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Absolute line intensities for photon pumping of x-ray lasers

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Absolute intensities of certain resonance lines which are considered to be promising candidates for matched-line (photoexcitation) pumping of x-ray lasers are measured. Most are in the 3–8-nm wavelength range. A promising Na^{9+} candidate at 1.1 nm is also tested. The measured radiation is emitted from high-density plasmas formed near the surface of laser-heated slab targets of selected materials. It is concluded that gain-length products of 3 can be achieved with a driving-laser irradiance of 10^{12} W/cm^2 and with characteristic dimensions in the 10–100-μm range. Also, among the one-, two-, and three-electron ions tested, an apparent decreasing $2p \rightarrow 1s$, $3d \rightarrow 2p$, $3p \rightarrow 2s$ order of preference for resonance pumping transitions may exist, subject to limitations of the present measurement accuracy. One $3d \rightarrow 2p$ transition in a neonlike ion is proven to be a particularly poor pumping candidate; however, the $3s \rightarrow 2p$ transition appears to be more promising.

I. INTRODUCTION

Absorption of intense line emission from a plasma at a matching wavelength represents a promising "flash-lamp" method of pumping plasma x-ray lasers.[1,2] The result is excitation of a specific upper laser state, population inversion, and gain. For efficient pumping and optimum gain, it is vital that the two transitions be of the same photon energy (wavelength) and spread (linewidth), and that the radiation coupling be over a solid angle as close to 4π steradians as possible. Some wavelength adjustments are possible through induced line broadening and shifting (e.g., Doppler streaming). Numerous combinations have been suggested and are summarized in a recent short review.[3] Induced excitation has been demonstrated by fluorescence;[4] however, vacuum-uv population inversion and gain by this method are yet to be demonstrated in the laboratory.

The intent herein is to present the measured radiative power from seven promising pumping lines extending in wavelength from 3.4–7.3 nm, as well as one at 1.1 nm. The output is determined from absolute line intensity measurements. These intensities are compared with each other, as well as with a simple model to estimate the characteristic dimensions for two-plasma configurations suitable for gain. Such evaluations and further optimization will, we hope, lead to selection of the most promising candidates for extended experiments directed towards population inversion and gain at progressively shorter wavelengths in the x-ray region.

The most promising candidates for pumping lines are thought to be the $2 \rightarrow 1$ resonance lines of hydrogenic and heliumlike ions and the $3 \rightarrow 2$ resonance lines of lithiumlike (and perhaps neonlike) ions, all generally observed to be intense in laser-produced plasmas. Considering these transitions, as well as the wavelength range available for absolute intensity measurements and the available driving-laser flux, eight pumping combinations involving mostly ions of similar ionization potential were chosen for this analysis and are indicated in Table I. Here the subscripts refer to transitions in a three-level lasing-ion system between an initial or final level o, a pumped upper laser level u, and a lower laser level l. Hence, absorbing and lasing transitions of quantum interval Δn and wavelength λ (in nm) are labeled ou and ul, respectively. Also, the subscript p in Table I refers to the pumping plasma. Wavelengths[10,11] are shown only to two significant figures for conciseness. The ratio $\Delta\lambda_p/\lambda_p$ (where

TABLE I. Some promising line matches for x-ray laser pumping.

Pump ion	Species	Δn_p	λ_p (nm)	$10^4(\Delta\lambda_p/\lambda_p)$		Lasing ion	Species	Δn_{ou}	Δn_{ul}	λ_{ul} (nm)	Ref.
B^{4+} [a]	(H-)	$2p \rightarrow 1s$	4.9	0.41	→	Ne^{9+}	(H-)	$2p \rightarrow 4d$	$4 \rightarrow 3$	19	3,5,6,7
C^{5+} [b]	(H-)	$2p \rightarrow 1s$	3.4	0.30	→	Mg^{11+}	(H-)	$2p \rightarrow 4d$	$4 \rightarrow 3$	13	3,5,6,7
Al^{10+}	(Li-)	$3p \rightarrow 2s$	4.8	0.42	→	Mg^{8+}	(Be-)	$2s \rightarrow 4p$	$4 \rightarrow 3$	20	3,7,8
Si^{11+} [c]	(Li-)	$3p \rightarrow 2s$	4.1	11	→	B^{4+}	(H-)	$1s \rightarrow 3p$	$3 \rightarrow 2$	26	3,7
Si^{11+} [c]		$3d \rightarrow 2p$	4.4	6.6	→	Mg^{9+}	(Li-)	$2s \rightarrow 4p$	$4 \rightarrow 3$	18	3,7
S^{6+}	(Ne-)	$3d \rightarrow 2p$	6.0	1.3	→	Ne^{7+}	(Li-)	$2s \rightarrow 5p$	$5 \rightarrow 3$	20	3,7
S^{6+}		$3s \rightarrow 2p$	7.3	1.5	→	Al^{5+}	(O-)	$2p \rightarrow 4d$	$4 \rightarrow 3$	44	7
Na^{9+} [d]	(He-)	$2p \rightarrow 1s$	1.1	2.4	→	Ne^{8+}	(He-)	$1s \rightarrow 4p$	$4 \rightarrow 3$	23	1,3,9

[a] BN.
[b] Graphite.
[c] SiO_2.
[d] NaF, NaCl targets.

$\Delta\lambda_p = |\lambda_p - \lambda_{ou}|$) is a figure of merit for the line matches, ideally equaling zero. Early references for each match in Table I are listed in the last column.

II. MODEL

Without a resonant cavity, the achievement of meaningful net gain $\exp(GL) \gtrsim 20$ (i.e., $GL \gtrsim 3$) over a lasing plasma of length L is largely governed by a high density N_o of ions available for pumping, as well as a high fractional population N_u/N_o of the upper laser state, i.e., the product $N_o(N_u/N_o)$ in the gain-length product relation:

$$GL = (N_u\sigma_{ul} - N_l\sigma_{lu})L = N_o\left(\frac{N_u}{N_o}\right)\sigma_{ul}IL \ , \tag{1a}$$

where

$$I = \left(1 - \frac{N_l}{N_u}\frac{\sigma_{lu}}{\sigma_{ul}}\right) = \left(1 - \frac{N_l}{N_u}\frac{g_u}{g_l}\right) \tag{1b}$$

is the population inversion factor (assumed ~ 1 for efficient pumping). Here σ_{ul} and σ_{lu} are the cross sections for stimulated emission ($u \rightarrow l$) and absorption ($l \rightarrow u$), respectively, and likewise g_u and g_l are the upper and lower laser state statistical weights.

The maximum values of the density N_o in Eq. (1) can be set by the opacity $N_o r_L \sigma_{ol}$ of the $o \rightarrow l$ absorption transition, where r_L is the short dimension in the laser plasma. This absorption coefficient must be $\lesssim 5$ to minimize radiative trapping and assure reasonably rapid radiative depletion of the lower level population density, for quasi-cw operation.

For matched-line photon pumping at a frequency ν in a quasiequilibrium plasma where the pumping rate dominates over collisions, the ratio N_u/N_o in Eq. (1a) can be approximated by

$$\frac{N_u}{N_o} \approx \frac{N_\nu \sigma_{ou} c}{2A_u} \ , \tag{2}$$

where σ_{ou} is the photoabsorption pumping cross section from level o to u, N_ν is the pumping photon density in the lasing volume, and

$$2A_u = \frac{16\pi c}{\lambda_{ou}^3}\left(\frac{g_o}{g_u}\right)\left(\frac{\Delta\nu}{\nu}\right)_{ou}\sigma_{ou} \tag{3}$$

approximates the total decay rate for all transitions (hence the factor of 2) out of level u for a $o \rightarrow u$ resonance frequency ν_{ou}, a linewidth $\Delta\nu_{ou}$, and a wavelength λ_{ou}.

Because the goal here is to measure and interpret pumping flux, the photon density N_ν $[=(W_p/h\nu)\times(\Omega/4\pi)(a_L c)^{-1}]$ subtended by a solid angle Ω (defined by the lasing plasma of area a_L and the distance from a point in the pump plasma) should be expressed in a measurable emission quantity such as the source-plasma radiated power W_p. For the important case of elongated, rodlike pumping and lasing plasmas, both of length L and separated by a distance r_s, the result of integrating the line density W_p/L over the pumping plasma length is

$$N_\nu = \frac{W_p}{h\nu_{ou}}\frac{1}{4\pi c}\frac{\eta}{r_s L} \ , \tag{4}$$

where η is a geometrical factor of order unity. For $L \approx 10r_s$, η ranges from ~ 3 near the center of the rod to ~ 1.5 at the ends. This same result is obtained for a coaxial configuration in which the pump plasma is the core (this geometry promises larger output because of the increased volume of the lasing plasma). In fact, the form of Eq. (4) remains valid for a number of other geometries, including the spherical shell case and the congruent-plasma cylindrical and spherical cases [using $N_\nu = (W_p/h\nu)\ (r/c)V^{-1}$ for a volume V], with appropriate values of $\eta = 1$–3 and with L a characteristic size of the plasmas. Combining Eqs. (1)–(4) with $\eta = 3$ for a typical geometry, with $N_o r_L \sigma_{ol} = 5$, and using $\sigma_{\rm abs} \propto \lambda f$, $\sigma_{ul} = \sigma_{lu}(g_l/g_u)$ from detailed balancing, we arrive at

$$\frac{W_p}{r_L r_s} = \frac{64\pi^2 hc^2}{3I}\left[\frac{1}{\lambda_{ou}^4}\frac{\lambda_{ol}}{\lambda_{ul}}\left(\frac{g_o f_{ol}}{g_l f_{lu}}\right)\right]\left(\frac{\Delta\nu}{\nu}\right)_{ou} \ , \tag{5}$$

for $GL = 3$ (the f's represent absorption oscillator strengths). To be noted especially is the very strong inverse dependence ($\propto \lambda_{ou}^{-3}$ for $\lambda_{ol} \approx \lambda_{ou}$) on pumping wavelength, which therefore should be made as long as possible. As to be expected, Eq. (5) shows that short laser wavelengths λ_{ul} and weak ($g_l f_{lu}$ low) laser transitions require increased pumping power. Likewise, strong ($g_o f_{ol}$ large) decay transitions reflect high pumping power needs through a decreased $N_o r_L$ opacity limit. Also to be noted in Eq. (5) is the absence of the parameter L, which results from specifying the desired gain product GL in Eq. (1). It should be noted that this model assumes an exact wavelength match for emission or absorption coupling which in reality would not likely be obtained, and a larger photon density might be required, depending on the mismatch, the plasma conditions, and the dynamics.

Equation (5) is evaluated for the cases of interest here, assuming $I = 1$ and Doppler broadening[12] at an ion temperature of one-third the ionization potential for producing the lasing ion from the next lower species. The results are listed in Table II from which a value of a characteristic (mean) dimension $r = \sqrt{r_L r_s}$ is determined, using the listed values of W_p, measured as described below.

III. EXPERIMENT

The intensities were determined from microdensitometer traces of the Kodak 101 film used. All film densities were in the linear range[13] below 0.5. The spectrograph used was a 1-m-focal-length grazing incidence instrument with a 1200-grooves/mm grating. This instrument viewed the plasma along an axis parallel to the target surface. Previous experience showed that the temporal dependences of the resonance line intensities measured here in similar wavelength (3–7 nm) and spatial regions are essentially the same. The 1.05-μm Nd-glass driving laser was operated for most cases at an output energy of 20 J, a pulse length of 10 ns and focused to a 0.5-mm-diameter spot (10^{12} W/cm^2) on the slab targets. The plasma was first viewed under optically thin conditions at 3 mm from the target for the absolute emission, and then extended to the surface region for the output power results reported in Table II. For the short-wavelength (1.1-nm) Na^{9+} emission, the power radiated was measured for NaF (and NaCl) solid targets irradiated by a 100-J, 4-ns Nd-glass laser, operated at 1.05 μm, and point focused to 80 μm for a target irradiance of 5×10^{14} W/cm^2.

TABLE II. Pumping power calculated ($W_p/\bar{r}^2$) and measured (W_p) and characteristic size $\bar{r}$.

Pump ion	Δn_p	$\lambda_p \approx \lambda_{ou}$ (nm)	$10^{-10}(W_p/\bar{r}^2)$ (W cm^{-2})	$10^{-4}W_p$ (W)	$\bar{r}$ (μm)
B^{4+}	$2p \rightarrow 1s$	4.9	0.026	3.9	120
C^{5+}	$2p \rightarrow 1s$	3.4	0.13	6.5	71
Al^{10+}	$3p \rightarrow 2s$	4.8	2.7	2.0	8.6
Si^{11+}	$3p \rightarrow 2s$	4.1	2.0	1.3	8.1
Si^{11+}	$3d \rightarrow 2p$	4.4	2.0	2.6	11
S^{6+}	$3d \rightarrow 2p$	6.0	4.1	0.32	2.8
S^{6+}	$3s \rightarrow 2p$	7.3	1.1	1.3	11
Na^{9+} [a]	$2p \rightarrow 1s$	1.1	31	120 (2500)[b]	20 (90)[b]

[a] For increased driving laser flux.

[b] Measured in near-normal direction to the slab target.

The other targets were not tested at this flux level so that no results are available on scaling of output with increased target irradiance, an interesting independent topic for further investigations (e.g., possible secondary effects such as increased expansion velocity and decreased density).

The absolute intensities in the soft-x-ray region were obtained essentially from a branching ratio technique using the $n=7$ level in the hydrogenic C^{5+} ion, knowing that the intensity $I_{7\text{-}j}$ of a $7 \rightarrow j$ transition is given by

$$I_{7\text{-}j} = N_7 A_{7\text{-}j} h\nu_{7\text{-}j} \ , \tag{6}$$

where $A_{7\text{-}j}$ and $h\nu_{7\text{-}j}$ are the transition probabilities[14] and the photon energy, respectively. Measurement of the absolute intensity standardized to a calibrated deuterium lamp at 343.4-nm wavelength (for $j=6$) resulted[15] in $N_7 = 1.5\times10^{12}$ cm^{-3} at an irradiance of 10^{12} W/cm^2 from a laser driver, producing a plasma on a graphite slab target and measured at 3 mm from the surface in vacuum. Hence, the absolute intensities for the $7 \rightarrow 2$ transition at 11.0 nm and the $7 \rightarrow 1$ transition at 2.58 nm are known from Eq. (6). Between these two limits, the relative instrumental sensitivity was determined by other series branching ratios in carbon ions. Using relative intensities between 3 mm and the target region, this calibration led to the power outputs listed in Table II for the pump lines in this wavelength region, corresponding to the targets indicated in Table I. In addition, the most intense line in the spectrum, namely, the $C^{4+}(2 \rightarrow 1)$ resonance line at 4.03 nm, was determined[7] to be saturated at a level corresponding to a brightness temperature of ~ 30 eV, consistent with an expanding laser-produced plasma.

For the $Na^{9+}(2 \rightarrow 1)$ pumping line at 1.1 nm, extension by an additional branching ratio was necessary. For this, the $3p \rightarrow 2s$ intensity for the same heliumlike ion at 6.36 nm was obtained as above, and the branching ratio of transition probabilities and photon energies were again used to obtain the intensity for the $3 \rightarrow 1$ line at 0.94 nm, sufficiently close to the pump line to obtain that emission by comparison of measured relative intensities.

IV. RESULTS

The results of the pumping power measurements are compared in Table II with estimates from Eq. (5) to yield $\bar{r} = \sqrt{r_L r_s}$, which is a characteristic dimension for the plasma combination. For the present rather modest driving-laser power density on slab targets, dimensions of ~ 10–100 μm are determined with a factor-of-2 estimated overall accuracy. Such dimensions are typically found in laser-produced plasmas, particularly in elongated configurations on slabs, fibers,[16] or foils.[17]

The scaling relations derived above indicate that higher pumping power output would permit larger dimensions or a shorter pumping wavelength or both. An example is the Na-Ne combination, the results for which are included in Table II. Because of the shorter (1.1-nm) pump wavelength, a driving laser of power more than 10 times as large was used and a characteristic dimension of 20 μm was first derived. Furthermore, an independent measurement[18] using a calibrated x-ray crystal spectograph which viewed the plasma along an axis more nearly normal to the slab yielded $W_p \approx 25$ MW (within a factor of 2), or $\bar{r} = 90$ μm which represents a ~ 20 times higher pump power from the surface-crater region compared to that viewed tangentionally by the grazing incidence spectrograph. No detailed study of this suggested angular dependence was possible. However, a similar but smaller (≤ 5 times) anisotropy has been previously observed[19] with thin Fe foil targets and was attributed to geometric and opacity effects peculiar to the directions of of view. It is possible that the emission for the other species could also increase similarly when at or imbedded in the target surface, the amount depending on the element and ionization species. Most likely, this 25-MW pumping power can be increased by factors of 10–100, mainly with a shorter wavelength and pulse in the driving laser and possibly with limited-mass (foil) targets.[20–22]

Equally as meaningful as the absolute power results are the relative merits of the various pumping transitions and ionic species tested. For the hydrogenic, heliumlike, and lithiumlike ion data, all intensities are within a factor of 3 of blackbody saturation, when compared to the already saturated C^{4+} resonance line.[7] For these ions, it can be concluded that the pumping line intensity from $2 \rightarrow 1$ and $3 \rightarrow 2$ transitions probably can be driven also to the saturation level by a higher power laser. It appears that the $2p \rightarrow 1s$ transitions are somewhat favored over the $3d \rightarrow 2p$ and particularly over the $3p \rightarrow 2s$ transitions, as might be expected considering population distributions over structured atomic levels; however, this difference is not decisive, particularly with the present limited accuracy of measurement.

The one neonlike candidate tested, namely, S^{6+}, proved disappointing, even with attempts in this case to improve the intensity by lowering (through defocusing) the driving laser power in order to compensate for a somewhat lower ionization potential. The spectrographic data showed the $3s \rightarrow 2p$ line to be more intense than the $3d \rightarrow 3p$ line, most likely because of the $3s$ population acquired from the $3p$ level with a closed $2s$ shell. The intensity (compared to saturation) is still low for both compared to that of the other candidates, and may be indicative of the general undesirability of a neonlike pump ion with a large number of lines for the number of electrons.

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