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Progress in Excimer Laser Technology and Applications

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ABSTRACT

Significant advances have been made in the development of excimer lasers operating from 157 nm in the VUV to the visible with powers from watts to terawatts. This paper discusses the status of excimer laser technology and describe present and emerging applications in various areas such as in materials processing, photolithography, high intensity laser sources, remote sensing, and medicine.

I. Introduction

Excimer lasers generate short pulses of high power ultraviolet (UV), vacuum ultraviolet (VUV), and visible radiation. These lasers may be pumped by optical sources or electrically by discharge with UV or x-ray preionization schemes and electron beam excitation of high pressure (1 to 5 atm) rare gas-halogen mixtures [1,2]. The excitation produces an electrically excited molecule, usually known as an *excimer*, by a three body collision process. High excitation-rate pumping of several 100 kw/cm^3 to several MW/cm^3 is necessary to efficiently produce sufficient gain of the excimer laser of interest in relation to absorption. When the molecule undergoes a radiative transition to its repulsive electronic state, radiation is emitted and the molecule dissociates. Fig. 1 shows the potential energy curves of the XeF excimer. Intense laser emission can be generated by ArF (193 nm), KrF (249 nm), $XeCl$ (308 nm), and XeF (351 and 485 ± 35 nm) with peak powers of 10^5 to 10^7 W for 10-50 ns and terawatt power levels for subpicoseconds. In addition, the same technology can be used to obtain laser emission from $N_2(C-B)$ at 337 nm, N_2^+ $(B \rightarrow X)$ at 428 nm, and F_2 at 158 nm. Typical wall plug efficiencies range from 1.5 to 5%.

The rare gas halide excimer lasers have many attractive features that include:

- Short wavelengths (193, 248, 308, 351, and 485 nm);
- High powers;
- High efficiency;
- Short pulse duration;
- High pulse repetition rate;
- Good spectral and spatial characteristics.

II. Rare Gas–Halide Excimer Laser Technology

The critical elements are the pulsed power, selection of the excimer medium, basic kinetics, optical system, gas management, and materials compatibility [3,4].

1. Discharge Pumping

The discharge pumping technology is well suited to excite high-repetition-rated excimer lasers, which can operate so far with laser output energies of several milijoules per pulse to the joule level at repetition rates of up to several kHz.

A pumping rate on the order of 1 GW per liter of discharge volume is necessary to efficiently produce excimer laser gains. The discharge resistance of the excimer discharge load is typically around 0.2 ohms. Therefore, a typical voltage of 20 kV results in discharge currents of as high as 100 kA. This high current cannot be switched by thyratron switches due to the heavy loadings of the switch. Hence, the primary low power and long pulse is produced in a primary circuit and then compressed in a secondary circuit to a high power level and short pulse, which can efficiently pump excimer lasers. Discharge pumping circuits developed so far are mainly classified into a capacitor transfer circuit, pulse forming line (PFL) circuit, magnetic pulse compressor circuit, or spiker-sustainer circuit, which are shown schematically in Fig. 2.

The spiker sustainer circuit is the most advanced excitation circuit for excimer lasers. A low-impedance PFL sees initially the discharge as being an open load so that the reflection of the voltage pulse develops as a result of the impedance mismatch. To eliminate this unfavorable voltage reflection, the high-voltage high-impedance pulser (spiker) initially breaks down the laser gas mixture and then the other supply (sustainer) maintains the condition of the discharge plasma with an electron energy distribution suitable for efficient pumping of excimer lasers. This scheme is somewhat more complicated than a PFL circuit, but a higher overall electrical efficiency is achieved because of impedance matching for such a circuit. When a saturable inductor is used in series with a thyratron switch, the current-rise rate through the thyratron is reduced so as to minimize the energy dissipation in the thyratron. The allowable current rise rate for a thyratron is less than $10^{11}\ As^{-1}$.

(a) Preionization Technology

To initiate a volumetrically uniform avalanche discharge in a 2-4 atm rare gas-halogen mixture, preionization of the high-pressure mixture prior to the initiation of the main discharge is essential. Spatial uniformity of the preionization in the rare gas-halogen mixture is the most important issue. Especially, its uniformity perpendicular to the discharge electric field is of importance. A preionization electron number density on the order 10^6 to 10^{12} cm^{-3} is experimentally utilized, and is dependent on the preionization strength and gas mixtures.

The simplest and most convenient preionization technology is UV photo-preionization using a photo-electron emission process. The UV photons are generated by means of a pin-arc discharge and a dielectric surface discharge. X-ray preionization technology has also successfully been applied to high-pressure excimer lasers and is used effectively in large-scale excimer lasers.

(b) High-Repetition-Rate Operation

High-repetition-rate operation of excimer lasers is desirable for high average-power generation. For this purpose thyratrons have been commonly used as switching elements. The operational performance of such switches is one of the repetition-rate limiting issues.

Because of rapid progress in both pulsed power technology and laser gas purification, operation at repetition rates of up to 5 kHz and average laser powers at the 1 kW level have been demonstrated separately.

For achieving long operational lifetimes for an excimer laser exciter, an all-solid-state circuit appears to be a future promising solution. High-power semiconductor switches are now commercially available, but maximum ratings such as hold-off voltage, peak current, and current-rise rate cannot yet fulfill the stringent switching requirements necessary for efficient excimer laser excitation. Therefore, additional voltage transformer circuits and a magnetic pulse compression circuit are needed.

In addition to a high-repetition-rate exciter, gas purification and aerodynamic technologies such as a fast gas flow circulation system and an acoustic damper at repetition rates exceeding the multi-kilohertz range are required to realize long-life high-repetition-rated operation of excimer lasers. A long operational shot life of up to 10^8 shots has already been demonstrated to date.

2. Electron–Beam Pumping

The pulsed-power technology involved in the efficient and spatially uniform generation of intense (high current-density) relativistic e-beams from relatively large aperture diodes has advanced significantly in recent years. The e-beam generation technology is scalable in energy so that high-energy excimer lasers are more readily reliazable with e-beam pumping than with discharge pumping.

The relevant components of a cold cathode e-beam diode for excimer laser pumping are shown schematically in Fig. 3. The e-beam diode consists of a cold cathode to which a pulsed high voltage (negative polarity) of 30 to 1500 ns duration is applied, a thin foil anode, and a pressure foil separating the high-pressure laser gas mixture from the vacuum diode chamber. These foils are made of 50 μm to 75 μm thick titanium foil or aluminized polyimide film. A support structure for the foils is a Hibachi-like assembly to withstand the pressure differential between the high pressure of the laser gas mixture and the diode chamber which is typically evacuated to very low pressures of less than 10^{-3} torr. Intense relativistic electron beams with current densities of 10 to 1000 A/cm^2 and electron energies of 0.3 to 2 MeV can be used to pump excimer lasers. Such high current-density e-beams can be generated from cold cathode-type diodes made of carbon felt, or multi-blades of tantalum or titanium foils, which can enhance the local electric field strength up to typically 1 MV/cm. The operational characteristics of such a cold cathode-type diode obeys the Child-Langmuir law.

A variety of e-beam-pumped excimer laser configurations have been operated successfully. Four major geometries are shown schematically in Fig. 4. The relative merits of these geometries can be evaluated in terms of energy scaling, the aspect ratio of the pumped region, uniform pumping, e-beam utilization, and output performance for a given pulsed high-voltage generator. To improve the e-beasm energy deposition into laser mixtures, an external magnetic confinement field can be applied. The depth of the pumped region is determined by the electron penetration depth for a given electron accelerating voltage.

III. Control of Excimer Laser Radiation

1. Temporal Characteristics

Excimer lasers have typical pulsewidths of 10 to 150 ns at repetition rates to several kHz and extend today from subpicoseconds to microsecond pulse durations. Long pulse excimer lasers with reduced intensity can efficiently deliver excimer laser energies through quartz fibers.

2. Spatial Properties

Because of the high gain and wide gain bandwidth of excimer lasers, high-Fresnel-number stable resonators, which are commonly used to efficiently extract the available laser energy, produce a laser output with high spatial divergence. A low divergence laser beam from excimer lasers can be obtained by using a confocal unstable resonator with a large magnification or by simply using a low-Fresnel-number stable resonator. The use of an unstable resonator can efficiently extract laser output with high beam quality, while the use of low-Fresnel-number stable resonators results in a decrease in the available laser energy. Rare gas-halide laser beams with divergencies within a factor of one or two of their diffraction limits are obtainable without difficulty by the use of high-magnification confocal unstable resonators.

3. Spectral Characteristics

With the exception of XeF, rare gas halide excimers have excited states with large binding energies and relatively flat ground states. They thus give rise to emission spectra of relatively narrow spectral width ($\Delta\lambda \sim 1$ nm) near the peak of their fluorescence. In the case of $XeF(C \rightarrow A)$, the wavelength is tunable over a wavelength range of almost 100 nm [5].

The linewidth of excimer lasers can be reduced, and tunability can be realized using gratings, prisms, etalons, or some combination of these elements in the laser resonator. Spectral linewidths of less than 10^{-3} nm have been achieved. The ability to extract energy in a narrow bandwidth depends upon the distribution of energy among the vibrational levels of the excimer and the rate of vibrational relaxation within the manifold. The output power of narrow-bandwidth excimer lasers is usually much too low for most applications, and further amplification is generally necessary. The wavelength capability of excimer lasers can be considerably extended by a number of techniques, including optical pumping of other laser media (e.g., dyes) and stimulated Raman scattering in high pressure gases (e.g., H_2, D_2). (See Fig. 5 and Ref. 6.)

4. Injection Control

For some applications requiring high power levels, narrow bandwidths and low spatial divergence injection control by means of a tunable or fixed wavelength low power laser can be utilized. To fulfill these requirements, use of either a master-oscillator/power amplifier system or an injection-locked resonator is the most convenient method (Fig. 6). Input pulse can be generated by a master oscillator typically consisting of a cavity with a line-narrowing function. An input power level on the order of 10^{-3} or less of the output laser level is effective. High-power laser extraction is more readily achievable as a by-product with injection locking than without injection because the injected seed pulse assists the build-up of the intracavity flux and reduces energy loss. This effect is more effective in shorter duration lasers.

In injection-locked excimer lasers, an input pulse can be generated by an entirely different kind of laser. When radiation generated from a dye laser, an Ar ion laser, a frequency-doubled YAG laser, a semiconductor laser, or an excimer laser is injected into an excimer amplifier after one or more stages of frequency conversion, a highly controlled laser pulse with desirable spectral, spatial, and temporal beam quality can be amplified to a high energy level. For example, use of a cw single-mode dye laser provides a narrow band seed signal whose wavelength is tunable over the bandwidth of the excimer laser. A system of this type has been demonstrated for the $XeF(C \rightarrow A)$ laser [7]. The extremely wide bandwidth of 80 nm of this laser was continuously tuned with a linewidth of less than 0.001 nm. (See Figs. 7 and 8.) Beam divergencies within a factor of three of the diffraction limit were achieved.

IV. Applications

Numerous excimer laser applications in science, industry, and medicine have been reported. The major science areas include:

- Spectroscopy and nonlinear optics;
- Pumping of other lasers;
- Ultrashort pulse amplification;
- Laser fusion.

A number of potential industrial and medical applications are in various stages of development.

- Materials processing,
  - Microelectronics,
  - Polymers and ceramics,
- Photochemistry;
- Isotope separation and enrichment;

- ∗ Remote sensing,
- ∗ Medicine,
  - Cardiology,
  - Ophthalmology.

1. Microelectronics

In optoelectronics excimer lasers have been shown to be useful in material processing of semiconductors, polymers, ceramics, glass, metals, and compound materials. This includes ablation, etching, deposition, doping, annealing, and lithography [8,9].

In laser direct-writing applications, scanning focused laser beams are used to "write" small scale structures as metal interconnects, high-aspect-ratio via holes in microelectronic chips and chip packages. UV light is necessary because of its ability to initiate specific photochemical reactions, to form submicrometer structures, and to couple effectively with semiconductor components. Also, UV light offers the potential for high spatial resolution in a process of interest. This property is especially important in applications relevant to future DRAM chip generations with increased integration density. Furthermore, materials often respond differently to UV light, thus allowing the observation of effects such as laser ablation. A further use of high repetition rate UV lasers is in the growth of thin film materials (e.g., diamond, superconductors).

(a) Photolithography

The excimer laser is an ideal light source for ultraviolet photolithography, primarily because of its high spectral brightness, which can easily provide exposure doses two orders of magnitude greater than provided by conventional light sources. Although this has been recognized for some time, a principal difficulty has been the unavailability of photoresist materials suitable for use in the ultraviolet region of the spectrum (200-350 nm). However, new concepts in polymeric materials and in photosensitive compounds provide the basis for a new generation of photoresist materials that can be tailored for response in the 200-350 nm region. The coupling of these photoresist materials with excimer laser light sources should provide sufficient resolution and throughput to serve a high fraction of the microelectronics industry output for the next 10 years.

(b) Etching and Deposition

Several potential applications of lasers in integrated circuit fabrication involve only surface heating and melting (i.e., in the absence of photochemical reactions). In two examples, large areas of a wafer are exposed to pulsed UV excimer radiation. In one case, a metal surface in a multi-level interconnect structure is melted in order to planarize it, prior to subsequent processing. In the second example, the silicon wafer surface is melted to incorporate adsorbed dopant into the melted zone. These applications demand long pulse widths (~ 100 ns) to prevent ablation, good spatial uniformity ($\sim \pm 1\%$) that does not degrade over the operating life of the laser, and corresponding pulse energy reproduceability.

(c) UV Photo-Assisted Epitaxy

This has come to encompass a wide range of materials including the $II-VIs$, $III-Vs$, Si, and Ge. Photoassisted epitaxy can take the form of a photoenhanced thermal decomposition of precursors bringing about a UV modulation of the epitaxial properties or a purely photolytic deposition process.

Optimization of these processes will require a choice of laser and precursor that are matched not only for efficient absorption but also to ensure the most suitable decomposition path. The laser wavelength must also avoid photochemical reactions with products that may cause the incorporation of impurities.

To exploit the many advantages of UV laser assisted photoepitaxy, versatile laser sources will be required that will produce more than one wavelength, enabling one to "switch" from one specific reaction process to another and hence modulate the layer composition, etc. For the exploitation of UV lasers in epitaxy, it is essential that the lasers have excellent beam stability and uniformity, and that the instrument be reliable.

(d) Diagnostics: Gas Phase and Surface

Many applications of laser diagnostic techniques to semiconductor processing have been developed. As an example, atomic oxygen and chlorine have been detected by two-photon laser-induced fluorescence (LIF). Narrow band, injection-locked excimers permit high resolution optical spectroscopy.

Laser diagnostic techniques in direct semiconductor fabrication technology may be used for etch endpoint detection and may provide many advantages, such as cost, sensitivity, and selective measurement of defined areas, over conventional endpoint measurement techniques. For example, As has recently been detected by exciting the atom *in situ* with 193 nm (ArF) radiation. This simple excitation technique may also provide endpoint detection for exposure of n-type Si films with very high sensitivity.

With the availability of reliable excimer lasers, an increasing number of fundamental studies of inorganic as well as organic reactions are being made. The major barrier to a "renaissance" in effort is laser cost and complexity of operation. The absence of a viable industrial photochemical process (except for Vitamin D synthesis) inhibits more wide-spread research. However, it is anticipated that commercial applications of UV lasers in photochemistry will become prevalent in the next 10 years. The most likely candidate appears to be the uranium isotope separation process being pursued in Japan. Less likely, but also promising, is the development of a process for europium separation (under investigation in at least two countries). Other potential processes, which have been shown to be feasible in the laboratory but not yet developed at the industrial level, include platinum metal separation, synthesis of fine pharmaceuticals, and purification of high value industrial chemicals, such as silane or polymer precursors.

2. Materials Processing of Polymers and Ceramics

Processing materials with UV and VUV laser light is another important application of excimer lasers. This includes photoablative processing of polymeric materials, laser driven etching, etc., for electronics applications [10]. Fig. 9 shows the sheet conductivity of irradiated polyimide and PBI samples as a function of laser shots on the sample for three different fluences. The sheet conductivity of polyimide increases from $10^{-15}\,\Omega^{-1}\,\mathrm{cm}^{-1}\,\mu\mathrm{m}$ for the nonirradiated sample to $1\,\Omega^{-1}\,\mathrm{cm}^{-1}\,\mu\mathrm{m}$ after 6000 shots [11]. Equally important is the processing of metals, ceramic composites, and optical materials.

Potentially important applications exist in shaping ceramics and composite (metal matrix and ceramic matrix composite) materials. The use of UV lasers leads to smoother, stronger, and more predictable surfaces for laser shaped ceramics and composites.

3. Ultrashort Pulse Amplification

The broad bandwidth afforded by excimer lasers makes these systems promising candidates as amplifiers of ultrashort laser pulses. Although direct generation of ultrashort excimer laser pulses by using either active or passive mode locking is limited to its pulsewidth on the order of or slightly shorter than 1 nsec due to the short gain duration, successful ultrashort pulse generation using visible dye lasers and their wavelength conversion technologies have made it possible to generate high-power subpicosecond UV and visible laser radiation using excimer gain media as amplifiers [12-14]. The highest laser peak power of 4 TW in a 390 fsec pulse has been obtained by amplifying a frequency tripled short pulse dye laser generated in a synchronously pumped dye laser stage by three discharge pumped KrF amplifiers and an e-beam pumped KrF amplifier [15]. The shortest excimer laser pulse of 80 fs was obtained by amplifying a frequency doubled short pulse dye laser which was generated in a distributed feedback dye laser cavity pumped by a cavity quenched dye laser pulse.

Since the storage time of excimer lasers are 1-2 nsec, saturation energy is typically 1-2 $\mathrm{mJ/cm^2}$, which is much lower than the values of other high-power lasers. Therefore, an amplifier with large aperture is required to amplify a laser pulse to very high energy levels. In such a system, amplified spontaneous emission (ASE) generated from the final large aperture amplifier tends to reduce the aspect ratio of the output laser pulse.

When focusing subpicosecond excimer laser pulses generated in these systems, intensities reaches well above $10^{17}\ \mathrm{W/cm^2}$, and with future developments power densities of $10^{20}\ \mathrm{W/cm^2}$ will be attainable with laboratory sized systems. High-order harmonic generation induced by short pulse excimer laser pulses can occur at wavelengths down to the XUV wavelength region (see Fig. 8). Recently, a 9.9 nm pulse was obtained by 25th-harmonic generation with a picosecond KrF laser [16]. An ultrashort UV probe continua was also generated through self-phase modulation (SPM) induced in high peak power excimer lasers focused into high-pressure gases. A TW-level excimer laser will be able to generate continuum pulse in the XUV spectral range.

4. Medical Applications of Excimer Lasers

Medical applications of UV lasers are currently found in the following areas:

1. Laser ablation of tissue, generally involving an excimer laser and, at present, most attractive for use in ophthalmology and cardiology [17];

2. As radiation sources in the 250-420 nm range for photomedicine experiments involving large areas of tissue;

3. Selective, pulsed UV laser photochemistry of biological molecules.

Of the three, the use of the UV laser in cardiology and laser angioplasty has received the most attention. Atherosclerotic disease is treated by two major methods: heart by-pass surgery and balloon angioplasty with over 200,000 procedures per year. Although both techniques have a good success rate, it has been recognized that additional, less invasive methods ought to be developed to complement the two primary treatments. To this end, the concept of a laser vaporizing atherosclerotic plaque was suggested in the early 1980's. This appeared attractive, since one could actually remove the obstruction rather than displace it, as is done with balloon angioplasty.

Pulsed UV excimer ablation showed promise in that, with the proper choice of wavelength, pulse energy, and pulse width, one could achieve precise vaporization of all tissue types of plaque without thermal damage at the edges of the tissue. However, problems with mutagenic effects at UV wavelengths, healing — if a given technique removes the plaque, will the vessel become reoccluded in a short time or will it stay open — and the question of whether the laser steerable catheter fiber optic can be engineered in such a way that the typical physician can use it safely, are being addressed presently.

Aside from laser angioplasty, other promising applications of UV lasers lie in ophthalmology. The use of excimer lasers in re-sculpting the eye has been demonstrated (see Fig. 13). This will make it possible to eliminate eye glasses in the future.

V. Acknowledgments

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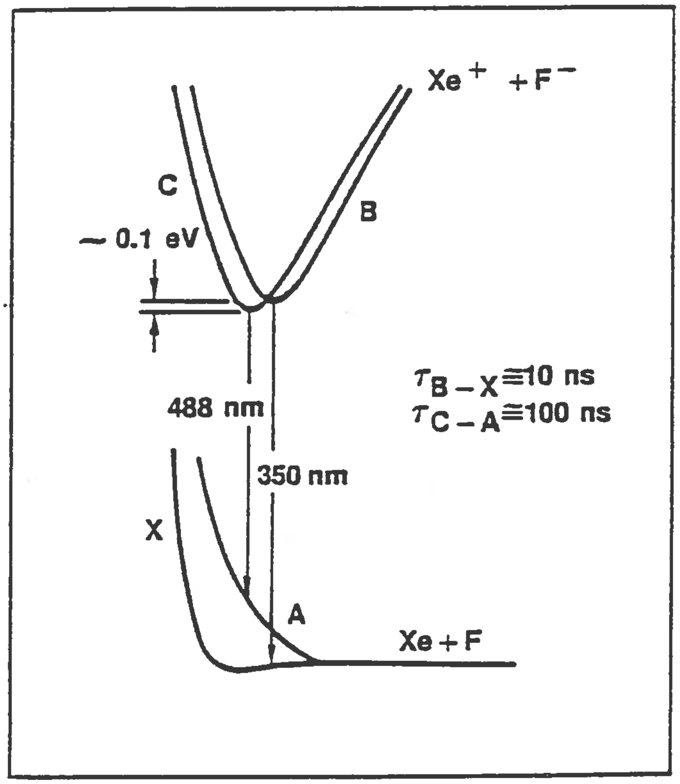

Fig. 1: Energy as a function of interatomic separation for the excimer XeF, showing both the B→X and the C→A transitions.

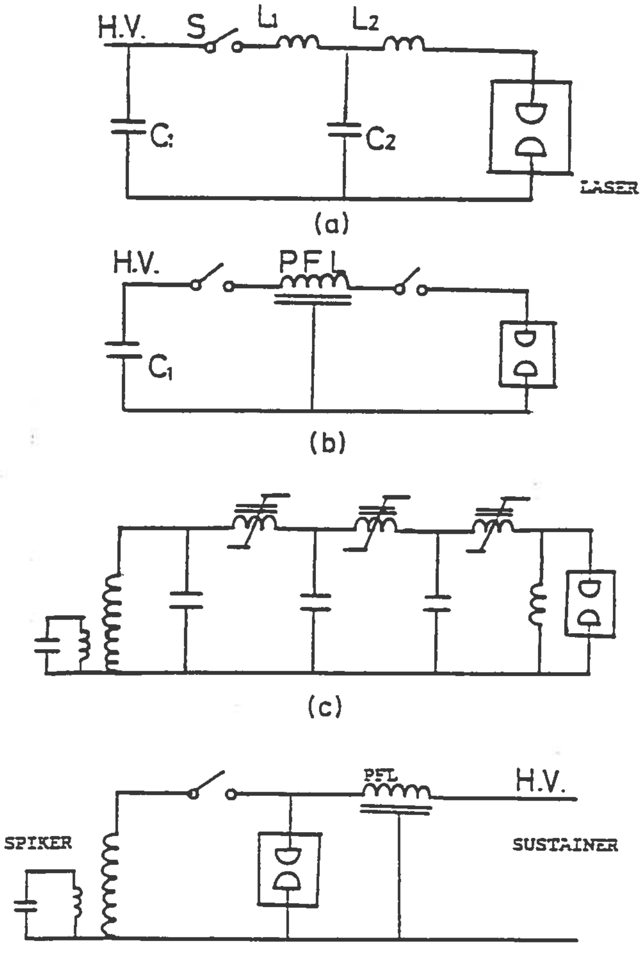

Fig. 2: Schematic of widely used excimer laser excitation circuits. (a) Charge transfer circuit, (b) pulse forming line (PFL) circuit, (c) magnetic pulse compressor circuit, and (d) spiker sustainer circuit.

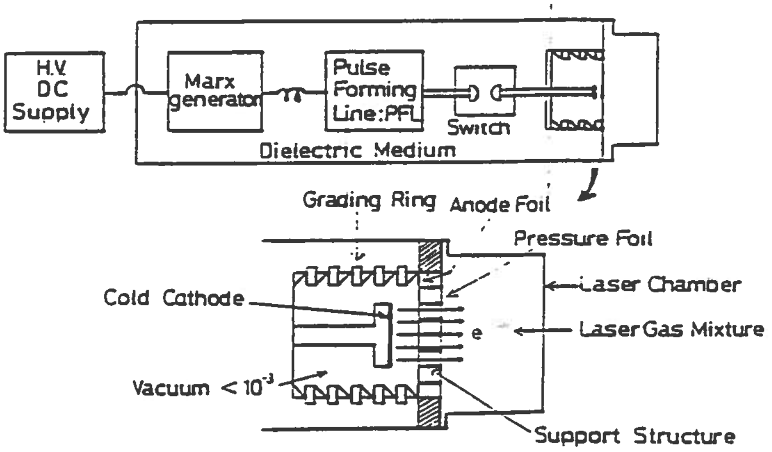

Fig. 3: Schematic diagram of a typical e-beam pumping system for excimer lasers. [Ref. 2]

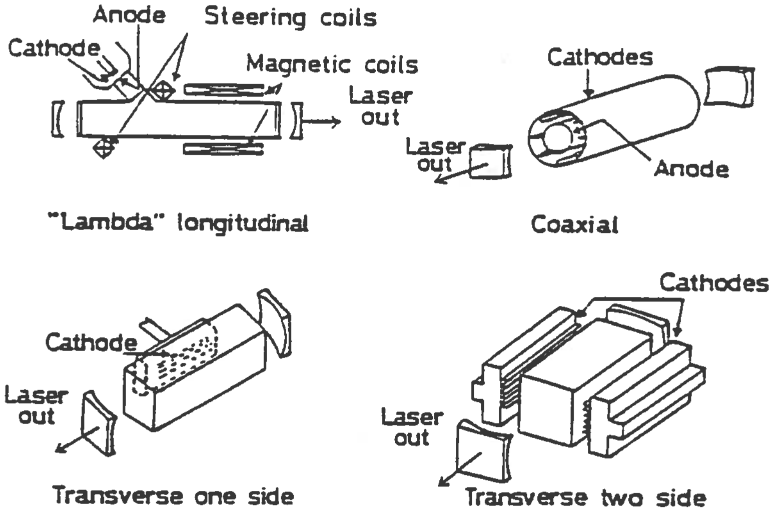

Fig. 4: Schematic of four major layouts for e-beam pumped excimer lasers. [Ref. 2]

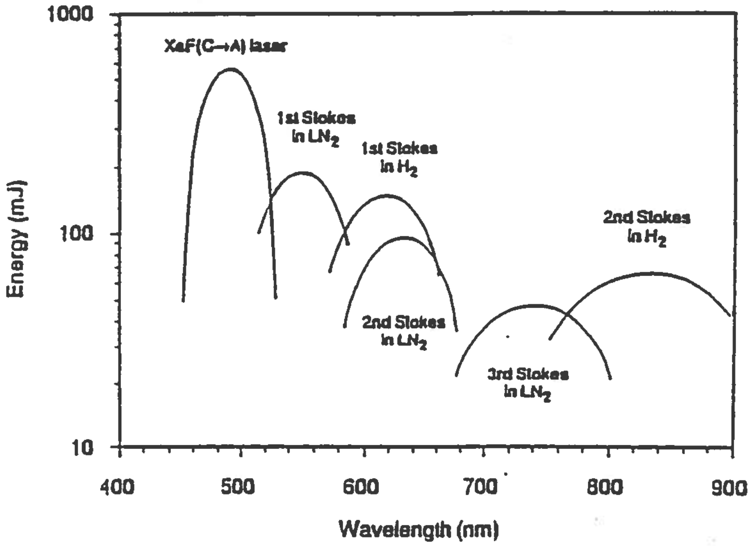

Fig. 5: Spectral range accessible by the XeF(C→A) excimer laser and Stokes lines in liquid nitrogen and hydrogen. [Ref. 6]

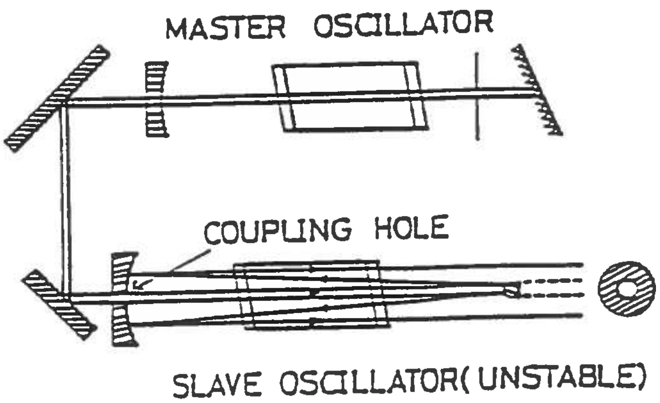

Fig. 6: Schematic of an injection-locked resonator (unstable). [Ref. 2]

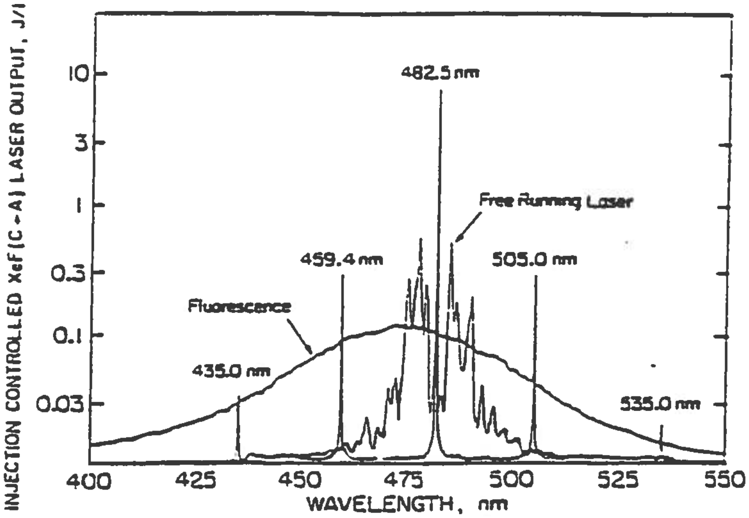

Fig. 7: Qualitative comparison of superimposed time integrated spectra of the XeF(C→A) fluorescence, the injection-controlled output of five separate shots at several wavelengths, and a typical free-running oscillator spectrum, all for representative conditions. [Ref. 5]

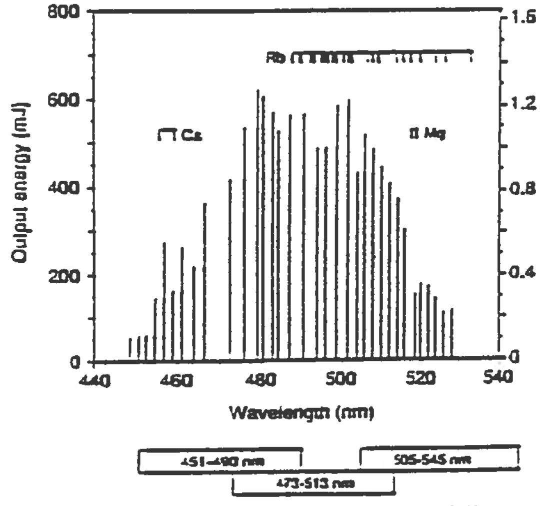

Fig. 8: Output energies for the injection-controlled XeF(C→A) laser between 450 and 530 nm showing a tuning bandwidth of 50 nm FWHM centered at 490 nm. The dye laser injection intensi was 2 MW/cm^2 with a spectral linewidth of 0.005 nm. The wavelengths shown were chosen not to coincide with narrow-band atomic absorptions occurring in this spectral region. Three different resonator coatings and four injection laser dyes were used to span the XeF(C→A) tuning bandwidth

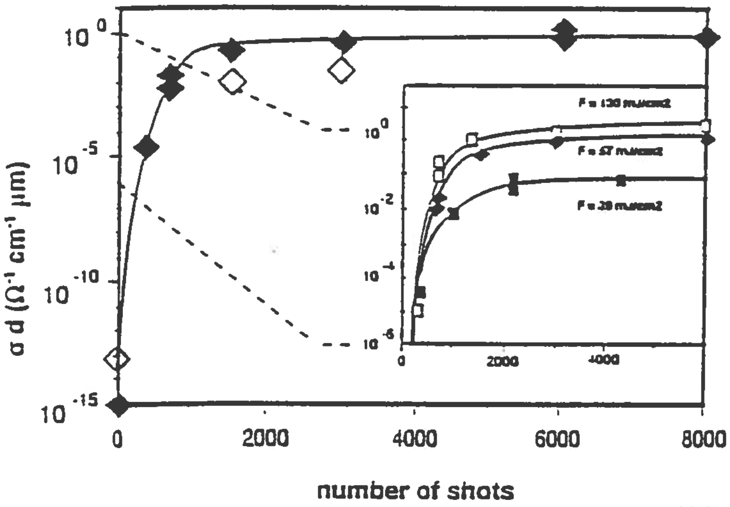

Fig. 9: Sheet conductivity for polyimide (◆) and PBI (◇) as a function of the number of laser shots for a fluence of 57 mJ/cm^2. The conductivi changes over 15 orders of magnitude for polyimide The inset shows that the saturation sheet conduct ity depends on the fluence per laser shot.

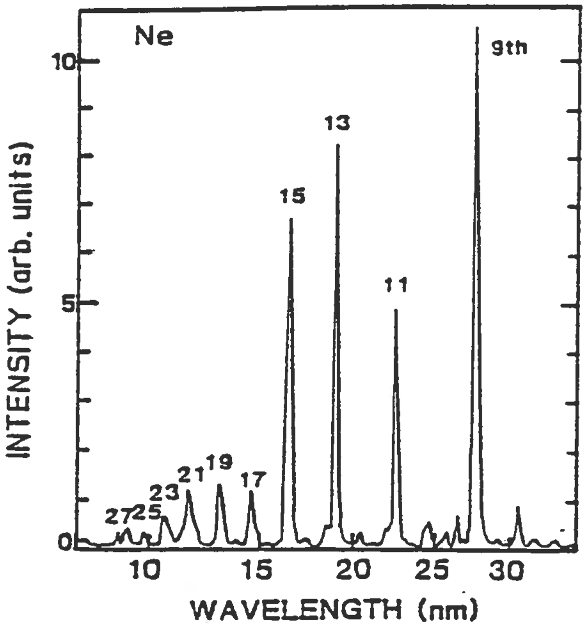

Fig. 10: Harmonic spectrum for Ne. A 100-mm-thick boron filter was used. The laser intensity was 4 x 10^{17} W/cm^2, at a gas density of $\sim$7 x 10^{17}cm^{-3}. [Ref. 15]

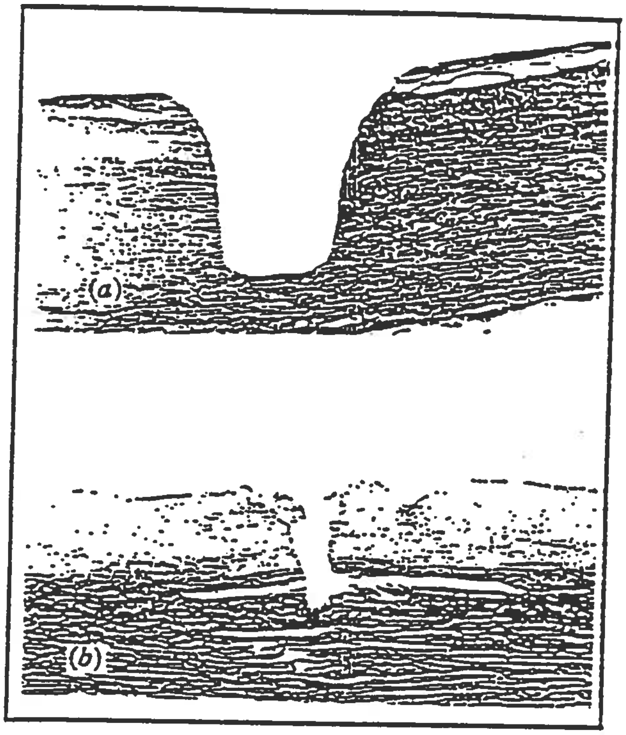

Fig. 11: Segments of human aorta ablate with (a) an excimer laser, and (b) a pulsed Nd:YAG laser (7 ns pulses). The excimer laser ablation is seen to be clean and has no thermal or acoustic damage, while the Nd:YAG laser ablation is seen to show only tearing, characteristic of acoustical damage.

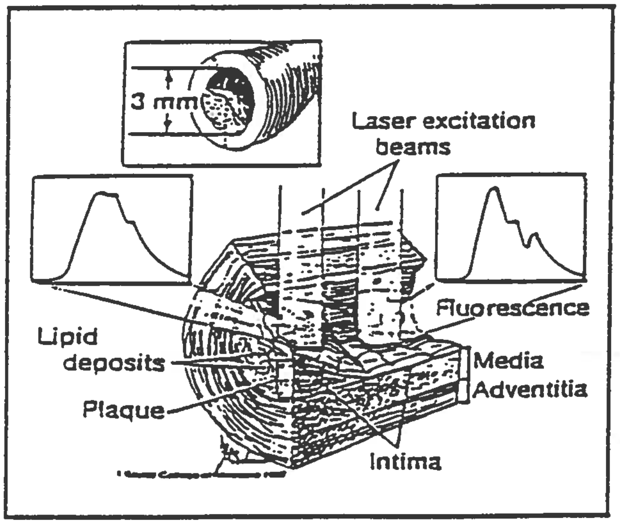

Fig. 12: An illustration of a segment of artery showing hypothetical differences in laser induced fluorescence from healthy and atherosclerotic tissue.

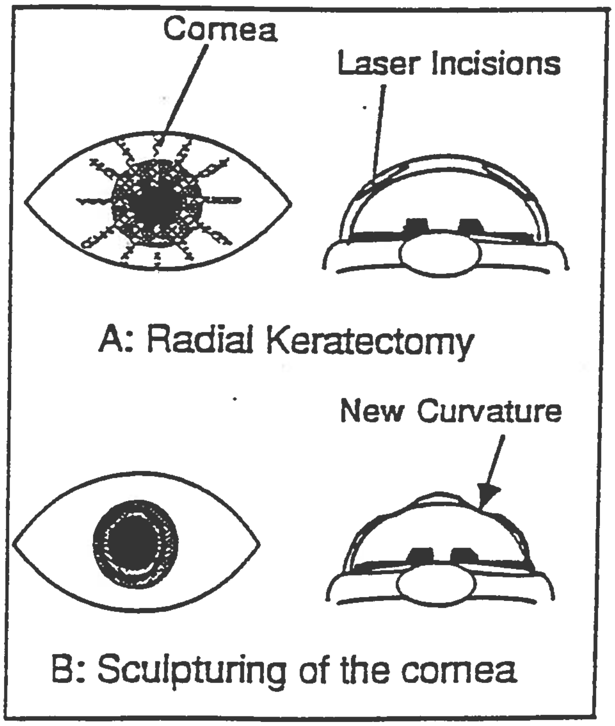

Fig. 13: Illustrations to show the two photorefractive correction procedures. In radial keratectomy (a), shallow radial incisions are made from the center allowing the cornea to relax, and thus decrease in curvature. In sculpturi of the cornea (b), concentric circles are grafted in the cornea to increase or decrease the curva of the cornea. In this figure the concentric circles are used to increase the curvature of the cornea.