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Optical Pumping Experiments on Next Generation Light Sources

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ABSTRACT

Laser-based plasma spectroscopic techniques have been used with great success to determine the line shapes of atomic transitions in plasmas, study the population kinetics of atomic systems embedded in plasmas, and look at the redistribution of radiation. However, the possibilities for optical lasers end for plasmas with $n_e > 10^{22}\text{cm}^{-3}$ as light propagation is severely altered by the plasma. The construction of the Tesla Test Facility (TTF) at DESY (Deutsche Elektronen-Synchrotron), a short pulse tunable free electron laser in the vacuum-ultraviolet and soft X-ray regime (VUV FEL), based on the SASE (self amplified spontaneous emission) process, will provide a major advance in the capability for dense plasma-related research. This source will provide 10^{13} photons in a 200 fs duration pulse that is tunable from $\sim$ 6 nm to 100 nm. Since an VUV FEL will not have the limitation associated with optical lasers the entire field of high density plasmas kinetics in laser produced plasma will then be available to study with the tunable source. Thus, one will be able to use this and other FEL x-ray sources to pump individual transitions creating enhanced population in the excited states that can be easily monitored. We show two case studies illuminating different aspects of plasma spectroscopy.

Keywords: Laser produced plasma; X-ray spectroscopy, Free electron laser

1. INTRODUCTION

The next light generation sources, based on self amplified spontaneous emission (SASE), can play a critical role in moving the field of plasma spectroscopy substantially forward. Advances in the area of plasma spectroscopic techniques require a fast-rising short-duration source of high-energy photons, and a pump rate larger than competing rates. Currently no VUV laser or synchrotron-based light source has the required flux. However, the next generation light sources, which will be tunable, narrow-band x-ray sources with very short pulse duration, will achieve the necessary intensity. Indeed, for all the plasma-based research the individual bunch photon number is the essential quantity and one finds a > 10 order of magnitude enhancement in this quantity that make these the most promising source for plasma-based research. Under construction at DESY (Deutsche Elektronen-Synchrotron) is a short pulse tunable free electron laser (FEL) in the vacuum-ultraviolet and soft X-ray regime (VUV) based on the SASE FEL process. The pulse length will be of order 200 fs and tunable down to 6 nm providing 10^{13} photons per pulse. This VUV FEL will provide a test bed to field a variety of experiments investigating plasma conditions not previously accessible.

There is great interest in the higher temperature dense plasma regime, i.e., plasmas with electron densities $n_e \geq 10^{22}\text{cm}^{-3}$. In any experiment where a high intensity laser, e.g., $I_{\text{laser}} \geq 10^{12}\text{W/cm}^2$, irradiates a solid target there will be a region of the solid that is hot and near solid densities. Lasers with wavelengths $> 0.25\mu$m do not directly heat the solid as they can not propagate beyond the critical electron density $\sim 10^{21}\text{cm}^{-3} \times (1\mu\text{m}/\lambda_{\text{laser}})^2$; however, heat flow from the surface efficiently generates the hot dense medium.[1] The spectroscopic information derived from these plasmas provides, on the one hand, diagnostic information about the plasma itself, while on the other hand we can investigate, using spectroscopy, our understanding of the mechanisms at play in the creation of the plasma and the interaction of the atoms/ions with the plasma in which these are embedded. Here the next generation source will provide the ability to explore laser pump-probe techniques for high density plasmas that have been used in low density plasmas to measure line shapes, observe radiation redistribution, and determine the kinetics processes.[2–6] In the next section we discuss two optical pumping experiments that can be performed with an x-ray FEL. Here we restrict ourselves to the VUV FEL case and by doing this show clearly that these facilities will provide critical steps towards understanding the physical processes occurring in a broad area of finite temperature near solid density matter.

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2. RESONANT PHOTOPUMPING OF A LINE TRANSITION IN A DENSE PLASMA

2.1. Background

Since the creation of high density laser produced plasmas there have been virtually no quantitative in situ measurements of the kinetic rates or the populations. This is a major impediment to progress, as population kinetics of highly stripped ions is a complex problem. The complexity derives from the large number of states that must be considered in a model and the detail to which one must incorporate these states. The situation is made yet more difficult by the rapid time evolution and large spatial gradients.[1]

Indeed, much of the effort to improve understanding of the dense plasma regime has been focused on target design and advanced diagnostic development. However, difficulties in determining the level populations and/or the kinetic rates remain, therefore, the interest has never been met with substantial improvement in experiments. The import of the next generation FEL-based sources for high energy density plasma experiments is that one can use these x-ray sources to pump individual transitions in a plasma creating enhanced population in the excited states that can be easily monitored. The idea has been used in lower density plasmas with visible lasers and can, with the x-ray FEL, be employed to advance the study of high density plasmas.[7,8]

Variations on the idea of pumping individual transitions in high energy density plasma include the selective pumping of the wings of a line transition to observe redistribution within the line profile and pumping of selected transitions to attempt to understand the inversion mechanisms for the production of laboratory x-ray lasers. In all of these applications tests of the theoretical developments in the areas of atomic processes, kinetics model creation, line shape formation, and x-ray gain studies would be the first of their kind as there are currently no available probes.

There are several constraints on the x-ray source for it to be useful as a laser probe of the high energy density regime. First, the probe must be tunable and this is easily satisfied. Second, the line width of the pump must be such that it can pump entire line profiles and also be capable - for studies of redistribution within line profiles - of pumping parts of the line profile. Again, these conditions will be readily met. Finally, we need to have a pump that can move enough population from one state to another so that the population changes can be monitored. This last requirement can be verified by looking at the radiative pumping rate, R_{LU}, due to the source compared to the spontaneous emission rate, A_{UL}, of the transition being pumped. This is proportional to the number of photons per mode and is given by[9]

$$\frac{R_{LU}}{A_{UL}} = 6.667 \times 10^{-22} \frac{g_U}{g_L} \lambda_{\text{Å}} I_o(\mathrm{W/cm^2}) \frac{[,]}{\delta_\lambda \Delta_\lambda} \tag{1}$$

where g represents the statistical weights of the upper and lower states, $\lambda_{\text{Å}}$ and I_o are the photopumping source wavelength and intensity. The δ_λ and Δ_λ are the bandwidths of the x-ray source and the line shape of the transition being pumped, respectively, while [,] represents the minimum of the two. Two important insights emerge when evaluating the equation. First, if we conservatively assume $I_o \sim 10^{14}\mathrm{W/cm^2}$ and $[,]/\delta_\lambda\Delta_\lambda \sim 0.001$ we find that the ratio is approximately 1 for λ_L of 10 Å. This number is at least 10^3 larger than can be obtained by using a plasma source to pump a transition. Second, the ratio does not increase with decreasing source wavelength, indicating that large numbers of photons per mode will not be available as we move toward shorter wavelengths so that somewhat longer wavelength will be extremely useful. This is due to the fact that the spontaneous rate has a strong inverse dependence on wavelength. Of course, matching, or at least controlling source bandwidth can have salutary effects as indicated by the equation above.

The possibilities provided by probing are illustrated with the simulation of a sample that is irradiated by a short pulse laser. This creates a laser-produced plasma that will evolve on the time scales of the duration of the short pulse laser. The experiment to be performed will use a short-pulse optical laser to heat a small dot of material deposited on a CH backing. The laser spot is larger than the dot, creating a plasma blowoff from the material of interest that is constrained laterally, i.e., in the direction along the target surface, by the more rapidly expanding, usually lower Z, surrounding material. At some time in the expansion we irradiate the expanding plasma as indicated in Fig. 1, which shows a schematic of the experimental configuration. For the VUV FEL we find that optical pumping of the He-like 1s2l - 1s3l is quite efficient as the large ionization potential for

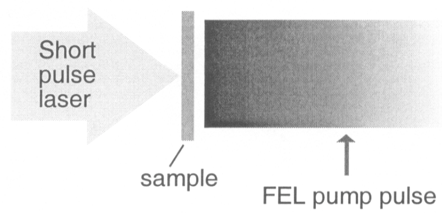

Figure 1. The schematic for the plasma spectroscopy photopumping experiments. The 100 fs short pulse visible laser impinges on a planar sample. After laser ablation causes a plasma to form the photopump is tuned to an ion resonance

the He-like species provides a broad range of temperature and density conditions wherein the population of the He-like ground state, $1s^2$, is the dominant species. This, in turn, leads to large optical depths in the resonance transition $1s^2$ - $1s2l$ creating population in the $1s2l$ state, which then leads to substantial population movement when the VUV FEL pumps this state.

The sample can be made of several low Z elements that satisfy the conditions that the transition energy, ΔE, falls in the tunable spectral range of the VUV FEL. Thus, using simple hydrogenic scaling we find that

$$\Delta \mathrm{E} \sim (Z-1)^2 \times 13.6 \text{ eV}(1/4 - 1/9) \tag{2}$$

so that for an VUV FEL with an upper spectral energy of 200 eV, or equivalently 60 Å, the range of possible atomic numbers, Z, is $\in [6, 11]$. The VUV FEL has a limited energy range in which it can operate and be tuned to specific line transitions. We have shown that a n = 2 to 3 transition is easily obtained for elements from C to Na and in section 2.2 we show the interesting features of this transition. One interesting feature is that the energy used to excite the 2 to 3 transition is sufficient to further ionize the n = 3 level stripping out the upper level creating H-like ions. However, if we consider a n = 3 to 4 level transition the energy requiement to pump the transition is not sufficient to ionize the n = 4 level.

The importance of development of an VUV probe is that it can potentially propagate through solid density matter, because critical density n_{cr} for a 100-Å laser can theoretically probe plasmas of solid densities and above.

2.2. K-Shell Fluorine Case

In the first example we study fluorine occurring in the He-like ion stage in the late time development of the expanding plasma. The He-like plasma is generated by a 1 J, 600 ps at 1μm wavelength focused to an intensity of 10^{12} W/cm^2. The time-dependant temperature and density was modeled using the rad/hydro simulation code LASNEX[10] giving rise to a plasma that expands $\sim 100\mu$m/ns. The FEL is tuned to the 1s2s ^{1}S - 1s3p ^{1}P transition that has a transition energy of 133.4 eV. There are two interesting points about this scheme. First, the ambient emission of the transition is small as the plasma is in steady-state at electron temperature, T_e, of 80 eV and n_e of 5×10^{20}cm^{-3}. Second, the population from the 1s2s ^{1}S is pumped to the 1s3p ^{1}P state and then can be ionized due to the photon pump. This occurs since the pump energy is greater than the ionization potential of the 1s3p ^{1}P level. However, in the single photon limit the population can not move beyond the ground state of H-like F. Thus, the population is pumped in a closed atomic system and the ability to model the response will be relatively straightforward.

The ambient emission spectrum from experiments of this type have been observed and the results of a radiation transfer simulation[11] coupled with a detailed atomic level calculation computed with the HULLAC suite of codes[12] including charge states from bare to Be-like and using the time-dependant plasma parameters are shown in Fig. 2 at three times in the evolution of the plasma: 1) at the initiation of the photopump that starts at 1 ps; 2) 50 fs later, i.e., at 1.05 ps, while the pump is on; and, 3) at 800 fs after the photopump is off at 1.38 ps. The most notable feature of the spectrum at 1.05 ps is the substantial decrease in the emission in

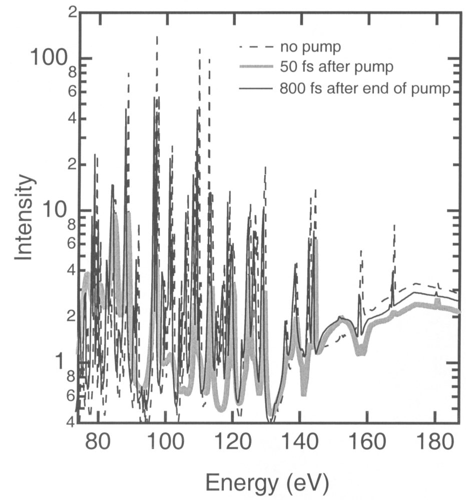

Figure 2. The logarithm of the emissivity versus spectral energy for an exploding Z=9 foil plasma. The plasma is photopumped resonantly with the He-like 1s2l -1s3l starting at 1 ps after the plasma-creating 100 fs visible laser. Three times are shown 1) the time of initiation of the photopump at 1 ps, dashed line; 2) 50 fs later during the photopump, solid thick gray line; and 3) 800 fs after the photopump is off, thin solid black line.

the spectral region as the population is being pumped to the hydrogenic ground state. The later time spectrum indicates that the relaxation is sufficiently slow that one does not, on these time scales, reproduce the ambient plasma emission. Thus, the evolution of the system will provide detailed information on the kinetics of this system.

2.3. K-Shell Boron Case

In the second example we show what occurs when we pump the He-like B resonance transition 1s^2 ^{1}S - 1s2p ^{1}P at 205 eV. Here the situation is distinct from the previous example as the pump can now photoionize the atom through to the fully stripped ion. This implies that the recombination to He-like will not occur during the pump as there will be no H-like species. In Fig. 3 we show the emission at three spectral energies as a function of time. We note that during the FEL pump, which is also shown for reference, the intensities at the He-like 1s^2 ^{1}S - 1s2p ^{1}P and H-like 1s-3l transitions are enhanced; however, we see in Fig. 4 that this is due to continuum scattering of the pump. As the pump starts to decrease at 200 fs off there becomes a rapid rise in the H-like 1s-3l intensity due to recombination from the fully stripped ion. On the other hand, the He-like 1s^2 ^{1}S - 1s2p ^{1}P intensity rises slightly as the pump drops due to residual recombination but decreases dramatically when it goes to zero. This latter behavior is due to the fact that the pump is resonant so that when it goes to zero the spontaneous decay rapidly depletes the n= 2 state.

In Fig. 4 we show the actual spectra for various times in the history of the system. The spectra have been displaced in energy and shifted slightly to make the pattern visible. Note that the spectrum at 200 fs is due to continuum emission and is actually much more intense than the spectrum at 300 fs, see Fig. 3. It is clear from the emission spectra shown in Fig. 4 that one could use a fraction of the FEL pump to generate observable signals. The detailed information that can be obtained from these measurements would provide unique constraints on the complex processes necessary to construct a complete kinetics model for highly charged ions. Indeed, we chose to use as an example the K-shell spectra as it is easily interpretable; however, the generation of L-shell and M-shell models are also of importance and raise the level of complexity substantially. That is, the need for experiments that can provide basic information on the processes necessary to build kinetics models is independent of the atomic number.

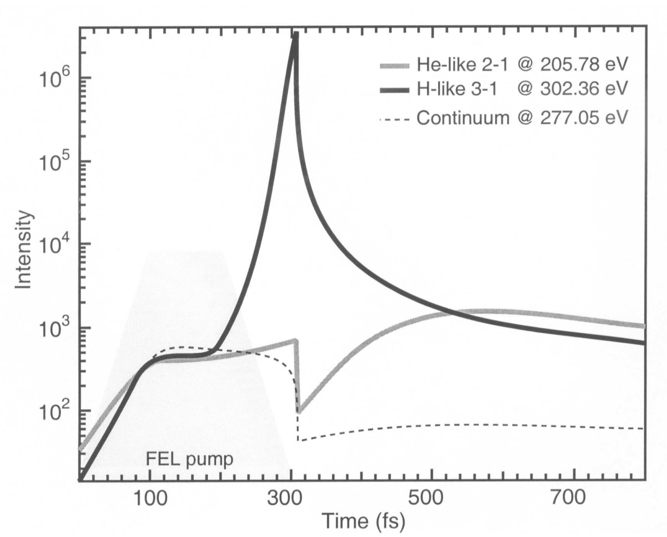

Figure 3. The absolute intensity versus time from the start of the FEL pulse for three energies. The solid black curve is intensity at the energy of the H-like B n=1 to 3 transition, the solid gray curve is the intensity at the He-like B $1s^2$-1s2p ^1P transition where the photopump is tuned, and the dashed curve is the intensity of the continuum at 277 eV. For reference the shape of the FEL pulse is shown in gray.

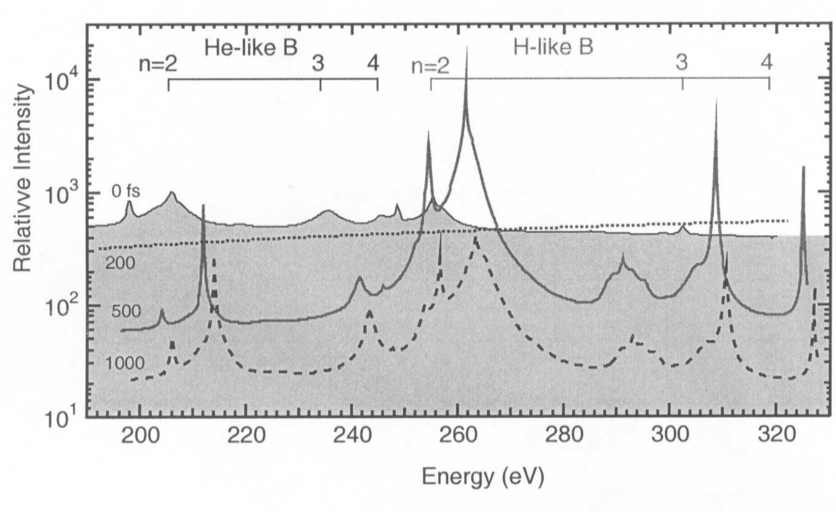

Figure 4. He- and H-Like B spectra shown as relative intensity versus energy in eV. The spectra are labeled for the various times relative to the initiation of the FEL pump at 0 fs. To illustrate the spectral shapes the spectra are shifted in energy and displaced in intensity. See Fig. 3 for the absolute intensity as a function of time. For reference the transitions of the He- and H-like transitions are indicated for the 0 fs spectra

3. CONCLUSIONS

The mechanisms involved in the formation of a plasma and the details of the kinetics processes can be illuminated by using a laser to selectively photopump levels and redistribute radiation. In a particularly intriguing possibility one will be able to study the formation of laboratory x-ray lasers that currently depend on kinetics processes.[13–15] Thus, one could disentangle the plasma production from the inversion-forming processes that lead to the x-ray lasing. It is clear that numerous aspects of plasma spectroscopy have been severely constrained by a lack of data. The 4th generation sources will provide a substantial improvement in the development of our understanding of intrinsic line shape formation, level shifts, radiation transfer, and detailed kinetics processes. The combination of the short pulse length, the tunable wavelength, the repetition rate, and the energy per pulse will provide unique data derived from these plasma-based experiments a major advance in our knowledge in this area

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REFERENCES

1. M. N. Rosenbluth and R. Z. Sagdeev, eds., *Handbook of Plasma Physics, vol. 3, Physics of Laser Plasmas*, Amsterdam:Elseviert, 1991.
2. C. A. Back, R. W. Lee, and C. Chenais-Popovics, "Measurement of resonance fluorescence in a laser-produced al xii plasma," *Phys Rev Letts* **63**, pp. 1471–1474, 1989.
3. C. A. Back and C. C.-P. R. W. Lee, "Photopumping and fluorescence in a laser-produced plasma. i. experimental results," *Phys Rev A* **44**, pp. 6730–6742, 1991.
4. J. A. Koch, R. W. Lee, J. Nilsen, J. C. Moreno, B. J. MacGowan, and L. B. D. Silva, "X-ray lasers as sources for resonance-florescence experiments," *Appl Phys B* **58**, pp. 7–11, 1994.
5. A. Bar-Shalom, M. Klapisch, and J. Oreg, "Electron collision excitations in complex spectra of ionized heavy atoms," *Phys Rev A* **38**, pp. 1773–1784, 1988.
6. M. E. Foord, S. H. Glenzer, R. S. Thoe, K. L. Wong, K. B. Fournier, J. R. Albritton, B. G. Wilson, and P. T. Springer, "Accurate determination of the charge state distribution in a well characterized highly ionized au plasma," *J Quant Spectrosc Radiat Transfer* **65**, pp. 231–241, 2000.
7. D. D. Burgess and C. H. Skinner, "Dye laser induced fluorescence of plasmas and its application to measurement of lower state decay rates," *J Phys B* **7**, pp. L297–L301, 1974.
8. G. T. Razdobarin, V. V. Semenov, L. V. Sokolova, I. P. Folomkin, V. S. Burakov, P. Misakov, P. A. Naumenkov, and S. V. Nechaev, "An absolute measurement of the neutral density profile in the tokamak plasma by resonance fluorescence on the ha line," *Nuc Fusion* **19**, pp. 1439–1446, 1979.
9. R. C. Elton, *X-ray Lasers*, San Diego: Academic Press, 1990.
10. G. B. Zimmerman and W. L. Kruer *Comments on Plasma Physics for Controlled Thermonuclear Fusion* **2**, pp. 51–61, 1975.
11. H. A. Scott and R. W. Mayle, "Glf - a simulation code for x-ray lasers," *Appl Phys B* **58**, pp. 35–43, 1994.
12. K. B. F. et al *J Phys B: At Mol Phys* **35**, pp. 3347–3364, 2002.
13. G. Tallents, ed., *X-Ray Lasers 1990*, Bristol: IOP Publishing, 1990.
14. D. L. Matthews and D. C. Eder, eds., *X-Ray Lasers 1994: Fourth International Colloquium*, New York: AIP Conference Proceedings 332, 1994.
15. S. Svanberg and C. G. Wahlstrom, eds., *X-Ray Lasers 1996*, Bristol: IOP Publishing, 1996.