theory developed by Madey (1971) was derived from quantum mechanics. However, Planck's constant h vanished from the results in the appropriate limit (laser wavelength large compared with the Compton wavelength) and it was subsequently shown that the same results could be derived from classical mechanics. (Hopf, et al., 1976). Since then, the classical theory has proved to be much more powerful and useful, able to describe nonlinear effects (such as pulse slippage) and plasma effects (Colson and Ride, 1979; Kwan, et al., 1977). Equally important, the classical theory has led to insights on ways to improve the efficiency using rf-accelerator techniques (Kroll, et al., 1979a). Accordingly, we shall adopt the classical point of view here.

Basically, a free electron laser consists of a relativistic electron beam in a periodically alternating, static, transverse magnetic field, called a "wiggler". The period of the alternations is typically a few centimeters, and the length of the wiggler of the order of a few meters. The field strength is typically a few kilogauss. A relativistic electron beam is injected into this magnetic field, the electrons execute forced transverse oscillations, as shown in Fig. 1. When a laser beam is propagated parallel to the electron beam, the oscillating transverse electric field of the laser can do work on the transversely oscillating electrons. When, for example, the laser field is phased relative to the electron oscillations as shown in Fig. 1, the electric field pushes down on the electrons as they are rising and pushes up on them as they are descending. Thus, the electrons are always moving in opposition to the laser field and are decelerated by the interaction.

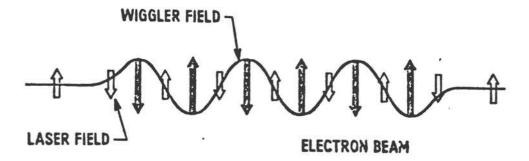


Fig. 1. Schematic diagram illustrating the basic principle of a free electron laser. As the electrons pass through the free electron laser, the periodically varying force of the wiggler magnet (solid arrows) acts on the electrons, forcing them to oscillate transversely. The oscillating field of the laser (open arrows) does work on the oscillating electrons to retard (as shown) or accelerate them, depending on the relative phase of the electron and laser oscillations.

Since electrons cannot exchange energy with a static magnetic field, the energy they lose appears in the laser field. This constitutes the basic mechanism by which energy is transferred from the electron beam to the laser beam. Since the electric field of the laser is rather weak, even for very powerful laser beams, significant energy transfer can accumulate only after many transverse oscillations. This resonance condition is expressed by the equation

$$\lambda_{L} = \frac{\lambda_{W}}{2\gamma_{R}^{2}} \left[ 1 + \left[ \frac{eB\lambda_{W}}{2\pi mc} \right]^{2} \right]$$
 (1)

where  $\lambda_W$  is the wiggler wavelength,  $\lambda_L$  is the laser wavelength, and  $\gamma_R$  is the resonant energy, that is, the energy of an electron travelling at the resonant velocity. Not too surprisingly, the width of the resonance is given by the approximate expression,

$$\frac{\Delta \lambda_{\rm L}}{\lambda_{\rm L}} = 2 \frac{\Delta \gamma}{\gamma_{\rm R}} \cong \frac{1}{2N} , \qquad (2)$$

where N is the number of periods in the length of the wiggler.

Although the basic mechanism for exchange of energy between the laser and electron beams is simple to understand, the production of net gain or absorption is more subtle. The difficulty is that the electrons enter the wiggler in a steady stream, that is, at all phases of the laser radiation. Since the energy gained or lost by the electrons depends on the phase, there is no net gain or

loss. To understand this, recall that in the example discussed above (see Fig. 1), the electron entered the wiggler at that phase of the laser field for which the electrical force was downward when the electron was oscillating upward, upward when the electron was moving downward, and so on. Thus, the electron was decelerated. However, an electron entering the wiggler half an optical period later (or earlier) will experience just the opposite electrical forces and will be accelerated. Thus, there is no energy exchange averaged over many random electrons. This must be so since there is no radiation from a steady current. The only radiation produced is incoherent, spontaneous emission due to the discrete nature of the electrons. This emission is precisely that obtained from synchrotron sources (Kunz, 1979). Net coherent radiation becomes possible when the electrons are formed into bunches, coherently phased with respect to the laser waves. To see how this occurs, we recall that some of the electrons are accelerated by the interaction with the laser, while others - half a wavelength ahead or behind - are decelerated. As the faster ones catch up to the slower ones, they form into bunches spaced at the laser wavelength, which, in the electron frame of reference, is equal to the wiggler wavelength [see Eq. (1)]. With the electron beam formed into coherent bunches, it is now possible to extract net coherent energy. After catching up to the slow electrons, the fast electrons will continue past them, advancing in phase relative to the laser oscillations until they reach the phase, half a wavelength ahead, where the slow electrons started. At this phase the fast electrons will experience deceleration and move backward. Over a period of time, therefore, the electrons will oscillate back and forth, (relative to the mean electron motion) a distance of the order of half an optical wavelength. These longitudinal motions are called synchrotron oscillations. Since they are the result of work done on the electrons over many transverse "wiggler" oscillations, the synchrotron oscillations have a much longer period. The transverse oscillations occur over lengths in the laboratory frame of the order of a few centimeters (the wiggler period). By comparison, even at very high laser powers (of the order of gigawatts) the synchrotron oscillations take place over distances of the order of a meter, in the laboratory frame, corresponding to several tens of wiggler oscillations.

The synchrotron oscillations are analogous to the motions of particles rolling on a sinusoidal potential surface, as shown in Fig. 2. The effective potential, called the ponderomotive potential, is stationary in the rest frame of electrons moving at the resonant velocity. It is therefore moving in the laboratory frame at the velocity corresponding to the resonant electron energy given by Eq. (1):

$$\gamma_{\rm R}^2 \cong \frac{\lambda_{\rm W}}{2\lambda_{\rm L}} \,. \tag{3}$$

The period of the potential is just the optical wavelength in the electron rest frame, which, by Eq. (1), is also the wiggler wavelength in this reference

frame. Using this picture we can more easily understand how net gain and energy extraction occur. When resonant electrons are injected into the wiggler, they are initially at rest in the ponderomotive potential. They are then accelerated and decelerated, but, by symmetry, their average velocity in the resonant frame remains zero for all time. Thus, there is no gain and no net energy extraction for resonant electrons. When electrons are injected above the resonant energy, so that they are moving to the right in Fig. 2, some are slowed down. In fact, those electrons which are initially in the trapped (shaded) regions of Fig. 2 oscillate about the resonant velocity and - after some time - possess, on the average, just the resonant energy. Thus, they have lost the difference between their initial energy and the resonant energy. The energy lost by the electrons appears in the laser field as net gain. Similarly, electrons injected below the resonant energy are accelerated, on the average, toward the resonant energy. These electrons gain energy at the expense of the laser field, corresponding to net absorption rather than gain.

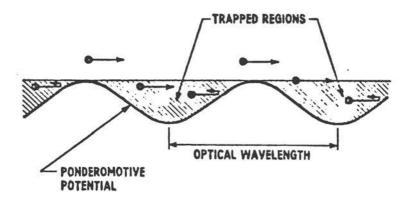


Fig. 2 Schematic diagram of the electrons moving in the ponderomotive potential. Depending on the phase of the electrons with respect to the optical field, the electrons are accelerated or decelerated. The electrons in the trapped regions oscillate longitudinally at the synchrotron frequency and, after many periods, lose all memory of their initial velocity.

Since electrons injected above the resonant energy can, at most, be decelerated to resonance, on the average, the net energy extraction must be less than the difference between the initial energy and the resonant energy. However, the width of the resonance in the wiggler interaction is given by Eq. (2), so that the net extraction efficiency  $\eta_X$  is limited by the expression

$$\eta_{\rm X} \lesssim \frac{\Delta \gamma}{\gamma_{\rm R}} \cong \frac{1}{4N} \,.$$
(4)

Since the number of periods in the wiggler is typically of the order of 100, the extraction efficiency is limited to a few tenths of a percent.

A way to exceed this limitation has recently been proposed by Morton and his coworkers (Kroll, et al., 1979b). As discussed above, near-resonant electrons become trapped in the ponderomotive potential and oscillate about the resonant energy  $\gamma_R$ . Morton's proposal, which is based on techniques used in rf linear accelerators (linacs), is to decrease the value of  $\gamma_R$  by decreasing the wiggler wavelength along the length of the wiggler [see Eq. (1)]. Provided that the decrease takes place slowly enough (that is, adiabatically compared with the synchrotron oscillations), the electrons will continue to oscillate about the decreasing resonant energy. Thus, they will lose, on the average, an amount of energy corresponding to the decrease in the resonant energy from the beginning to the end of the wiggler. Unfortunately, the length of the wiggler is limited by diffraction spreading of the laser beam. Since the decrease, or "taper", of the resonant energy must take place slowly, this places a limitation on the total deceleration of the electrons. Another limitation on energy extraction is imposed by the requirement that the electrons be trapped in the ponderomotive potential. When the "buckets" of the ponderomotive potential, to use rf linac terminology, are decelerated by tapering the wiggler period, some of the electrons spill out, just like water spilling from an accelerated cup, and become detrapped. The faster the buckets are decelerated, the fewer electrons are trapped. The optimum rate of deceleration corresponds to about 40% trapping of the electrons (Brau and Cooper, 1979). This result is well known in rf-linac theory, where it is called the condition of maximum phase flux. Despite these limitations it is projected that more than 10% of the electron energy might be extracted in this way.

Such "tapered" wigglers face several practical difficulties. In the first place, very high laser power is required to trap the electrons. For example, to trap 40% of the electrons and decelerate them 15%, corresponding to a net extraction efficiency of 6%, a peak laser power of 1.5 GW is required (Brau and Cooper, 1979). Because of the dependence of diffraction on wavelength, this result is independent of the laser wavelength, as first pointed out by Kroll. A further disadvantage of tapered wigglers is their very low gain, typically about an order of magnitude smaller than that of comparable uniform wigglers (Brau, 1980). The small-signal gain is low because the electrons, which are not trapped in the small-signal limit, remain near resonance only for a short distance near the wiggler entrance. The large-signal gain is low because of the large saturation flux required to trap the electrons. Because of these difficulties, the concept of a tapered wiggler has not yet been experimentally demonstrated, although several experiments are now being undertaken to do so. Further, tapered wiggler schemes can be used as amplifiers.

Up to this point, the discussion has addressed the behavior of individual electrons in the laser and wiggler fields. When this is a satisfactory approximation, the laser is said to be in the "Compton" regime. However, at sufficiently high electron densities, electron-electron interactions must be taken into account. (Kwan, et al., 1977; Kroll and McMullin, 1977). When plasma

effects dominate, the laser is said to be in the "Raman" regime. Plasma oscillations will become excited in the electron beam when the plasma frequency is large compared with the rate of passage of the electrons through the wiggler. For a cold (monoenergetic) electron beam, this condition may be expressed in the form

$$\frac{\omega_{\rm p}\tau}{\gamma^{3/2}} > 1 , \qquad (5)$$

where  $\omega_p$  is the plasma frequency and  $\tau$  is the transit time of the electrons through the wiggler. The factor  $\gamma^{1/2}$  is used to Lorentz-transform the square root of the electron density (which appears in  $\omega_p$ ) from the laboratory frame to the electron rest frame, where the plasma frequency must be computed. The remaining factor of  $\gamma$  is used to Lorentz-transform the plasma frequency back into the laboratory frame, where  $\tau$  is measured. Under conditions when Eq. (3) is satisfied, negative-energy plasma waves can become strongly excited. The resonant laser frequency is then shifted downward from the value given by Eq. (1) by the plasma frequency, and the gain and extraction efficiency may be significantly enhanced. For uniform wigglers, the saturated extraction efficiency in the presence of plasma effects is given by the approximate formula (Sprangle, et al., 1979),

$$\eta_{\rm X} \cong \frac{1}{4N} \frac{2}{\pi} \frac{\omega_{\rm p} \tau}{\gamma^{3/2}} \,. \tag{6}$$

Comparing this with Eq. (2) we see that the saturated extraction efficiency is enhanced by the factor  $(2\omega_p\tau/\pi\gamma^{3/2})$ , which, by Eq. (3), is large in the Raman regime. Tapered wigglers may also be used in the Raman regime to further improve the energy extraction efficiency. Extraction efficiencies in excess of 20% are predicted.

### APPENDIX III

Developments Since the Final Write-up of the Report (February, 1981)

Since the committee deliberations in December 1980 and the final write-up of the report a number of developments have taken place in the field of free electron lasers. These are summarized below. (May, 1982)

- 1. Output Pulse Width determination The free electron laser output from the Stanford University 3.2 $\mu$ m laser setup has been analyzed for an accurate determination of the pulse width, pulse shape, energy, power, and spectrum (Benson et al, 1982). Using an optical pulse autocorrelation scheme and second harmonic generation in a LiNbO3 crystal, Benson et al. were able to measure the pulse width as a function of detuning of the optical cavity length from that determined by synchronous condition set by the electron bunch repetition rate. Under exactly synchronous cavity length condition, a free electron laser pulse width of  $1.5\pm0.3$  psec was measured. Simultaneous measurements of the frequency spectrum indicated that the output pulses were nearly transform limited. Under these conditions, maximum peak power output of 400 kW was estimated. On detuning the optical cavity from the synchronous length, an increase in the pulsewidth and a corresponding decrease in the spectral width was measured. The measurements allow significant comparisons to be made between theory and experiments.
- 2. Improvements in the mm-Wavelength Free Electron Laser Using an improved electron beam of low emittance, Parker et al (1982) have reported significant improvements in the millimeter wavelength free electron laser. In the collective interaction mode, now achieved by obtaining physical conditions where the Debye length in the electron beam is shorter than the laser wavelength, they report a peak power output of  $\sim 30 \text{ MW}$  in a  $\sim 20 \text{ nS}$  pulse corresponding to a peak energy output of  $\sim 0.68 \text{ J}$ . The laser was operated at a single pass amplifier which amplified spontaneous emission. An energy extraction efficiency of  $\sim 2.5\%$  was obtained and the experimental findings appear to substantiate theoretical calculations.
- 3. Tapered Wiggler Experiments Using a 2.25% magnetic field taper, an experiment for amplifying  $10.6\mu m$  CO<sub>2</sub> laser radiation in a free electron laser amplifier was successfully carried out. Boehmer et al (1982) measured an optical gain of 2.7% and an energy extraction efficiency of 0.07% for a CO<sub>2</sub> laser input power of  $\sim$  20 MW. These numbers are seen to be in reasonable agreement with the theory. In particular, it is noteworthy that the gain and the energy efficiency values measured for the tapered wiggler are an order of magnitude larger than that would have been obtained for a uniform wiggler with otherwise identical conditions.

- 4. Storage Ring Experiments Initial experiments on a wiggler placed within a storage ring were designed to measure optical gain at 4880A radiation obtained from an argon to ion laser. Very promising results are obtained which show a gain of  $4.3 \times 10^{-2}$  % per pass (Deacon et al, 1981).
- 5. Proposed Improvements in Far Infrared Free Electron Lasers It can be shown that when using electron beam sources such as microtrons, FR linacs, etc. where the electron beam current is in the form of short duration bunches, walk-off between the optical pulse and the relativistic electron bunch results in a limitation on the maximum length of wiggler. Such a restriction limits the maximum single pass gain and the power extraction efficiency. Shaw and Patel (1981) have proposed the use of discrete optical filters in the far infrared region to slow down the optical pulse in order to increase the synchronous interaction distance. They show theoretically that in the far infrared region, such methods will prove advantageous in significantly increasing the gain and power output.

As can be seen, significant advances have taken place in the area of free electron lasers since the Committee's deliberations in December 1980 and since the report which was written in February 1981. These developments support the conclusions already arrived at in the committee discussions and indicate that the newly proposed free electron lasers at Los Alamos National Laboratory, University of California, Santa Barbara, Brookhaven National Labs and Bell Labs (among others) will be successfully implemented in the coming one to two years.

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