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Applications of X-ray lasers utilizing plasmas that are only a few times ionised

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Abstract. With the advent of tabletop X-ray lasers that operate at high repetition rate more emphasis is being put on finding useful applications for these lasers. The 14.7 nm Ni-like Pd X-ray laser at Lawrence Livermore National Laboratory is being used to do many interferometer experiments. As detailed quantitative comparisons are done between experiments and code simulations it is clear that some of the assumptions used to analyse the experiments need to be modified as one explores plasmas that are only a few times ionised. In the case of aluminium plasmas that have been analysed with interferometers there has been some unusual behaviour where the fringe lines bend the wrong way. In this work we will discuss how the index of refraction for aluminium is far more complicated than generally assumed because there are significant contributions to the index from the continuum and line structure of the bound electrons that can dominate the free electron contribution and even cause the index to be greater than one. We will also discuss some potential applications of the high repetition rate Ne-like Ar X-ray laser at 46.9 nm. In particular we will present modelling that shows how the Ar laser could be used to modify the absorption coefficient of a helium plasma and allow one to study the kinetics of plasmas with very low temperatures of a few eV. We will also discuss frequency doubling of the 46.9 nm laser.

1. Introduction

With the availability of table-top X-ray lasers that operate at high repetition rate more emphasis is being put on using these lasers in applications. A particularly useful application that many laboratories are pursuing is the use of X-ray laser based interferometers to study plasmas and other materials. At Lawrence Livermore National Laboratory, the 14.7 nm Ni-like Pd X-ray laser is being used to do many interferometer experiments. As detailed quantitative comparisons are done between experiments and code simulations it is clear that some of the assumptions used to analyse the interferometer experiments need to be modified as one explores plasmas that are only a few times ionised. In the case of aluminium plasmas that have been analysed with interferometers there has been some unusual behaviour where the fringe lines bend the wrong way. The basic assumption since the earliest days of x-ray laser interferometers using the Ne-like Y laser at 15.5 nm at NOVA is that the index of refraction can be calculated from the free electron density and that the bound electrons do not contribute to the index of refraction.

In this work we will discuss how the index of refraction for aluminium is far more complicated than generally assumed because there are significant contributions to the index from the continuum and line structure of the bound electrons that can dominate the free electron contribution and even change the sign of the gradient of the index with respect to the plasma density. We will also show how Al is not unique and how the bound electrons can have significant contributions for any material.

We will also discuss some potential applications of the high repetition rate Ne-like Ar X-ray laser at 46.9 nm. In particular we will present modelling that shows how the Ar laser could be used to modify the absorption coefficient of a helium plasma and allow one to study the kinetics of plasmas with very low temperatures of a few eV. We will also discuss frequency doubling of the 46.9 nm laser.

2. Index of refraction for Al plasmas

Recent experiments [1] at the COMET laser facility at LLNL and the Advanced Photon Research Center [2] at JAERI have observed anomalous behaviour of fringe lines in interferometer experiments of Al plasmas observed at late time when the plasma is cooling. These experiments used the 14.7 nm Ni-like Pd laser and 13.9 nm Ni-like Ag laser, respectively. The basic assumption for many years has been that the index of refraction can be calculated from the free electron density and that the bound electrons do not contribute to the index of refraction. The formula is $n = (1 – N_{elec} / N_{crit})^{1/2}$ where N_{elec} is the electron density of the plasma and N_{crit} is the plasma critical density. In the Al plasmas the fringes were observed to bend the wrong way, suggesting that the electron density is negative or the index of refraction is greater than one. This was observed at late time when the plasma is cooling and may be only a few times ionised. It was recognized that neutral Al has a negative contribution to the index of refraction due to the L3 absorption edge at 73.2 eV.

For neutral materials the complex index of refraction $n = 1 – \delta – i \beta$. β is related to the absorption coefficient while $1 – \delta$ is the real part of the index of refraction that is relevant for interferometers. The coefficient $\delta = 0.5 \times f1 \times (N_{ion} / N_{crit})$ where N_{ion} is the ion density and N_{crit} is the plasma critical density. For an ionised plasma the contribution to the index of refraction due to free electrons uses the same formula for δ but f1 is replaced by the number of free electrons per ion. For most neutral materials $f1 > 0$ but for Al at 14.7 nm and 13.9 nm $f1 = -1.4$ and $–0.8$, respectively. This raised the interesting question of whether Al was unique and what the index is for singly ionised Al and other nearby charge states.

The other interesting question is whether the X-ray laser can propagate through the partially ionised Al plasma. To address this question we used a Hartree-Slater code to calculate the absorption coefficient f2 for singly and double ionised Al and compared this with the published data [3] for neutral Al in Fig. 1. Keep in mind that $\beta = 0.5 \times f2 \times (N_{ion} / N_{crit})$. If one considers a plasma with an ion density of 10^{20} cm^{-3}, then the mean free path for 14.7 nm X-rays is only 24 µm for neutral Al but jumps to 512 and 1060 µm for singly and doubly ionised Al. This is because the absorption edge for Al moves from 73.2 eV for neutral to 91.7 and 105.4 eV for singly and doubly ionised Al. The result is that the X-ray laser goes from being highly absorbed by neutral Al to being on the less absorptive side of the edge for the partially ionised Al. This suggests that neutral Al does not likely play a large role in the anomaly because the plasma would be very absorptive. However the question then becomes what is the index of refraction for the partially ionised Al.

Table 1. Contributions to optical constant f1 at photon energy 84.46 eV for Al.

Ion	L3 edge (eV)	Line	Continuum	Free	F1-total
+0	73.1	2.61	-3.46	0.00	-0.85
+1	92.36	-0.70	-4.48	1.00	-4.19
+2	105.4	-2.91	-2.63	2.00	-3.54
+3	119.99	-3.16	-1.64	3.00	-1.80
+4	153.71	-2.49	-0.66	4.00	0.84
+5	190.47	-1.17	-0.29	5.00	3.54
+6	241.43	-0.57	-0.13	6.00	5.30
+7	284.59	-0.20	-0.06	7.00	6.73
+8	330.21	0.03	-0.03	8.00	8.00
+9	398.57	0.19	-0.02	9.00	9.18

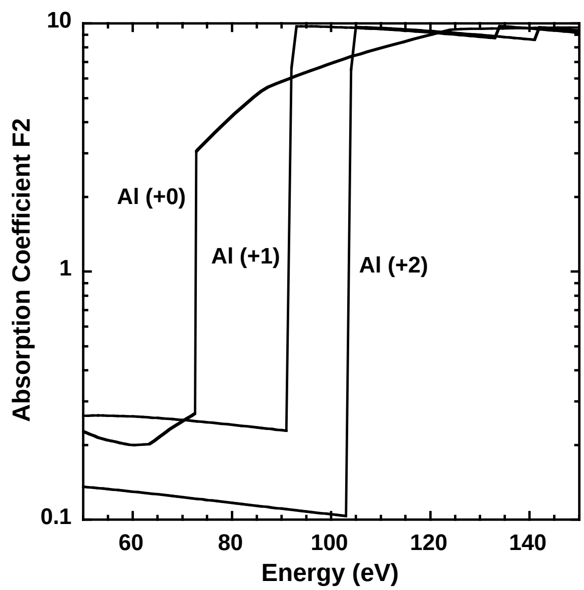


Fig. 1. Absorption coefficient f2 vs photon energy for neutral Al(+0), Al(+1), and Al(+2).

To address this we calculated the continuum absorption cross sections for each ionisation stage using a Hartree Slater code. The energies were adjusted to make sure the L3 edges agree with the experimentally measured edges for singly, doubly, and triply ionised Al. We then add the absorption for the lines below the L3 edges to the continuum absorption. For Al (+1) and Al (+2) we used the measured line positions and oscillator strengths from Refs. 4 and 5. For Al (+3) the n=2 to n=3 and 2p to 4s line positions and line strengths were used from Ref. 6. Using the absorption coefficient f2, the real part of the index, f1, is then derived using the Kramers-Kronig dispersion relation. This part of the index is the bound electron contribution so we then add the number of free electrons to determine a total value for f1. Ref. 3 has a description of the Kramers-Kronig dispersion relation. Keep in mind that the line contributions are extremely sensitive to the line positions and should include the effects of line broadening. The calculation uses the best data we have available but still has a lot of uncertainty associated with it. Table 1 shows our best calculation of the partial and total f1 value for each ionisation stage of Al for a 14.7 nm X-ray.

From Table 1 one can see immediately that there can be large contribution to f1 from both the absorption lines and from the continuum absorption edges. If one looks at the ratio of f1-total to the number of free electrons this is the same as the ratio of the measured electron density to the actual electron density when we use the usual analysis that only considers the free electron contribution. One can greatly underestimate the electron density for low ionisation states. In fact one needs to reach Al (+7) before the contribution from the bound electrons becomes less than a 10% effect.

To understand how important the line contribution can be let us look at Al (+2), which has a large line contribution. Looking at the data in Ref. 4, there is a strong line measured at 89.89 eV with oscillator strength, f_{osc}, of 0.377. The incremental contribution to f1 from a single line, $\Delta f1(E) = f_{osc} / [(E / E_0)^2 - 1]$, where E_0 is the energy of the line. For our X-ray laser at 14.7 nm (84.46 eV), $\Delta f1 = -3.22$. For the Ag laser at 13.9 nm (89.25 eV), $\Delta f1 = -26.6$. The value of $\Delta f1$ is very sensitive to the exact position of the line and its line width as you approach resonance.

One needs to remember that this problem is very complex and these results are only approximate. For all these calculations we assume the plasma is only in the ground state of the particular ionic stage while a real plasma will have a distribution of population in excited states.

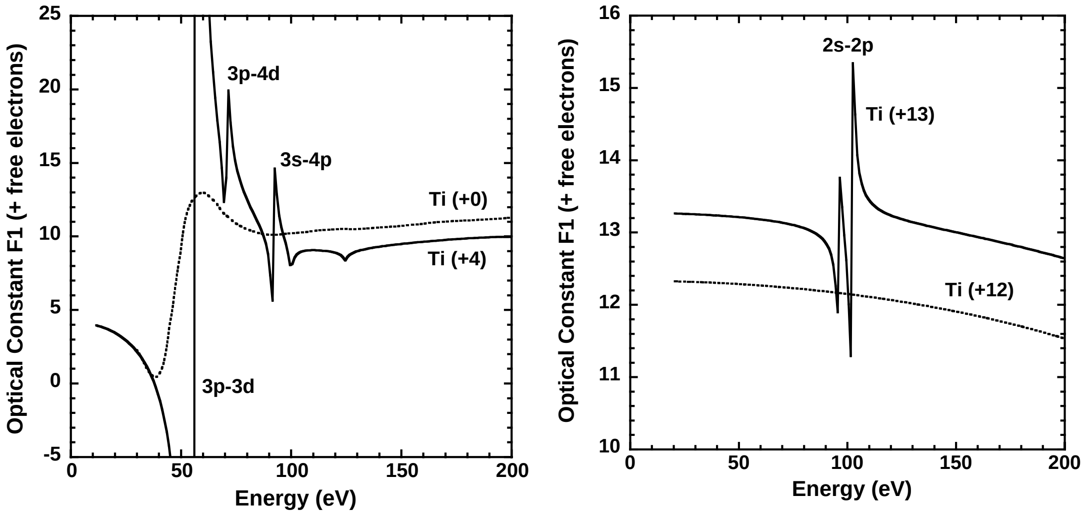
3. Index of refraction for a Ti plasma

Having shown that the bound electrons can have a significant contribution to the index of refraction for an Al plasma the question remains as to whether this issue is important in other materials that do not have neutral absorption edges near the X-ray laser energy. If one looks at the ionisation energies across the periodic chart a typical material has its first ionisation about 5 to 10 eV. As it continues to ionise there will almost always be an edge near the X-ray laser energy that needs to be considered.

For Ti we decided to calculate f1 for Ti (+4), Ti (+12), Ti (+13) and compare these with neutral Ti. For each ionisation stage we calculated the photoionization cross-sections with our Hartree-Slater code. Using the Grant code [7] we then calculated oscillator strengths for the strongest lines and added these to the absorption cross section. We then estimated the term f1 by using the Kramers-Kronig dispersion relation and then added the free electrons to calculate a total value for f1.

In Fig. 2 we compare our estimated f1 for Ti (+4) with neutral Ti. Ti (+4) is interesting because it is a stable sequence with closed 3s and 3p subshells. It has an ionisation potential of 98 eV. From Fig. 2 one sees that at 84.46 eV, f1 = 10.3 for neutral Ti and still has a value of 11.0 for Ti (+4). For Ti (+4) we expect the total f1 = 4 so in an actual experiment we would overestimate the electron density by 11/4 or 175% when you take the ratio of these numbers.

If we now look at Ti (+12) where the ionisation potential is 788 eV one would expect all the absorption edges and lines to be at much higher energy than the X-ray laser energy and therefore the bound electrons should make a very small contribution. That is what we estimate in the calculation shown in Fig. 3. Now if we do the same calculation of f1 for Ti (+13) we discover that there are 2 weak lines 2s-2p lines at 96 and 102 eV that perturb the value of f1, as shown in Fig. 3. At 84.46 eV the effect is small but as you approach the resonances the effect from these small lines can be significant.



Figs. 2 (left) and 3 (right). Optical constant f1 versus photon energy for neutral Ti(+0), Ti(+4), Ti(+12), and Ti(+13).

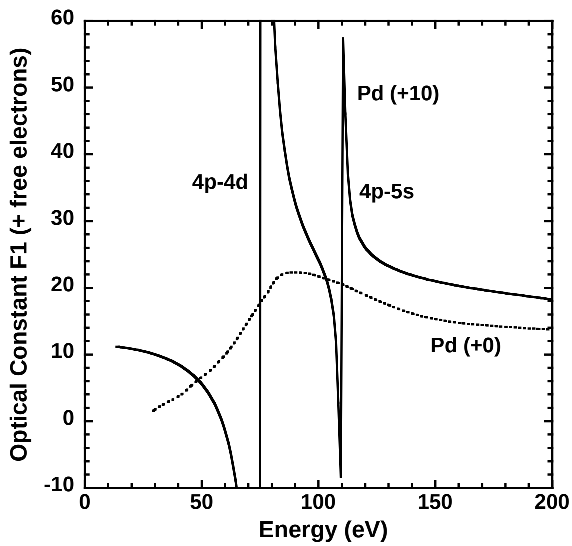


Fig. 4. Optical constant f1 versus photon energy for neutral Pd(+0) and Pd(+10).

4. Index of refraction for a Pd plasma

In order to better understand the physics of the Ni-like Pd X-ray laser, experiments have been done to look at the electron density of the Pd as prepared by the prepulse. Taking Pd (+10) as an example, we calculate the optical constants and compare with neutral Pd using the same procedure as described for Ti above. Pd (+10) has an ionisation potential of 239 eV so we might expect a significant abundance of this ion for plasmas below 100 eV. Figure 4 shows the total value of f1 including free electrons for neutral Pd (+0) and Pd (+10). At 84.46 eV, neutral Pd has f1 = 22 while Pd (+10) has f1 = 43. In both cases we would greatly overestimate the electron density of the plasma. For the Pd (+10) one can see that the strong lines, especially the 4p – 4d line estimated to be at 75 eV with oscillator strength of 7, have a large contribution to f1.

5. Studying kinetics of He plasma with Ar XRL

The Ar X-ray laser at 46.9 nm (26.46 eV) has been available for many years and offers a high repetition source with 1 mJ output. This laser has been used in many interferometer experiments but we would like to propose several other potential applications. Since neutral He has its photoionization edge at 24.59 eV we first looked at whether we could use the Ar X-ray laser to ionise neutral He and drive a laser in neutral He on either the 3d – 2p line at 667.8 nm or the 2p – 1s line at 58.4 nm as shown in Fig. 5. If we look at driving a He gas with ion density of 10^{18} cm^{-3} with an Ar X-ray laser with output of 10^{10} W cm^{-2} in a 1 ns pulse the calculations show that we quickly make a 2 eV plasma of He. The calculations do not show any gain on these lines because the plasma is too hot and the upper states tend to collisionally ionise very rapidly. There may be other conditions where gain could be achieved. However we can easily ionise the neutral He plasma which suggested that we could study the kinetics of the plasma recombining or even watch the absorption change as a function of time.

In Fig. 6 we show the absorption coefficient of He versus energy at several different times for the plasma created by driving a He gas with ion density of 10^{18} cm^{-3} with an Ar X-ray laser with output of 10^{10} W cm^{-2} in a 1 ns pulse. For simplicity we assume a square pulse that turns on at time 0 and turns off at 1 ns. In Fig. 6 one observes that the K edge at 24.59 eV is quickly bleached away by a factor of 100 as we almost completely ionise the neutral He with the Ar X-ray laser. At 1 ns the singly ionised K edge at 54.4 eV dominates the absorption. After 10 ns the He has recombined sufficiently that the absorption from the neutrals start returning. It would be very interesting to do experiments to probe a He plasma at different times and measure the absorption coefficient as a function of time. Alternatively one could watch the emission of the $K\alpha$ line at 58.4 nm versus time to study the recombination process in the He plasma. Figure 7 shows the absorption coefficient (dotted line) at 46.9 nm and the $K\alpha$ emission at 58.4 nm versus time for the plasma conditions described above. One could also do interesting experiments to bleach a channel in the He gas using the Ar X-ray laser.

6. Frequency doubling the Ar XRL

The Ar X-ray laser at 46.9 nm (26.447 eV) looks like a promising candidate for trying some two-photon absorption experiments. As shown in the last section it should be fairly easy to ionise a He gas. In singly ionised He there is a very good two-photon absorption resonance from the 1s ground state to the 6d excited state as shown in Fig. 8. The 6d state is nearly degenerate with the 6s and 6p states. We estimate the two-photon absorption is off resonance by only –0.011 eV for the 1s – 6d transition. For the two-photon absorption process the 2p state is used as the intermediate state. However the intermediate state is far off resonance so this does reduce the absorption rate significantly. Using the oscillator strengths for the 1s – 2p – 6d transition we estimate the two photon absorption rate $W = 10^{-24} \times I^2$ sec^{-1} per ion where I is the laser intensity of the Ar X-ray laser in W cm^{-2}. For $I = 10^{10}$ W cm^{-2} and an ion density of 10^{18} cm^{-3} the two photon absorption rate would be 10^5 per nsec per cm^3. This assumes a line width of 10^{-7} eV for the two-photon resonance. This would be followed by the 6p – 1s spontaneous emission at 23.4 nm. Other longer wavelength lines would also be emitted. This calculations are quite preliminary but do suggest that some interesting two photon effects might be observed using the Ar X-ray laser with a singly ionised He plasma, especially if we had a higher intensity X-ray laser.

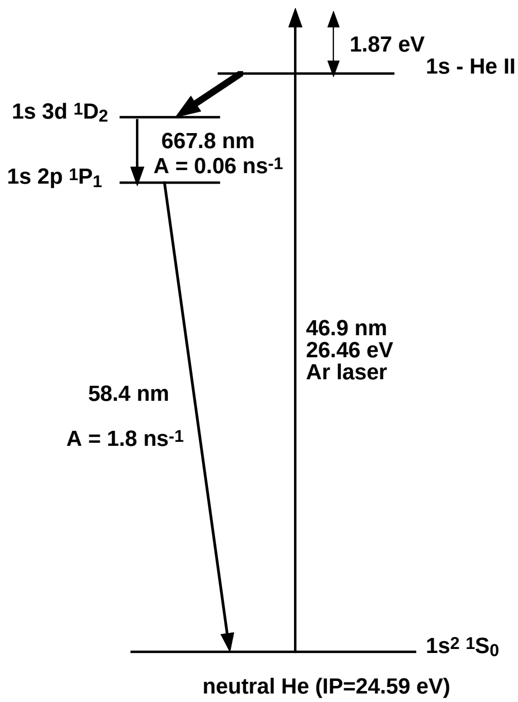


Fig. 5. Energy levels of He I showing potential laser lines.

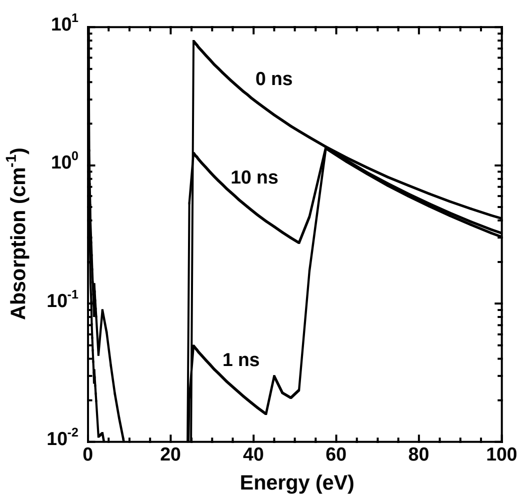


Fig. 6. Absorption spectra of He at different times before and after illumination by 1 nsec duration Ar X-ray laser.

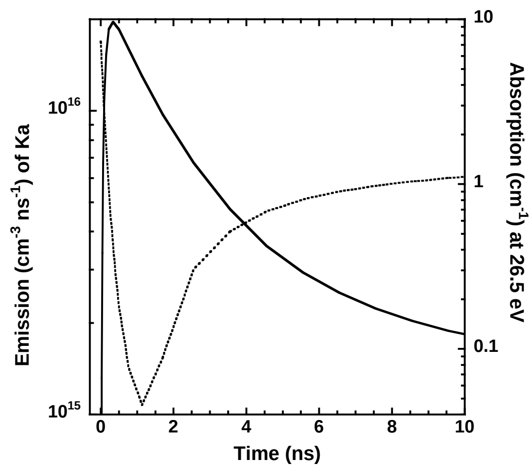


Fig. 7. Emission of He K-α line (solid) and absorption of Ar X-ray laser line (dotted) in He plasma versus time.

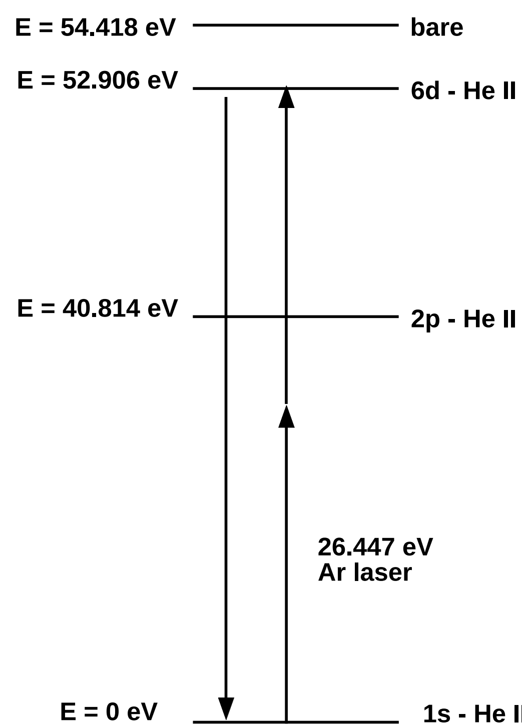


Fig. 8. Energy level diagram of He II showing two-photon absorption of Ar X-ray laser.

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