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## Applications of soft x-ray lasers.

C. H. Skinner

Princeton University, Princeton, NJ 08543

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### ABSTRACT

The high brightness and short pulse duration of soft x-ray lasers provide unique advantages for novel applications. Imaging of biological specimens using x-ray lasers has been demonstrated by several groups. Other applications to fields such as chemistry, material science, plasma diagnostics, and lithography are beginning to emerge. We review the current status of soft x-ray lasers from the perspective of applications, and present an overview of the applications currently being developed.

### 1. INTRODUCTION

Applications to conventional lasers have certainly changed the way we live, although it took time for the most widespread applications to be developed. In fact, it took a surprising number of years for the laser itself to be developed, considering that Einstein<sup>1</sup> first described the principles of stimulated emission in 1916. Though the concept of feedback oscillation was known to engineers and the technology of some lasers existed long before the demonstration of the ruby laser in 1960, the information was fragmented between different groups of people and lasers had to wait for the invention of the maser before the possibilities became obvious.<sup>2</sup> For conventional lasers the most significant applications were found between the boundaries of disparate fields, subjects as different as lasers and printing which few would have thought to be connected at all before the invention of laser printers.

We are in the midst of an on-going revolution in soft x-ray sources, optics, and applications that has been the subject of several recent reviews<sup>3,4</sup> and conferences.<sup>5</sup> X-ray lasers now exist at wavelengths from 3.5 to 32nm and have produced saturated output close to the diffraction limit. Their peak brightness is extremely high with equivalent black body radiation temperatures of up to 6 GeV, *seven* orders of magnitude brighter than the spontaneous emission lines from the same plasma. The unparalleled brightness of soft x-ray lasers together with their short pulse duration offers many opportunities for applications in diverse fields such as biological imaging and photochemistry. A workshop dedicated to exploring x-ray laser applications was held in 1992 and the proceedings<sup>6</sup> describes many applications in detail. We present here an overview of current and emerging x-ray laser systems from the perspective of applications and review the range of applications currently being considered and developed for x-ray lasers.

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## 2. CURRENT X-RAY LASER PERFORMANCE

The current performance of x-ray lasers is shown in Table 1. The high peak brightness of x-ray lasers as compared to other sources is emphasized here, however this should not be taken to imply that x-ray lasers are universally 'better'. Applications needing a high average power will be more suited at present to synchrotrons or laser produced plasmas. Harmonic generation is also beginning to emerge as a practical source in the XUV region. However, for applications demanding the highest brightness sources at x-ray wavelengths, the x-ray laser is ideal.

A crucial factor for applications is the availability and cost of these devices. Presently, the x-ray lasers with the highest brightness are dependent on large scale Nd lasers at inertial confinement facilities that are expensive and hard to access. However a major effort to develop 'tabletop' x-ray lasers is in progress at several laboratories. The key issue here is the overall x-ray laser efficiency. This can be clearly seen in conventional lasers where the most widespread applications use CO<sub>2</sub> and diode lasers that are also the most efficient. The overall efficiency of x-ray lasers is a product of three factors: the driver efficiency, the efficiency in generating gain, and the efficiency in extracting energy from the laser and we will discuss these factors in turn.

A class room sized Nd laser has been designed to pump collisionally excited lasers.<sup>7</sup> This design takes advantage of the latest advances in laser technology to generate 1kJ beam energy at a much lower cost than NOVA. It is interesting here to see the strong correlation of efficiency and cost in the design. Increased

X-ray laser wavelength	≈	Brightness photons/ mm <sup>2</sup> mrad <sup>2</sup> sec (0.01%BW)
23 nm (Ne-like Ge)	≈	10 <sup>24</sup>
20 nm (Ne-like Se)	≈	10 <sup>24</sup>
18.2 nm (H-like C)	≈	10 <sup>21</sup>
15 nm (Ne-like Y)	≈	10 <sup>24</sup>
4.5 nm (Ni-like T)	≈	10 <sup>21</sup>
For comparison:		
equivalent radiation temp.	≈	Mev-Gev
NOVA Nd laser (1.06μm)	≈	5 x 10 <sup>19</sup>
Advanced Light Source	≈	10 <sup>20</sup>
Short Pulse	≈	70 psec - 30 nsec
Output energy	≈	1-8 mJ (15-23 nm)
	≈	10μJ (4.5 nm)
Spectral linewidth ( $\Delta\lambda/\lambda$ )	≈	10 <sup>-4</sup>
Repetition time	≈	20 sec. - 90 min.
Energy efficiency	≈	10 <sup>-9</sup> - 10 <sup>-5</sup>

Table 1, Current X-ray Laser Parameters

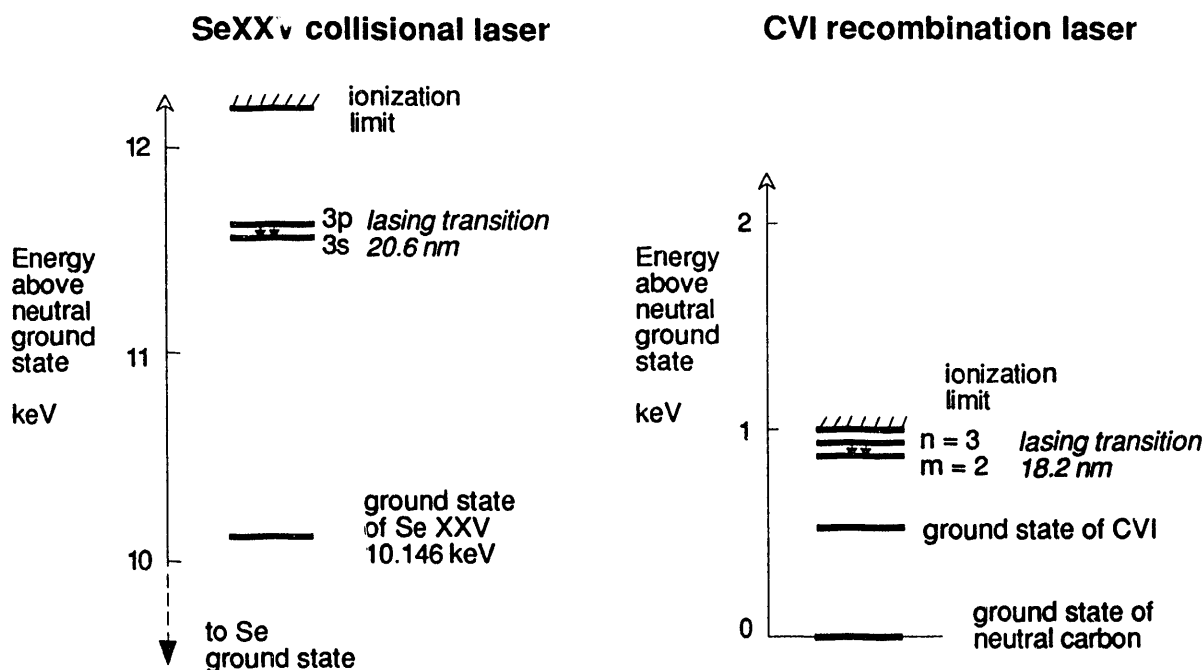


Fig. 1 X-ray Laser "Quantum Efficiency"

efficiency from techniques of multipass, chirped pulse amplification and improvements in optics and energy storage in the glass amplifiers all make the system more affordable for applications. A very different approach is to use a capillary discharge to efficiently generate a plasma appropriate for a x-ray laser and progress in this direction is reported in these proceedings.<sup>8</sup>

Although the recombination pumped systems have not yet equaled the high brightness achieved by collisional x-ray lasers they are very attractive in terms of their higher quantum efficiency in producing gain. This is illustrated in Fig. 1 which shows that the total potential energy needed to produce the upper lasing level in the CVI recombination laser is an order of magnitude lower than that needed for the Se XXV collisional laser. An additional advantage for recombination systems is the faster scaling to shorter wavelengths with increasing nuclear charge,  $Z$ , since the lasing transition involves a change in principal quantum number. The obstacle in achieving higher brightness in recombination systems has been the difficulty in maintaining a high gain coefficient over multi-centimeter plasma lengths. This could potentially be due to inhomogeneities in the driver laser beam, refraction of the x-rays out of the gain region, or the development of plasma instabilities.

At Princeton, work is proceeding to develop a tabletop recombination x-ray laser at 18.2 and 13.5 nm. High gain (up to  $7.1\text{cm}^{-1}$  in a 6 mm target) has been measured with a pump laser energy of only 4J.<sup>9</sup> Two separate targets are being used to double the gain-length. This technique has already been demonstrated in collisional systems<sup>10</sup> and should enable us to overcome any deleterious effects that occur at long plasma lengths. Recently, in Li-like sulfur, gain of  $2.5\text{cm}^{-1}$  on the 5g-4f transition was maintained when the plasma length was increased from 1 to 2 cm and work to increase this to 4 cm is in progress.<sup>11</sup> Another approach to the problem of maintaining gain over longer plasma lengths in small scale systems is to develop a tabletop collisional pumped system and progress in this direction is reported in these proceedings.<sup>12</sup>

The third issue in x-ray laser efficiency is the efficient extraction of the potential energy of the inverted population in a x-ray laser pulse. Significant progress has been made in double passing the laser beam through the gain medium with multilayer mirrors and in addressing problems of mirror damage.<sup>13</sup> The development of a negative branch unstable resonator cavity for x-ray lasers would enable a micro-radian beam divergence and extend the current high brightness of x-ray lasers by several orders of magnitude.<sup>14</sup> However, a key issue here is in extending the duration of the x-ray laser pulse to permit many round trips in the cavity and since the input driver energy is limited, this will also be dependent on improvements in the efficiency of generating gain.

## 2. X-RAY LASER APPLICATIONS.

The development of applications of x-ray lasers is following the same path as applications of conventional lasers with the first applications being to a preexisting field. For example, in 1960 Xe lamps were used to repair detached retinas, however the advantage of the newly invented ruby laser in this procedure was quickly recognized. The more surprising applications of conventional lasers, such as CD players and supermarket scanners came much later. In the same way the most developed application of x-ray lasers is to imaging, building on the experience gained using synchrotrons in soft x-ray microscopy, however more novel applications will surely emerge as x-ray lasers become more accessible. Developing x-ray laser applications is inherently an interdisciplinary activity and a workshop was held in 1992 with the specific aim of bringing together leaders in different fields to explore the opportunities and issues of x-ray laser applications.<sup>6</sup> This overview will summarize a sample of some of the work presented there and elsewhere.

### 2.1 X-ray Microscopy.

X-ray microscopy offers the opportunity of obtaining images of cells at resolutions much higher than available from light microscopy and free from possible artifacts<sup>15</sup> due to the intensive specimen preparation necessary in electron microscopy. The potential for x-ray lasers to be used in biological imaging of living cells was recognized early on<sup>16</sup> and the first images of cervical cancer cells obtained using a x-ray laser were reported in 1990.<sup>17</sup> Images obtained with a x-ray laser showing the effects of protein tagging of sperm were presented in this conference.<sup>18</sup> X-ray imaging of biological specimens has also been performed with synchrotrons and laser produced plasmas.<sup>19</sup> The special advantage of x-ray lasers in this field lies in their extremely high brightness. To obtain a high resolution image of a moderate contrast object it is necessary to absorb or scatter a large number of energetic x-ray photons. For example; to image a 50 x 50 x 200 nm pixel, with 4.3 nm photons and a 10% combined imaging and detection efficiency, the number of photons absorbed is  $\approx 10^4$  and the associated heating can be more than enough to boil water and destroy the specimen. In synchrotron based x-ray microscopy, with an exposure time of the order of seconds to minutes, this problem is alleviated by convective cooling which limits the temperature rise. However the potential for radiation damage to migrate to an adjacent pixel during the exposure needs to be carefully checked. The advantage of both x-ray lasers and laser produced plasmas is that the exposure time can be short enough that the image is "inertially confined" i.e., obtained in a psec - nsec exposure time before features in the specimen have time to move one resolution element. The appropriate time scale was investigated by London<sup>20</sup> who found a maximum pulse length of 50-220 psec to obtain a hologram of a biological sample in an aqueous environment with  $r = 50$  nm resolution. This limit scales rapidly with  $r^3$ , the third power of the desired resolution, and puts extreme demands on the source brightness at the highest resolutions. X-ray lasers

with a source brightness equivalent to a radiation temperature in the MeV to GeV range are an obvious choice. In addition to the extremely high brightness, the monochromaticity and directionality are ideally suited to x-ray optics such as zone plates and Schwarzschild objectives and to three-dimensional imaging. For holographic imaging the coherence of x-ray lasers is essential.<sup>21</sup> Current limitations of x-ray lasers are the absence of continuous tunability (although a large number of discrete lines are available) and the difficulty of access for biologists. The latter issue is motivating a major effort to develop tabletop x-ray lasers.

Besides conventional transmission x-ray microscopy it is also possible to form an image from reflected x-rays with potential application to high resolution inspection of lithographic products and the observation of surfaces (or membranes) of biological cells. A proof-of-principle experiment at Princeton has demonstrated imaging of a test grid in reflected x-rays from a soft x-ray laser beam.<sup>22</sup> Chemical contrast has been demonstrated in absorption microscopy by exploiting the sharp edges in the x-ray absorption spectra.<sup>23</sup> The same sharp variations of optical constants that form edges in x-ray absorption spectra can also provide chemical contrast in reflection microscopy. For example, calcium pyrophosphate deposits in articular cartilage are associated with chronic osteo-arthritic disease. Fig. 2 shows the large difference between the reflectivity of calcium and protein using 18.2 nm light at 10° incidence.<sup>24</sup>

The reflection imaging x-ray laser microscope will have important applications in materials sciences for example in the characterization of semiconductor heterojunctions, for investigating buried interface structures, and the detection of precipitates inside silicon surfaces. The electronic properties of heterojunctions, namely metal-, semiconductor-, and insulator-semiconductor interfaces, depends critically on the morphology of the interface. This morphology can be drastically affected by chemical reaction and interdiffusion that take place during interface formation or during subsequent processing steps. Extensive mixing at the interface introduces new chemical phases with electronic properties different from those of the substrate and overlayer. Chemical reactions induce defects that can act as recombination centers and affect carrier flow or band bending at the interface. Thus a detailed characterization of the structure of these interfaces is extremely useful. Very few techniques are suitable for investigating buried interface structures. Most of them such as transmission electron microscopy, are destructive in the sense that the sample must be drastically modified, thinned down or cleaved. The reflection microscope, however, will require no sample preparation and the optimal wavelength range: 10-20 nm, makes soft x-ray lasers the ideal source. It should be particularly powerful in the visualization of features and contrasted interfaces buried under overlayers 10-20 nm thick. A resolution of 50 nm is insufficient to provide an 'atomic' picture but should provide valuable information on large scale inhomogeneities that exist at some interfaces. The microscope can also be applied to the detection of precipitates inside silicon surfaces. During annealing of high dose implants, (such as As or P) clusters can form within the near surface region of silicon when the dopant concentration exceeds solid solubility. These clusters do not contribute to the desired electrical conductivity and can nucleate other defects during subsequent processing, so their detection is very important. Fig 3 illustrates the difference in reflectivity at 18.2 nm of pure silicon and silicon with a 10 nm thick layer of As buried 10 nm below the surface.

## 2.2 Plasma Diagnostics.

Plasma regions at the high electron densities ( $\approx 10^{21} \text{cm}^{-3}$ ) have been inaccessible to plasma diagnostic techniques based on conventional lasers since the plasma frequency is close the laser frequency. Recently X-ray lasers have been applied to probe this regime and high resolution images and electron density profiles are reported elsewhere in these proceedings.<sup>25</sup> Although these plasmas emit copious x-rays, the plasma self emission is completely negligible compared to the x-ray laser intensity.

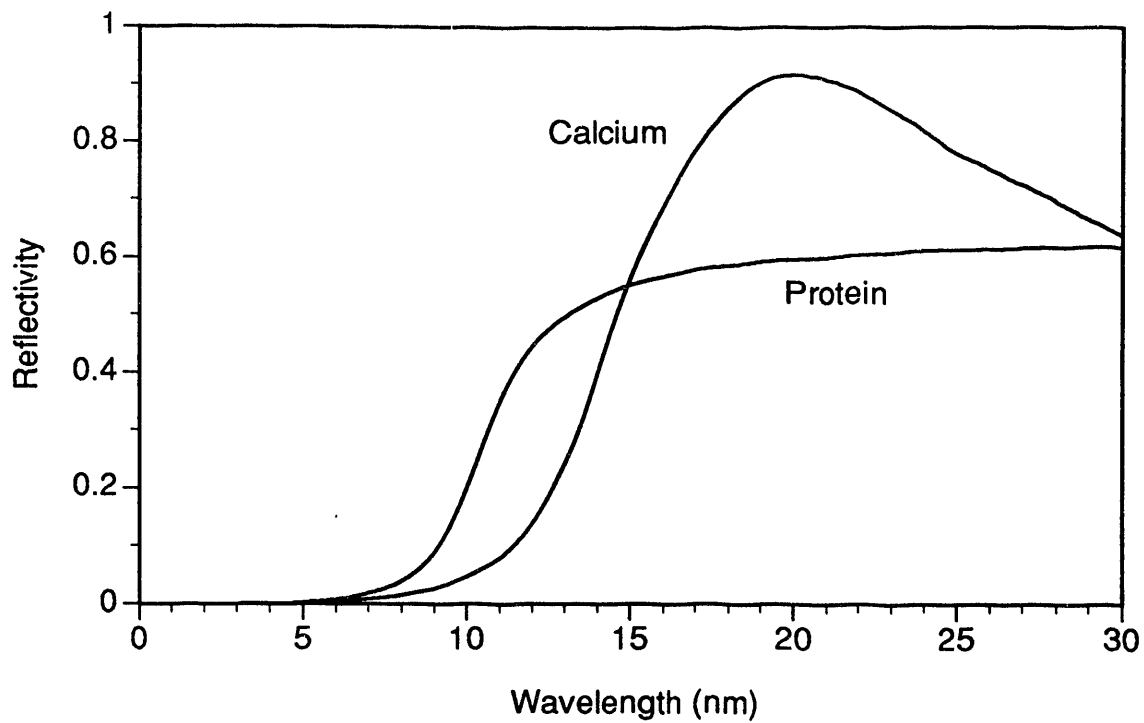


Fig. 2 Reflectivity at 10° incidence showing chemical contrast in reflection soft x-ray microscopy.

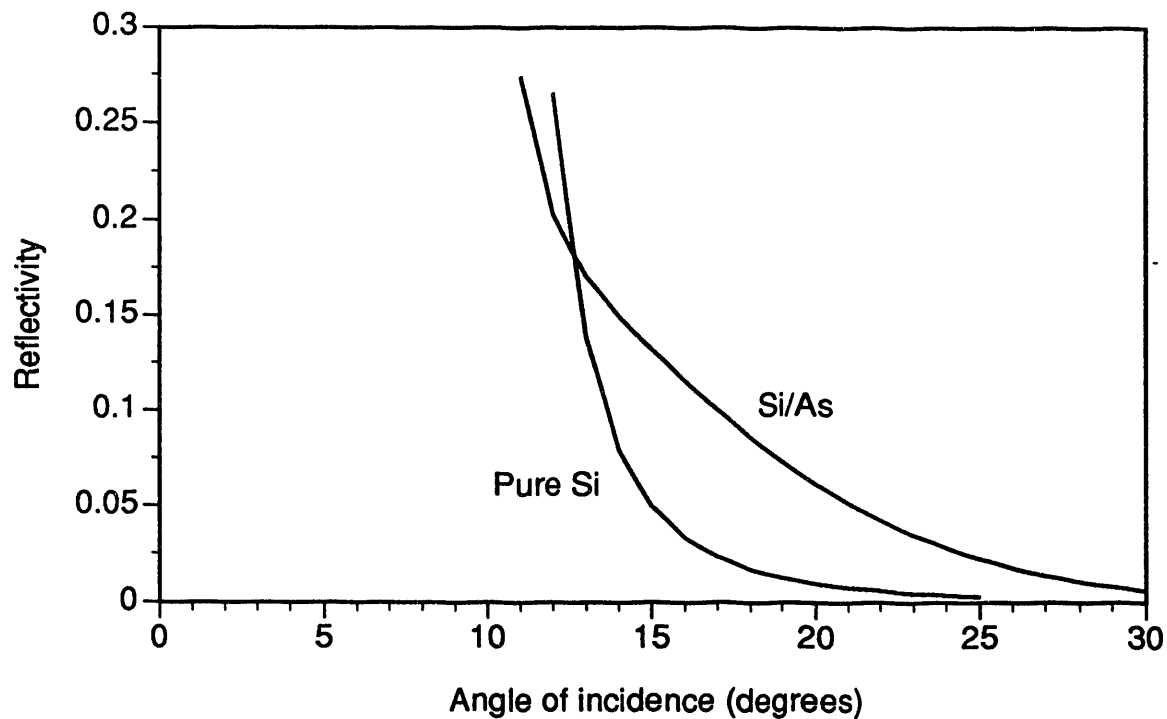


Fig. 3 Reflectivity of silicon with a 10 nm layer of As buried 10 nm below the surface and for comparison the reflectivity of pure silicon.



### **2.3 Non-linear x-ray optics, Resonance fluorescence, Atomic physics codes**

Harmonic generation and frequency conversion in the x-ray regime can be studied with x-ray lasers.<sup>26</sup> Besides extending the science of non-linear optics this also offers the possibility of producing shorter x-ray wavelengths and continuously tunable coherent x-rays that would extend the range of potential applications. Other physics applications to resonance fluorescence are being explored.<sup>27</sup> An application to the development of atomic physics codes used in plasma modelling was reported at the conference.<sup>28</sup> Several unique features in the Ne-like Ti laser at 32 nm had led to the suggestion that it was photo-pumped by emission from C-like and N-like Ti.<sup>29</sup> However, the incorporation of hyperfine structure in emission lineshapes was found to be essential in modelling the performance of the Ti laser and it is now believed to be collisionally pumped.

### **2.4 Photoionization Dynamics and the Chemistry of Clusters.**

Applications to chemistry are emerging particularly for soft x-ray lasers in the 20-50 eV range. One example is to photoionization dynamics. The Frank-Condon principle separates the treatment of nuclear and electronic motion in molecules, however this principle breaks down in molecular photoionization due to trapping of the photo-electron in the molecular potential. It has been suggested that the high brightness, pulsed nature of soft x-ray lasers may be exploited in pump/probe experiments in which a specific molecular state is prepared with a pulsed IR laser and then photoionized with the x-ray laser.<sup>30</sup> Other possible experiments include studies of the chemistry of transient molecular clusters.<sup>31</sup> Clusters bridge the gap between molecules and solids and the majority are produced by laser vaporization of a target material in a pulsed carrier gas making a continuous source of x-rays such as a synchrotron inappropriate.

### **2.5 Applications to Microelectronics.**

The semiconductor fabrication industry has seen an inexorable progression to optical lithography using ever shorter wavelengths to meet the demands for increasing miniaturization of devices. X-ray lithography will be required to progress beyond the intrinsic limits of optical lithography. The minimum acceptable print rate of  $1\text{cm}^2\text{sec}^{-1}$  leads to a requirement on average x-ray power on the silicon wafer of 20 mw that is most easily met, at present, by laser produced plasmas.<sup>32</sup> High efficiency discharge pumped x-ray lasers may be a potential contender in the future, however there are other lithographic applications where current x-ray lasers could play an important role. The high spatial and temporal coherence of present x-ray lasers will be very useful in x-ray interferometric testing of the extreme precision aspheric optics needed for projection x-ray lithography. X-ray laser microscopy can also be applied to the inspection of masks and wafers at a fabrication facility. A combined holographic and reflection microscope has been proposed for such an application.<sup>33</sup> The high energy density and  $\approx 50\text{ nm}$  focal spot of focussed x-ray lasers could be applied to micro machining, such as connecting or disconnecting individual circuits on future custom ICs with sub  $0.1\mu\text{m}$  features.

Holographic nano-lithography on semiconductors has also been proposed.<sup>34</sup> Up to now electronic device operation can be largely described by classical physics, however quantum effects become important when feature sizes decrease below 100 nm. Posts 6 - 7 nm diameter and 50 nm high have been fabricated by electron beam lithography, however quantum confinement has been difficult to observe due to electrostatic fields due to surface states. An alternative is a periodic array of holes however exact periodicity is difficult to

maintain with e-beam fabrication. A coherent x-ray laser may be able to produce a highly periodic structure with dimensions below 100 nm in a way exactly analogous to the present fabrication of holographic gratings with visible lasers.

### 3. PROSPECTS FOR THE NEAR FUTURE.

X-ray lasers are in a transition phase between the successful generation of ultra high brightness beams at a range of x-ray wavelengths and their broad acceptance by a community of users as practical and useful tools for a variety of applications. In the near term, progress in extending the duration of x-ray laser gain and in developing hardened x-ray mirrors will result in the achievement of cavity oscillation and diffraction limited output beams. An exciting recent development has been work on recombination x-ray lasers pumped via optical field ionization that potentially operate on transitions to the ground state.<sup>35</sup> The order of magnitude higher quantum efficiency offers the prospect of high repetition rate x-ray lasers pumped by small scale low energy driver lasers ideal for many applications.<sup>31</sup> Some exciting experimental results in this area have been reported recently.<sup>36</sup> We also may anticipate important biological insights arising from the application of x-ray laser microscopy to hydrated specimens. Increasing demands for further miniaturization in microelectronics will continue to stimulate applications to microlithography. As the precedent of conventional lasers shows, the most novel applications are likely be found in the boundaries between completely disparate fields. The most crucial issue for applications is the development of small scale 'tabletop' x-ray lasers that can be comfortably accommodated at individual researchers' laboratories and within their research budget. The rate of progress in this regard in the U. S. has suffered from recent funding cuts. The precedent of conventional lasers and applications makes it clear however, that provided sufficient funds are made available, there will be many unanticipated discoveries and surprisingly useful applications of these devices.

### 4. ACKNOWLEDGMENTS

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