Modeling the gain of inner-shell X-ray laser transitions in neon, argon, and copper driven by X-ray free electron laser radiation using photo-ionization and photo-excitation processes

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Abstract

Using an X-ray free electron laser (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, an inner-shell X-ray laser was demonstrated at 849 eV in singly-ionized neon gas several years ago. It took decades to demonstrate this scheme, because it required a very strong X-ray source that could photo-ionize the 1s (K shell) electron in neon on a timescale comparable to the intrinsic Auger lifetime in neon of 2 fs. In this paper, we model the neon inner shell X-ray laser under similar conditions to those used in the XFEL experiments at the SLAC Linac Coherent Light Source (LCLS), and 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 also show how the XFEL could be used to photo-ionize 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 very useful for doing high-resolution spectroscopy and for studying non-linear process in the X-ray regime.

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1. Introduction

In the 1960's scientists at Bell Laboratories, Duguay and Rentzepis, proposed using photo-ionization to create an X-ray laser on the inner shell K-\(\alpha\) line in sodium vapor [1]. A decade later in the 1970's Ray Elton [2] from the Naval Research Laboratory discussed the challenges of making quasi steady state inner-shell K-\(\alpha\) lasers in Si, Ca, and Cu. In 2003 Lan and colleagues modeled XFEL radiation driving gain on the He-\(\alpha\) and Ly-\(\alpha\) lines in He [3,4]. 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 [5].

Another approach pursued in the 1970's 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 by Vinogradov and colleagues [6,7]. This scheme used the strong Na He-\(\alpha\) line at 1127 eV to resonantly photo-pump the Ne He-\(\gamma\) line and lase at 53.7 eV (23.1 nm) on the 4f-3d transition in He-like Ne. This scheme
was studied extensively and numerous experiments were done to try to demonstrate lasing and measure gain [8]. Weak gain [8] was inferred in several experiments. However 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. The resonant photo-pumped scheme selectively pumps a transition so 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-\(\alpha\) 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-fs) the resonant photo-excitation could reduce the XFEL flux requirements by several orders of magnitude. We then 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 photo-ionizes 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 photo-ionizes the 2p electron and creates lasing on the strong 3d-2p transitions near 1 keV.

2. Modeling the inner-shell Ne laser

Fig. 1 shows the pumping mechanism used in the LCLS experiments that demonstrated lasing on the inner-shell neon laser. The XFEL beam is tuned above the K-edge of neutral Ne I and photo-ionizes the 1s electron. This creates an excited state of singly-ionized Ne II that has a missing 1s electron. This excited state can Auger decay to Ne III or lase to the ground state of Ne II by emitting an X-ray on the 2p-1s transition at 848.6 eV [9]. The experiment starts with a neon gas that is in the Ne I ground state. The lower laser state is initially unoccupied. The natural lifetime of the laser transition is 135 fs based on calculations done with the multi-configuration Dirac-Fock (MCDF) atomic physics code of Grant et al. [10] for similar transitions in Mg assuming the oscillator strength is the same for Ne. This is consistent with the Auger branching ratio of 98.2% calculated in the work of Aksela [11].

The notation [KL] signifies the fully occupied K shell, 1s\(^2\), and the L shell, 2s\(^2\)2p\(^6\), configurations while the notation (1s) represents a 1s hole in the K shell and (2p) represents a 2p hole in the L shell. The challenge with this scheme is that the Auger lifetime of the upper laser state is 2.3 fs [11] so pumping this scheme requires a very short pulse duration in the fs regime. The Auger process is an auto-ionizing process that causes rapid ionization as one of the L-shell electrons fills the 1s hole while a second L-shell electron is ionized.

Fig. 2 shows the resonant photo-pumping mechanism for driving the inner-shell neon laser. In this case, the XFEL is tuned to the 1s-3p transition in Ne I at 867.63 eV [12] creating a large population in the [KL] (1s) 3p or 1s2s\(^2\)2p\(^5\)3p level. This level can then lase to the lower [KL] (2p) 3p or 1s\(^2\)2s\(^2\)2p\(^5\)3p level by emitting X-rays on the 2p-1s transition centered at 848.96 eV. Because of the splitting in the lower level, there are 5 X-ray lines emitted that are spread over the energy range from 848.67 to 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 fs that implies a linewidth of 0.3 eV on the lasing transition. The linewidth, \(\Delta E\), equals \(h\Gamma/2\pi\) where \(h\) is the Planck's constant and \(\Gamma\) is the inverse lifetime or destruction rate of the transition which is determined primarily by the Auger lifetime. 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 instead of Ne II. 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 Mb compared to 0.3 Mb for the photo-ionization scheme. In this paper we show how

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**Fig. 1.** Energy level diagram for the photo-ionization driven inner-shell neon X-ray laser showing lasing on the 2p-1s line at 848.6 eV in Ne II.

**Fig. 2.** Energy level diagram for the photo-excitation driven inner-shell neon X-ray laser showing lasing on the 2p-1s line at 849.96 eV in Ne I.
we can take advantage of this much larger excitation rate to reduce the drive requirements on the XFEL source.

A simple atomic model of the levels shown in Figs. 1 and 2 was created. For Ne I the model includes the ground state, five singly-excited 3p states for the lower laser levels, and a single (1s) 3p state for the upper laser level for the photo-excitation lasing scheme. For Ne II the model includes the two 1s2s2p5 states and the 1s2s2p5 state as well as a single composite state for all the 1s2s2p3s states where 3l is a single 3s, 3p, or 3d electron. The final state is the 1s2s2p0 upper laser level for the photo-ionization lasing scheme. For Ne III there is a single ground state that is the final state for the Auger process from the 1s2s2p0 upper laser level. The atomic data from the NIST Atomic Spectra Database [13] is used in the model for the energy levels together with the data mentioned above.

The kinetics code Cretin [14] was used to model the kinetics and gain of the system under various conditions. The calculations use an ion density of $2 \times 10^{19}$ ions/cm$^3$ which corresponds to a pressure of 565 torr in Ne gas. This is similar to the conditions used in the Ne experiments [5] at LCLS. For the baseline case the XFEL beam is assumed to have $10^{12}$ photons in a 0.9 eV linewidth 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-fs FWHM while keeping the number of photons constant. The XFEL was designed to have a bandwidth of 0.1% as assumed in this paper even though the actual bandwidth is larger by a factor of 5—10 at the present time. The bandwidth has minimal impact on the photo-ionization mechanism but the strength of the photo-excitation mechanism is inversely proportional to the bandwidth. A challenge with the photo-excitation scheme is understanding the validity of the kinetics model and how to include the photo-excitation rate in the linewidth calculation of the gain. Currently the stimulated rate is included in the kinetics but not in the linewidth which means there are no Stark sidebands or broadening. The XFEL energy is set at 875 eV for modeling the photo-ionization scheme and 867.6 eV for modeling the photo-excitation scheme. For the 100-fs nominal pulse tuned to the 1s-3p line in Ne I we estimate the Rabi frequency [15] to be 1 eV which is comparable to the linewidth of the XFEL beam. The XFEL intensity is $1.4 \times 10^{17}$ W/cm$^2$.

Using an XFEL pulse with a 100-fs FWHM duration, Fig. 3 shows the predicted gain versus time for both schemes. The peak of the XFEL pulse is defined at time $= 0$. To understand the sensitivity to the XFEL flux, a series of calculations were done by using a multiplier between 1.0 and 0.001 on the nominal XFEL flux described above. For the photo-ionization scheme using the nominal intensity with a multiplier of 1.0, we predict a peak gain of $44 \text{ cm}^{-1}$ at a time of 81 fs before the peak of the XFEL drive. This is comparable to the inferred gain of $61-70 \text{ cm}^{-1}$ in the LCLS experiment [5]. The inverse photo-ionization rate is 20 fs at the time of the peak gain and reaches a minimum of 3.3 fs at the peak of the XFEL pulse.

In contrast the peak gain of $62 \text{ cm}^{-1}$ for the photo-excitation scheme occurs 128 fs before the peak of the XFEL. The big difference between the behaviors 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 $12 \text{ cm}^{-1}$ at $t = -12$ fs compared with $0.7 \text{ cm}^{-1}$ for the photo-ionization scheme. For both 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-fs duration XFEL driving the Ne gas as shown in Fig. 4. For the nominal XFEL flux, the peak gains are $910 \text{ cm}^{-1}$ at 0.5 fs before the peak of the XFEL drive for the photo-ionization case and $703 \text{ cm}^{-1}$ at 1.1 fs before the peak of the XFEL drive for photo-excitation. As the flux intensity is reduced, the gain for photo-ionization drops quickly but one notices that the peak gain for the photo-excitation scheme drops from $703 \text{ cm}^{-1}$ to $639 \text{ cm}^{-1}$ as the XFEL drive flux is reduced by a factor of 100. Also the peak of the gain
moves to a time of $-0.2$ fs, indicating near optimum drive conditions.

The absorption of the XFEL needs to be considered in both cases. At the time of the peak gain, the absorption coefficient for Ne absorbing the XFEL increases from 0.3 to 4.4 cm$^{-1}$ as the XFEL flux is reduced by a factor of 100 for the photo-ionization case. For the photo-excitation case, the XFEL is more strongly absorbed by the 1s-3p line, with the coefficient increasing from 6 to 14 cm$^{-1}$ as the XFEL flux is reduced by a factor of 100. In the LCLS experiments the Ne gas had a length of 0.28 cm.

As the XFEL flux drops further the gain drops more quickly and occurs after the peak of the XFEL flux, indicating that the flux is below ideal drive conditions. This figure shows that the photo-excitation mechanism offers potential to reduce the XFEL drive by two orders of magnitude 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.

Fig. 4. Gain versus time for the 2p-1s line in the neon X-ray laser driven by a 1-fs duration XFEL comparing the photo-ionization (top) and photo-excitation (bottom) mechanisms. The XFEL intensity is varied by using multipliers of 1.0 (nominal), 0.1, 0.01, and 0.001.

3. Modeling inner-shell Ar and Cu lasers

With 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. A promising candidate to consider is neutral argon gas, which has fully occupied K and L shells as well as 3s and 3p sub-shells. Fig. 5 shows the energy level diagram for using an XFEL above the L-shell edge of neutral Ar I to create an L-shell hole in singly-ionized Ar II. If the XFEL is 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 is 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 possible to tune the XFEL from low to high energy and watch the 3s-2p lasing turn on followed by lasing on both lines.

Fig. 6 shows the energy level diagram for using an XFEL above the L-shell edges of neutral Cu I to create a L-shell (2p) hole in singly-ionized Cu II and create lasing on the strong 3d-2p lines at 928 and 948 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 analogous to the Ne case discussed in the previous section. One advantage of Cu over Ar is that the M-shell is full and therefore one can obtain lasing on the 3d-2p lines that have larger emission rates than the 3s-2p and 3p-2s transitions used in Ar.

Fig. 5. Energy level diagram for the photo-ionization driven inner-shell argon X-ray laser showing lasing on the 3p-2s and 3s-2p lines in Ar II.

Fig. 6. Energy level diagram for the photo-ionization driven inner-shell neon X-ray laser showing lasing on the 3d-2p lines in Cu II.
To model this we created a simple atomic model of Cu with the Cu I ground state, 4 levels in Cu II as shown in Fig. 6, and 2 levels in Cu III. The calculations assume a Cu gas with an ion density of $2 \times 10^{19}$ ions/cm$^3$, so the binding energies from Ref. [16] differ from the binding energies by Bearden [9], which appear to be for Cu at solid density and therefore differ from the atomic values by the value of the work function. We assumed the Auger lifetime for the LMM transitions was 2.3 fs, similar to the KLL rates in Ne based on Ref. [17]. In the modeling the absorption oscillator strength for the 948 eV line is half of the 928 eV line and the calculated gain tends to be about half of the 928 eV line. As a result we only plot the gain for the 928 eV line in Fig. 7.

To model the gain we assume the nominal XFEL beam has $10^{12}$ photons at a photon energy of 1 keV in a 1.0 eV linewidth focused to a 1-μm diameter. For the pulse duration we compare a Gaussian shape with 100-fs FWHM to a 10-fs FWHM. Fig. 7 plots the gain of the 928 eV line versus time for the cases where the 100-fs XFEL pulse is reduced in intensity by multipliers varying from 1 to 0.01. Time $= 0$ is defined as the peak of the XFEL pulse. Using the nominal intensity with a multiplier of 1.0, we predict a peak gain of 31 cm$^{-1}$ at a time of 101 fs before the peak of the XFEL drive. As the intensity is reduced by a factor of 10, the peak gain drops to 22 cm$^{-1}$. Reducing the intensity by a factor of 100, gives a peak gain of 8.9 cm$^{-1}$ at 21 fs before the peak of the XFEL drive.

Keeping the same $10^{12}$ photons, we reduced the pulse duration to 10 fs and looked at the sensitivity of the gain for the 928 eV line versus time as a function of XFEL drive intensity. The peak gain for the nominal case is now 136 cm$^{-1}$ but a tenfold reduction in intensity only drops it to 126 cm$^{-1}$ and moves the peak closer to zero time as shown in Fig. 7. A further tenfold reduction (multiplier = 0.01) still yields a peak gain of 84 cm$^{-1}$ that now coincides with the time of peak XFEL drive. This suggests we could use a much smaller XFEL system or drive gain in a much larger volume if we have the shorter 10 fs pulse available. At the same time we predict a peak gain of 43 cm$^{-1}$ on the 948 eV line. Future work will look at even shorter pulse durations.

4. Conclusions

In this paper, we model the neon inner shell X-ray laser that demonstrated the Ne II lasing at 849 eV under similar conditions to those used in the experiments at LCLS. We discuss 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, the pulse duration, and the linewidth of the XFEL to better understand how to optimize this inner shell laser by understanding the tradeoffs 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-fs) 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 $n = 3\rightarrow 2$ transitions in materials such as Ar at 232 and 310 eV and Cu at 928 and 948 eV.

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References


