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Demonstration of Resonant Photopumping of MoVII by Mo XII for a VUV Laser Near 600 Å.

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Abstract

We present data of experiments on the resonant photopumping of Mo VII by Mo XII as a method of generating a coherent VUV source near 600 Å. The experiment is based on a scheme proposed by Feldman and Reader¹ in which the $4p^6 - 4p^56s$ transition in Mo VII is resonantly photopumped by the $5s^2S_{1/2} - 4p^2P_{1/2}$ transition in Mo XII. Results of the laser produced plasma experiments show the successful enhancement of the population of the Mo VII $4p^56s$ upper lasing level when pumped by an adjacent Mo XII plasma. No enhancement was seen in a control experiment where the Mo VII plasma was pumped by a Zr X plasma. Improvements of the intensity of the Mo XII pump source, achieved using an additional pump laser, lead to the generation of a population inversion for the VUV transition.

I. Introduction

Since the invention of soft X-ray lasers^{2,3} research has focused on developing shorter wavelength lasers with a recent record at 35.6Å laser⁴. A result of this trend is that the important spectral region between 400Å and 900Å has not received sufficient attention. Although the development of VUV lasers near 900 - 1000Å using Auger decay^{5,6} was a very impressive achievement and may provide a path to the region below 900 Å, altogether there is a very little research on the development of VUV lasers for this spectral region. Such lasers, particularly those operating near 600Å, are expected to be very useful as their photon energy (~20 eV) is close to molecular bonding energies.

Detailed spectroscopic studies of the electronic properties of clusters, radicals, and molecules in molecular beam apparatus require a high brightness, narrow linewidth source for high resolution at these photon energies. At present, one may choose one of two primary sources; synchrotrons, which possess sufficient brightness (but not narrow linewidth), are often too inconvenient to use due to their size and lack of availability except at large facilities; discharge lamps, although capable of narrow linewidth at low pressure, exhibit poor brightness under those conditions. A laser with photon energies in the 10 to 30 eV range would be an ideal source to replace these two given its potential size, resolution and brightness. A second area of research for which a laser in this spectral region would be useful would be as a photoionization source for mass spectroscopy. Photons around 20 eV are ideal for single photon ionization of most constituents found in molecular beams. Higher energy sources such as present soft x-ray lasers are too energetic and would cause complete ionization of the constituent atoms if used. At present, electron bombardment ionizers are commonly used in mass spectrometers. A high repetition rate, 600Å laser would have the capability of improving the ionization rate by a factor of 100 over existing systems.

Only a few proposals for generating a laser at these frequencies exist. One of them is based on conversion techniques utilizing an existing input laser⁷. There is a proposal for

harmonic generation using Free Electron Lasers, but these still suffer from technological immaturity⁸. There was one report of a collisionally pumped system demonstrating gain near 600Å, ⁹ but no one has reproduced this work. Finally there is proposal¹⁰ for a photoionization pumped laser in Cs III which would operate at 638Å.

Another potential technique for producing a laser at these wavelengths is resonantly photopumped lasers. Photopumping has recently been the subject of great interest both for the generation of lasers, as a spectroscopic tool for determining temperature and density conditions, and for studying transport in high density plasmas¹¹⁻¹³. Photopumping as a means of generating population inversions suitable for obtaining lasing action in the soft x-ray wavelengths was first proposed independently by Norton and Peacock¹⁴ and Vinogradov, Sobel'man, and Yukov¹⁵ in 1975. One UV wavelength laser incorporating a resonant photopumping process has been demonstrated. This was the work by Qi et al.¹⁶⁻¹⁸ which reported lasing action for several CIII lines near 2000 Å pumped by a Mn IV plasma. The shortest lasing line was at 2163Å. In addition, researchers have identified several wavelength matches for ion pairs that may act as potential lasing schemes¹⁹⁻²¹ and work has been carried out for several of them²²⁻²⁵.

There are, to our knowledge, two proposals for producing lasers near 600Å^{22,27} using resonant photopumping. Both these schemes suffer from pump-absorbing line wavelength mismatches. It has been suggested that by counter streaming two plasmas these mismatches could be reduced by the Doppler shifts induced by bulk plasma motion²⁸. Recently Feldman and Reader¹ reported the most promising line resonance proposal for a laser near 600Å. Their proposal uses an experimentally verified line coincidence between Mo XII and Mo VII as a way of producing a resonantly photopumped laser near 600Å. This scheme has several distinct advantages: the transition overlap is quite good, both the pump and absorbing lines have large oscillator strengths, the lower level 5p of the lasing transition has a fast decay rate, and the pump and lasing plasmas can be optimized separately. In the following sections, we will discuss our experimental progress in demonstrating the feasibility of this photopumping scheme.

II The MoXII-MoVII photopumping scheme

The scheme proposed by Feldman and Reader¹ employs the $4s^2 5s\ 2S_{1/2} - 4s^2 4p\ 2P_{3/2}$ transition in gallium-like Mo XII at 136.499Å to pump the krypton-like MoVII $4s^2 4p^6\ 1S_0 - 4s^2 4p^5 6s\ 1/2[1/2]_1$ transition at 136.507Å to produce a population inversion and gain on one of the several $4s^2 4p^5 6s - 4s^2 4p^5 5p$ transitions near 600Å. The existence of this resonance was demonstrated by Feldman and Reader through a detailed spectroscopic study of laser and spark produced Mo plasmas with the 10.7 m grazing and normal incidence spectrometers at NIST¹. An simplified energy level diagram for this system is shown in Fig. 1. In this figure, the pump transition is shown along with other transitions whose intensities were commonly monitored during our experiments. For Mo VII, several excited configurations with numerous levels are represented by the boxes and the approximate wavelengths of the associated transitions are indicated. Since resonant photopumping should only enhance the population of the single 6s level, transitions from other levels can be monitored to determine if only the resonant level is being pumped. This also helps differentiate if enhancement in the population of a particular level is due to resonant photopumping or due to some other pumping process such as collisional excitation.

In addition to the good wavelength match, this proposal has several inherent advantages compared to other potential line resonance schemes. Both the pump and absorbing transitions are to the ground level and are characterized by large oscillator strengths. Calculations²⁹ established that the decay rates for transitions from the lower level 5p in Mo VII were indeed sufficient to expect that a population inversion could be achieved for the 5p-6s transitions. Both ions are of the same element which simplifies target design and construction. A survey of the reflectivity of elements near 600Å indicates that several easily obtained metals such as iridium and platinum have above a 20% reflectivity near 600Å making these suitable for designing cavity mirrors. Finally one of the best soft x-ray wavelength mirrors constructed, a molybdenum-silicon multilayer, has high measured reflectivity near the 135Å pump wavelength^{30,31}. The

availability of such a mirror allows for the design of a pumping system to overcome some of the geometrical coupling losses that are associated with photopumping schemes.

III. Experimental Setup

A block diagram illustrating the experimental apparatus is shown in Fig. 2. Included in this figure is a drawing of the typical target arrangement used in our experiments. The primary diagnostics were visible and VUV spectrometers. These were used in cooperation with a Macintosh based data acquisition and analysis system.

The first of the two lasers used in these experiments was a Lumonics Model 610, 10 J 150 ns Transverse Atmospheric Excitation (TEA) CO₂ laser. The optics consisted of a beam splitter and cylindrical and spherical lenses for producing the cold absorbing and hot pump plasmas respectively. The system provided for both line and spot focus patterns on target with power densities between 10^7 and 10^9 W/cm². In addition, a beam delay unit capable of optically delay half of the beam by between 0 and 135 ns with respect to the undelayed half of the beam was constructed. An optical delay ensured a more reproducible delay between the formation of the pump and the Mo VII plasma than could be achieved with the synchronization of separate lasers.

The ruby laser system, which was introduced in the later experiments aimed at improving the pump intensity, consisted of a Korad Q-switched oscillator and two amplifier units. It provided a 3 J, 20 ns beam on target which was focused with a 25 cm spherical lens.

VUV spectra were recorded with a 1.5 m grazing incidence McPherson Model 252 spectrometer with a microchannel plate (MCP) and Reticon detector array. It was fitted with a 600 grooves/mm grating with a 5 degree blaze angle. The system was capable of monitoring spectra from 300 to 700Å in first order. The grating is about 60% efficient in second order, effectively extending its range to 150Å. The UV-visible spectrometer was a 0.5 m Ebert-Fastie type Jarrel-Ash air monochromator with a 1200 gr/mm normal incidence grating. This instrument was used primarily for calibration of the VUV instrument. The data acquisition system has been detailed previously³².

To facilitate temporal studies of the plasmas, a MCP gating circuit was designed and implemented. Gating of image intensifier tubes has been used in several applications to provide temporal resolution of plasma evolution³³⁻³⁸. Our system had two modes. The first mode permitted integration of the signal from the time the laser fired to a predetermined cutoff point. This system ensured there was no loss of data when we experienced jitter problems with the lasers. The second mode provided for a windowed sample which was produced by applying a voltage pulse of variable position in time, with respect to the trigger of the laser, and of variable width to the MCP.

IV. Initial Target Studies

Numerical calculations by Klapisch et. al²⁹ indicated that gain could be achieved through resonant photopumping of a plasma with a density of 10^{18} cm^{-3} and electron temperatures below 10 eV. The model used for these calculations²⁹ assumed a steady state coaxial pumping arrangement produced by a spark source. The low temperature requirement for the absorbing plasma prevents collisional excitation of the lower (5p) levels. Producing such a highly ionized but cold plasma is similar to the requirements encountered during the investigation of recombination x-ray lasers. It has been shown both theoretically³⁹ and experimentally³ that a highly ionized low temperature plasma can be formed in rapidly recombining laser produced plasma. The relevance of Klapisch et al²⁹ model to such an experimental setup has been previously discussed⁴⁰. The key omission of this work²⁹ was the neglect of recombination effects which were important in our experimental setup. In addition, the assumption of a spark generated plasma implies geometrical scale lengths much different than those encountered with the laser produced plasmas. However, comparisons between the two experimental cases indicated that key plasma parameters given above represent suitable conditions for a recombining laser produced plasma.

The start of this investigation began with experiments aimed at determining suitable conditions for producing both the Mo XII pump plasma and the Mo VII lasing plasma. These studies focused on a spectroscopic evaluation of radiatively cooled expanding

plasmas produced by normally incident laser on a variety of target geometries⁴¹. These early experiments were done with a time integrating spectrometer system. Initial tests to demonstrate resonant photopumping were negative. The hypothesis for this negative result was mismatch between the time of the peak Mo XII line emission and the time that the pumped plasma reached a peak population of the Mo VII ground state.

V. Plasma Evolution Studies

a) The Pump ("hot") Plasma

A mask was positioned so that the spectrometer viewed a chord of plasma 2 mm above the target. This position was chosen since the time integrated results indicated that there was strong Mo XII emission at this position. In addition, the ratio of the Mo XII line emission to the continuum radiation was much larger for the plasma 2 mm above the surface than the ratio nearer the target surface. Minimizing the amount of continuum radiation was necessary to reduce the flux of radiation that could photo-ionize or photo-excite ions in a laser plasma.

Figure 3 is a plot of the intensity, in arbitrary units, as a function of time for three spectral lines, all observed from the pump plasma. The intensities plotted are for two of the Mo XII $4s4p^2 - 4s^24p$ transitions, and a Mo VII $4f-4d$ transition. From these results, it was determined that the peak Mo XII line emission occurred at approximately 125 ± 10 ns after the start of the laser pulse, and that the temporal full width at a half maximum (FWHM) of this profile was 80 ± 10 ns. The errors are associated with errors in reading the crossings, and in the jitter of the trigger pulses. Comparing the intensities of the Mo XII lines to intensity of the Mo VII line, one can note more than an order of magnitude difference. This is expected since during recombination the plasma would have to cool through several ionization stages prior to reaching Mo VII. The peak of the Mo VII emission from the pump plasma occurs much later than 250 ns. This was of concern because Mo VII ions formed by recombination in the pump plasma had the potential of resonantly exciting transitions in the Mo VII laser plasma which could adversely affect the buildup of a population inversion. From these measurements it is possible to estimate

the plasma bulk velocity, and hence the plasma temperature. As an upper limit to the plasma bulk velocity, it is possible to take time at which radiation is first observed, approximately 30 ns, as the time it takes the plasma to expand 2 mm. This gives a sound speed of 5×10^6 cm/s. For Mo XII, this translates into a temperature of about 200 eV. As the ionization potentials for Mo XI and XII are 209 and 230 eV respectively, the estimated temperature is consistent with the observation of these lines in the spectra. From a similar experiment where the mask was moved closer to the target surface, no delay was measured. This supports the belief that the initial temperatures are in fact larger.

b) The Mo VII ("cold") Plasma

With the establishment of the time of the peak Mo XII emission, and the other temporal characteristics of the hot plasma a similar study for the cold plasma was conducted. The experimental geometry, data collection, and analysis techniques used were similar to the previous hot plasma experiments, except that now the focusing optic was replaced with a cylindrical lens. Use of the cylindrical lens fulfilled two goals: the production of an elongated lasing plasma and the creation of a plasma dominated by ions in lower ionization stages. The cold plasma was studied in two spectral regions, the 400Å range and the 650Å range, so that the behavior of the population of the various levels of Mo VII could be studied in detail. The intensities for several lines of interest are plotted in Figs. 4 (a) and (b).

The first piece of information gleaned from these scans was that there was indeed a temporal mismatch between the Mo XII peak emission time at 125 ns in the hot plasma and the peak Mo VII ground state density in the cold plasma. To identify the time of the peak Mo VII ground state density, we assumed that for a rapidly recombining plasma at low temperatures (the type expected for laser produced plasmas), there is not a significant amount of collisional excitation from the ground level of an ion. Population of the excited levels of an ion is due mainly to recombination into these states. Thus the time of the maximum density of the ground state for a particular ion can be considered to

coincide with the observation of the peak line emission for the next lower ionization state. For these experiments, the peak Mo VII ground state was assumed to occur near the time of the observation of the peak Mo VI line emission. It was determined that this peak occurs about 200 nsec after the laser hits the target(see Fig. 4b). This time-dependent intensity was obtained from an averaging of two Mo VI lines, each one with a slightly difference in the location of their peaks. Thus the measured mismatch between the two desired plasma conditions was about 75 ns. This confirmed the hypothesis that the lack of observed enhancement was due to temporal mismatch.

c) Cold plasma pumped by a Mo XII plasma

From this time resolved data it is also possible to estimate a temperature for the cold plasma. The time at which radiation is first observed 2 mm above the target surface is about 120 ns. This corresponds to a bulk plasma speed of approximately 2×10^6 cm/s, and a temperature of about 20-30 eV. This value is significantly higher than the limit of 10 eV required by Klapisch's et al²⁹ calculations for the observation of gain. However, this is the temperature as the plasma first expands to 2 mm. It is expected that as the plasma expands further, additional cooling will result in a temperature below 10 eV.

Prior to the discussion of the experimental results for the resonant photopumping experiments, it is possible to make an estimate of the relative cooling and recombination rates for the "cold" plasma. It was shown that a low electron temperature plasma with a high density of Mo VII ions would be required. This implies that for the lasant plasma the cooling rate must be significantly faster than the recombination rate. Although an accurate measurement of the density could not be done due to experimental limitations, it is possible to use reasonable estimate for the density and come to an order of magnitude approximation for these rates.

The adiabatic cooling rate can be estimated by considering the ratio of the measured plasma bulk velocity V_B to the plasma scale length L , where T is the electron temperature

$$\frac{\dot{T}}{T} = \frac{V_B}{L} \quad (1)$$

The three body recombination rate can be estimated from the expression of Zel'dovich and Raizer⁴²

$$\frac{N_{3\text{-body}}}{N} = N_e^2 \frac{8.75 \times 10^{-27} Z^3}{T_e^{9/2}} \quad (2)$$

where in the Eq. (2) $\ln(\Lambda)$ the coulomb logarithm has been replaced by unity, N is the ion density, N_e is the electron density, and Z is the ion charge. The radiative recombination rate is taken from Seaton⁴³. With these equations, a comparison of the three rates for both the lasing and pump plasmas can be made and is summarized in Table 1.

The estimate of the density range was motivated by the upper limit on the plasma density (10^{19} cm^{-3}) which is due to the critical density for the CO_2 laser. The scale length for both these plasmas was 2 mm.

From Table 1 one can see that for the lasing plasma, even at the upper density limit, the cooling rate is still an order of magnitude larger than the recombination rate. This indicates that it should be possible to cool the plasma faster than the Mo VII ions are lost through recombination.

The other point for the "cold" plasma is that radiative and three body recombination are about equally important to the evolution of the plasma. The estimated recombination times can be compared to the observed recombination times. Assuming that primary source of Mo VI ions in the excited states is recombining of Mo VII ions, we can roughly estimate from the temporal difference between the peak intensity of Mo VII and Mo VI lines that the recombination time for the Mo VII ions is about 30 ns. We neglected the contribution of the collisional excitation to Mo VI line intensity because their excitation energy is significantly larger than the plasma temperature. However the estimated recombination time (see Table 1) is about 1 μs at the higher density. The time evolution of the line intensities indicates that the actual recombination rates have been underestimated implying that the recombination rates are close to the estimated cooling rates. However, the cooling rates have also been underestimated by the neglect of radiation cooling which is significant for high Z plasmas.

With the identification of the magnitude of the temporal mismatch, corrections utilizing the beam delay optics were implemented to achieve the necessary synchronization. The first target arrangement for these experiments had the two Mo pieces shown in Fig. 2 separated by a distance of 3mm. The results for the key lines are shown in Fig. 5. These plots compared the intensity for the cold plasma alone (when the laser beam used to create the pump plasma was blocked) and the case when the cold plasma was pumped by the adjacent Molybdenum plasma. Figs. 5 (a) and (b) show the intensities for a 6s-5p and a 4f-4d transitions respectively. Both show about a factor of 2 enhancement in peak intensity at the time of the peak of the Mo XII pump emission. Comparing this to the other Mo lines, Figs. 5 (c) and (d) show representative intensities for Mo VI and Mo VIII, respectively. There is no significant increase of the observed intensities for the pumped case as compared to the "cold plasma" alone. These results indicate that only the populations of the upper Mo VII levels are being enhanced. The spread of the points is due to the fluctuations from shot to shot.

As a check on the results and conclusions of the previous experiment, a control experiment was conducted for the same target parameters. This accomplished by replacing the pump plasma target in Fig. 2 with a geometrically equivalent piece of Zirconium. The use of a Zirconium target tests for any excitation and enhancement produced by collisional pumping or broadband photopumping from the continuum. By using Zirconium, only the resonance between Mo VII and Mo XII is removed. Using such a pump plasma eliminates consideration of non-resonant effects as an explanation for the observed enhancement. The results of this experiment are easily summarized in Fig. 6 and support the conclusion of the observation of resonant photopumping. A survey of these plots in Fig. 6 shows that there is no significant change in the measured intensity for any of the Mo VII transitions, nor is there a change in the intensity of the other lines from Mo VI and VIII ions that were shown in Fig. 5.

A comparison of two spectra, both at the peak emission time for Mo XII pump, are shown in Fig. 7. These spectra again compare the two cases of (a) the "cold plasma"

alone , and (b) the "cold plasma" pumped by a Mo XII plasma. Several key features are noted for this spectra. All the Mo VII 6s-5p transitions in this region are enhanced. For the two lines near 645Å, it is important to realize that the 645.417Å line is on the shoulder of the peak identified in Fig. 7 (a), and that the 645.93Å line is between the two peaks as indicated. In the case of the spectra for the pumped shot, these two lines are enhanced to become the two prominent peaks identified. In addition, there is a wavelength shift in the peak of the 677Å line. For a cold plasma alone, the peak is at 677.78Å, which is the wavelength expected for the second order 4f-4d transition. In the pumped spectra, the peak is shifted to 677.70Å which is the wavelength of the 6s-5p transition that is blended with this line. A careful study of these spectra indicated that the lines showing the most significant enhancement were the lines whose upper level is the photopumped $4p^5 6s [1/2, 1/2]_1$ level. The other key line whose behavior is noteworthy is that of the 5p-4d transition at 674.41Å. This line can be seen in the spectra for the "cold plasma" alone, but is not enhanced and is so small in the pumped case so as not to be seen. This is suggestive that these experimental conditions were successful in populating the 6s levels while collisional processes were sufficiently low so as not to affect the 5p populations greatly. Comparing further these two spectra, it is also clearly seen that the intensities of the other Mo ions, and the intensities of both the lower lying transitions of Mo VII such as the $4p^5 4d-4p^6$ transition at 654.54Å, are equivalent for the pumped and non-pumped cases. A similar evaluation of the spectra for the Zr pumped experiments showed no significant change in the measured line intensities that is not due simply to shot to shot fluctuations. The spectra and the plots combined show that for this experiment, there are no observable effects. The Zirconium pump plasma is sufficiently distanced from the cold plasma so that there is no observed enhancement by collisions, by broadband continuum photopumping, or by resonant photopumping as expected. This is essentially a null experiment that support the conclusions that the enhancements in intensity seen in the photopumping experiments were solely a result of resonant photopumping.

VI. Two Laser Pump Enhancement Studies

The successful demonstration of resonant photopumping described in the previous section demanded the maximum capabilities of all the equipment in laboratory, especially the CO₂ laser. As the long term goal for this research was to produce an operating laser near 600Å, it was clear that improvements were required before further work could continue. As pointed out in earlier discussions, one of the most significant problems with the experimental geometry was the poor optical coupling of the two plasmas. As it required 60 % of the available laser energy just to produce a 200 μm spot size on target, it was not possible to improve on the spatial size of the Mo XII pump using the CO₂ laser alone. We therefore decided to use an additional shorter wavelength laser together with the CO₂ laser in order to increase the pump flux.

A 3 J, 20 ns ruby laser was aligned into the target chamber. This two laser combination could be exploited in three ways to improve the pump flux. With the significantly larger power densities that can be produced by the Ruby laser, the spot size on target that would give the equivalent power density as the CO₂ laser ($2 \times 10^{10} \text{ W cm}^{-2}$) has a radius of 0.47 mm thus improving the geometrical coupling of the "hot" and "cold" plasmas. The shorter wavelength of the ruby laser at 0.6943 μm compared to 10.6 μm for the CO₂ laser means that a factor of one hundred higher plasma density can be reached with the ruby laser owing to the higher critical absorption density. Finally, by utilizing the two lasers together, pre-forming a plasma with one laser, and then heating it with the second could also result in an increase in the plasma temperature and hence the pump strength. Initial tests showed that the Mo XII line emission from a plasma produced by the ruby laser only was too weak compared to the background continuum radiation produced from such a dense plasma, hence it would not be a suitable pump source. Two laser interaction experiments, where a plasma was created by one laser and heated by the second, was much more successful in increasing the intensity of the Mo XII pump lines. Since the relative timing of the two lasers was a key factor in this interaction, the spectrometer was refitted with the electronics to make it an integrating

system. This eliminated problems with the delay times between the two laser beams, and the MCP gating circuit. The two lasers were aligned so that the spots overlapped at the target surface. The mask was placed 2 mm above the surface, as this was the best location for the CO₂ laser alone. Spectra was recorded for various delays between the time the ruby laser and the CO₂ laser hit the target. A temporal calibration was made to ensure that the cable and detector response delays were accounted for in measuring the temporal separation between the arrival of the two lasers on target. The maximum measured 4 times improvement in the Mo XII line emission occurred for the case where the ruby laser was fired approximately 175 ns prior to the CO₂ laser. This result can be explained by the fact that in this case the CO₂ laser can initially interact with a plasma with a matching density, rather than with a solid. Spectra recorded for quite different temporal separations between the lasers, both larger and smaller, were identical to spectra for a CO₂ laser alone. While completing this temporal scan, care was taken to move the target to a new position every five shots as examination of the targets showed that significant damage occurred after about 10 shots invalidating the recorded data. The ruby laser was incident at a 45° angle with respect to the target normal. With further experiments, it was found that by using a contoured target, the 4 times increase in line emission could be further augmented. Converting the spectrometer back to the time resolving mode allowed the pump plasma's evolution to be studied. A plot of the Mo XII emission at 336.63Å is shown in Fig. 8 for the case of the ruby laser formed and CO₂ laser heated plasma as well as the plot for the case when the plasma was formed only by the CO₂ laser. There is a 10 times enhancement in the maximum line emission observed for the contoured target using both lasers; one also notices that the peak occurs at an earlier time.

VII. Resonant Photopumping Experiments

With the improvements in the Mo XII source, a new series of resonant photopumping experiments were conducted with the aim of increasing the observed population enhancement discussed in Section V. The first experimental runs were conducted in the

650Å region to look for an increase in the enhancement of emission from the Mo VII 6s levels. Fig. 9 shows the intensities for 3 of the Mo VII 6s - 5p transitions. The "C" designation is for the intensity measured for the "cold plasma" alone, and the "R" designation is for the measured intensity for the lines when the Mo XII pump plasma created by the Ruby and CO₂ (RC) laser combination was present. These lines all show a 4-6 times enhancement when the pump source is present compared to the cold plasma alone, and this enhancement occurs at the time of the peak Mo XII emission from the delayed beam. The most strongly enhanced line from a 6s upper level is the 6s1/2[1/2]₁ - 5p 1/2[3/2]₂ transition at 645.42Å. This line is expected to exhibit the largest increase since the 1/2[1/2]₁ level is the one that is directly populated by the pump.

Considering next the behavior of the other Mo VII levels, Figs. 10 (a) and (b) are the intensity plots versus time for the 317.5 and 338.9Å (635 and 677Å in second order) Mo VII lines that could be collisionally enhanced by the population of the 6s manifold, and in Fig. 10 (c) is plotted the intensity for the 5p-4d transition that is observable in this region at 674.41Å. The two potentially collisionally populated lower levels show only a small amount of enhancement, discounting the one particular point for the 635Å line. These show that the density and temperature of the new Mo VII "cold plasma" are lower, thus decreasing the collisional enhancement of the population of the lower levels compared that observed in the previous experiments. This represented improved conditions for the future experiments aimed at measuring gain. In addition, in Fig. 11 (a) is shown the line intensity versus time for the 4p⁵4d-4p⁶ transition of Mo VII at 327.27Å (654.54 Å in second order), and in Figs. 11 (b) and (c) are shown the line intensities for two Mo VI transitions. There is no significant difference in the measured intensities for any of these lines for the "cold plasma" alone and a "cold plasma" pumped by the Mo XII ("hot plasma"). Figure 12 shows two representative spectra for (a) the "cold plasma" alone, and for (b) the "cold plasma" pumped by the Mo XII line recorded at the peak time of the Mo XII line emission at 150 ns. The enhancement of the 6s-5p lines (colored in black) is clearly seen. Lines with upper levels in the 4f or 4d² manifold are seen to show small

enhancements from collisional mixing with the pumped 6s levels, and the remainder of the lines are approximately equal for the two spectra.

VIII. Relative Population Inversion Calculations

The focus of this work has been on using resonant photopumping to create a population inversion for Mo VII 6s-5p transitions when pumped by a adjacent Mo XII plasma. The previous sections detail the best enhancement achieved with the present experimental arrangement, but the question remains: was this population enhancement sufficient to produce an inversion and hence gain?

At present, this evaluation can not be done in absolute terms since the absolute ion populations are unknown. What can be calculated, however, is a relative population inversion. Recasting the standard expression for a population inversion one obtains

$$\frac{N_2}{N_1} \frac{1}{g_2} - \frac{1}{g_1} > 0 \quad (3)$$

where N_i is the ion density of the level i , and g_i is the statistical weight of level i . The ratio N_2/N_1 can be determined from line intensity ratio's in the experiment. Assuming an optically thin plasma the line intensity I_{lu} is as given below

$$I_{lu} = N_u A_{ul} h\nu_{lu} F(x) \quad (4)$$

where the subscripts "u" and "l" refer to the upper and lower levels, the subscript "lu" refers to the transition itself. A_{ul} is the spontaneous transition probability, and $F(x)$ is a geometrical factor that accounts for the line integral and collection optics. One may write

$$\frac{N_2}{N_1} = \frac{N_{6s}}{N_{5p}} = \frac{I_{6s-5p} \lambda_{6s-5p} A_{5p-4d}}{I_{5p-4d} \lambda_{5p-4d} A_{6s-5p}} \quad (5)$$

N_{6s} is the population of the upper 6s level and, N_{5p} the population of the lower 5p level of a lasing transition; the function $F(x)$ was assumed to be the same for two close lines. For complete accuracy, the particular 5p-4d transition used in the calculation should have for its upper level the same level as lower level of the 6s-5p transition. In the spectral region of interest near 650Å, there is an insufficient number of 5p-4d transitions to allow measurement of the correct pairs of line intensities. An assumption can be made for these

calculations that the populations of the 5p sublevels as well as the 6s sublevels are close to that in LTE due to the small energy gaps and collisional population redistribution.. However, it was shown²⁹ that deviations of the 5p and 6p sublevel populations from that of Boltzmann are quite significant. In experimental check of this situation, the relative population densities for the 6s $1/2[1/2]_1$ level (giving rise to the 645.417Å line) and the 6s $3/2[3/2]_2$ level (giving rise to the 638.922Å line) levels for the non photopumped experiments were compared. Typical values for the intensities would give a ratio of 0.743 for N_{645}/N_{638} , whereas a calculation for the Boltzmann populated levels with an assumption of a temperature of 20 eV would give a ratio of 0.51. Therefore the relative population densities of the levels of interest were used directly from line intensity measurements with the reminder that this does introduce some error into the calculations since we are not using perfectly matched line pairs. The values for the transition probabilities were supplied by Joe Reader at NIST and were calculated with the Cowan code with relativistic Hartree-Fock exchange (HXR) wave functions⁴⁴. Among the lines observable in the 650Å region, the two 6s - 5p transitions at 638.922Å and 645.417Å, and the one 5p - 4d transition at 674.419Å can be reliably used. The results are presented in Table 2. In Table 2, the intensities are the peak line intensities at the line center. The population ratios tabulated were calculated using Eq. 5 where the measured intensity of the 674.419Å line was used to evaluate the lower level 5p population. The last two columns are the calculated relative population inversion using expression 3, where g_l is the statistical weight of the appropriate lower level of each transition. The three shots selected were taken at a time that corresponds to the peak pump emission. Results show that there are two shots where the system is just slightly absorbing, (the calculated inversion being slightly less than zero), and one shot for which the inversion threshold was crossed. Given the errors associated with the neglect of the spatial density profiles for the line integrated measurements, which tends to mask increasing of population inversion and those associated with not measuring the lower level populations directly, which could either decrease or increase the measured ratios, what can be safely concluded

from these calculations is that the system at present is at the verge of having an inverted population for the 6s levels relative to the 5p levels when the pump is present. A similar calculation for the unpumped case resulted in all the lines showing stronger absorption (negative inversion).

IX. Conclusions

The work presented in this paper details the first demonstration of a population inversion produced by the resonant photoexcitation of Mo VII by a Mo XII pump source. It was found that suitable pump and the lasant plasmas could be formed at a distance of 2 mm above the target surface for properly selected power densities on target. Time resolved spectroscopic studies of the pump and lasant plasmas identified a separation between the time of the peak Mo XII line emission, and the peak of the Mo VII ground state density. This temporal mismatch was compensated for with an optical delay network, the result being the demonstration of population enhancement of the Mo VII 6s level due to photoexcitation by Mo XII. Experiments with a Zirconium target, which produced no enhancement in the Mo VII observed intensities, support the conclusion that the observed effects are due to resonant effects. Work aimed at increasing the intensity of the pump source utilizing a ruby laser demonstrated that a pre-formed ruby produced plasma and subsequently heated by the CO₂ laser results in a 10 times increase in the observed Mo XII emission. The use of this stronger pump source has resulted in a further increase in the measured enhancement of the Mo VII levels population. With this increased enhancement, calculations of the relative population inversions indicate that this amount of enhancement has been sufficient to produce a population inversion for this system as a result of resonant photopumping.

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Table Captions

Table 1: Recombination and cooling rates for the pump and lasant plasmas.

Table 2: Intensities and relative population inversion calculations for 6s-5p transitions

Figure Captions

Fig. 1: Representative Energy Level Diagram for Mo XII and Mo VII.

Fig. 2: Experimental Block Diagram. showing a blowup of the target configuration.

Fig. 3: Measured intensity for Mo XII and Mo VII lines for the pump plasma.

Fig. 4: Line intensities for several Mo ions in the (a) 400 Å and (b) 650 Å regions for the cold plasma.

Fig. 5: Line intensity for Mo ions for a cold Mo plasma alone (C) and when pumped by a Mo XII plasma (P) for (a) Mo VII 638.92 Å, (b) Mo VII 677.78 Å, (c) Mo VI 637.17 Å, and (d) Mo VIII 647.88 Å.

Fig. 6: Line intensity for Mo ions for a cold Mo plasma alone (C) and when pumped by a Zr X plasma (Z) for (a) Mo VII 638.92 Å, (b) Mo VII 677.78 Å, (c) Mo VI 637.17 Å, and (d) Mo VIII 647.88 Å.

Fig. 7: Spectra from (a) a "cold Mo plasma" alone, and (b) a "cold Mo plasma" pumped by an adjacent Mo XII plasma. The plasmas were separated by 3mm.

Fig. 8: Comparison plot of the temporal Mo XII line emission for a ruby and CO₂ laser combination to that for the CO₂ laser alone.

Fig. 9: Intensities for a "cold plasma" alone "C", and a Mo XII pumped plasma "R" for Mo VII 6s-5p transitions at (a) 638.92 (b) 645.42, and (c) 693.17 Å.

Fig. 10: Intensities for a "cold plasma" alone "C", and a Mo XII pumped plasma "R" for Mo VII transitions with an (a) 4d², (b) 4f, and (c) 5p upper level.

Fig. 11: Intensities for a "cold plasma" alone "C", and a Mo XII pumped plasma "R" for (a) a Mo VII 4d- 4p transition and (b) and (c) Mo VI transitions.

Fig. 12: Spectra at the Mo XII peak emission time near 650 Å. (a) for the "cold plasma" alone, and (b) for the "cold plasma" by the Mo XII RC combination plasma.

	Pump Plasma	Lasant Plasma
T_e (eV)	200	25
C_s (cm/s)	5×10^6	2×10^6
N_e (cm ⁻³)	$10^{17} - 10^{18}$	$10^{17} - 10^{18}$
$\dot{T}/T$ (s ⁻¹)	2.5×10^7	10^7
$\bullet$ 3 body N/N (s ⁻¹)	1 - 100	$10^4 - 10^6$
$\bullet$ radiative N/N (s ⁻¹)	$5 \times 10^5 - 5 \times 10^6$	$10^5 - 10^6$

Table 1

Shot Number	Intensity (Arbitrary Units)			N_u/N_{674}		$\frac{N_u/N_{674}}{g_u} - \frac{1}{g_l}$	
	638	645	674	638	645	638	645
14356	52	106	15.5	0.26	0.48	-0.09	-0.04
14360	54	119	16.5	0.29	0.58	-0.08	-0.008
14374	155	119	8.5	1.53	1.05	+0.16	+0.15

Table 2

Figure 1

Figure 2

Blowup of target assembly

Figure 3

Figures 4(a) & (b)

Fig. 5.

Fig. 6.

Fig. 7

Figure 8

Fig 9

Fig. 10

Fig 11

Figure 12

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