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Comparing the gain of the Ne K- α inner-shell X-ray laser using the XFEL to drive the kinetics with photo-ionization versus photo-excitation

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Abstract. Over the last four decades many photo-pumped X-ray laser schemes have been proposed. However, demonstrating these schemes in the laboratory has proved to be elusive because of the difficulty of finding a strong resonant pump line or X-ray source. With the advent of the X-ray free electron laser (XFEL) at the SLAC Linac Coherent Light Source (LCLS) we now have a tuneable X-ray laser source that can be used to replace the pump line or X-ray source in previously proposed laser schemes and allow researchers to study the physics and feasibility of photo-pumped laser schemes. Many of these photo-pumped schemes are driven by photo-excitation from a resonant line source but others are driven by photo-ionization from a strong non-resonant X-ray source. Three years ago an inner-shell X-ray laser was demonstrated at 849 eV (1.46 nm) in singly ionized neon gas using the XFEL at 960 eV to photo-ionize the 1s electron in neutral neon followed by lasing on the 2p – 1s transition in singly-ionized neon. In this paper we model the neon inner shell X-ray laser under similar conditions to those used at LCLS. We investigate how we can improve the efficiency of the neon laser and reduce the drive requirements by tuning the XFEL to the 1s-3p transition in neutral neon in order to create gain on the 2p-1s line in neutral neon. We explore the sensitivity to the drive intensity, pulse duration, and line-width of the XFEL to better understand how to optimize this inner shell laser by understanding the trade-offs between using photo-ionization versus photo-excitation to drive gain in these systems. We also discuss how photo-ionization of L-shell electrons can be used to create lasing on n=3-2 transitions in materials such as Ar and Cu.

1 Introduction

Since the earliest days of the laser, scientists have proposed schemes to achieve lasing at shorter wavelengths. Five decades ago, working at Bell Laboratories where the laser was invented, Duguay and Rentzepis proposed using photo-ionization to create an X-ray laser on the inner shell K- α line in sodium vapour [1]. Forty-years ago Ray Elton [2] discussed the challenges of making quasi steady state inner-shell K- α lasers in Si, Ca, and Cu. In 2011 the dream of demonstrating an inner-shell X-ray laser was realized at the SLAC Linac Coherent Light Source (LCLS) when the X-ray free electron laser

(XFEL) at 960 eV was used to photo-ionize the K-shell of neutral neon gas and create lasing at 849 eV in singly ionized neon gas [3].

Another early approach for creating X-ray lasers was the idea of a resonantly photo-pumped laser where a strong emission line in one material could be used to photo-excite a transition in another material and create lasing. A classic example of this scheme is the Na-pumped Ne X-ray laser scheme proposed 40 years ago by Vinogradov and colleagues [4-5]. This scheme used the strong Na He- α line at 1127 eV to resonantly photo-pump the Ne He- γ line and lase on the 4f – 3d transition at 23.1 nm in He-like Ne. This scheme was studied extensively and numerous experiments were done to try to demonstrate lasing and measure gain [6]. While weak gain [6] was inferred in several experiments the difficulty with this type of scheme was creating a sufficiently strong pump line. With the availability of strong XFEL sources the pump line in the traditional photo-pumped schemes can be replaced with an XFEL that is tuned to the appropriate resonance. Since the resonant photo-pumped scheme selectively pumps a transition it offers the potential for higher gain and lower drive intensity than the photo-ionization pumping.

In this paper we look at the advantages and challenges of using the XFEL to resonantly photo-pump the 1s-3p line in neutral neon as a mechanism for creating gain on the K- α line in Ne and compare this with the photo-ionization pumping that has already been demonstrated. We show that with the use of a sufficiently short XFEL pulse (1-fsec) the resonant photo-excitation could reduce the XFEL flux requirements by several orders of magnitude.

2 Modelling the inner-shell Ne laser

Figure 1 shows the pumping mechanism used in the LCLS experiments that demonstrated lasing on the inner-shell neon laser. A strong XFEL beam tuned above the K-edge of neutral Ne I photo-ionizes the 1s electron. This creates an excited state of singly ionized Ne II that has a missing 1s electron. This excited state then lases to the ground state of Ne II by emitting an X-ray on the 2p – 1s transition at 848.6 eV. The experiment starts with neon gas that is all in the Ne I ground state so the lower laser state is initially unoccupied. The natural lifetime of the laser transition is 135 fsec. However the challenge with this scheme is that the Auger lifetime of the upper laser state is 2.3 fsec so pumping this scheme requires a very short pulse duration in the fsec regime.

Figure 2 shows the resonant photo-pumping mechanism for driving the inner-shell neon laser. The XFEL is tuned to the 1s – 3p transition in Ne I at 867.63 eV creating a large population in the 1s2s²2p⁶3p level. This level can

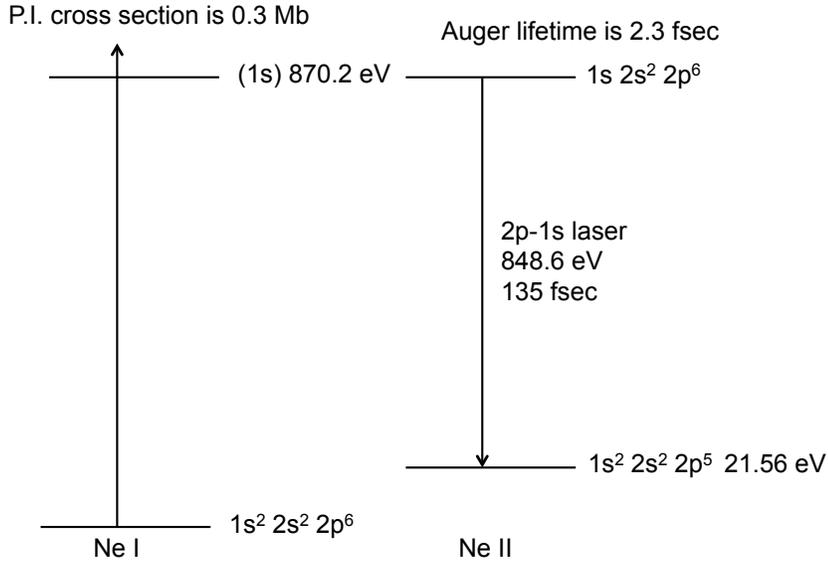


Fig. 1. Energy level diagram for the photo-ionization driven inner-shell neon X-ray laser.

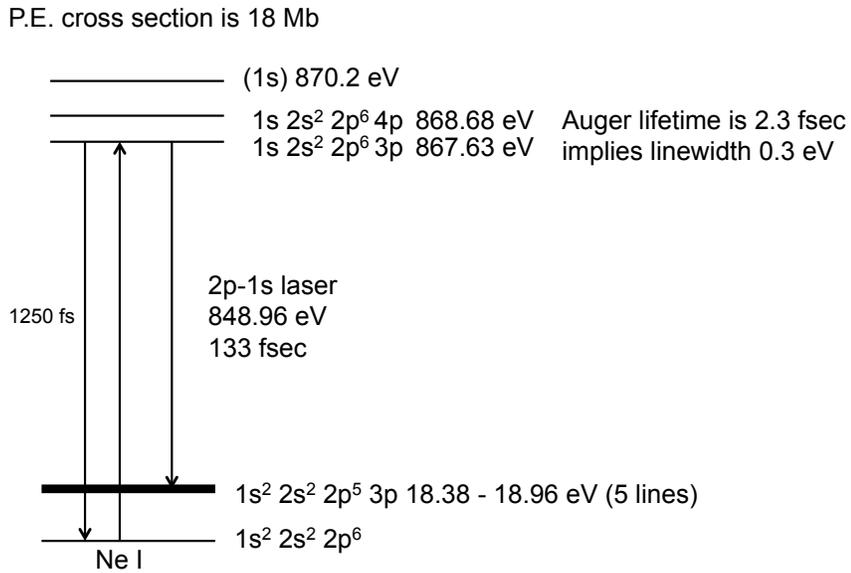


Fig. 2. Energy level diagram for the photo-excitation driven inner-shell neon X-ray laser.

then lase to the lower $1s^22s^22p^53p$ level state by emitting X-rays on the $2p - 1s$ transition centered at 848.96 eV. Because of the splitting in the lower level state there are actually 5 X-ray lines emitted that are spread over a range of 848.67 - 849.25 eV. We calculate the total gain by summing the gain of the 5 lines. The upper laser level has a similar Auger lifetime of 2.3 fsec that implies a line-width of 0.3 eV on the lasing transition. The photo-excitation scheme also requires a short pulse drive because of the very short Auger lifetime. The difference between this scheme and the photo-ionization scheme is that lasing is now in neutral Ne I. The lasing energies differ by about 0.4 eV. The potential advantage of this scheme is that the photo-excitation cross-section is about 18 Mbarns compared to 0.3 Mbarns for the photo-ionization scheme. The question we want to address in this paper is how we can take advantage of this much larger excitation rate to reduce the drive requirements on the XFEL source.

To model the photo-ionization and photo-excitation schemes we created a simple atomic model of the levels shown in Figs. 1 and 2. We then used the kinetics code Cretin [7] to model the kinetics and gain of the system under various conditions. For the baseline XFEL beam we assume the XFEL beam has 10^{12} photons in a 0.9 eV line-width focused to a 1- μm diameter. For the pulse duration we compare a Gaussian shape with 100-fsec full-width half-maximum (FWHM) to a 1-fsec FWHM. The XFEL was designed to have a bandwidth of 0.1% as we are assuming even though the current bandwidth is larger by a factor of 5-10. The bandwidth has minimal impact on the photo-ionization mechanism but the strength of the photo-excitation mechanism is inversely proportional to the bandwidth. One challenge with the photo-excitation scheme is understanding the validity of the kinetics model and how to include the photo-excitation rate in the line-width calculation of the gain. Currently the stimulated rate is included in the kinetics but not in the line-width which means there are no Stark sidebands or broadening. The XFEL energy is set at 875 eV for modelling the photo-ionization scheme and 867.6 eV for modelling the photo-excitation scheme.

Starting with a 100-fsec duration XFEL pulse, Fig. 3 shows the predicted gain versus time for both schemes. The time scale is set so that zero time is at the peak of the XFEL pulse. To understand the sensitivity to the XFEL flux a series of calculations were done using a multiplier between 1.0 (nominal) and 0.001 on the nominal XFEL flux described above. For the photo-ionization scheme we predict peak gain of 44/cm at $t=-81$ fsec for the nominal case with multiplier of 1.0. In contrast the photo-excitation scheme has peak gain of 62/cm at -128 fsec for the nominal case. The big difference between the behaviour of the two schemes is that the gain of the photo-excitation scheme falls much slower as the XFEL flux is reduced. With a multiplier of 0.001, which corresponds to 10^9 photons in the beam, the peak gain is still 12/cm at -12 fsec as compared with 0.7/cm for the photo-ionization scheme. For both

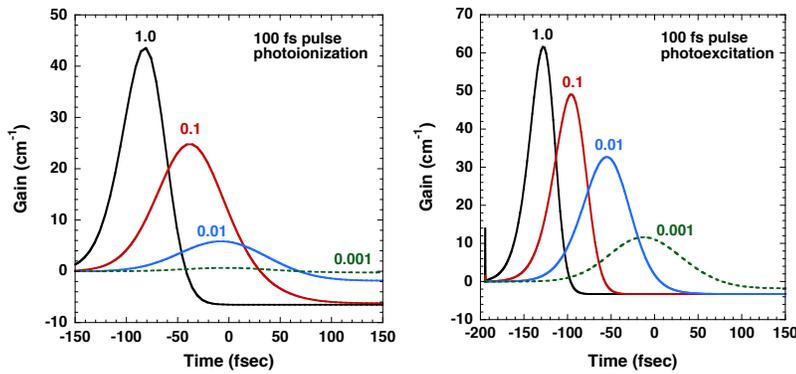


Fig. 3. Gain versus time for the neon X-ray laser driven by a 100-fs duration XFEL comparing the photo-ionization and photo-excitation mechanisms. The XFEL intensity is varied by using a multiplier between 1.0 (nominal) and 0.001.

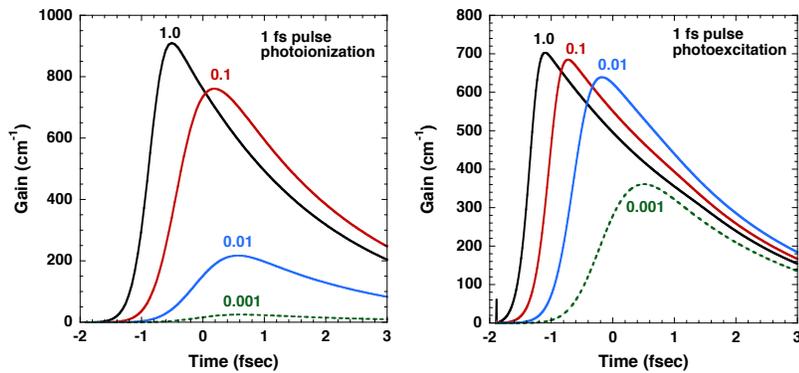


Fig. 4. Gain versus time for the neon X-ray laser driven by a 1-fs duration XFEL comparing the photo-ionization and photo-excitation mechanisms. The XFEL intensity is varied by using a multiplier between 1.0 (nominal) and 0.001.

schemes the peak gain starts before the peak of the XFEL pulse and moves closer to $t=0$ as the flux is reduced. To optimize the XFEL drive one wants the peak gain to occur at the peak of the XFEL drive pulse, otherwise some of the XFEL drive is not being used.

Now consider a 1-fsec duration XFEL driving the Ne gas as shown in Fig. 4. For the nominal XFEL flux the peak gains are 910/cm at -0.5 fsec for the photo-ionization and 703/cm at -1.1 fsec for photo-excitation. As the flux intensity is reduced the gain for the photo-ionization drops quickly but one notices that the peak gain for the photo-excitation scheme drops from 703/cm to 639/cm as the XFEL drive flux is reduced by a factor of 100. Also the peak of the gain moves to a time of -0.2 fsec, indicating near optimum drive conditions. As the XFEL flux drops further the gain drops more quickly and occurs after the peak of the XFEL flux indicating the flux is below ideal drive conditions. This figure shows that the photo-excitation mechanism offers the potential to reduce the XFEL drive by one to two orders of magnitude as compared with the photo-ionization mechanism. This advantage could enable smaller facilities to drive inner shell X-ray lasers or allow facilities such as LCLS to drive even higher energy X-ray lasers with the current XFEL fluxes.

3 Modelling Ar and Cu L-shell X-ray lasers

Given the success of the K-shell neon X-ray laser it should be possible to demonstrate inner-shell X-ray lasers in other principal shells such as the L and M shells. One promising candidate to consider is neutral argon gas. Figure 5 shows the energy level diagram for using an XFEL above the L-shell edge of neutral Ar I to create a L-shell hole in singly ionized Ar II. If an XFEL was tuned between the two L-edges at 248 and 326 eV one could create a 2p hole that would result in lasing on the 3s-2p transition at 232.7 eV. If the XFEL drive was tuned above the L-edge at 326.3 eV then one would have holes in both the 2s and 2p shells that would result in lasing on the 3p-2s transition at 310.6 eV as well as the 3s-2p transition. It would be very interesting to tune the XFEL from low to high energy and watch the 3s-2p lasing turn on followed by lasing on both lines.

Figure 6 shows the energy level diagram for using an XFEL above the L-shell edge of neutral Cu I to create a L-shell hole in singly ionized Cu II and create lasing on the strong 3d-2p line at 922 eV. As an alternative, photo-excitation of the 2p-4d transition in Cu I would also create lasing on the 3d-2p line in Cu I.

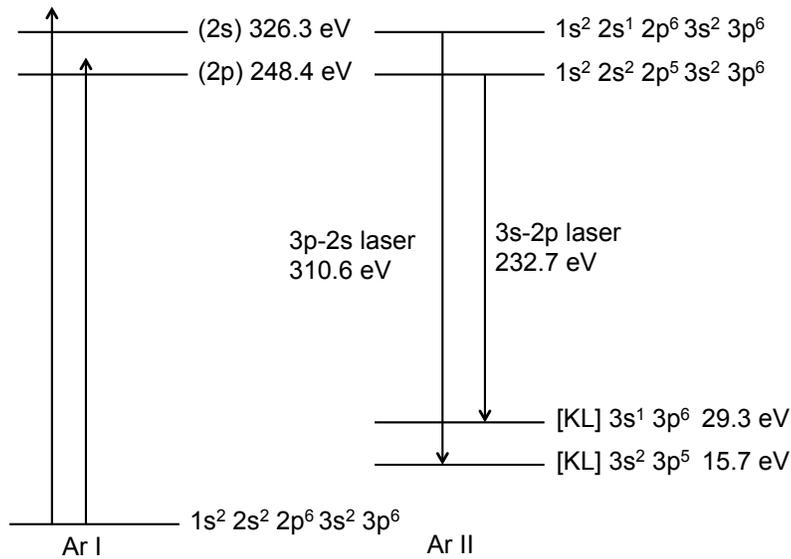


Fig. 5. Energy level diagram for the photo-ionization driven inner-shell argon X-ray laser.

Photo-excitation of 2p to 4d level would also drive 3d-2p laser

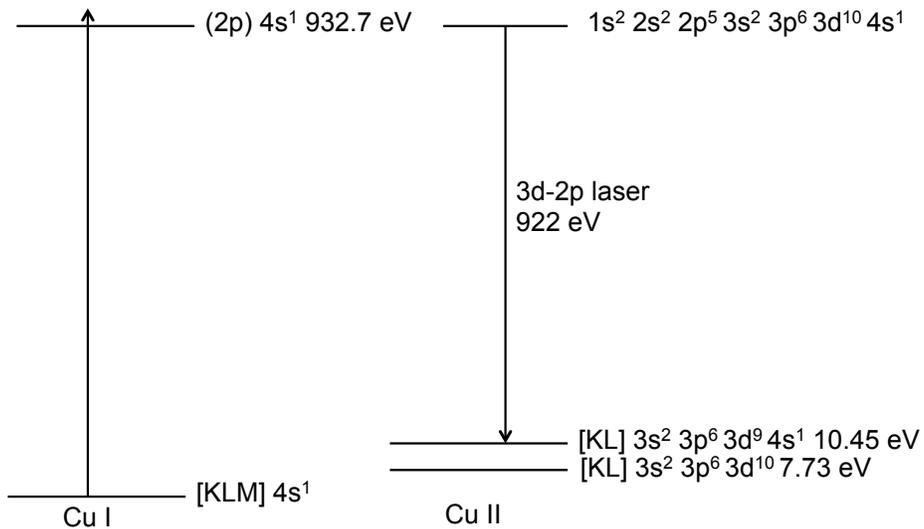


Fig. 6. Energy level diagram for the photo-ionization driven inner-shell copper X-ray laser.

4 Conclusions

In this paper we model the neon inner shell X-ray laser under similar conditions to those used at LCLS. We show how we can improve the efficiency of the neon laser and reduce the drive requirements by tuning the XFEL to the 1s-3p transition in neutral neon in order to create gain on the 2p-1s line in neutral neon. We present the sensitivity to the drive intensity, pulse duration, and line-width of the XFEL to better understand how to optimize this inner shell laser by understanding the trade-offs between using photo-ionization versus photo-excitation to drive gain in these systems. We show that with the use of a sufficiently short XFEL pulse (1-fsec) the resonant photo-excitation could reduce the XFEL flux requirements by several orders of magnitude. We also discuss how photo-ionization of L-shell electrons can be used to create lasing on n=3-2 transitions in materials such as Ar at 232 and 310 eV and Cu at 922 eV.

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References

1. M. A. Duguay and P. M. Rentzepis, *Appl. Phys. Lett.* **10**, 350–352 (1967).
2. R. C. Elton, *Appl. Opt.* **14**, 2243- 2249 (1975).
3. N. Rohringer, D. Ryan, R. A. London, M. Purvis, F. Albert, J. Dunn, J. D. Bozek, C. Bostedt, A. Graf, R. Hill, S. P. Hau-Riege, and J. J. Rocca, *Nature* **481**, 488–491 (2012).
4. A. V. Vinogradov, I. I. Sobelman, and E. A. Yukov, *Sov. J. Quantum Electron.* **5**, 59–63 (1975).
5. J. Nilsen, J. H. Scofield, and E. A. Chandler, *Appl. Opt.* **31**, 4950 – 4956 (1992).
6. J. Nilsen and E. Chandler, *Phys. Rev. A* **44**, 4591-4598 (1991).
7. H. A. Scott, *JQSRT* **71**, 689–701 (2001).