

## Electron collisional excitation in F-like selenium

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Electron collisional excitation of the low-lying  $1s^2 2s^2 2p^5$  and  $1s^2 2s 2p^6$  states of F-like selenium to the singly excited  $M$ -shell states is studied using a relativistic distorted-wave model and multiconfigurational relativistic Hartree-Fock bound states. Results are presented for all 2-3 transitions from the low-lying  $2s^2 2p^5 \ ^2P_{3/2}$  and  $\ ^2P_{1/2}$  levels, and also the  $2s 2p^6 \ ^2S_{1/2}$  level. We find a number of strong dipole-allowed  $2p$ - $3d$  cross sections with peak values near threshold in excess of  $10^{-20} \text{ cm}^2$ , and derive Gaunt factors which are in good agreement with values used in the literature (0.15–0.20) for most strong transitions. Very strong monopole  $2p$ - $3p$  excitation cross sections have been important in soft-x-ray laser theory, and are found to be as strong as the largest dipole-allowed cross sections for F-like selenium. Theoretical output powers for the strong lines of the  $2p$ - $3s$ ,  $2p$ - $3d$ , and  $2s$ - $3p$  transition arrays are computed and presented for plasma conditions of  $N_e = 3 \times 10^{20} \text{ cm}^{-3}$  and  $T_e = 1.0 \text{ keV}$ . These results are compared in detail for proportionality against  $gf$  values for each array separately, as a test of how well line intensities might be judged from  $gf$  values in the absence of detailed theoretical intensity results. We find that for the  $2p$ - $3s$  and  $2p$ - $3d$  arrays, the intensities are in fair agreement with  $gf$  values within the array, while the agreement is much poorer in the case of the  $2s$ - $3p$  array. The two weaker arrays  $2p$ - $3s$  and  $2s$ - $3p$  are found to radiate more per unit  $gf$  than the  $2p$ - $3d$  transition array, in agreement with earlier observations in the Ne-like sequence. Theoretical line positions are tabulated for all strong 2-3 lines, and found to be in good agreement with experimental results for most strong transitions. Gains on the 3-3 transitions in between 2 and  $4 \text{ cm}^{-1}$  are predicted for four lines under the plasma conditions quoted. Such conditions are similar to those of the recent extreme-uv laser experiments at the Lawrence Livermore National Laboratory, yet no F-like 3-3 amplification has yet been observed for F-like transitions. The discrepancy is currently a mystery.

### I. INTRODUCTION

More than a decade after the initial proposals suggesting electron collisional excitation as a mechanism to pump lasers in the extreme-uv (xuv) and soft-x-ray regimes,<sup>1–6</sup> and following extensive examination and modeling efforts,<sup>7–16</sup> theoretical and experimental efforts at the Lawrence Livermore National Laboratory lead to the experimental observation of  $3p$ - $3s$  amplification in Ne-like selenium at 206 and 209 Å (Ref. 17) using a laser-driven exploding-foil target.<sup>18</sup> The early proposals suggested that the dominant excitation mechanism in pumping this type of laser should be direct  $2p$ - $3p$  electron collisional excitation by ambient hot thermal electrons. While there is still little reason to believe that such direct excitation does not occur (this direct excitation drives a line near 182 Å with high theoretical gain), it is currently thought that recombination plays the dominant role in the development of population inversions of the highest-gain lines at 206 and 209 Å. The 182-Å line is observed routinely in recent experiments. However, the experimentally determined gain is still less than that predicted at the time of optimum target conditions for both gain and plasma uniformity.

Although direct experimental determination of sequence abundances and overall ionization balance is not yet in hand, our theoretical model predicts very substantial F-like populations, and such a model is not incon-

sistent with the strong 3-2  $L$ -shell spectra observed experimentally. In order to obtain comparisons between the detailed kinetics model of selenium and the observed experimental spectra, it has been necessary to develop a kinetics model of the complex F-like ion. Our earlier comparisons of this model against low-resolution 3-2 spectra taken from an exploding-foil target were presented in Ref. 18, and further comparisons will be published elsewhere.

Having developed a kinetics model for F-like selenium, it is natural to enquire whether  $3p$ - $3s$  population inversions occur among the singly excited F-like levels. The question becomes even more relevant, given the comments above concerning the large abundance (40–50 %) of F-like ions calculated to be present during much of the lifetime of the plasma. For example, if theoretically large gains are predicted for F-like  $3p$ - $3s$  transitions, and if it is established by 3-2 x-ray spectroscopy that substantial F-like population is present, then either F-like  $3p$ - $3s$  amplification should be observed experimentally, or else there must be an excellent reason as to why such amplification is absent.

The situation at present appears to be that a large F-like population exists over much of the plasma's lifetime and spatial extent, and that theoretically the resulting gain on F-like  $3p$ - $3s$  transitions should be high enough to be manifestly obvious, and that no such amplification has yet been observed experimentally.<sup>19</sup> Precisely why this is true is not well understood, although candidate explanations

include possible resonant line absorption and plasma turbulence effects.

One further anomaly which may perhaps provoke some thought is that when the theoretical xuv spectrum is produced, using the results of the plasma hydrodynamic simulations and detailed atomic physics models, the resulting spectra in the vicinity of the 3-3 lines of F-like to Na-like selenium are calculated to be very rich, due to contributions from many Ne-like and F-like 3-3 lines. Many lines are calculated to be fully as bright, when viewed on-axis, as the dominant Na-like selenium resonance lines, including lines which are inverted as well as noninverted lossy lines. Once again the experimental spectra are at variance with the theoretical modeling results, for reasons which remain poorly understood. Many of the lines which seem to be missing have optical depths (or gain lengths) of near or less than unity axially (the laser plasmas have dimensions of one to several centimeters axially, and of roughly 100  $\mu\text{m}$  transversely).

The problems with missing lines in the axial spectra have eluded quantitative explanation for several years now, and we shall not explain it in the present paper. One cannot help but wonder if possibly some nonselective absorption or scattering mechanism is not present which has the effect of simply removing all of the relatively weak lines of low optical depth from the spectrum. If scattering due to small-scale density fluctuations were present with a scattering coefficient of 5–10  $\text{cm}^{-1}$  at early times, and much less later on, then one might envision a scenario wherein the Ne-like lines emerge late in time (400–500 psec after the peak of the 450-psec optical pulse) where little or no F-like gain remains. Theoretically, the Ne-like gain outlasts the F-like gain considerably. Due to the high theoretical F-like gain, which is considered near the end of the present paper, one seems pressed towards concluding that something is not quite right with axial xuv beam propagation in the exploding-foil plasma while the optical laser pulse is present.

The purpose of the present paper is to examine the electron collisional excitation process in F-like selenium, to begin exploring the 2-3 x-ray spectra theoretically, and to discuss briefly the development of  $3p$ - $3s$  population inversions and gain under conditions relevant to the selenium exploding-foil amplifier. We shall not explore radiative emission in the 3-3 spectra in this paper, although such a study is now within our grasp theoretically.

In Sec. II we discuss theoretical methods used in our structure calculations, and present results for wavelengths and oscillator strengths for the strong 2-3 transitions. The relativistic distorted-wave model is reviewed briefly in Sec. III, and the 2-3 collisional excitation cross sections are tabulated and discussed in Sec. IV. Section V is devoted to an investigation of theoretical output powers of the 2-3 transitions, and gain predictions for  $3p$ - $3s$  candidate laser transitions are explored briefly in Sec. VI.

## II. STRUCTURE CALCULATIONS, WAVELENGTHS, AND OSCILLATOR STRENGTHS

The structure calculations described here are based on multiconfigurational relativistic Hartree-Fock wave func-

tions with the Breit interaction included in the  $\omega=0$  limit. We have employed YODA, an atomic physics package of Hagelstein and Jung,<sup>20</sup> which calculates energy levels, oscillator strengths, photoionization cross sections, collisional cross sections, and Auger rates for a restricted angular-momentum coupling scheme. YODA has been used extensively for the support of non-local-thermal-equilibrium (NLTE) kinetics model development at Lawrence Livermore National Laboratory (LLNL) for several years, and was employed in the construction of models used for the design efforts described in Ref. 18.

The multiconfigurational wave functions which serve as the starting point for the distorted-wave collisional calculations were computed using a single set of basis orbitals ( $1s_{1/2}, 2s_{1/2}, \dots, 3d_{5/2}$ ) computed from a single spherically averaged relativistic Hartree-Fock self-consistent field calculation with fractional orbital occupation. The specific configuration used in this calculation was the fictitious configuration with occupation numbers given as follows:

$$\begin{aligned} & 1s_{1/2}, 2.00 ; 2s_{1/2}, 1.27 ; 2p_{1/2}, 1.27 ; \\ & 2p_{3/2}, 3.82 ; 3s_{1/2}-3d_{5/2}, 0.127 . \end{aligned} \quad (2.1)$$

The use of these particular fractional occupational numbers deserves a modicum of comment. The total  $L$ -shell occupation for the ground states  $1s^2 2s^2 2p^5$  and  $1s^2 2s 2p^6$  is 7, and for the singly excited  $M$ -shell states is 6. If our structure calculations were based on nonorthogonal single-electron wave functions, then we should do well to include  $L$ -shell orbitals computed both with 6 and 7  $L$ -shell electrons present, and include configurations corresponding to the appropriate single-electron orbitals. This approach would be able to include relaxation effects more accurately than the present model. If the orbitals must be orthonormal, then a choice must be made as to precisely which orbitals are to be used, as the final answers will show some sensitivity to the choice. One possibility is to put 6.5 electrons in the  $L$  shell, and split the remaining half electron between the  $M$ -shell states, giving 0.10 electrons per  $M$ -shell orbital. The above choice of occupation numbers gives 6.36 electrons in the  $L$  shell and 0.64 electrons in the  $M$  shell, which is not so far from a 50%-50% split. Beyond this, it makes little sense to discuss the algorithm by which YODA computes default fractional occupations, and our results will be slightly biased towards the states with more  $2s$  and  $2p_{1/2}$  vacancies, although effects on cross sections are no more than a few percent.

The multiconfigurational structure calculation includes 113 states, which is the total for all  $L$ -shell and  $M$ -shell ground and singly excited states of F-like selenium. In order to check whether a larger configuration-interaction (CI) calculation would change the results given the basis orbitals, we calculated energy levels for 1622 states which included the 113 low-lying states as well as all  $3l$ / $3l'$  doubly-excited states. Although some shift in optimized total energies was observed, relative level shifts were about 0.1 eV. In comparing against experimentally determined wavelengths (which we shall discuss shortly), we found a systematic shift of about 1 eV for most 2-3 transitions, and this shift seemed most likely to be due to correlation























$2s^22p^{5/2}P_{1/2}$  and  $2s2p^{6/2}S_{1/2}$  states in Tables VII and VIII. Cross sections in excess of  $10^{-20}\text{ cm}^2$  are observed for near-threshold excitation of two dipole-allowed transitions with large  $f$  values, and these are (2-57) and (2-58) with oscillator strengths of 0.984 and 0.770, respectively.

In short-wavelength-laser research, electron monopole transitions play a key role (at least theoretically) in the production of  $\Delta n = 0$  population inversions. For example, in neonlike selenium, the excitation of a  $2p$  electron from the ground state  $2s^22p^{6/1}S_0$  to  $2s^22p^{5/3}p J=0$  states is calculated to drive population inversions on a number of  $3p-3s$  transitions. The strongest of these transitions is near 182 Å, and amplification has been observed now on this line. The mechanism by which gain is produced on this line is believed to be direct excitation, in contrast to that of the 206- and 209-Å lines, which are believed to be populated predominantly through recombination channels. The question might be asked as to whether analogs of this strong monopole excitation exist in F-like selenium, and, if so, whether collisionally pumped  $3p-3s$  transitions in F-like selenium might be observed.

The monopole transitions in question would occur from the  $2s^22p^{5/2}P_{3/2}$  level to states of the form  $2s^22p^{4/3}p J=3/2$ . There are eight such states (levels 9, 17, 18, 20, 24, 27, 33, and 46). Of these states, one of them is amply favored in terms of collisional strength, and that is level 33 ( $[2s^22p_{1/2}2p_{3/2}^3]_23p_{1/2} J=3/2$ ) which has a near-threshold excitation cross section of  $1.7 \times 10^{-20}\text{ cm}^2$  at 200 eV above threshold. This cross section is larger than the dipole cross section for (1-56) of  $1.0 \times 10^{-20}\text{ cm}^2$  discussed above. One should expect level 33 to be the upper  $3p$  state of a monopole-excited  $3p-3s$  inversion driven from the ground state  $2P_{3/2}$ .

Six states of the form  $2s^22p^{4/3}p J=1/2$  occur which might serve as candidates for strong monopole excitation, including levels 13, 16, 19, 23, 38, and 45. In this case, the strong monopole excitation is split between two levels, namely levels 38 ( $[2s^22p_{1/2}2p_{3/2}^3]_23p_{3/2} J=1/2$ ) and 45 ( $[2s^22p_{3/2}^4]_03p_{1/2} J=1/2$ ), for which the near-threshold excitation cross sections are  $1.9 \times 10^{-21}$  and  $4.3 \times 10^{-21}\text{ cm}^2$ , respectively, from level 2. In terms of candidate monopole excited  $3p-3s$  laser transitions, one might look for levels 38 and 45 as potential upper laser states. However, the quantity which is more important in terms of driving a population inversion than the excitation cross section is the product of the lower-state statistical weight  $g_i$  and the cross section  $\sigma_{ij}(E)$ . This product for excitation to levels 38 and 45 is less than one-fifth of that for the transition (1-33) considered above. One would expect that level 33 would be a much better candidate upper laser state than either levels 38 or 45, based on this simple consideration alone.

One might enquire about monopole excitation from the third F-like level (the  $2s2p^{6/2}S_{1/2}$ ), given the set of cross sections of Table VIII. From the point of view of xuv lasers and  $3p-3s$  inversions, the monopole excitation process is not of particular interest, as the upper  $3p$  electron will be able to radiatively decay rapidly back to the  $L$  shell, by virtue of the initial  $2s$  hole. But it is this difference which makes it interesting in terms of dielectronic recombination physics. The reason for this can be seen

from a consideration of quantum-defect theory, from which a linear relation exists between the threshold excitation cross section and the dielectronic capture rate into states corresponding to the excited state plus an additional highly excited Rydberg electron. If one starts from the ground state  $2s^22p^6$  Ne-like level, then the capture process is dominated by  $2p-3d$  excitation leading to Na-like states of the form  $2s^22p^53dn$ , and these states may be stabilized by  $3d-2p$  radiative decay. Although there is a large monopole  $2p-3p$  excitation cross section, and a correspondingly large capture rate into  $2s^22p^53pn$  doubly excited levels, no stabilizing  $3p-2s$  radiative decays are possible because the  $2s$  shell is already fully occupied.

The situation starting from the F-like  $2s^22p^{5/2}P$  levels is similar, in that although substantial capture occurs into the  $2s^22p^43pn$  doubly excited neonlike levels, no stabilizing  $3p-2s$  decay may occur. From the  $2S_{1/2}$  state, however, the capture into  $2s2p^53pn$  levels can be followed by  $3p-2s$  radiative decay, and hence a new capture channel is opened. Based on this, one might expect the dielectronic recombination rate coefficients from the  $2S_{1/2}$  level to be larger than the recombination rate coefficients from the  $2P$  levels, simply due to the existence of an additional strong recombination channel.

The largest collisional excitation cross section from the  $2s2p^{6/2}S_{1/2}$  level is a  $2p_{1/2}-3p_{1/2}$  monopole transition to level 99 ( $[2s2p_{1/2}2p_{3/2}^4]_13p_{1/2} J=1/2$ ), with a near-threshold cross section of  $5.7 \times 10^{-21}\text{ cm}^2$ . This cross section is comparable in magnitude to that of any dipole cross section from the  $2S_{1/2}$  level.

## V. THEORETICAL INTENSITIES OF F-LIKE 2-3 LINES

From the 2-3 electron collisional cross sections presented in Sec. IV, one can compute theoretical values for absolute line emission. In Tables X–XII, we present results for the strong 2-3 transitions of F-like selenium under conditions similar to those found in the selenium exploding-foil laser target when the plasma is thought to be near optimal conditions for the development of xuv gain. The electron density is taken to be  $3.0 \times 10^{20}\text{ cm}^{-3}$  and the electron temperature assumed is 1.0 keV. The calculation was carried out using our current selenium xuv laser design model, which incorporates the F-like collisional data which is the subject of the present work, as well as the detailed Ne-like cross sections presented in Ref. 48. The model includes very detailed atomic physics for the F-like through Na-like sequences, and multicore “hydrogenic” physics in the neighboring sequences, computed originally using Morgan’s XATOM code,<sup>49</sup> and upgraded substantially over the years with improved rate coefficients from YODA<sup>20</sup> and a variety of other sources. The dielectronic recombination model is based on Refs. 50 and 51, and has been extended to all sequences modeled in a detailed and consistent fashion (Hagelstein). Direct dielectronic recombination into the F-like excited states is included in the intensity results under discussion. This design model will be documented at greater length elsewhere.







## VII. SUMMARY AND CONCLUSIONS

We have analyzed theoretically the 2-3 electron collisional process in F-like selenium using a multiconfigurational relativistic distorted-wave model, and have tabulated a complete set of 2-3 cross sections for excitation from the  $2s^22p^{5/2}P_{3/2}$ , and  $2s^22p^{5/2}P_{1/2}$ , and  $2s2p^{6/2}S_{1/2}$  levels. The cross sections have been used to derive Gaunt factors as a check on the accuracy of empirical methods in highly stripped F-like systems, and have found agreement with Gaunt factor values in the literature (0.15–0.20) for many of the strong  $2p$ - $3d$  transitions. Dipole-allowed and monopole electron collisions have the largest theoretical cross sections. A very strong monopole excitation cross section is found for the  $2s^22p^{5/2}P_{3/2} - [2s^22p_{1/2}2p_{3/2}]_23p_{1/2} J = 3/2$  transition, with a value of  $1.7 \times 10^{-20} \text{ cm}^2$  at 200 eV above threshold.

We have calculated wavelengths and intensities of the 2-3 F-like selenium transitions, and have examined the use of  $gf$  values as approximations to line intensity in spectroscopic analysis. We have compared our theoretical wavelengths with the experimental results of Gorden *et al.*<sup>25</sup> and have found good agreement for many identified transitions. Some of the weaker lines are in disagreement with our results, and seem to be inconsistent with the accurate  $2P_{1/2}$ - $2P_{3/2}$  splitting of Edlen.<sup>21</sup> We have tabulated predicted output powers for all strong  $2p$ - $3s$ ,  $2p$ - $3d$ , and  $2s$ - $3p$  transitions, and compared in detail the output powers relative to the line strength  $gf$ . We found fair agreement within the transition arrays  $2p$ - $3s$  and  $2p$ - $3d$ , and observed a systematic reduction of output power relative to  $gf$  for lines originating from the  $2S_{1/2}$  state. Agreement was poorer for  $2s$ - $3p$  lines. We noted that the two weaker arrays ( $2p$ - $3s$  and  $2s$ - $3p$ ) are expected to radiate more per unit  $gf$  than the strong  $2p$ - $3d$ , in qualitative agreement with early astrophysical observations in Ne-like Fe.<sup>52</sup>

The gains on the 3-3 transitions in F-like selenium have been examined briefly, and four transitions are predicted to have gains between 2 and 4  $\text{cm}^{-1}$  under conditions thought to occur near the optimum for LLNL selenium exploding-foil laser targets. Direct collisional excitation from the ground states is calculated to be the principal excitation mechanism for most of the high-gain lines. We have tabulated theoretical wavelengths and  $gf$  values for these candidate laser transitions.

The exploding-foil selenium laser experiments yield 3-2 spectra in which F-like emission is manifestly obvious, but in which there is no discernible 3-3 emission. Detailed comparisons between the present theoretical results and experimental observations will be presented elsewhere; however, the model seems to be in agreement on the 2-3

spectra. The absence of observed gain on the F-like 3-3 lines, coupled with predictions presented here and found in the design and analysis simulations of Rosen, results in a mystery.

Where are the missing lines? We simply do not know whether our theoretical models are incorrect in some fundamental way, or whether some interesting physics not modeled is at work. Possible explanations include resonant line absorption or axial beam scattering by density fluctuations. One might propose a scenario in which very strong scattering occurs while the incident optical laser pulse is present (for which there is currently no direct evidence), and which relaxes some time after the optical laser intensity has fallen. The time history of the F-like gain is such that it falls off faster than the Ne-like lines, which tend to hang up in our present model, hence one might propose that the Ne-like lines themselves emerge only very late in time, thereby providing a solution to our mystery. Forthcoming absolutely timed measurements will help to resolve these matters in the near future.

The possibility of designing a laser which amplifies F-like 3-3 transitions is of interest. Currently, the F-like 3-3 spectroscopy is not well understood in selenium, and the observation of gain on a number of easily identifiable  $3s$ - $3p$  or  $3p$ - $3d$  transitions would make a significant difference in our understanding of a very complex spectrum. Such a laser could readily be designed with current techniques, and tested in experiments similar to ongoing selenium exploding-foil work.

*Note added in proof.* Measurements of the absolute timing of the selenium laser emission at 206 and 209 Å have been completed as this paper was being proofed. The measurements indicate that the emission occurs early with respect to the incident laser pulse, and therefore the speculations concerning the timing of the laser pulse which appear in the text are incorrect.

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