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FLUORESCENCE AND SMALL-SIGNAL GAIN AT 2177 AND 2163 Å IN CIII BY RESONANT PHOTOEXCITATION WITH 310 Å, MnVI LINE RADIATION

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Abstract - Line radiation at 310 Å from a laser-produced Mn plasma was used to resonantly photoexcite the 2s 1S - 4p 1p0 transition in CIII ions in a vacuum-arc discharge. Enhanced fluorescence, up to a factor of 180, was observed on the 4p-3d line at 2177 Å. Enhanced fluorescence was also observed on other 4-3 lines in CIII, because the photoexcited 4p population is collisionally redistributed among all the n = 4 levels. A 72-level, collisional-radiative model of CIII was constructed, including the resonant photoexcitation. Measured values of electron density and temperature were used in the model. Theoretical predictions of enhanced fluorescence agree well with the measurements. Small-signal gain coefficients of ~0.1 cm⁻¹ were theoretically predicted. Single-pass gain measurements show gains as high as 0.4 cm⁻¹ on the 4p-3d, 2177 Å line and the 4f-3d, 2163 Å line. This CIII-MnVI photoexcitation scheme is a prototype for soft x-ray lasers, using higher Z, isoelectronic analogs.

I. INTRODUCTION

There are many approaches to soft x-ray lasers. Population inversion mechanisms include direct collisional excitation, three-body or dielectronic recombination, broadband inner-shell photoexcitation, and resonant photoexcitation. Recently, high gain at 206 and 209 Å was demonstrated [1] in a Ne-like, Se laser-produced plasma. Inversion between the 2p⁵ 3p and 2p⁵ 3s levels in Ne-like Se appears to be driven by a combination of direct collisional excitation from the 2p⁶ ground state as well as dielectronic recombination from the F-like 2p⁵ ground state. The search for a three-body recombination pumped laser has produced noteworthy results [2] in CVI at 182 Å, in a laser-produced magnetically confined plasma. A third approach to soft x-ray lasers is by resonant photoexcitation of ions in one plasma by intense line radiation from ions in an adjacent plasma. This approach was proposed in 1975 by Vinogradov, Sobelman, and Yukov [3], and by Norton and Peacock [4]. Several other resonant photoexcitation schemes have been proposed [5] and analyzed [6], and some experiments [7] have been reported, in Ne-like ions. Krishnan and Trebes [8] proposed a class of photoexcited lasers in Be-like ions. Quasi-cw lasers with wavelengths from 2177 Å in CIII to 213 Å in MgIX were identified, in an isoelectronic sequence.

Figure 1 illustrates the principle of resonant photoexcitation. MnVI line radiation at 310.182 Å pumps the CIII ions from the 2s² ground state to the 2s4p 1p⁰ level. For a sufficiently strong Mn pump, inversion occurs between 4p and 3d, and a laser is possible at 2177 Å. Collisions in the CIII plasma may rapidly thermalize the n = 4 level populations. In this case, gain is also possible on other n = 4-3 transitions as shown in the figure. This CIII-MnVI laser scheme is a prototype for soft x-ray lasers in higher Z, isoelectronic analogs. Qi, Kilic, and Krishnan reported [9] enhanced fluorescence on several 4-3 transitions in CIII ions in a vacuum-arc discharge, pumped by MnVI line radiation from a Mn, laser-produced plasma.

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This paper describes further experiments on the prototype CIII-MnVI scheme. Measured fluorescence due to photoexcitation is compared with the predictions of a 72-level, collisional-radiative model. This model for CIII was constructed using an atomic data base provided by Dr. W.L. Morgan of Lawrence Livermore Laboratory. The model predicted gains of ~0.1 cm\(^{-1}\) at 2177 Å, 2163 Å, and 1894 Å in CIII. Using an optimized discharge geometry, small-signal gains (~0.4 cm\(^{-1}\)) were measured, using a single-pass reflection technique.

II. FLUORESCENCE MEASUREMENTS

Figure 2 is a schematic drawing of the experimental arrangement. The CIII plasma is produced in a vacuum-arc discharge between a hollow carbon anode and a 6 mm diameter solid C cathode as shown. The interelectrode separation is 100 mm. The cathode is located 5 mm off the optical axis of a 0.5 m VUV spectrometer, which was used to measure fluorescence and gain on the n = 4-3 lines of CIII. The C cathode is biased negatively by up to ~6 kV by a pulse-forming network. A vacuum-arc discharge is triggered by focusing a 15 J/150 MW CO\(_2\) laser onto the cathode. A typical discharge current waveform is shown in Fig. 3(a). The rise-time is 15 µs, the flat-topped width about 50 µs, and the peak current is 6400 A. The discharge plasma so produced consists mostly of CIII ions, with an electron density \(n_e \sim 10^{16} \text{ cm}^{-3}\) and temperature \(T_e \sim 4 \text{ eV}\). The Mn pump is produced by focusing another 15 J/150 MW CO\(_2\) laser to a 2 mm x 23 mm line focus on a Mn slab as shown. The line focus is 7 mm off the spectrometer axis. Typically, the Mn laser-produced plasma is produced 36 µs after discharge initiation.

Typical enhanced fluorescence measurements are shown in Fig. 3. The horizontal time scale in the figures is 10 µs/div. Figure 3(b) shows the spontaneous emission on the 4p \(1p^0\) - 3d \(1D\) line at 2177 Å. The trace shown is an average over five successive discharges. Figure 3(c) shows the enhanced fluorescence at 2177 Å, when the Mn laser-produced plasma was created, 36 µs after discharge initiation. To capture this enhanced fluorescence, the sensitivity of the detection electronics was decreased by a factor of 45. When the Mn plasma was created without a C discharge, Fig. 3(d) shows that there is indeed some spurious radiation at 2177 Å (line or continuum) from the Mn plasma which is detected. When this spurious background is subtracted from the fluorescence in Fig. 3(c), the net enhanced fluorescence is 180...
times the spontaneous emission, i.e., optical pumping increases the 4p 1p0 population by a factor of 180. Such enhancements were found to be very reproducible over hundreds of shots. The typical duration of the enhanced fluorescence (FWHM) was about 0.5-1 μs.

Similar enhancements were then made at several other n = 4-3 wavelengths in CIII. The net enhanced fluorescence on the 4s 1S - 3p 1p0 line at 1894.5 Å, and the and a self-similar hydrodynamic expansion model, the initial Mn plasma temperature was estimated to be 35 eV. The brightness temperature of the MnVI pump line will be somewhat lower, and was assumed to be 20 eV. The effective solid angle for photoex-
Fig. 4. Theoretical calculations. (a) Populations of the 4s, 4p, and 4f singlet levels in CIII vs $n_e$ for $T_e = 4$ eV (dashed curves); enhanced populations of 4s, 4p, and 4f when the 4p $1p^0$ level is photoexcited (full curves). The MnVI pump line was assumed to have a brightness temperature of 20 eV.

(b) Predicted small-signal gain coefficients vs $n_e$ for the 2177, 2163, and 1894 Å lines in CIII ($T_e = 4$ eV).

citation was assumed to be $\pi$ steradians. With this brightness temperature and assuming Doppler line profiles for MnVI and CIII, stimulated absorption and emission were included in Eq. 1 for the 4p level, to give the enhanced $n = 4$ populations in CIII. As shown by the full curves in Fig. 4(a), significant enhancement occurs for all $n_e < 10^{16}$ cm$^{-3}$. It should be recalled that the measured value of $n_e$ in the CIII discharge at a current of 6400 A was about $7 \times 10^{15}$ cm$^{-3}$ over the first 25 mm from the cathode. For this value of $n_e$, Fig. 4 shows a population enhancement of 4p by about a factor of 140 while the 4f and 4s levels are enhanced by about a factor of 70 and 90, respectively, over their spontaneous values. These predicted enhancements agree remarkably well with the measurements described earlier, over the same distance downstream from the cathode.

For $n_e$ between $10^{14}$ and $10^{16}$ cm$^{-3}$, the enhanced $n = 4$ populations are high enough to invert them with respect to the $n = 3$ levels. Gain is thus possible on many 4-3 transitions. Typical small-signal gains are plotted in Fig. 4(b), vs $n_e$, for a fixed $T_e$ of 4 eV. The gains shown are for the 4s-3p, 1894.5 Å, 4p-3d, 2177 Å, and 4f-3d, 2163 Å lines. At the densities actually observed in discharges of about 6400 A, gains of about 0.1 cm$^{-1}$ are predicted for 2163 and 2177 Å. If the 23 mm long Mn plasma provided uniform photoexcitation over a 20 mm length of the CIII discharge, overall, single-pass gains of 0.2, or 22%, were expected. Single-pass gains of 100% per pass, with a gain coefficient of 0.4 cm$^{-1}$, were measured at both the 2177 and 2163 Å wavelengths, as described in the next section.

IV. SINGLE-PASS GAIN MEASUREMENTS

For gain measurements, the mirror behind the hollow anode (see Fig. 2) was adjusted to reflect rays from the pumped region of the CIII plasma back onto themselves. Irises were placed on either side of the discharge, to ensure that the solid angles subtended by the reflecting mirror and the spectrometer gathering optics were identical. On successive shots, the spontaneous emission and enhanced fluorescence were measured with and without the reflecting mirror. The ratio $R$ of the measured line intensities with and without the mirror is given by:

$$R = 1 + r e^{-\tau}$$

where $r$ is the net reflectivity of the vacuum window and reflection mirror, and $\tau$ is the integrated absorption coefficient along the entire optical path length. For
4f $^1P^0$ - 3d $^1P$ line at 2163 Å was found to be about a factor of 70 above the spontaneous emission. Enhanced fluorescence was also measured on the 4d $^3D$ - 3p $^3P^0$ line at 1620 Å and the 4f $^3P^0$ - 3d $^1P$ line at 1923 Å. These measurements confirm that the photoexcitation of the 4f $^1P^0$ state is accompanied by rapid collisional exchange of this population with other n = 4 levels.

The CII, 4p $^2P^0$ - 3s $^2S$ line at 2174 Å and the CIII, 2p$^2$ $^1P^0$ - 2s2p $^1S$ line at 2297 Å were also studied. No fluorescence was observed, confirming the selectivity of the resonant photoexcitation. As an additional check, a laser-produced Al plasma was substituted for the Mn plasma. No fluorescence was observed at any of the CIII, 4-3 wavelengths, thus ruling out collisional excitation by electrons from the laser-produced plasma, or broadband photoexcitation. These fluorescence measurements confirmed the general predictions [10] of the kinetics of resonant photoexcitation in CIII.

To further quantify the kinetics and to estimate small-signal gain under optimum CIII discharge conditions, a detailed collisional-radiative model was developed. This model is described in the next section.

### III. COLLISIONAL-RADIATIVE MODEL FOR CIII

A collisional-radiative model for CIII was developed in two steps. First, the relative abundances of different ground state populations were calculated, ignoring all excited states. Then, using the calculated ground states of CIII and CIV, a 72-level model was constructed for CIII. Such an approach is valid when the excited state populations are very small relative to the ground states. Electron density $n_e$ and $T_e$ are essential inputs to such a model. $n_e$ and $T_e$ were measured spectroscopically at five axial locations up to 25 mm from the cathode. Density was in the range $6 \times 10^{15} \leq n_e \leq 1.2 \times 10^{16}$ cm$^{-3}$. $T_e$ was uniform at 4 eV. These measurements will be described elsewhere.

Processes considered in the ionization balance included collisional ionization [11], three-body recombination [12], radiative recombination [13], and dielectronic recombination [14,15]. For $n_e = 10^{16}$ cm$^{-3}$ and $T_e = 4$ eV, 70% of the ions are in the CIII ground state, and 30% in the CII ground state.

Starting with the ground state populations of CIII and CIV, a 72-level, collisional-radiative model was developed for CIII. In this model, all singlet and triplet states of the outer electron configurations, with principal quantum numbers $n = 2$ to $n = 6$ were included. For $n = 7$ to 10, single lumped states distinguished only by principal quantum number were included. For all levels considered from $n = 2$ to 6, energy levels, transition oscillator strengths and collisional excitation/de-excitation rates were provided by Dr. W.L. Morgan of Lawrence Livermore Laboratory. For $n = 7$ to 10 levels, hydrogenic rates [16] were used. The generalized rate equation for the jth level is as follows.

$$
\frac{dn(j)}{dt} = -n(j)[S(j)n_e + \sum_{j>i} A(j,i)n_i + \sum_{j>i} C^d(j,i)n_e + \sum_{j>i} C^e(j,i)n_e]
+ n(i)[\sum_{j<i} A(i,j) + \sum_{j<i} C^d(i,j)n_e + \sum_{j<i} C^e(i,j)n_e]
+ n_g^{CIV}[\alpha(j) + \beta(j)n_e]
$$

$n_g^{CIV}$ is the CIV ground state density, $S(j)$ is the collisional ionization rate [17-19], $C^d(j,i)$ and $C^e(j,i)$ are the collisional de-excitation and excitation rates, $\beta(j)$ is the radiative recombination rate [20], and $\alpha(j)$ is the three-body recombination rate, which was determined from the ionization rate by detailed balancing. For $n_e > 10^{15}$ cm$^{-3}$, some of the lower lying (resonance) lines can be optically trapped. Holstein escape factors [21,22] were used to account for the effects of opacity on all levels. Since the relaxation times for excited levels are much shorter than the time scales for evolution of the CIII and CIV ground states, the excited state populations attain quasi-steady-state values.

Figure 4(a) shows typical quasi-steady-state level populations vs $n_e$. The dashed lines on the figure show the absolute population density of the 4s, 4p, and 4f singlet states as a function of $n_e$, for a fixed $T_e = 4$ eV. The full curves on the figures are the enhanced populations of these levels, when the 4p $^1P^0$ level is resonantly photoexcited by MnVI line radiation. The absolute intensity of the MnVI pump line was not measured. Based on measurements of absolute continuum intensity...
Fig. 5. Single-pass gain at 2177 and 2163 Å in CIII. (a) Enhanced fluorescence at 2177 Å, without reflecting mirror; (b) fluorescence at 2177 Å, with reflecting mirror; (c) enhanced fluorescence at 2163 Å, without reflecting mirror; (d) fluorescence at 2163 Å, with reflecting mirror.

The measured gain coefficients are significantly higher than those predicted by the collisional-radiative model (see Fig. 4). The discrepancy could be due to overestimation of the effects of opacity in the model. Streaming motion of the ions, smaller lateral plasma dimension than that assumed, and the effect of gradients will all reduce the opacity effects and correspondingly increase the predicted gains.

V. SUMMARY

A detailed theoretical and experimental study was presented of resonant photoexcitation in CIII ions by MnVI line radiation. Using an optimized discharge and pump geometry, enhanced fluorescence and small-signal gain on the 4p-3d, 2177 Å and the 4f-3d, 2163 Å transitions in CIII were measured. Gain coefficients of 0.4 cm⁻¹ were measured for both wavelengths. To compare with the experiments, a detailed, 72-level, collisional-radiative model was developed for CIII. An ionization balance first gave the CIII and CIV ground state populations, using measured values of \( n_e \) and \( T_e \). These ground state populations in turn yielded the populations of various excited states in CIII. Resonant photoexcitation was modeled by an assumed brightness temperature of 20 eV for the pump line. Good agreement was obtained between
experiment and theory. The high gains measured and their ~1 μs duration suggest that a Fabry-Perot cavity may be used to demonstrate laser oscillation at the 2177 and 2163 Å wavelengths. Such experiments are under way and will be reported elsewhere, along with further details on the experimental measurements and the atomic physics code.

VI. ACKNOWLEDGMENTS

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VII. REFERENCES