Femtosecond-pulse-driven electron-excited extreme-ultraviolet lasers in Be-like ions

S. M. Hooker and S. E. Harris

Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305

Received June 2, 1995

A suggestion for the generation of extreme-ultraviolet (XUV) laser radiation based on tunneling ionization and subsequent electron excitation of $\Delta n \neq 0$ transitions is described. The favorable scaling of the required intensity of the pump laser with the output XUV wavelength is compared with that exhibited by XUV lasers based on $\Delta n = 0$ transitions. Calculations for Be-like Ne predict significant gain at 14.1 nm.

The production of both a controlled ion state and electron energy distribution by tunneling ionization in intense femtosecond laser radiation is a promising approach for the generation of coherent short-wavelength radiation. The threshold nature of the tunneling ionization mechanism ensures that essentially all the target atoms may be converted to the desired ion state. In addition, the electron energy distribution produced by tunneling ionization may be controlled by adjustment of the polarization of the ionizing radiation.[1,2] In particular, the use of circularly polarized radiation results in the generation of extremely energetic electrons while a relatively cold ion temperature is maintained. This excitation scheme was the basis of a proposal for extreme-ultraviolet (XUV) lasers in eight-times-ionized rare gases.[3] Recently Lemoff *et al.*[4] demonstrated a small-signal gain of 13.3 cm^{-1} at 41.8 nm in Xe IX with this approach.

In this Letter we describe a new scheme for the generation of coherent XUV radiation based on electron excitation of Be-like ions. We discuss how the required intensity of the pump laser scales with the output wavelength and present a calculation of the gain in one such case, Ne VII, which has a predicted laser wavelength of 14.1 nm.

The inset in Fig. 1 shows a simplified energy-level diagram of the Be-like Ne VII ion. The proposed laser transition is $2s3p\ ^1P_1 \rightarrow 2p^2\ ^1D_2$, and both the lasing ion stage and the electrons that pump it are produced by tunneling ionization.

The weighted oscillator strength gf of the $2s^2\ ^1S_0 \rightarrow 2s3p\ ^1P_1$ pump transition is 0.514, which results in strong pumping by all electrons above the threshold energy. In contrast, excitation of the lower level involves a two-electron transition. Optically nonallowed transitions are pumped rapidly only by electrons close to the threshold energy, and hence the pumping of the lower laser level is expected to be slow.

Although the laser transition also appears to be a two-electron transition, strong mixing of the upper laser level with the nearby $2p3s\ ^1P_1$ level, together with the large transition energy, results in a relatively large weighted oscillator strength and Einstein-A coefficient. For example, in Ne VII the laser transition has $gf = 0.0833$ and an Einstein-A coefficient of 9.24×10^9 s^{-1}. Furthermore, for ion densities corresponding to a few millibars or above, radiative decay of the $2s3p\ ^1P_1$ level to the $2s^2\ ^1S_0$ ground state is strongly trapped.

We now discuss how, for an isoelectronic series, the required intensity of the driving laser varies with the output laser wavelength. To this end it is useful to express the energy E_{nl} of a level as $E_{nl} = -R_\infty(Z - S_{nl})^2/n^2$, where Z is the atomic number of the lasing ion stage, n is the principal quantum number, $l\hbar$ is the orbital angular momentum, and S_{nl} is the shielding factor of the active electron. The ionization energy E_I of the penultimate ion stage is given by $E_I = R_\infty(Z -$

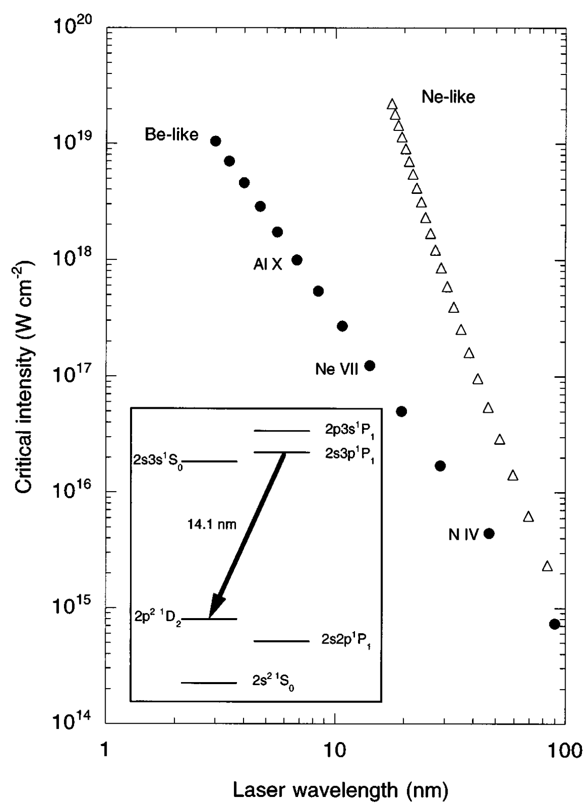

Fig. 1. Calculated critical intensity I_C for an ionization rate of 10^{14} s^{-1} as a function of the wavelength of the $2s3p\ ^1P_1 \rightarrow 2p^2\ ^1D_2$ transition in Be-like ions and the $2p^53p\ ^1S_0 \rightarrow 2p^53s\ ^1P_1$ transition in Ne-like ions. The inset shows the simplified energy-level diagram of Ne VII.

0146-9592/95/191994-03$6.00/0

$S_I)^2/n_I{}^2$, where S_I and n_I are the shielding factor and the principal quantum number of the ionized electron, respectively. The frequency ω of a given $n_2l_2 \rightarrow n_1l_1$ transition in the lasing ion may then be expressed in terms of the ionization energy E_I of the penultimate ion stage as

$$\frac{\hbar\omega}{R_\infty} = n_I{}^2\left(\frac{1}{n_1{}^2} - \frac{1}{n_2{}^2}\right)\left(\frac{E_I}{R_\infty}\right) + 2n_I\left(\frac{\delta_1}{n_1{}^2} - \frac{\delta_2}{n_2{}^2}\right) \times \left(\frac{E_I}{R_\infty}\right)^{1/2} + \left(\frac{\delta_1{}^2}{n_1{}^2} - \frac{\delta_2{}^2}{n_2{}^2}\right), \quad (1)$$

where $\delta_i = S_I - S_{n_il_i}$ is the difference in shielding factors and hence of order unity.

We see that, for laser transitions in which there is no change of principal quantum number ($\Delta n = 0$), such as in the Ne-like laser scheme, the first term in Eq. (1) vanishes and the output laser frequency ω is approximately proportional to $E_I{}^{1/2}$. The critical intensity I_C required for generation of the lasing ion stage varies[2] approximately as $E_I{}^3$, and hence for lasers based on $\Delta n = 0$ transitions I_C is approximately proportional to ω^6, a scaling that leads rapidly to high driving intensities at only moderately short wavelengths. However, for laser transitions in which there is a change in principal quantum number ($\Delta n \neq 0$), such as in the proposed scheme, the output laser frequency is approximately proportional to E_I, and hence I_C varies approximately as ω^3.

Figure 1 shows the calculated critical intensity for a tunneling ionization rate of 10^{14} s^{-1} as a function of the wavelengths λ of the $2s3p\ ^1P_1 \rightarrow 2p^2\ ^1D_2$ transitions in Be-like ions and the $2p^53p\ ^1S_0 \rightarrow 2p^53s\ ^1P_1$ transitions[5] in Ne-like ions based on the ionization data of Kelly[6] and the atomic physics code of Cowan.[7] In agreement with the above, for the Ne-like system I_C is approximately proportional to $\lambda^{-5.8}$, whereas for the Be-like ions $I_C \propto \lambda^{-2.8}$. The significantly improved scaling of the critical intensity for the Be-like scheme is apparent if we consider the intensity required for reaching the onset of the water window at 4.36 nm. For Be-like ions this critical intensity is approximately 3.4×10^{18} W cm^{-2}, 4 orders of magnitude lower than that required in the Ne-like system.

It is important that the lasing ion show stability against variation of the driving intensity, so that the desired ion stage may be generated over as large a volume as possible and also exhibit tolerance of shot-to-shot variation of the intensity of the driving laser. This requires the ionization energy of the lasing ion to be significantly greater than that of the penultimate ion stage, a criterion that may be met by use of a lasing ion in which the ground state has a closed shell, as in Xe IX, or a closed subshell, such as the closed $2s^2$ subshell of Be-like ions. For example, we calculate that more than 50% of Ne atoms will be converted to Ne VII for driving intensities between 1.3×10^{17} and 2.6×10^{17} W cm^{-2}.

We consider now the calculation of the gain of the $2s3p\ ^1P_1 \rightarrow 2p^2\ ^1D_2$ transition in Ne VII at 14.1 nm. The rate equations for the levels of interest are

$$\frac{dN_2}{dt} = R_2n_eN_0 - R_2^{\text{out}}n_eN_2 - \frac{N_2}{\tau_2^{\text{trap}}}, \quad (2)$$

$$\frac{dN_1}{dt} = R_1n_eN_0 - R_1^{\text{out}}n_eN_1 - \frac{N_1}{\tau_1^{\text{trap}}} + N_2A_{21}, \quad (3)$$

$$\frac{dN_0}{dt} = -(R_1 + R_2)n_eN_0, \quad (4)$$

where for each level i, N_i is the number density; τ_i^{trap} is the effective radiative lifetime, allowing for optical trapping; and R_i and R_i^{out} are the averaged electron excitation and deexcitation rates. In this notation the ground state of the ion is level 0, and the upper and lower laser levels are levels 2 and 1, respectively. The total electron density n_e is taken to be constant and for Ne VII is equal to six times the initial value of N_0. We calculated the values of τ_i^{trap}, using the theory of Holstein,[8] by approximating the gain region as an infinitely long cylinder of 30-μm diameter.

The electron rates are determined by the distribution of the residual electron energies after the driving pulse. Figure 2 shows the residual electron energy distribution calculated from the tunneling ionization formula[2] for an 800-nm pulse of Gaussian temporal profile with a peak intensity of 1.75×10^{17} W cm^{-2} and a FWHM of 30 fs. The six peaks in the electron distribution correspond to the removal of the six p electrons. The shape of the electron peaks is a function of the temporal profile of the driving pulse, and the pronounced feature at the highest electron energies corresponds to ionization of Ne^{5+} at the peak of the pulse, where the intensity is almost constant.

In calculating the rates for electron excitation we represented the electron distribution by six electron classes of energies 68.70, 232,0, 610.8, 1680, 2982, and 4780 eV. All electron cross sections were calculated with the Cowan code based on a modified plane-wave Born method,[7] which is estimated to be accurate to within 50%, even close to threshold. As expected, the excitation is highly selective. The calculated excitation rates per state, R_i/g_i, are 1.9×10^{-10} and 1.3×10^{-11} cm^3 s^{-1} for the upper and lower laser

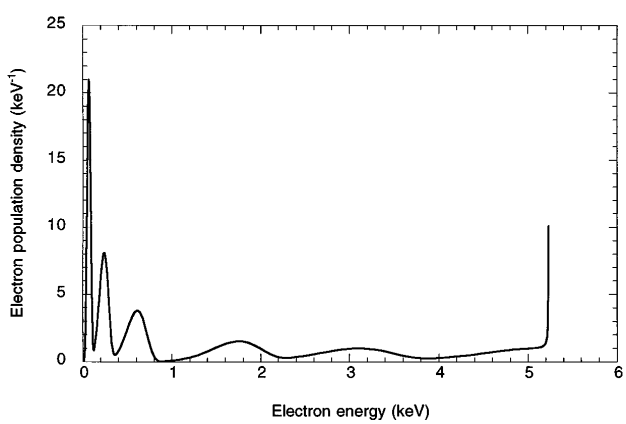

Fig. 2. Calculated distribution of electron energies for the ionization of Ne by an 800-nm Gaussian pulse with a peak intensity of 1.75×10^{17} W cm^{-2} and 30-fs (FWHM) duration.

Table 1. Parameters Used in the Calculation of Gain in Be-Like Ions[a]

Ion	Wavelength (nm)	R_2 (10^{-9} cm^3 s^{-1})	R_1 (10^{-9} cm^3 s^{-1})	R_2^{out} (10^{-9} cm^3 s^{-1})	R_1^{out} (10^{-9} cm^3 s^{-1})	τ_2^{trap} (ps)	τ_1^{trap} (ps)	T_{ion} (K)	A_{21} (ns^{-1})	Inversion (10^{14} cm^{-3})	Gain (cm^{-1})
N IV	46.8	1.1	0.72	370	44	330	3500	426	1.7	2.8	3.2
Ne VII	14.1	0.58	0.064	54	9.2	34	1500	3370	9.2	23	5.4
Al X	6.78	0.14	0.013	35	7.2	5.7	890	21700	26	4.4	0.16

[a]The inversion, the gain, and τ_i^{trap} are calculated for a pressure of 15 mbars.

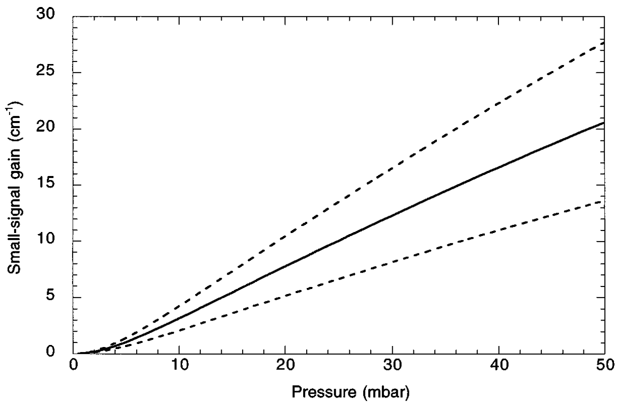

Fig. 3. Calculated small-signal gain coefficient for the $2s3p\ ^1P_1 \rightarrow 2p^2\ ^1D_2$ transition in Ne VII. The dashed curves indicate estimates of the upper and lower bounds of the gain.

levels, respectively. We obtained deexcitation rates by summing over all significant inelastic, including superelastic, collisions. These rates are summarized in Table 1.

As part of the tunneling process, at each ionization event the ions acquire a transverse momentum equal to that of the emitted electron. Ion–ion collisions then thermalize this momentum to yield an increased ion temperature T_{ion} in a time of the order 100 fs. By assuming that for each ion the electrons are emitted in random directions in the transverse plane we estimate that for Ne VII the ions gain an energy of 0.398 eV, which results in an ion temperature of 3370 K.

We estimated the gain cross section for the $2s3p\ ^1P_1 \rightarrow 2p^2\ ^1D_2$ transition by assuming a Lorentzian line shape with a width equal to the quadrature sum of the homogeneous and Doppler widths. At the high electron energies appropriate to the present case it is expected that the Stark width will be dominated by inelastic collisions, and so it may be calculated from the electron collision cross sections. This approach yielded values for the broadening coefficients within 30% of those calculated by evaluation of an effective Gaunt factor,[9] with the Cowan code cross sections yielding the larger widths.

Figure 3 shows the small-signal gain coefficient for the $2s3p\ ^1P_1 \rightarrow 2p^2\ ^1D_2$ transition in Ne VII as a function of the initial Ne pressure, calculated by numerical integration of the above rate equations. The main sources of error are expected to be errors in the calculated excitation rates of the upper and lower levels, which are estimated to be accurate to better than $\pm 50\%$. The figure shows bounds to the gain calculated by use of $(1 \pm 25\%)R_2$ and $(1 \mp 25\%)R_1$ for the excitation rates to the upper and lower levels, respectively. At pressures greater than approximately 450 mbars (not shown), electron collisions cause the gain to saturate at ~ 92 cm^{-1} as a result of the transition linewidth and level lifetimes being dominated by electron collisions.

In Table 1 we summarize the results of similar calculations for N IV and Al X, which have calculated laser wavelengths of 46.8 and 6.78 nm, respectively. For both ions the calculated gain is lower than that of Ne VII. The lower gain in N IV is due to the fact that the energies of the three electron classes produced by the ionization are relatively low, which leads to poor selective excitation and rapid quenching of the upper laser level. The primary reason for the low gain in Al X is the much greater ion temperature. We could reduce ion heating while maintaining a high excitation rate by employing elliptically polarized pump radiation. For the N IV and Al X systems the gains are calculated to saturate at 3.4 and 17 cm^{-1} for pressures greater than approximately 16 and 2000 mbars, respectively.

In conclusion, we have suggested a new scheme for the generation of XUV laser radiation. Since in this system $\Delta n \neq 0$, the necessary intensity of the driving laser scales approximately as ω^3 instead of as ω^6, as in $\Delta n = 0$ systems. Calculations for Be-like Ne predict significant gain at 14.1 nm.

The authors thank G. Y. Yin and P. T. Epp for many helpful discussions. This research was supported by the U.S. Army Research Office and the U.S. Air Force Office of Scientific Research.

References

1. P. B. Corkum, N. H. Burnett, and F. Brunel, Phys. Rev. Lett. **62,** 1259 (1989).
2. N. H. Burnett and P. B. Corkum, J. Opt. Soc. Am. B **6,** 1195 (1989).
3. B. E. Lemoff, C. P. J. Barty, and S. E. Harris, Opt. Lett. **19,** 569 (1994).
4. B. E. Lemoff, G. Y. Yin, C. L. Gordon III, C. P. J. Barty, and S. E. Harris, Phys. Rev. Lett. **74,** 1574 (1995).
5. For the Ne-like scheme the coupling of the lower level progresses from LS to jj as Z is increased. At high Z the lower level is more properly labeled $2p^5 3s$ $(3/2, 1/2)_1$.
6. R. L. Kelly, J. Phys. Chem. Ref. Data **16,** Suppl. 1 (1987).
7. R. D. Cowan, *The Theory of Atomic Structure and Spectra* (U. California Press, Berkeley, Calif., 1981), Secs. 8-1, 16-1, and 18-13.
8. T. Holstein, Phys. Rev. **83,** 1159 (1951).
9. J. D. Hey and P. Breger, in *Spectral Line Shapes*, B. Wende, ed. (de Gruyter, New York, 1981), pp. 191–200.