# Chapter 15
# Using the XFEL to Drive Gain in K-Shell and L-Shell Systems Using Photoionization and Photoexcitation of Inner Shell Transitions

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**Abstract** Six years ago, X-ray lasing on an inner shell transition was demonstrated at 849 eV (1.46 nm) in singly ionized neon gas using the XFEL at 960 eV to photoionize the 1s electron in neutral neon followed by lasing on the 2p-1s transition in singly ionized neon. That research was done at the SLAC Linac Coherent Light Source (LCLS) by a multi-laboratory team led by Nina Rohringer and published in the January 26, 2012, issue of Nature. It took many decades to demonstrate this scheme because it required a very strong X-ray source that could photoionize the 1s (K-shell) electrons in neon on a timescale comparable to the intrinsic auger lifetime in neon, which is typically 2 fs. In this chapter, we have shown how the XFEL could be used to photoionize L-shell electrons to drive gain on n = 3–2 transitions in singly ionized Ar and Cu plasmas. These bright, coherent and monochromatic X-ray lasers may prove to be very useful for doing high-resolution spectroscopy and for studying nonlinear process in the X-ray regime.

## 15.1 Introduction

Scientists have proposed schemes to achieve lasing at shorter wavelengths since the invention of the laser. In the 1960s, Duguay and Rentzepis proposed using photoionization to create an X-ray laser on the inner-shell K-α line in sodium vapour [1]. In the 1970s, Ray Elton [2] discussed the challenges of making quasi–steady-state inner shell K-α lasers in Si, Ca and Cu. The dream of demonstrating an inner shell X-ray laser was realized at the SLAC Linac Coherent Light Source (LCLS) in 2011 when the X-ray free electron laser (XFEL) at 960 eV was used to photoionize the K-shell of neutral neon gas and create lasing at 849 eV in singly ionized neon gas [3].

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An alternative 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 photoexcite a transition in another material and create lasing. A classic example is the Na-pumped Ne X-ray laser scheme proposed 40 years ago by Vinogradov et al. [4] and modeled by Nilsen et al. [5] that 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 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 photoionization pumping.

Recently, we looked 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 photoionization pumping that has already been demonstrated [7]. We showed that with the use of a sufficiently short XFEL pulse (1-fs) the resonant photoexcitation could reduce the XFEL flux requirements by two orders of magnitude. In this chapter, we look at how the inner shell X-ray laser can be extended to lasing on L-shell transitions in Ar and Cu. For Ar, we consider an XFEL pulse that photoionizes the 2p or 2s electrons and creates lasing on the 3s-2p or 3p-2s transitions. In the case of Cu, we consider an XFEL pulse that photoionizes the 2p electron and creates lasing on the strong 3d-2p transitions near 1 keV.

## 15.2 Modelling Ar and Cu L-Shell X-Ray Lasers

Starting with the neon X-ray laser based on photoionization of the K-shell, we look at extending inner shell X-ray lasers to the other principal shells beginning with the L and M shells. Neutral argon gas is the first promising candidate we investigate. Figure 15.1 shows the energy level diagram that uses XFEL above the L-shell edge of neutral Ar I to create an L-shell hole in singly ionized Ar II. Tuning an XFEL between the two L-edges at 250 and 326 eV, one could create a 2p hole that would lase on the 3s-2p transitions at 219 and 221 eV. If the XFEL drive was tuned above the L-edge at 326.3 eV then, one would create holes in the 2s and 2p shells simultaneously that would create lasing on the 3p-2s transitions at 310.4 and 310.6 eV in addition to the 3s-2p transitions. By tuning the XFEL from low to high energy, one could watch the 3s-2p lasing turn on followed by lasing on both sets of lines.

To model these photoionization schemes, we created a simple atomic model of the levels shown in Fig. 15.1. We used the Cretin code [8] to model the kinetics and gain of the system under various conditions. For the baseline XFEL beam, we assume that the XFEL beam has $10^{12}$ photons in a 0.1% linewidth focused to a

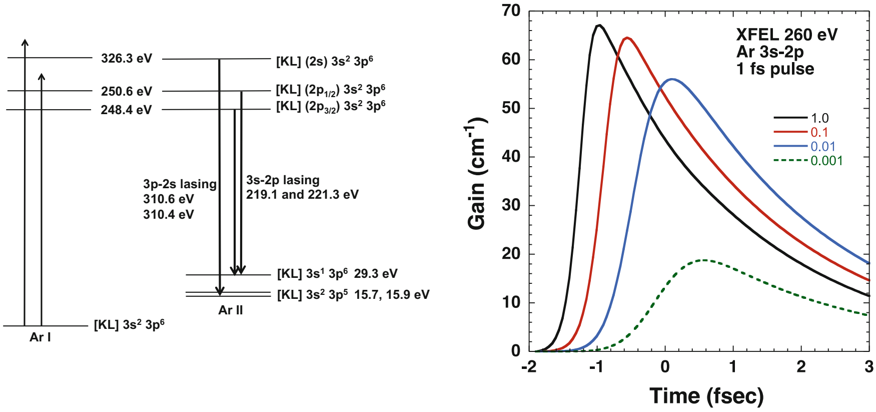

**Fig. 15.1** Energy level diagram (left) for the photoionization-driven inner shell argon X-ray laser. Gain versus time (right) for the 3s-2p line at 219 eV in the argon X-ray laser driven by the photoionization mechanism using a 1-fs duration 260 eV XFEL drive pulse. The XFEL intensity is varied using a multiplier between 1.0 (nominal) and 0.001. The peak of the XFEL pulse is defined as time = 0

1-μm diameter. In our previous work, we had studied the nominal LCLS conditions that used a 100-fs full-width half-maximum (FWHM) Gaussian pulse [7]. In this work, we examine using a shorter 1-fs FWHM pulse, which produces much higher gain than the 100-fs pulse used in the LCLS experiments [3, 7].

Figure 15.1 shows the gain versus time for the Argon 3s-2p X-ray laser line at 219 eV driven by a 260 eV XFEL with $10^{12}$ photons in a 1-fs pulse focused to 1-μm diameter spot. The 219 eV line has twice the gain of the 221 eV line. One observes that the peak gain of 67 cm$^{-1}$ falls very slowly to 56 cm$^{-1}$ as the XFEL flux is reduced by a factor of 100. By comparison, using a 330 eV XFEL pulse, the 3p-2s line at 310 eV has a peak gain of about 30 cm$^{-1}$.

Figure 15.2 shows the energy level diagram for using a 1-keV XFEL whose energy is above the L-shell edge of neutral Cu I to photoionize an L-shell hole in singly ionized Cu II that results in lasing on the strong 3d-2p lines at 928 and 948 eV. The gain at 928 eV is predicted to be about twice the gain at 948 eV. Fig. 15.2 shows the gain versus time for the Copper 3d-2p X-ray laser line at 928 eV driven by a 1000 eV XFEL with $10^{12}$ photons in a 10-fs pulse focused to 1-μm diameter spot. The gain peaks at 136 cm$^{-1}$ and falls slowly as the XFEL intensity is reduced. As an alternative, photoexcitation of the 2p-4d transition in Cu I would also create lasing on the 3d-2p line in Cu I and might require an even lower XFEL intensity.

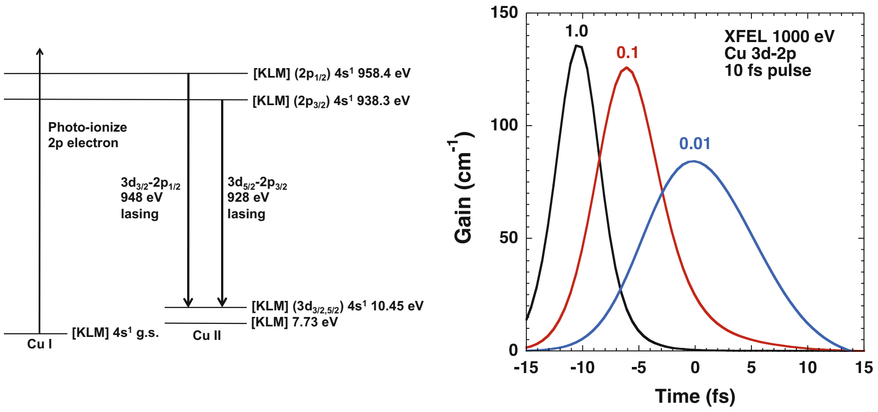

**Fig. 15.2** Energy level diagram (left) for the photoionization-driven inner shell copper X-ray laser. Gain versus time (right) for the 3d-2p line at 928 eV in the copper X-ray laser driven by the photoionization mechanism using a 10-fs duration 1000 eV XFEL drive pulse. The XFEL intensity is varied using a multiplier between 1.0 (nominal) and 0.01. The peak of the XFEL pulse is defined as time = 0

## 15.3 Conclusions

In this chapter, we have shown how the XFEL could be used to photoionize L-shell electrons to drive gain on n = 3–2 transitions in singly ionized Ar and Cu plasmas. We model those systems and estimate peak gain of 67 $cm^{-1}$ on the 3s-2p line at 219 eV in Ar and peak gain of 136 $cm^{-1}$ on the 3d-2p line at 928 eV in Cu. Substantial gain is predicted in other n = 3–2 lines in Ar at 221 and 310 eV as well as 948 eV in Cu. These bright, coherent and monochromatic X-ray lasers may prove to be very useful for doing high-resolution spectroscopy and for studying nonlinear process in the X-ray regime.

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## References

1. Duguay, M.A., Rentzepis, P.M.: Appl. Phys. Lett. **10**, 350–352 (1967)
2. Elton, R.C.: Appl. Opt. **14**, 2243–2249 (1975)
3. Rohringer, N., Ryan, D., London, R.A., Purvis, M., Albert, F., Dunn, J., Bozek, J.D., Bostedt, C., Graf, A., Hill, R., Hau-Riege, S.P., Rocca, J.J.: Nature **481**, 488–491 (2012)
4. Vinogradov, A.V., Sobelman, I.I., Yukov, E.A.: Sov. J. Quantum Electron. **5**, 59–63 (1975)
5. Nilsen, J., Scofield, J.H., Chandler, E.A.: Appl. Opt. **31**, 4950–4956 (1992)
6. Nilsen, J., Chandler, E.A.: Phys. Rev. A **44**, 4591–4598 (1991)
7. Nilsen, J.: Matter Radiat. Extremes **1**, 76–81 (2016)
8. Scott, H.A.: JQSRT **71**, 689–701 (2001)