Enhancement of short-pulse recombination-pumped gain by soft-x-ray photoionization of the ground state

J. P. Apruzese

Radiation Hydrodynamics Branch, Plasma Physics Division,
Naval Research Laboratory, Washington, D.C. 20375

D. Umstadter

Center for Ultrafast Optical Sciences, University of Michigan, Ann Arbor, Michigan 48109

Received February 23, 1995; revised manuscript received June 9, 1995

The gain achieved in lasing to the ground state following short-pulse field ionization by a pump laser is highly transient. It will usually persist for only tens of picoseconds because of the rapid filling and negligible emptying of the ground state. Employing a detailed atomic model of lasing in hydrogen, we show that the removal of ground-state population by an appropriate broadband ionizing radiation field can enhance and prolong the gain in such a laser.

1. INTRODUCTION

The rapid development and improvement of high-intensity, short-pulse lasers over the past several years[1] has stimulated much experimental and theoretical work aimed at demonstrating various applications of such devices. One of the most intriguing and frequently mentioned possibilities is the achievement of short-wavelength lasers, some of which terminate on the ground state of an ionic or an atomic species.[2–9] The recent demonstrations of optical guiding of intense laser pulses in a plasma channel,[6] of gain on the Lyman-α transition of H-like Li,[7] and of lasing at 41.8 nm in Xe IX by a femtosecond laser pulse[9] have further sharpened interest in this area. Extensive analysis of the laser kinetics of systems terminating on the ground state has been directed primarily at H-like[2,8] and Li-like[4] sequences. The filling of the ground state (which must be initially emptied) limits both the duration and the amount of gain. However, recent experimental results from studies of the interactions of ultrafast and intense lasers with solid targets may change this picture. The bright x-ray emission reported in these experiments[10–15] can have a picosecond rise time and may thus be used to change the inversion kinetics of Lyman-α by means of photoionization. In this paper we concentrate on the inversion kinetics of the Lyman-α line of neutral H. We demonstrate that photoionization of the ground state by a broadband soft-x-ray radiation field can, under some conditions, significantly prolong and enhance the expected gain. This occurs because the ground state can be depopulated on a time scale comparable with that required for filling it. Section 2 presents a description of the model, assumptions, and calculations that lead to this conclusion. In Section 3, specific results for gain calculated both with and without soft-x-ray ionization are presented.

2. BASIC CONCEPT AND MODEL

Recombination lasers work because the rate of three-body recombination to a bound state of principal quantum number n, per unit statistical weight, is approximately proportional to n^4/T_e. Thus, if one can set up a sufficiently cool plasma of appropriate density in which the dominant population mechanism for a particular species is three-body recombination, population inversions and gain are achievable. Soft-x-ray lasers driven in this fashion, which operate between excited states of various ions, have been demonstrated for some time (see Refs. 16 and 17 and references therein). It is a much more difficult task, however, to create and sustain an inversion between an excited state and the ground state of an ion or atom.[2,4,8] It is still true that the excited states can be populated at a greater rate than the ground state when three-body recombination is used, but, in contrast to excited-to-excited-state lasing, the (ground) lower laser level does not radiatively decay. The repopulation of the ground level therefore tends to proceed essentially unimpeded, and the gain lasts only as long as it takes to fill it to the point at which the inversion is quenched. Because transient recombination-pumped gain is both calculated and observed to exist even without depopulation of the ground state, it is possible that the introduction of a removal mechanism for the ground state to the plasma kinetics may increase the gain. As pointed out in Ref. 4, both the gain and the saturation lasing intensity are sensitive to the lower-level population in these lasers. With an expected gain duration in H of tens of picoseconds, it follows that, to affect this gain significantly, the ground state must also be depopulated on a similar time scale. Photoionization will be feasible as such a mechanism if the ionization time is also tens of picoseconds, i.e., the rate should be a few times 10^{10} s^{-1}. Ionization pumping has been calculated to be useful for generating gain in the Balmer-α transition of H-like ions,[18] but considerably different requirements and effects are expected for the currently considered Lyman-α application.

Consider a blackbody radiation field $B_\nu(T_r)$ that is incident upon an initially fully stripped H plasma over half the sky, i.e., 2π sr. Let σ_0 be the threshold cross section for ionization from the ground state ($\sigma_0 = 6.3 \times 10^{-18}$ cm^2). The threshold frequency is denoted by ν_0. We employ the usual approximation in which the ionization cross section varies as the inverse cube of the photon frequency. For H, such an approximation is accurate[19] to 10% for photon energies of up to 200 eV and to 35%, for up to 1 keV. The ground-state ionization rate that is due to such a radiation field is given by

$$W_{\text{ion}} = 2\pi\sigma_0 \int_{\nu_0}^{\infty} \left(\frac{\nu_0}{\nu}\right)^3 \frac{2h\nu^3 \mathrm{d}\nu}{c^2 h\nu[\exp(h\nu/kT_r) - 1]} \,. \tag{1}$$

In the present study only pump temperatures that are considerably greater than the ionization potential are considered; thus $\exp(h\nu/kT_r)$ may be approximated as $1 + h\nu/kT_r$. This results in considerable simplification and an analytic solution to the integral of Eq. (1). If the pump radiation temperature T_r and ionization potential χ are expressed in electron volts, the ionization rate, in inverse seconds, is given by

$$W_{\text{ion}} = 1.98 \times 10^{23} \sigma_0 T_r \chi^2 \,. \tag{2}$$

For a radiation pump temperature T_r of 100 eV, the ionization rate from the H ground state is calculated from Eq. (2) as 2.3×10^{10} s^{-1}. This rate is comparable with the inverse of the gain duration and suggests that a significant effect on the gain is possible. At the same radiation temperature, the ionization rates out of levels $n = 2, 3, 4, 5$ are 3.2×10^9, 9.7×10^8, 4.3×10^8, 2.2×10^8, s^{-1} respectively. The corresponding threshold cross sections, assuming statistical population of the angular momentum sublevels, are $(1.4, 2.2, 3.0, 3.7) \times 10^{-17}$ cm^2, respectively, for $n = 2, 3, 4, 5$. These rates from the upper levels are well below those from the ground state. This is a favorable feature of pumping by a Planck-shaped radiation field. It is undesirable to remove the excited states because recombination and cascade to and among these levels set up the population inversion with respect to the ground state. In the calculations described below, pump radiation fields with temperatures of 50 and 200 eV are also considered. In each case the radiation is assumed to be incident over 2π sr. Because the ionization rate, according to Eq. (2), is approximately proportional to the radiation temperature, the ionization rates for 50 and 200 eV are, respectively, half and twice those given above for 100 eV. The ion density and the electron temperature are specified and assumed constant, and the level populations for principal quantum numbers 1–5 are calculated by numerical integration of the fully time-dependent rate equations. These equations incorporate all significant collisional and radiative processes connecting the levels. Given the evolving populations, the time-varying gain in the Lyman-α line is calculated with an analytic fit to the Stark-profile cross section, as given in Ref. 8, where further discussion of the model may be found. Following Refs. 2, 4, and 8, the plasma is assumed to be initially fully stripped.

3. RESULTS AND DISCUSSION

Because the photoionization rate is independent of plasma density, but collisional processes are faster at a higher density, the perturbation on the level kinetics and the gain caused by an ionizing radiation field are greatest at low density and, at some high density, becomes negligible. At too low a density, however, little or no gain is achievable because there are too few amplifying ions in the line of sight and, also, three-body recombination is increasingly ineffective in establishing a population inversion. At some intermediate density, therefore, the photoionization enhancement may be significant in a regime in which the gain is still substantial. For H, our calculations indicate that such a density is $\sim 4 \times 10^{17}$ (ions or electrons) cm^{-3}. At a density of 10^{18} cm^{-3}, for instance, the effect on magnitude and duration of gain is slight because of the greater relative importance of electron collisional processes at this higher density. At lower densities, a significant positive effect on gain is calculated down to $\sim 10^{17}$ cm^{-3}, at which point the gain dwindles to a few inverse centimeters. Because the density of 4×10^{17} cm^{-3} illustrates all the features of the photoionization enhancement mechanism, in this paper we present detailed results for only that density.

Another important factor for determining the gain is the electron temperature. The extent to which electrons can be kept cold while the plasma is fully stripped to one charge state higher than that of the lasing species is a subject of considerable current discussion.[3–5,8] Heating processes include above-threshold-ionization energy, inverse bremsstrahlung, and stimulated Raman scattering. In estimating a reasonable assumed temperature we consult the results of Ref. 8 for residual electron energy. Because the three-body recombination rate varies as the inverse of the electron temperature, lower temperatures result in higher gain as long as the plasma is initially fully stripped. The electron temperature following field ionization depends on both the wavelength of the driving laser and its irradiance (see, e.g., Fig. 3 of Ref. 8). Figure 2 of Ref. 8, which is based on the tunneling ionization model of Ammosov, *et al.,*[20] shows that an irradiance of $\sim 5 \times 10^{14}$ W cm^{-2} is required for fully stripping H atoms to bare protons. For a frequency-doubled Nd:glass laser operating at a wavelength of 0.53 μm, an electron temperature of ~ 0.6 eV is expected at such an irradiance (Fig. 3, Ref. 8). This temperature is sufficiently low for recombination-pumped gain and is assumed in the calculations of Figs. 1 and 2.

Figures 1 and 2 show the gain in the Lyman-α line of H as a function of time following the full stripping of the plasma at an ion density of 4×10^{17} cm^{-3} and an electron temperature of 0.6 eV. The gain calculations are presented both with and without an assumed photoionizing radiation field. For the pumped cases, radiation temperatures of 50, 100, and 200 eV were assumed. Figure 1 shows the result with the ionizing radiation assumed constant in time, whereas Fig. 2 presents results with an incident radiation of 1-ps rise time and an exponential falloff of 100 ps. One recent experiment[15] indicates that, with a subpicosecond laser-pulse duration, the rise time of the x-radiation may be of the order of 1 ps, whereas the fall time can be varied from 1 to 100 ps

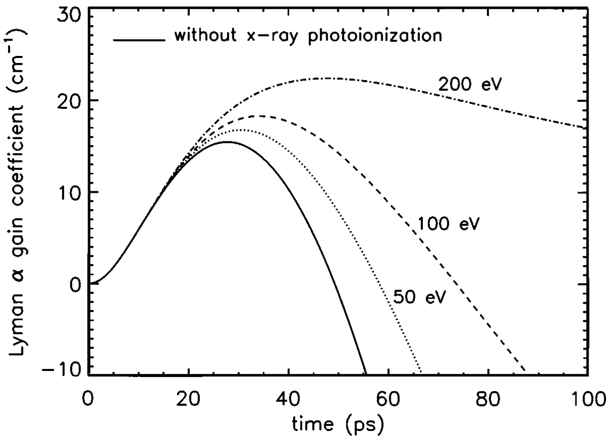

Fig. 1. Gain coefficient for the Lyman-α line of H is shown as a function of time for an initially fully stripped plasma of density 4×10^{17} ions cm^{-3}. The electron temperature is assumed to be 0.6 eV. The solid-curve result assumes that no photoionization is present; the other curves were calculated assuming photoionization over 2π sr by blackbody radiation fields of the indicated temperatures. The ionizing radiation is constant in time.

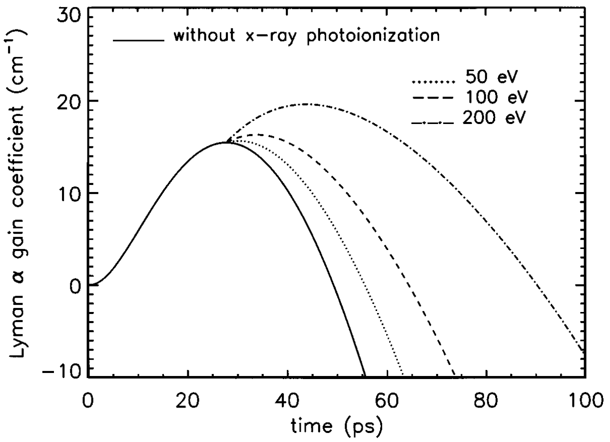

Fig. 2. Same as in Fig. 1, except that the ionizing radiation rises linearly in 1 ps and falls exponentially with an e-fold time of 100 ps. The peak intensity occurs at 28 ps.

by simply adjusting the incident laser flux. Large fractions, of the order of 10%, of the laser energy may be converted into a spectrum of x rays that approaches that of a blackbody at a temperature of several hundred electron volts.

In Fig. 2, the blackbody radiation is assumed to reach a maximum at $t = 28$ ps, the time of peak gain without incident radiation present. As expected from the discussion of Section 2, the magnitude and the duration of gain are noticeably enhanced in both cases, although the effect is more pronounced for the constant pump radiation. The higher gain is mostly due to reduction of the population of $n = 1$, as opposed to the enhancement of the population of other levels. As would be expected and is obvious from the figures, the more intense the ionizing source, the more favorable the effect. At $t = 40$ ps, the gain in the photoionized cases is sharply differentiated from the baseline case with no radiation assumed incident. In units of 10^{-2} of the total, the fractional populations of $n = 2$ at this time for the cases of no radiation, 50-, 100-, and 200-eV pumps are, respectively, 3.6, 3.6, 3.5, and 3.3. For the lower laser level ground state $n = 1$, the fractional populations are (in the same order and units) 0.72, 0.61, 0.53, and 0.40. These values pertain to the steady-state pumping case. Clearly the gain enhancement comes about from partial evacuation of the ground state rather than from the small effect on the excited levels. The bare proton fraction differs by less than 1%, remaining at ~92% of the total. As is consistent with the sensitivity of gain to population fraction, a small degree of differential ionization is capable of producing the significant changes in gain noted above.

One potential disadvantage of this approach to gain enhancement is that the ejected photoelectrons might heat the ambient cool electrons, thus reducing the recombination rates and spoiling the gain. However, this does not appear to be a significant problem in the cases considered above. The energy transfer rate[21] of the ejected hot electrons to the colder recombining ones varies as $\sim\epsilon^{-1.5}$, where ϵ is the energy of the hotter photoejected electrons. At a density of 4×10^{17} cm^{-3}, electrons of energy of a few hundred electron volts require several hundred picoseconds for transferring their energy. Consider cooler photoelectrons of energy ~50 eV. The total ionization rate per electron is nearly equal to the rate per ion, because the plasma is almost fully stripped. This rate is given by the sum of the ionization rates from each level multiplied by the population fraction of that level. It is typically $\sim 3 \times 10^{8}$ s^{-1}, and therefore in 10 ps only 0.003 of the ambient electrons will have been created by photoionization. Even if all their ~50-eV energy is transferred to the cold electrons during this time, the energy amounts to only (0.003)(50 eV) ~ 0.15 eV/cold electron. This result would not significantly affect the temperature or the gain.

An important issue is the possible scaling of this scheme to a higher atomic number Z and therefore to a lower laser wavelength. As long as the pump temperature well exceeds the ionization potential, Eq. (2) applies to the scaling of the ionization rate. Because the ionization potential χ varies as Z^2 but the threshold cross section[22] σ_0 varies as Z^{-2}, Eq. (2) indicates that the ionization rate scales favorably as Z^2. However, the radiative decay rates and collisional rates among bound states scale quite differently: the decay rate[23] varies as Z^4 and the collisional rates[24] vary as Z^{-3} (for a given electron density). Neither the radiative nor the collisional rates dominate the level kinetics that determine the achievable gain. The resulting populations are the product of both types of process. The gain is especially sensitive to these populations. In view of these considerations, there does not appear to be a simple or obvious analytic approach to the problem of atomic-number scaling of a radiatively pumped, transient recombination laser. Such a model may become feasible when further guided by numerical solutions for $Z \geq 2$.

ACKNOWLEDGMENTS

This research was supported by the U.S. Office of Naval Research and the National Science Foundation.

REFERENCES

1. G. Mourou and D. Umstadter, Phys. Fluids B **4,** 2315 (1992).

2. W. W. Jones and A. W. Ali, J. Appl. Phys. **48,** 3118 (1977).
3. N. H. Burnett and P. B. Corkum, J. Opt. Soc. Am. B **6,** 1195 (1989).
4. D. C. Eder, P. Amendt, and S. C. Wilks, Phys. Rev. A **45,** 6761 (1992).
5. S. C. Rae and K. Burnett, Phys. Rev. A **46,** 2077 (1992).
6. C. G. Durfee III and H. M. Milchberg, Phys. Rev. Lett. **71,** 2409 (1993).
7. Y. Nagata, K. Midorikawa, S. Kubodera, M. Obara, H. Tashiro, and K. Toyoda, Phys. Rev. Lett. **71,** 3774 (1993).
8. P. Pulsifer, J. P. Apruzese, J. Davis, and P. Kepple, Phys. Rev. A **49,** 3958 (1994).
9. B. E. Lemoff, G. Y. Yin, C. L. Gordon III, C. P. J. Barty, and S. E. Harris, Phys. Rev. Lett. **74,** 1574 (1995).
10. M. M. Murnane, H. C. Kepteyn, and R. W. Falcone, Phys. Rev. Lett. **62,** 155 (1989).
11. D. G. Stearns, O. L. Landen, E. M. Campbell, and J. Scofield, Phys. Rev. A **37,** 1684 (1988).
12. J. C. Kieffer, M. Chaker, H. Pepin, Y. Beaudoin, C. Y. Cote, J. P. Matte, G. Mourou, C.-Y. Chien, and S. Coe, Bull. Am. Phys. Soc. **37,** 1468 (1992).
13. D. Riley, L. A. Gizzi, F. Y. Khattak, A. J. Mackinnon, S. M. Viana, and O. Willi, Phys. Rev. Lett. **69,** 3739 (1992).
14. M. M. Murnane, H. C. Kapteyn, S. P. Gordon, J. Bokor, E. N. Glytsis, and R. W. Falcone, Appl. Phys. Lett. **62,** 1068 (1993).
15. U. Ellenberger, J. J. Workman, A. Maksimchuk, X. Liu, S. Coe, C.-Y. Chien, and D. Umstadter, Phys. Rev. Lett. **75,** 2324 (1995); J. Opt. Soc. Am. B **13,** 124 (1996).
16. S. Suckewer, C. H. Skinner, H. Milchberg, C. Keane, and D. Voorhees, Phys. Rev. Lett. **55,** 1753 (1985).
17. A. Carillon, M. J. Edwards, M. Grande, M. J. de C. Henshaw, P. Jaegle, G. Jamelot, M. H. Key, G. P. Kiehn, A. Klisnick, C. L. S. Lewis, D. O'Neill, G. J. Pert, S. A. Ramsden, C. M. E. Regan, S. J. Rose, R. Smith, and O. Willi, J. Phys. B **23,** 147 (1990).
18. D. G. Goodwin and E. E. Fill, Appl. Phys. B **50,** 177 (1990).
19. W. J. Karzas and R. Latter, Astrophys. J. Suppl. **6,** 167 (1961).
20. M. V. Ammosov, N. B. Delone, and V. P. Krainov, Sov. Phys. JETP **64,** 1191 (1986).
21. B. A. Trubnikov, in *Reviews of Plasma Physics* (Consultants Bureau, New York, 1965), Vol. 1, p. 105.
22. C. W. Allen, *Astrophysical Quantities,* 3rd ed. (Athlone, London, 1973), p. 96.
23. W. L. Wiese, M. W. Smith, and B. M. Glennon, *Atomic Transition Probabilities* (National Bureau of Standards, Washington, 1966), Vol. 1, p. ix.
24. D. H. Sampson and H. L. Zhang, Astrophys. J. **335,** 516 (1988).