Proposal for an extreme-ultraviolet Auger laser at 63.8 nm in Cs III

D. J. Walker, R. G. Caro,* and S. E. Harris

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

Received April 30, 1986; accepted July 21, 1986

A system is proposed in which Cs atoms photoionized by soft x rays from a laser-produced plasma undergo selective Auger decay, causing inversion and lasing at 63.8 nm in Cs III. Rate-equation calculations show that lasing should occur when a small (1–J) 532-nm pump laser is used. A similar system in Rb III is briefly discussed.

It has been shown that large densities of highly excited atoms can be produced as a result of photoionization by soft x rays emitted from a laser-produced plasma.1,2 By this technique, population inversions have been produced, and lasing in the visible and near-ultraviolet regions of the spectrum has been observed.3,4 In this paper we propose a system in which these x rays would photoionize Cs vapor, producing Cs II 4d105s5p6s ions. These ions would then undergo rapid, selective Auger decay, resulting in an inversion of the Cs III 4d105p5s6s configuration with respect to the Cs III 4d105p5p6s configuration and lasing at 63.8 nm. A similar system in which the Rb III 3d105p24s6 configuration is inverted with respect to the Rb III 3d105p44s configuration, resulting in gain at 49 nm, will be discussed briefly.

The key point in identifying systems such as those described in this paper is to note that Auger decay will be rapid and therefore selective when the two interacting electrons both make strong dipole transitions, one to a bound level and one to the continuum, and when the average distance between these electrons is small and hence the Coulomb interaction is large. This occurs when the two interacting electrons belong to the same n shell and when they have opposite spin. In this sense the present proposal is similar to that of the Zn III system of Mendelsohn and Harris.5 The present system has the additional advantage that the cross section for d-shell photoionization is larger than that of the other shells and therefore should be more efficient and less sensitive to electron deexcitation than was the Zn system.

We note that the use of Auger processes to create inversions in the extreme ultraviolet has been suggested earlier by McGuire6 and Krolik and Shapiro7 and has been demonstrated at visible wavelengths by Bokor et al.8

An energy-level diagram for the suggested Cs III laser system is shown in Fig. 1. The laser-produced x rays are assumed to have a blackbody spectral distribution with a temperature of 35 eV. Photoionization of Cs I by these x rays primarily produces 4d vacancies. According to the prediction of the RCN/RCG code,9 83% of these vacancies Auger decay to the Cs III 4d105s5p6s6s configuration with an average Auger rate of 4 × 1013 sec−1. Of these, 31% are predicted to go into the upper laser level 4d105s5p6s6p 5/2, therefore giving a total predicted yield to this level of 26%. About 1% of the initial vacancies are predicted to decay to the lower laser level 4d105s5p6p 3/2, and therefore the predicted degeneracy normalized inversion \[ \left[ \frac{N_u}{(\epsilon_s/\epsilon_l) N_l} \right] \] is about 24.5. This prediction is confirmed by experimental Auger electron spectroscopy.10 In Fig. 1, the level positions of Cs I and the lower two configurations of Cs II are taken from the tables of Moore.11 The energy of the laser transition at 63.8 nm is taken from Epstein and Reader,12 and the RCN/RCG code is used for the other level positions as well as for the calculated Auger rates and oscillator strengths.

In Rb III similar calculations predict that Auger decay should result in a degeneracy normalized inversion of the Rb III 3d104s4p5s6p 5/2 level relative to the Rb III 3d104s4p5p 3/2 level of a factor of 4.5 and that the wavelength of this transition is 49 nm. Experimental evidence supporting this prediction has been reported by Menzel and Mehlhorn.13

Although the photoionization process is predicted to produce a substantial inversion on the Cs III 4d105s5p6s6s 2D5/2– 4d105s5p6s 2P3/2 transition, many secondary processes can act to diminish or even destroy the inversion. To examine this problem, a rate-equation model has been used to predict the populations in a variety of levels in Cs I, Cs II, and Cs III under the conditions in which it is proposed to observe lasing on the 63.8-nm transition. The processes listed in Table 1 are those that have been considered of importance and that have been included in the model. The detailed mechanics of the rate-equation model are described elsewhere.14

Processes (1) and (2) of Table 1 are the photoionization and Auger decay processes that lead to population of Cs II and Cs III levels. The photoionization cross sections for the 4d, 5s, and 5p subshells of Cs I are obtained from the experimental work of Hecht.15 The photoionization cross section for the 6s subshell was obtained by an extrapolation of the data of Marr.16 The branching ratios for determining the distribution of the products of Auger decay among the Cs III levels were calculated with the RCN/RCG code. The RCN/RCG calculations also show that the near degeneracy of the Cs II 4d95s5p6s and Cs II 4d95s5p6p5d configurations does not result in appreciable mixing between levels of the Cs II 4d95s5p6s and Cs II 4d95s5p6p5d configurations that undergo Auger decay; hence this near degeneracy does not affect the Auger branching ratios. We note that one ambiguity exists: calculations show that the Cs III 4d105p6s6s configuration is nearly degenerate with the Cs II 4d95s5p6s6s configuration, and we are unable to predict which has the higher energy. Thus, if the Cs III configura-
Table 1. Processes Included in Rate-Equation Model

(1) Photoionization from Cs I 4p₁₀⁶₅s²₅p₆₆s to Cs II 4d¹₀⁶₅s²₅p₆₆s
    4d¹₀⁶₅s²₅p₆₆s

(2) Auger decay from Cs II 4d¹₀⁶₅s²₅p₆₆s to Cs III 4d¹₀⁶₅s²₅p₆₆s
    4d¹₀⁶₅s²₅p₆₅d
    4d¹₀⁶₅s²₅p₅₆s

(3) Radiative decay of Cs III 4d¹₀⁶₅s²₅p₆₅d to Cs III 4d¹₀⁶₅s²₅p₅₆s
    4d¹₀⁶₅s²₅p₅₆s

(4) Gain depletion by ASE

(5) Electron double ionization of Cs I 4d¹₀⁶₅s²₅p₆₆s

(6) Electron single ionization of Cs I 4d¹₀⁶₅s²₅p₆₆s
    Cs I 4d¹₀⁶₅s²₅p₆₅p
    Cs II 4d¹₀⁶₅s²₅p₆₅p
    Cs III 4d¹₀⁶₅s²₅p₆₅p
    Cs III 4d¹₀⁶₅s²₅p₆₅p

(7) Electron excitation of Cs I 4d¹₀⁶₅s²₅p₆₆s → 4d¹₀⁶₅s²₅p₆₅p
    Cs II 4d¹₀⁶₅s²₅p₆₅p → 4d¹₀⁶₅s²₅p₆₅p
    Cs III 4d¹₀⁶₅s²₅p₆₅p → 4d¹₀⁶₅s²₅p₆₅p

Table 2. Auger Branching Ratios and Decay Times of Levels That Decay Radiatively to the Lower Cs III Laser Level

<table>
<thead>
<tr>
<th>Designation</th>
<th>Auger Yield (%) from Cs II</th>
<th>Radiative Decay Time to Lower Laser Level (nsec)</th>
<th>Total Radiative Decay Time (nsec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cs III 4d¹₀⁶₅s²₅p₆₅p₆s 3D₁₂</td>
<td>23.0</td>
<td>0.41</td>
<td>0.41</td>
</tr>
<tr>
<td>4d¹₀⁶₅s²₅p₆₅p₆s 3S₁₂</td>
<td>11.0</td>
<td>0.38</td>
<td>0.05</td>
</tr>
<tr>
<td>4d¹₀⁶₅s²₅p₆₅p₆s 3P₁₂</td>
<td>6.8</td>
<td>2.0</td>
<td>2.0</td>
</tr>
<tr>
<td>4d¹₀⁶₅s²₅p₆₅p₆s 3P₂₂</td>
<td>4.7</td>
<td>2.7</td>
<td>2.7</td>
</tr>
<tr>
<td>4d¹₀⁶₅s²₅p₆₅p₆s 3P₃₂</td>
<td>4.0</td>
<td>1.1</td>
<td>1.1</td>
</tr>
<tr>
<td>4d¹₀⁶₅s²₅p₆₅p₆s 3P₄₂</td>
<td>3.9</td>
<td>0.45</td>
<td>0.45</td>
</tr>
<tr>
<td>4d¹₀⁶₅s²₅p₆₅p₆s 3P₅₂</td>
<td>3.2</td>
<td>0.47</td>
<td>0.26</td>
</tr>
<tr>
<td>4d¹₀⁶₅s²₅p₆₅p₆s 3P₆₂</td>
<td>2.6</td>
<td>0.053</td>
<td>0.03</td>
</tr>
<tr>
<td>4d¹₀⁶₅s²₅p₆₅p₆s 3P₇₂</td>
<td>2.2</td>
<td>1.0</td>
<td>1.0</td>
</tr>
</tbody>
</table>

fluorescent systems, particularly those with low aspect ratio, amplified spontaneous emission (ASE), process (4) of Table 1, plays an important role in determining the magnitude and duration of the gain of the system. This is because as the population inversion and the gain rise, spontaneous emission in directions other than the lasing direction causes other pumped atoms to radiate in random directions. This reduces the system gain without contributing to the output signal. For the rate-equation model used here, we assume a cylindrical volume of length L, diameter D, and aspect ratio L/D and calculate the stimulated lifetime of upper-laser-level atoms in the presence of the photon field of the surrounding atoms.

Processes (1)–(4) of Table 1 deal with the effects of interactions between photons and Cs atoms and ions; processes
during the 100-psec duration of the laser pulse. For longer pump pulses, these processes would become important. Curve c shows the effect of cascades from the levels listed in Table 2. The effect of these cascades is to reduce the duration and magnitude of the inversion by filling the lower laser level more quickly. Curves d–f show that ASE can dramatically reduce the inversion in systems of small aspect ratio, whereas for large aspect ratio the effect is smaller.

At a temperature of 270°C, corresponding to a Cs-vapor density of $2 \times 10^{14}$ atoms/cm$^3$, the Doppler width of the 63.8-nm transition is 0.23 cm$^{-1}$. A calculation of the hyperfine splitting of the upper laser level using Sobel’man’s$^{19}$ and RCN/RCG wave functions indicates that the hyperfine splitting is larger than the Doppler width and that only a single hyperfine component falls under the Doppler width. Assuming that each hyperfine component is populated in proportion to its degeneracy, the largest hyperfine component of the $4d^{10}5s^25p^66s^2D_{5/2}$ level receives 1/3.7 of the total population. (Therefore the Auger yield to this particular component is 6.2%). This, along with the gain cross section of $5.6 \times 10^{-13}$ cm$^2$ and the curves of Fig. 2, therefore implies a maximum gain on the 63.8-nm transition of $\exp(21)$ at an aspect ratio of 32. It should be noted that the model predictions are insensitive to the exact temperature of the soft-x-ray blackbody—the inversion drops to half of its peak value at 15 and 75 eV.

To summarize, we have suggested a method of using selective Auger decay to produce population inversion and gain in Cs III at 63.8 nm. Rate-equation calculations show that a 100-psec, 1-J pulse of 532-nm light should produce superfluorescent gain. An analogous system in Rb III at 49 nm was also briefly discussed.

ACKNOWLEDGMENTS

The authors acknowledge important discussions with J. Reader, R. W. Falcone, A. J. Mendelsohn, and J. F. Young.

The research here was supported by the Strategic Defense Initiative Organization, the U.S. Army Research Office, the U.S. Air Force Office of Scientific Research, the U.S. Office of Naval Research, and the Lawrence Livermore National Laboratory.

* Present address, Summit Technology, Inc., 150 Coolidge Avenue, Watertown, Massachusetts 02171.

REFERENCES AND NOTES

17. R. W. Falcone, Department of Physics, University of California, Berkeley, California 94720 (personal communication).