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D. C. Eder, P. Amendt, P. R. Bolton,
L. B. Da Silva, L. A. Hackel, R. A. London,
B. J. MacGowan, D. L. Matthews, M. D. Rosen,
R. E. Stewart, and S. C. Wilks

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Applications and source development for high-repetition rate x-ray lasers

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Lawrence Livermore National Laboratory, Livermore, CA 94550

ABSTRACT

Many applications in material science, chemistry, and atomic physics require an x-ray source that has a repetition rate of 1 Hz to a few kHz. In these fields, a very wide range of photon energies is of interest. One application is time-resolved surface photoelectron spectroscopy and microscopy where low energy ($< 1\mu\text{J}$) pulses are required to avoid space charge effects but high-repetition rates ($\approx 1\text{kHz}$) provide the high average power which is needed to obtain the desired resolution. In pump-probe experiments, it is desirable to have the repetition rate of the x-ray source be comparable to the repetition rate of the corresponding IR, optical, or UV laser. We show that the very high-repetition rate of synchrotrons (1-1000 MHz) results in an inefficient use of x rays for these types of experiments and that a kHz repetition rate x-ray laser would be an excellent source for many experiments. For some applications, a slower repetition rate of order 1 Hz is adequate provide the energy per pulse is larger ($\approx 1\text{ mJ}$). For example, in photoelectron spectroscopy of free clusters, an x-ray laser can provide the required large number of nearly mono-energetic photons during the short time the clusters can be probed in each formation cycle. In the context of source development, we discuss conventional collisional x-ray laser schemes with mJ output pulses that can operate at Hz repetition rates using a high average power driving laser. The near term prospects for high-repetition rate x-ray lasing for photon energies below 100 eV are very good with higher energy capabilities expected in the future. In addition, prospects of table-top size x-ray lasers with kHz repetition rates are presented.

1. INTRODUCTION

Many time-resolved and pump/probe experiments, as well as studies of short lived objects, require a source of x rays that have temporal properties that are not available from synchrotrons or current x-ray lasers. In the case of synchrotrons, the quasi-continuous temporal output property associated with very high-repetition rates ($> 1\text{ MHz}$) result in relatively low number of useable x rays for these experiments. In contrast, the majority of current x-ray lasers have very low repetition rates and do not provide the required high average power.^{1,2} The repetition rate of x-ray lasers is set by the driving laser and at most laboratories this is generally of order one shot per hour. Recent advances in optical laser drivers will allow a large increase in repetition rates to about 1 Hz in the next few years. These new drivers will be of modest size, fitting on large optical tables (6 m by 10 m), with of order 1 kJ per pulse. This energy is sufficient to pump conventional collisional x-ray

lasers with wavelengths down to approximately 12 nm (100 eV). Advanced x-ray laser designs using multiple or shaped pulses³⁻⁵ show promise of lower energy requirements and shorter wavelengths. In addition to this approach for increasing repetition rates using "conventional" x-ray laser schemes, new approaches to x-ray lasing using high intensity but low energy (<1 J) lasers are being tested. These approaches could provide μ J pulses at kHz repetition rates.⁶ In both of these cases, the average power is comparable to or greater than what will be available from the Advanced Light Source (ALS) synchrotron facility for a 10^{-4} bandwidth.⁷ The unique attributes of these new x-ray lasers can aid in the understanding of dynamical processes in surface layers, the structure and chemistry of free clusters, and general material properties studied with pump/probe techniques.

2. TIME-RESOLVED AND PUMP-PROBE EXPERIMENTS ON SURFACES

There has been a number of synchrotron radiation experiments on surfaces that could be extended or improved by using a high-repetition rate x-ray laser. We briefly discuss constraints associated with doing these experiments with synchrotrons and discuss one experiment studying time-resolved emission from GaAs surfaces. A feature of synchrotrons is that multiple beam lines can make use of the radiation from the circulating electrons. Multiple users allow for better utilization but if the properties of the ring are changed to suit one beam line, all of the users are affected. In particular, to conduct time-resolved or pump-probe experiments it is desirable to have the synchrotron operate with a single electron bunch as compared to using twenty or more bunches. The use of single bunches gives the largest number of photons per synchrotron pulse, but significantly reduces the time average number of photons available in all the beam lines. Even in single bunch mode, the repetition rate is still very high, for example, at the National Synchrotron Light Source (NSLS) at Brookhaven the repetition rate is 53 MHz in normal operation and 1.8 MHz in single bunch mode.⁸ However, the repetition rate of lasers used in pump-probe experiments is often in the kHz range, which means only a fraction of the synchrotron pulses are utilized.

Despite these problems some interesting experiments have been conducted at NSLS and other synchrotrons. In a set of experiments conducted at NSLS, Long and co-workers studied time-resolved photoemission from GaAs surfaces that were irradiated with 5 ns laser pulses.⁹ They used a 6 kHz copper vapor laser ($\lambda = 510$ nm) as the pump. Control of the laser pulses caused the synchrotron pulses to arrive at different times relative to the peak of the laser pulse allowing time-resolved spectra to be obtained. Their work showed that the broadening of core-level lines from the decomposed surface originates from nonuniform band bending and not from chemical disorder. The optimum x-ray source for this type of experiment would have a narrow bandwidth to resolve the core-level spectrum and a repetition rate that is comparable to that of the pump laser, i.e., 6 kHz. Tunability of the x-ray source is not required because the spectrum is determined by measuring the energy of the photoelectrons. The required energy per pulse (<1 μ J) is small because a large number of photoelectrons results in a significant space charge above the surface. For a large space charge, the energy of the ejected electrons is shifted and accurate measurement of core-level binding energies can not be obtained. This experiment and

many others requires photons with energies less than 100 eV. A high-repetition rate x-ray laser meets all of these requirements and would be an excellent source for time resolved and pump-probe experiments.

3. PHOTOELECTRON SPECTROSCOPY OF FREE CLUSTERS

In the study of short lived objects, it is essential that the x rays arrive during the time that the object is present in the detector. One example of short lived objects are free clusters that are produced by laser vaporization of a target material in a pulsed carrier gas. Cluster properties bridge the gap between the electronic properties of solids and those of molecules. They are important in the study of catalytic reactions and surface chemistry. While a few clusters, e.g., C_{60} and C_{70} , can be produced in macroscopic quantities, the majority can only be obtained by laser vaporization. This pulsed formation process leads to a short "duty cycle" (percentage of time that clusters are in the interaction region of a detector) that is generally only 10^{-4} , making quasi-continuous sources of x rays, e.g., synchrotrons, inappropriate. In addition, the low density of clusters requires x-ray pulses with energies in the mJ range to obtain even a few photoelectrons during a cluster formation cycle. Ultraviolet valence photoelectron spectroscopy of free clusters has already been demonstrated using ultraviolet lasers.¹⁰ The advantage of core-level x-ray photoelectron spectroscopy is that it will be possible to analyze more complex structures because core levels are simpler to interpret than valence levels. An exciting application of using core levels is to study how reactants bond to the clusters. As for the case of surface photoelectron spectroscopy, a wide range of materials are of interest with a corresponding range of binding energies for the core electrons. In particular, there are many elements that can be studied with photon energies less than 100 eV.

4. CONVENTIONAL X-RAY LASERS WITH HIGH REPETITION RATES

Current x-ray lasers are laser heated, high temperature, high density, elongated plasmas. Lasing has been established using different methods to populate the upper-laser level including collisional excitation from the ground state and collisional cascade following recombination.² For collisional excitation, Ne-like and Ni-like ions with 10 and 28 bound electrons, respectively, are widely used. X-ray lasers have been well characterized in the current, essentially single shot, mode of operation. For example, the output energy for a Ne-like Y laser at 15.5 nm (80 eV) has been measured to be 8 mJ in a 200 psec duration pulse¹¹ and the spectral line width of the Ne-like Se laser at 20.6 nm (60 eV) has been observed to be gain narrowed with a bandwidth ($\Delta\lambda/\lambda$) of less than 10^{-4} at saturation.¹² The output properties per pulse would be approximately the same if operated at a high-repetition rate.

Conventional x-ray lasers require relatively high pump energy (≈ 1 kJ/pulse). Recent advances in optical lasers, using multiple-pass zig/zag slab amplifiers combined with phase conjugators and pulse compressors, show that compact high energy, high-repetition rate lasers can now be built. We have a laser system under construction with an energy of 150 J per pulse that will provide approximately 150 pulses at 1 Hz followed by a 20 min cool

down time for the amplifier. This laser will have very good beam quality allowing for the possibility of creating a narrow line focus. This reduces the energy requirements necessary to achieve the required high intensity for x-ray lasing. This beam quality is achieved, in part, because the pulse after completing half of the passes through the amplifier is reflected by a stimulated Brillouin scattering (SBS) phase conjugator and sent back for the final passes through the amplifier.¹³ The phase aberrations that accumulated as a result of residual thermal distortion of the amplifier slab and phase errors in the ring optics are subtracted out as the pulse retraces the initial path after reflection by the phase conjugator. The pulse duration during amplification is approximately 10 nsec. Shorter pulses with the same energy can be created by using a SBS pulse compressor.¹⁴ To achieve shorter wavelength x-ray lasing, we have designed a 1 kJ per pulse system. This would provide 150 to 200 pulses between cool down times of order 20 minutes. If laser diodes are used to pump the slab, rather than flash lamps, a factor of 3 enhancement in the number of pulses is obtained. The repetition rate for a package of pulses (≈ 500 for diode pumping) is variable in the range of 1 to 10 Hz. The average repetition rate is found by averaging over the cool down time giving approximately 0.5 Hz using diode pumping. In addition to using a narrow line focus, another way to reduce the energy requirements of the driving laser is to use multiple or shaped pulses. Recent experiments in Ne-like Y using three 100 psec pulses separated in time, as compared to a single 500 psec pulse with the same intensity, measured comparable x-ray intensities but of shorter duration.⁴ In summary, the prospects are very high for creating an x-ray laser with mJ pulses and a repetition rate of order 1 Hz in the near future.

5. TABLE-TOP APPROACH TO X-RAY LASING

To achieve very high-repetition rates (≈ 1 kHz), x-ray laser schemes must be developed that require much less energy per pulse to keep the average power requirements on the driving laser at a reasonable level. One approach is the optical-field-ionized (OFI) laser on which recombination following optical field ionization by a high-intensity ($\approx 10^{17}$ W/cm²) short duration (≈ 100 fsec) laser pulse creates the population inversion. High efficiencies have been calculated for wavelengths ≥ 10 nm in Li-like ions.^{6,15} For example, field-ionizing Ne to the He-like ionization stage allows for lasing in the $3d_{5/2} - 2p_{3/2}$ transition in Li-like Ne at 9.8 nm. The short pulse duration and the longitudinal focus geometry give low input energy requirements (< 1 J) despite the need for a high intensity pulse. Driving lasers with this energy per pulse currently have Hz repetition rates but 100 Hz or even kHz lasers should be available in the future. The calculated x-ray output energy per pulse is in the μ J range giving an averaged power that is comparable to conventional x-ray lasers with mJ pulses running at 1 Hz. The rapid and smaller pulses of the OFI plasma x-ray laser are important for photoelectron spectroscopy of surface, as discussed above. Experiments are in progress at a number of laboratories to test this scheme over a range of wavelengths from 10 to 30 nm. Time-integrated experiments at the RIKEN laboratory in Japan showed exponential growth over short lengths in H-like Li at 13.5 nm that is suggestive of lasing.¹⁶ However, length studies are difficult in the confocal geometry of this scheme. The predicted short duration of lasing compared to the recombination time scale makes time resolved measurements essential. Subsequent time resolved measurements in H-like Li at

Berkeley also showed some exponential growth for short lengths but efforts to see exponential growth over longer lengths with corresponding large gain-length products has not been obtained at this time.¹⁷ A major issue that needs to be addressed for this x-ray laser scheme is the propagation of the longitudinally focused pump beam including the effects of refraction. The field ionization leads to an electron density profile that is similar to the transverse profile of the laser beam in overall width, but the detailed shape depends on the ionization thresholds of the different ionization stages. The corresponding electron density gradient can result in significant refraction of the ionizing pulse which limits the length of pumped plasma. The use of larger focal spots reduces refraction, but increases the energy requirements of the scheme.¹⁵ Another requirement of this x-ray laser scheme is that the electron temperature immediately following ionization be low to allow for rapid recombination. Low electron temperatures are calculated following field ionization if short (≤ 100 fsec) pulses are used to minimize secondary heating mechanisms. Continuing experiments will address these issues. There are some other approaches to table-top x-ray lasing including using lower Z Ni-like ions, recombination schemes using a series of pulses, and schemes based on capillary discharges.² Most of these systems could operate at high-repetition rate when developed. While the prospect for table-top lasing with large gain lengths and kHz operation is not as assured as increasing the repetition rate of conventional x-ray laser schemes, it remains high.

6. SUMMARY

There are many new applications, requiring a range of wavelengths, where a high-repetition rate is desired. One technique where x-ray lasers can play an important role is photoelectron spectroscopy. The narrow bandwidth that is inherent in x-ray lasers is required to achieve resolution of core levels and the discrete tunability can be used to study different levels. Continuous tunability, not yet available for x-ray lasers, is not required because the measurement of the energy of the ejected electron combined with a known photon energy determines the binding energy of the core level. In surface photoelectron spectroscopy, low energy pulses are required to control space charge effects and high-repetition rates are required to obtain the desired resolution. In photoelectron spectroscopy of free clusters, a lower repetition rate is acceptable provided the individual pulses have more energy. The prospects for having x-ray lasers with the desired output properties for these new applications in the near future are high. In addition, the advent of table-top size x-ray lasers will allow these experiments to be conducted at small laboratories and universities.

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