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Effect of dielectronic recombination on the kinetics of neonlike selenium

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In the recent successful demonstration of a soft-x-ray laser in neonlike selenium, the $3p$-$3s$, $J=0$-1 transition, which had been predicted to show very high gain, did not appear to be amplified. Most previous calculations have assumed that the dominant pumping mechanism is electron collisional excitation and have neglected the effects of ionization and recombination on the kinetics of neonlike levels. In this work, we studied the effects of collisional ionization and three-body, radiative, and dielectronic recombination, connecting the three fluorinelike ground states to the lowest 89 levels of neonlike Se. Our results show that dielectronic recombination is a significant process for populating the $J=2$ upper laser levels, and must be considered to model accurately the excited-state kinetics of the neonlike ion.

The recent successful demonstration of a soft-x-ray laser in neonlike selenium[1] raised a number of interesting puzzles. Perhaps the most intriguing is the fact that the $2p^53p$-$2p^53s$ ($J=0$-1) transition near 183 Å, which was predicted to have very high gain in all previous calculations,[2–4] did not show significant amplification.

The mechanism for creating the population inversion has been assumed to be collisional excitation from the neonlike ground state. For this reason, several predictions[2,3] have been based on kinetic models which included only neonlike states and did not explicitly consider ionization or recombination from nearby ionization stages. Even calculations[4] which included other ionization stages treated dielectronic recombination in a simple way and neglected its effect on the populations of excited levels.

In this paper, we consider the effects of ionization and recombination, including detailed dielectronic recombination rates, which couple the fluorinelike ground states and the neonlike levels. We show that dielectronic recombination is an important mechanism for populating the $n=3$ excited states of the neonlike ion, and that it has significant effects on the $2p^53p$-$2p^53s$ inversion kinetics. However, it is not in itself enough to explain the apparent lack of amplification of the $J=0$-1 transition observed in the experiments.[1]

We constructed a kinetic model for selenium which included the $2s^22p^6$ ($J=0$) neonlike ground state, the 36 $n=3$ and the 52 $n=4$ excited states, and the three fluorinelike states $2s^22p^5$ ($J=\frac{1}{2},\frac{3}{2}$) and $2s2p^6$ ($J=\frac{1}{2}$). Energies and radiative rates were computed using configuration interaction wave functions in intermediate coupling. For the $n=2$ and $n=3$ states of Ne-like selenium, we used the atomic structure code SUPERSTRUCTURE,[5] for the $n=4$ states of Ne-like ion and the F-like states, the multiconfigurational Dirac-Fock code YODA.[6]

All collisional excitation rates were calculated by averaging collision strengths over a Maxwellian electron distribution. For the $n=2$-3 transitions, the distorted wave code DSW[7] was used to compute L-S scattering amplitudes, which were transformed to intermediate coupling using JAJOM.[8] The $n=2$–4 collision strengths were calculated directly in intermediate coupling using the relativistic distorted wave code MCDW.[6] The electron collision strengths for the electric dipole allowed excited-state–excited-state transitions in the Ne-like ion, and the $2s$-$2p$ transitions in the F-like ion were calculated using the classical path method.[9] Finally, the collision strengths for transitions among the fine-structure levels of a given $n=3$ configuration [e.g., $(2p^53p)\,J \rightarrow 2p^53p\ J'$] were computed with DSW and JAJOM.

Ionization and three-body recombination rates were calculated using the scaled hydrogenic rates of Sampson and coworkers.[10] Radiative recombination rates were obtained using the prescription of Weisheit, Tarter, Scofield, and Richards.[11] All these rates are given by simple formulas which do not take into account the detailed structure of the states.

For the plasma conditions relevant for the neonlike selenium laser,[4] the dominant recombination process is dielectronic recombination, which we treated in full detail for all the $(3l3l')$ and $(3l4l')$ doubly excited states of Ne-like selenium. The dielectronic recombination coefficients were calculated in the isolated resonance approximation.[12] The Auger and radiative rates of the individual doubly excited states were evaluated in intermediate coupling with configuration interaction using the multiconfiguration Dirac-Fock method.[12,13] The contributions from the higher autoionizing states (e.g., $3lnl'$ with $n>5$ and $nln'l'$ with $n,n'>4$), which are estimated to be 30%–40% of the total rate in the case of Na-like Se,[14] were neglected in the present calculations. The contribution from $\Delta n=0$ (Coster-Kronig) transitions peaks at a much lower temperature ($T_e=60$ eV) and is only 2% of the total rate at $T_e=1$ keV.

Table I gives the total dielectronic recombination coefficients, including the contribution from the $3l3l'$ and $3l4l'$ doubly excited states, for the three lowest levels of fluorinelike selenium. At $T_e=1.0$ keV the estimated total dielectronic rate coefficients, including other high-n intermediate

TABLE I. Theoretical dielectronic recombination coefficients (in 10^{-11} cm^3/sec) for Se^{25+}.

Initial core	Temperature (eV) 500	1000	2000
$2s_{1/2}$	2.55	1.96	1.04
$2p_{1/2}$	2.75	2.22	1.20
$2p_{3/2}$	2.42	1.94	1.06

states, are in the range $2.8–3.5\times10^{-11}$ cm^3/sec which is consistent with the value used by Rosen *et al.*[4] in the design of the selenium soft-x-ray laser targets. However, our dielectronic rate coefficients for Se^{25+} are about an order of magnitude larger than the value used by Apruzese *et al.*[15] in their recent analysis of the Livermore experiments.[1] The dielectronic rate coefficients used in Ref. 15 were obtained by using autoionization rates derived from extrapolation of calculated electron-impact excitation cross sections.[16] This extrapolation procedure has been found to introduce large errors in the calculation of Auger rates for low-lying doubly excited states.[17] In the case of the Li-isoelectronic sequence, large discrepancies were also found between results obtained from *ab initio* calculations[18] and using the extrapolation procedure.[16] It should be noted that, for Ne-like ions, the dielectronic rates from the present multiconfiguration Dirac-Fock method[14] agree with other detailed Hartree-Fock calculations[19–21] within 20%.

To study the excited-state kinetics, we constructed rate equations and solved them in steady state for the level populations. We then calculated the relative inversion density for a given transition as

$$y_{UL}=\nu_U-g_U\nu_L/g_L\ ,$$

where ν_U (ν_L) and g_U (g_L) are the relative population and statistical weight, respectively, of the upper (lower) state of the transition. (Relative population is defined as the fraction of neonlike ions in a particular excited state.) We then calculated the gain coefficient of a Doppler broadened line from the expression

$$\alpha=\frac{\lambda^3A}{8\pi}\left(\frac{M_i}{2\pi kT_i}\right)^{1/2}y_{UL}n_{\rm Ne}\ ,$$

where λ and A are the wavelength and radiative decay rate, respectively, of the lasing transition, M_i and T_i are the mass and temperature, respectively, of the ion, and $n_{\rm Ne}$ is the total density of Ne-like ions. We have taken the plasma parameters for our calculations to be those characteristic of the neonlike selenium laser plasma:[4] $n_e=5\times10^{20}$ cm^{-3}, $T_e=1000$ eV, $T_i=400$ eV, and $n_{\rm Ne}=5\times10^{18}$ cm^{-3}. We assumed that the fraction of Ne-like ions is 0.25.

The importance of dielectronic recombination in determining the kinetics of the Ne-like excited states, of course, depends on the relative numbers of F- and Ne-like ions present. Models which include the F-like ionization stage will predict a F- to Ne-like ratio ($n_{\rm F}/n_{\rm Ne}$). In our case, this ratio cannot be expected to be entirely correct since we have made no attempt to accurately determine the ionization balance. However, it should be noted that we found a reasonable $n_{\rm F}/n_{\rm Ne}$ ratio, i.e., our value agreed within a factor of 2 with that obtained from steady-state ionization balance calculations[22] for $1\times10^{20}<n_e<8\times10^{21}$ cm^{-3}. We also varied $n_{\rm F}/n_{\rm Ne}$ artificially between 0.1 and 3.0 by introducing a multiplier for the collisional ionization rates.

The relative effects of cascade and dielectronic recombination on the inversion kinetics are shown in Table II, where we compare results for models with different processes included. (For this comparison the collisional ionization multiplier is taken to be one.) Column 1 shows results obtained with a model containing only the Ne-like ground state and the $n=3$ excited states (Ne37). In this case all excited-state population comes directly or indirectly from the Ne-like ground state. Column 2 shows results for

TABLE II. Effects of $n=4$-3 cascades and dielectronic recombination on gain coefficients (cm^{-1}) of some $2p^53p$-$2p^53s$ transitions in neonlike selenium.

Transition	Wavelength (Å)	$n_{\rm F}/n_{\rm Ne}$:	Ne37 0.0	Ne89 0.0	Model Ne37/F3 0.8	Ne89/F3 0.6
$J=0$-1	182.4		14.3	13.4	13.9	13.0
$J=2$-1	209.8		11.4	13.0	13.8	15.3
$J=2$-1	206.4		9.7	11.5	12.1	13.6

a model containing the $n=2$, 3, and 4 Ne-like states (Ne89). A comparison of columns 1 and 2 shows that collisional and radiative cascades alone increase the gain coefficient of the $J=2$-1 transitions by 15%–20%. Column 3 shows the results for a model (Ne37/F3) which adds the three F-like ground states to the Ne37 model, as well as ionization and recombination between the two ion stages. In this case, dielectronic recombination rates are summed over the ($3l3l'$) excited states only. By comparing columns 1 and 3, we see that, even for a relatively small value of $n_{\rm F}/n_{\rm Ne}$, dielectronic recombination alone increases the gain coefficient of the $J=2$-1 transitions by about 20%–25%, and slightly decreases the $J=0$-1 gain. Finally, column 4 gives results for a model (Ne89/F3), which includes both dielectronic recombination through the ($3l3l'$) and ($3l4l'$) Ne-like doubly excited states, and cascades from the $n=4$ Ne-like excited states. In this case, the gain coefficient of the $J=2$-1 transitions increases by 35%–40% relative to the Ne37 case. The $J=0$-1 transition decreases by about 10%, mostly due to an increase in population for the $2p^53s$ lower level.

Figure 1 shows the effects of varying $n_{\rm F}/n_{\rm Ne}$ on the gain coefficients of the three transitions. When the number of

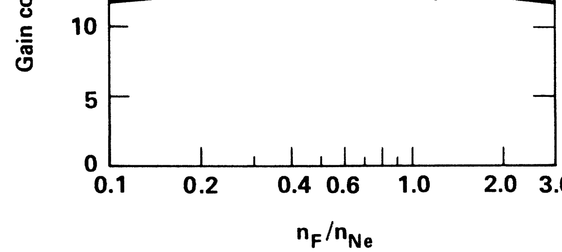

FIG. 1. Gain coefficient (in cm^{-1}) of the three relevant $2p^53p$-$2p^53s$ transitions in Ne-like selenium as a function of $n_{\rm F}/n_{\rm Ne}$. 1, $J=0$-1 transition at 182.4 Å; 2, $J=2$-1 transition at 209.8 Å; 3, $J=2$-1 transition at 206.4 Å.

F-like ions is small, the $J=0$-1 transition has about the same gain coefficient as the larger of the two $J=2$-1 transitions. As n_F/n_{Ne} increases the gains of the $J=2$-1 transitions increase, and that of the $J=0$-1 decreases slightly. This is because dielectronic recombination favors states with high statistical weight. Thus, the $J=2$ states receive substantial population directly from the F-like ground state. The recombination rate into the $J=0$ state is smaller, and does not compete with the very large collisional excitation rate from the Ne-like ground state. The small drop in gain for the $J=0$-1 transition occurs because the $2p^53s(J=1)$ lower level is also being populated by dielectronic recombination. We see from this figure that n_F/n_{Ne} must be very large for the gain of the $J=0$-1 transition to be significantly reduced.

Determining the actual ionization balance of the x-ray laser plasma is an extremely difficult problem because it is not in steady state, and there are temporal and spatial gradients in the plasma during the amplified spontaneous emission. We have no direct measurement of the ionization balance, and the predictions of different codes do not agree. Based on qualitative information provided by time-resolved, $n=2$-3 x-ray spectra recorded during the experiments,[1,4] we believe that n_F/n_{Ne} is between 0.5 and 2.0.

Finally, Table III shows the results of varying the electron temperature in the calculation. We see that the relative inversion densities of the $J=2$-1 transitions rise substantially compared to that of the $J=0$-1 transition as the temperature rises. This is due primarily to the fact that the proportion of F-like ions rises rapidly with temperature.

These simple calculations cannot be expected to produce quantitative predictions for the gain coefficients. However, they clearly demonstrate that a model containing only the $n=2$ and $n=3$ states of Ne-like selenium is inadequate to calculate accurate level populations for the Ne-like excited states. The $2p^53p$ $(J=0)$ state is populated almost entirely by the rapid collisional excitation from the Ne-like ground state, and its population is not significantly affected by cascade from higher levels or by dielectronic recombination.

TABLE III. Effect of electron temperature on the relative inversion density (10^{-3} cm^{-3}) for some $2p^53p$-$2p^53s$ transitions of neonlike selenium.

T_e (eV)	Transition $J=0$-1 (182.4 Å)	$J=2$-1 (209.8 Å)	$J=2$-1 (206.4 Å)
500	0.65	0.71	1.4
1000	2.5	3.1	5.7
2000	4.2	6.4	11

However, the $2p^53p$ $(J=2)$ upper laser states [and, to a lesser extent, the $2p^53s$ $(J=1)$ lower states] *are* populated by collisional cascade from more highly excited states of the Ne-like ion (e.g., the $2s^22p^53d$, $2s2p^63p$, and $2s^22p^54l$ states) and by dielectronic recombination from the F-like ground states, as well as by direct collisional excitation from the Ne-like ground state. As a result, the gain coefficients of the transitions involving these states are sensitive to the ionization balance, i.e., n_F/n_{Ne}.

Finally, it is clear that dielectronic recombination alone does not explain the experimental result[1] that the $J=0$-1 transition did not exhibit significant amplification. The inversion density of this transition is reduced only slightly by this mechanism for reasonable values of n_F/n_{Ne}. In order to explain the absence of this line, a mechanism is required which either reduces the population of the $2p^53p$ $(J=0)$ upper state or absorbs the emitted photons.

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[1]D. L. Matthews *et al.*, Phys. Rev. Lett. **54**, 110 (1985).
[2]A. V. Vinogradov and V. N. Shlyaptsev, Kvantovaya Elektron. (Moscow) **7**, 1319 (1980) [Sov. J. Quantum Electron. **10**, 754 (1980)], and references therein.
[3]U. Feldman, A. K. Bhatia, and S. Suckewer, J. Appl. Phys. **54**, 2188 (1983); U. Feldman, J. F. Seely, and A. K. Bhatia, *ibid.* **56**, 2475 (1984).
[4]M. D. Rosen *et al.*, Phys. Rev. Lett. **54**, 106 (1985).
[5]W. Eissner, M. Jones, and H. Nussbaumer, Comput. Phys. Commun. **8**, 270 (1974).
[6]P. L. Hagelstein, Ph. D. thesis, Lawrence Livermore National Laboratory Report No. UCRL-53100, 1981 (unpublished); P. L. Hagelstein and R. Jung (unpublished).
[7]W. Eissner and M. J. Seaton, J. Phys. B **5**, 2187 (1972).
[8]H. E. Saraph, Comput. Phys. Commun. **1**, 232 (1970); **3**, 256 (1972).
[9]A. Burgess and H. P. Summers, Mon. Not. R. Astron. Soc. **174**, 345 (1976).
[10]D. L. Moores, L. B. Golden, and D. H. Sampson, J. Phys. B **13**, 385 (1980); L. B. Golden and D. H. Sampson, *ibid.* **13**, 2645 (1980), and references therein.
[11]J. C. Weisheit, C. B. Tarter, J. H. Scofield, and L. M. Richards, J. Quant. Spectrosc. Radiat. Transfer **16**, 659 (1976).
[12]M. H. Chen, Phys. Rev. A **33**, 994 (1986).
[13]M. H. Chen, Phys. Rev. A **31**, 1449 (1985).
[14]M. H. Chen (unpublished).
[15]J. P. Apruzese, J. Davis, M. Blaha, P. C. Kepple, and V. L. Jacobs, Phys. Rev. Lett. **55**, 1877 (1985).
[16]V. L. Jacobs, J. Davis, J. E. Rogerson, M. Blaha, J. Cain, and M. Davis, Astrophys. J. **239**, 1119 (1980).
[17]I. L. Beigman, L. A. Vainstein, and B. N. Chichkov, Zh. Eksp. Teor. Fiz. **80**, 964 (1981) [Sov. Phys. JETP **53**, 490 (1981)].
[18]L. J. Roszman (unpublished).
[19]L. J. Roszman, Phys. Rev. A **20**, 673 (1979).
[20]J. N. Gau, Y. Hahn, and J. A. Retter, J. Quant. Spectrosc. Radiat. Transfer **23**, 147 (1980).
[21]S. Dalhed, J. Nilsen, and P. Hagelstein, Phys. Rev. A **33**, 264 (1986).
[22]Y. T. Lee (unpublished).