Was the Livermore X-Ray Laser Pumped by Recombination?

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We demonstrate that recombination pumping following rapid radiative cooling of an overstripped selenium plasma can quantitatively account for the observed gains of the $J = 2$ to 1 Sexxv transitions in the recent successful Livermore x-ray-laser experiments. The absence of gain in the $J = 0$ to 1 transition, hitherto unexplained, is fully consistent with this scenario.

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Recently, Rosen, Matthews, and co-workers at Lawrence Livermore National Laboratory achieved a noteworthy success in the demonstration^{1,2} of a soft x -ray laser utilizing $3p-3s$ transitions in neonlike selenium (Se XXV). This success was, however, accompanied by one unexplained result: The $J = 0$ to 1 lasing transition at 183 \AA , predicted to have the highest gain, was absent, while the $J = 2$ to 1 transitions at 206 and 209 \AA exhibited gain coefficients of 5 ± 1 cm⁻¹. These gain predictions were based on the assumption that collisional excitation of the transitions from the Se XXV ground state in a plasma of electron density 5×10^{20} cm⁻³ and temperature 1 keV would be the dominant pumping mechanism.

In this Letter we demonstrate that an alternative picture accounts quantitatively for the absence of observed gain in the $J=0$ to 1 transition as well as for the gains achieved in the $J = 2$ to 1 transitions. The picture is as follows: The selenium plasma reached a temperature near 1 keV and was stripped through the neonlike stage into higher charge states so rapidly that the finite photon transit time prevented amplification.

Subsequent rapid radiative cooling produced copious recombination back to the neonlike stage during which the gain was observed. The radiative recombination preferentially pumped the $2p^{5}3p$ J = 2 levels (mainly due to their high relative statistical weights), resulting in the observed gains.

We now present and discuss our calculations which support this picture.

An atomic model of highly ionized selenium has been constructed. It includes the thirteen most relevant excited (j, j) levels of Sexxv in addition to the ground states of the other ionization stages. The excited levels above 3d through $n = 6$ are consolidated by principal quantum number. The various states are coupled by electron collisional excitation, deexcitation, and ionization, spontaneous radiative decay, and radiative, dielectronic, and three-body recombination. The techniques used for our rate calculations are described elsewhere.³⁻⁹

The most important atomic rates and energy levels are detailed in Fig. 1 of Ref. 1. The only noteworthy difference with our calculations is in the dielectronic-

FIG. 1. Gain coefficients in the Sexxv $J = 2$ to 1 transitions at 206 and 209 Å plotted as a function of time for a recombining plasma of temperature 0.2 keV and electron density 5×10^{20} cm⁻³. The gain in the $J = 0$ to 1 transition at 183 A never exceeds 0.25 cm⁻¹ and is not plotted. The plasma is assumed to be 65% fluorinelike and 35% oxygenlike at $t = 0$. Also shown are the ground-state ionic fractions of oxygenlike, fluorinelike, and neonlike selenium.

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recombination rate coefficient for Se xxvI to form neonlike Sexxv at ¹ keV. We obtain this rate coefficient by interpolating detailed calculations for nickel $(Z = 28)^6$ and krypton $(Z = 36)^{10}$ Our rate coefficient is 3.4×10^{-12} cm³ s⁻¹, an order of magnitud cient is 3.4×10^{-12} cm³ s⁻¹, an order
smaller than the $(1-5) \times 10^{-11}$ cm³ s used in the model presented in Ref. 1. Since krypton is so close to selenium in atomic number, and the nickel rate coefficient differs from the krypton rate coefficient by less than a factor of 2, interpolation errors are very small for the above rate coefficient for selenium. Similar previous calculations for iron,⁵ including the relevant fluorinelike stage, have received substantial experifluorinelike stage, have received substantial experi-
mental corroboration.^{11–13} Moreover, according to Ref. 1, dielectronic recombination was "modeled in a crude way" for their calculations. In the present model the medium is assumed to be optically thin. For the plasma geometry used in Ref. 1, optical depth has only a moderate effect on the gain¹ and may actually increase the gain¹⁴ through $2p-3d$ line trapping followed by cascade to the $3p$ levels. All level populations are calculated with use of a time-dependent collisional radiative model and the numerical algorithm of Young and Boris.¹⁵

In Refs. 1 and 2 temperatures of 1 keV and electron densities of $5 \times 10^{20} - 1 \times 10^{21}$ cm⁻³ are cited as the conditions under which collisionally pumped gain was achieved in the two $3p-3s$ transitions. Let us assume that the temperature of 1 keV is initially achieved at a slightly higher electron density of 3×10^{21} cm Under such conditions our time-dependent model calculations show that Se xxv is stripped to higher stages in an e-folding time of 45 ps. This is smaller than the 100 ps reported in Ref. ¹ as the minimum duration required for the neonlike plasma to produce amplification during the 40-ps photon transit time through the 1.2-cm path. Our order-of-magnitude-smaller dielectronic-recombination rate is a key factor producing this brief ionization time. Using the rate of Ref. 1, we find a substantial fractional equilibrium population of Se XXV, in agreement with Ref. 1. It is therefore likely that the neonlike state was stripped through before any significant collisionally pumped lasing could build up. The actual ionization time is probably smaller than 45 ps since we have not included the levels above $n = 6$ or photon pumping of the excited states. The importance of line photon pumping and ionization from excited states in enhancing plasma ionization is stressed in detail elsewhere.¹⁶

The second argument supporting the recombination picture is the importance of radiative cooling of such plasmas, occurring primarily through the collisional excitation of line radiation. A (conservative) linear extrapolation of previous detailed calculations for calcium $(Z = 20)$ and nickel $(Z = 28)^6$ results in a minimum cooling rate, from 1.0—0.² keV, of

 2.4×10^{-26} W cm³. At an electron density of 10^{21} cm^{-3} this is equivalent to 150 keV/ion ns. If there are 25 stripped electrons at 1 keV associated with each ion, cooling these electrons to 0.2 keV requires $(\frac{3}{2})$ (25) (0.8) keV = 30 keV/ion, which would require 200 ps. The rate quoted above is the minimum for 1.0—0.² keV, and the true cooling time is closer to 130 ps. The above cooling rate is just 3% of the blackbody limit for emission from two sides of a rectilinear plasma with dimensions $0.01 \times 0.01 \times 1.2$ cm³ and of temperature 0.2 keV. At a temperature of ¹ keV this cooling rate is only 5×10^{-5} of the Planck limit. We note that our cooling time is in very good agreement with he 100-ps estimate of Seely et al ¹⁷ for selenium, for the same electron density and temperature considered here. This estimate is consistent with the recombination observed in a carbon-selenium plasma as described in Ref. 17. The following sequence of events is therefore quantitatively supportable: The selenium plasma was stripped so rapidly through the neonlike stage during the middle of the driving laser pulse that no lasing could occur. Toward the end of the laser pulse efficient radiation cooling lowers the temperature from \sim 1 to \sim 0.2 keV in just over 100 ps, setting the stage for recombination back to neonlike Sexxv. We now discuss the remaining question: Do the calculated gains to be achieved during recombination match those observed according to Refs. ¹ and 2?

In accordance with the above calculations and discussion we assume that within 100 ps of the end of the driving laser pulse, the selenium plasma has cooled to \sim 0.2 keV, and the electron density, as reported in Ref. 1, is reduced by expansion to 5×10^{20} cm⁻³. The following time-dependent calculation was performed in accordance with this picture. At $t = 0$ the temperature is 0.2 keV and electron density is 5×10^{20} cm⁻³. We assume that stripping past the neonlike stage has resulted in 65% fluorinelike and 35% oxygenlike ions. The plasma then proceeds to recombine. The temperature and density are held constant during the 250 ps duration of the calculation. Further reduction in the ion and electron temperatures which occur during this time would increase the gain through faster recombination and smaller line Doppler widths and would therefore tend to offset reductions in gain due to expansion of the plasma. The principal results of this calculation—the gains in the 206- and 209- \tilde{A} J = 2 to ¹ transitions —are plotted in Fig. ¹ along with the ionic stage fractions as a function of time. The gain in the missing $J = 0$ to 1 transition at 183 A never exceeds 0.25 cm^{-1} during the calculation and is therefore not plotted. Given the many uncertainties in laser-plasma-interaction physics, we fully concur with the criterion of Ref. ¹ that 50% agreement is as good as can be reasonably expected. Figure ¹ reflects just such agreement of our interpretation with the reported (5 ± 1) -cm⁻¹ gain¹ in these transitions, along with the absence of gain in the $J = 0$ to 1 transition. The duration of gain (\sim 200 ps) agrees with that reported in Ref. 2. Radiative and three-body-recombination cascade processes tend to populate the $3p J = 2$ states more than the $J = 0$ states because of their greater statistical weights.¹ It is precisely this effect which is responsible for the missing $J = 0$ gain. The x-ray laser was probably pumped exclusively by recombination accelerated by strong radiative cooling. We emphasize that the dominant pumping recombination processes are ordinary three-body and radiative recombination. Population of the upper lasing levels occurs by direct radiative recombination from the flourinelike stage as well as by cascade from the higher levels, which themselves are populated by radiative and three-body recombination. We also note that total stripping through the neonlike stage, though likely, is not required for recombination-pumped lasing to dominate. As long as the radiative cooling is sufficiently rapid to inhibit collisional excitation before amplification builds, recombination gain in the $J = 2$ to 1 transitions dominates. A similar calculation in which the plasma was initially assumed to be 10% oxygenlike, 65% flourinelike, and 25% neonlike yielded gain versus time curves virtually identical to those of Fig. 1. However, we find that the temperature must not exceed 0.25 keV in order to prevent collisional excitation of 'the $3p(\frac{1}{2},\frac{1}{2})_0$ level and subsequent (unobserved) gain in the $J=0$ to 1 transition.

In summary, we have developed and quantitatively justified a plausible sequence of events in the successful x-ray laser experiments reported in Refs. 1 and 2 which has the important virtue of explaining the observed gains in the $J = 2$ to 1 transitions and the absence of gain in the $J = 0$ to 1 transitions. The picture we invoke is that of a selenium plasma which is stripped so rapidly through the neonlike stage that collisionally pumped lasing could not be achieved. This was followed by efficient radiative cooling to 0.2 keV, accompanied by recombination back to the neonlike stage, during which the observed lasing occurred.

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