Possibilities for achieving X-ray lasing action by use of
high-order multiphoton processes

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Abstract

We consider some possible mechanisms for producing gain in the 10 nm spectral region. They involve the creation of a population inversion in a confined plasma column by selective excitation of multicharged ions via absorption of many (>10) ultraviolet photons. Specific treatment is made of Kr-like ions pumped by a KrF excimer laser.

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1. INTRODUCTION

Recent experimental studies have demonstrated that an atom can absorb many photons with high probability under readily achievable conditions of laser intensity. Boyer et al.¹ report cases of multiple ionization of atoms by ArF laser radiation ($\lambda=193$ nm) at intensities of 10^{14} - 10^{15} W/cm². The minimal number of photons which must be absorbed to produce the observed stages of ionization is as high as 49. Lomare et al.²⁻³ utilize the fundamental ($\lambda=1064$ nm) and harmonic radiation of a Nd-YAG laser at lower power levels (10^{12} - 10^{13} W/cm²) to observe ionization of noble gases requiring up to 68 photons. Other ionization experiments involving absorption of 10 or more infrared or visible photons are reported in Refs. 4-7. The specific mechanisms by which such high-order multiphoton processes occur are not well understood. In most cases which have been studied, no resonant intermediate state can be identified in the transition from neutral atom to ion.

The above-cited studies all observe ionization, and up to the present there do not appear to have been any experiments which have seen discrete excitation by absorption of large numbers of photons. This may be due in part to the fact that high intensity laser sources have limited ranges of tunability, and that most neutral atoms cannot absorb more than about five visible or UV photons without ionizing. However, there are many cases known in which significant population of an excited state is produced by absorption of a few photons. One such case is particularly relevant to the topic of this paper. Boyer et al.¹ have observed stimulated emission at 93 nm in Kr gas irradiated by intense ArF laser light. The mechanism involved appears to be as follows: ground state Kr, with electronic configuration $4s^2 4p^6$, is excited

to a $4s4p^64d$ state by absorption of four 193 nm photons:

This establishes a population inversion of the $4s4p^64d$ state with respect to the $4s^24p^54d$ state, and stimulated emission occurs on a transition

In this paper, we consider the prospects for using similar schemes to produce population inversions in ions leading to gain at shorter wavelengths. The basic idea is to excite either one subvalence electron (as in Eq. 1), or two valence electrons, of a highly ionized atom. The excitation of two valence electrons is especially attractive for the development of an X-ray laser because of the faster progression to shorter wavelength with increasing atomic number Z compared to the case of excitation of one subvalence electron. The radiative transition then occurs as in Eq. (2), or by radiation of one of the two excited electrons. We deal specifically with the case of Kr-like ions excited by a KrF excimer laser ($\lambda \sim 250$ nm). This case is not necessarily an optimal one, but it is one with a good deal of spectroscopic data available for preliminary analysis, and it is also suited to our present experimental program utilizing a powerful picosecond laser.⁸

Evaluation of the feasibility of the multiphoton approach requires consideration of several aspects of atomic and plasma physics. These are dealt with in subsequent sections. Section II treats the appropriate energy level structure of ions in the Kr isoelectric sequence, from Kr to Cd^{+12} .

Where possible, we utilize experimental data, but since this is rather sparse for highly-excited states of multicharged ions, we rely extensively on scaled Hartree-Fock calculations. Estimates are made of the oscillator strengths for the prospective super-radiant transitions. In Section III we discuss the multiphoton absorption process and the possible effects of a very strong laser field. There is an obvious need for more experimental and theoretical work on these subjects, and we believe this is the source of the largest uncertainties in our considerations. The important points of plasma physics, such as the production and characterization of the active laser medium, are deferred to a future paper. Their treatment will be based primarily on a present experimental approach, which involves production of a plasma by irradiation of a solid target by a kilojoule CO₂ laser, the plasma being confined by a 10 T magnetic field. Previous experiments on this apparatus have demonstrated one-pass gain of up to 6.5 at $\lambda = 18.2$ nm in a rapidly recombining plasma.⁹ Section IV summarizes our conclusions.

2. ENERGY LEVELS AND RADIATIVE TRANSITIONS IN THE Kr ISOELECTRONIC SEQUENCE

We shall consider processes of the type

$$X + N\gamma_L + X^{**} + X^* + \gamma, \quad (3)$$

where X is an ion in the ground state $4s^2 4p^6$; γ_L is a photon from the KrF laser (248 nm); X^{**} is a core- or multiply-excited state, specifically of either the $4s 4p^6 5p$ or $4s^2 4p^4 5s^2$ configurations; X^* is an excited state, and γ is a soft x-ray photon. Whatever specific states are chosen, the energy of γ will increase with the stage of ionization of X . We shall deal here with the Kr isoelectronic sequence up to Ca^{12+} .

Most of the experimental spectroscopic information on this sequence comes from the studies of Reader and coworkers.¹⁰⁻¹³ Their work deals with spectra in the sequence between Rb^+ and Mo^{6+} . The next member of the sequence is radioactive Tc^{7+} , which has never been investigated. The next five members of the sequence have been studied, though not to the same extent as Kr-Mo . Even-Zohar and Fraenkel¹⁴ have observed some lines in the spectra of Ru^{8+} , Rh^{9+} , and Pd^{10+} ; the Dublin group has looked at Ag^{11+} (Ref. 15) and Cd^{12+} (Ref. 16). There do not appear to have been any studies of higher members of the sequence. As the quality of experimental data (and of our present analysis) differs considerably between the core- and multiply-excited transitions, we now discuss them separately.

A. Transitions Involving 4s Excitation

We consider excitation of states of the $4s4p^65p$ configuration. Our interest in these states is due, first, to their being accessible by five-photon absorption by Kr at wavelengths within the tuning range of the KrF excimer laser, and second, to the existence of good spectroscopic data on them for several members of the isoelectronic sequence. However, there may be advantages associated with other states of the type $4s4p^6n\ell$, and some of these will be discussed briefly. We discuss first the ionization limits $4s4p^6$ and $4s^24p^5$, which determine both the maximum energy required to excite a 4s electron to a Rydberg state and the maximum photon energy of prospective lasing transitions. The three energies we shall consider as a function of ionization stage are: i) the ionization energy $4s^24p^6-4s^24p^5\ ^2P_{3/2}$, i.e., the first ionization limit; ii) the interval $4s^24p^5\ ^2P_{3/2}-4s^24p^5\ ^2P_{1/2}$; and iii) the ionization energy $4s^24p^6-4s4p^6$.

The first ionization energy is of course well-known¹⁷ in Kr, and has been established¹⁰ with an uncertainty of 200 cm⁻¹ or less for Rb⁺ through Mo⁶⁺ by means of quantum defect analysis. The basis for a quantum defect analysis is less well founded in Ru⁸⁺-Pd¹⁰⁺, and apparently nonexistent (at present) in Ag¹¹⁺ and Cd¹²⁺. We therefore have adopted an extrapolation procedure, which appears to be quite reliable and which may find application in other problems of high-ionization spectra. We compute¹⁸ a Hartree-Fock ionization energy I_{HF} from the difference of Hartree-Fock total energies of the 4s²4p⁶ and 4s²4p⁵ configurations (plus a suitable adjustment for the spin-orbit interaction, as described in the caption to Fig. 1). This is plotted vs the experimental ionization energy for the first seven members of the Kr sequence in Fig. 1. It is seen that the experimental and Hartree-Fock values approximately satisfy a linear relationship, which a least-squares fit finds to be

$$I_{\text{expt}} = 6743.4 \text{ cm}^{-1} + 1.007833 I_{\text{HF}}. \quad (4)$$

We do not fully understand why a linear relationship of this type should hold so well along the isoelectronic sequence. Its form is reminiscent of some of the results of Z^{-1} perturbation theory,¹⁹ which, however, are based on conditions that do not apply here. We tend to think of the intercept on the right hand side of Eq. (4) as a constant electron correlation energy, and the non-unity scaling factor as providing relativistic corrections. Comparison of the fit represented by Eq. (4) with actual experimental data is given in Table 1. Equation (4) can also be used to predict the electron affinity of Br: it gives a value of 3.286 eV, as compared to the experimental value²⁰ of 3.364 ± 0.004 eV.

The 4s²4p⁵ ²P_{3/2}-²P_{1,2} interval has been experimentally determined for all ions in the sequence except Tc⁸⁺. The experimental uncertainties increase

with ionization stage because observations are made at shorter wavelengths. We utilize the conventional Slater-Condon parameter, ζ , which is equal to two-thirds of the 3/2-1/2 interval, and we find once again that a linear relationship obtains between experimental and Hartree-Fock¹³ values of ζ (Fig. 2):

$$\zeta_{\text{expt}} = 26.4 \text{ cm}^{-1} + 1.1016 \zeta_{\text{HF}}. \quad (5)$$

Table 2 shows the comparison of this fit with the experimental data. It should be noted that the data for Ag and Cd have relatively large experimental uncertainties, and we shall utilize our fitted values for those ions in subsequent discussion.

The $4s^2 4p^6 - 4s 4p^6$ ionization energy is known experimentally only from Kr to Mo⁶⁺ (Ref. 10-13, 17). The usual means of determining this energy is via observation of the transitions $4s^2 4p^5 - 4s 4p^6$; it is interesting to note that Even-Zohar and Fraenkel¹⁴ report that these lines were not observed for Ru, Rh, and Pd in their experiment. Thus, in this case an extrapolation method is required to get any information at all about the elements above molybdenum. As in the previous two examples, we compare experimental and Hartree-Fock values in the known range and find a good fit to be given by

$$I_{\text{expt}} = -24524 \text{ cm}^{-1} + 1.01403 I_{\text{HF}}. \quad (6)$$

Table 3 gives numerical values. The scaling factor in Eq. 6 is presumably larger than that of Eq. (4) because of the greater effect of relativity upon a 4s than a 4p electron. It would probably be preferable to use a relativistic theory, such as the Dirac-Fock approximation, as the basis for extrapolation.

The preceding discussion establishes the energies of the first three ionization limits with uncertainties of (apparently) a few hundred cm⁻¹. We

now consider the excitation energy $4s^24p^6-4s4p^65p$. This can be written as

$$E = I - (Z_{\text{eff}}^2/n^{*2})\text{Ryd} , \quad (7)$$

where I is the $4s^24p^6-4s4p^6$ ionization energy, Z_{eff} is the ionization stage plus 1, and n^* the effective principal quantum number of the $5p$ electron. Since the ground $4s^24p^6$ state has total angular momentum $J=0$ and even parity, the $4s4p^65p$ state must be excited by an odd number of photons and have $J=1$. For simplicity, we shall designate the two possible $J=1$ states as $^3p^o$ (lower energy) and $^1p^o$ (higher energy), although they depart from LS coupling at high ionization stages. To determine the excitation energies, we need to find the values of n^* . This is done as follows:

The energy difference between $^1p^o$ and $^3p^o$ states can be expressed in terms of the Slater-Condon parameters $G^1(4s,5p)$ and ζ_{5p} . In Ref. 10, this difference was determined experimentally for Mo^{6+} , and effective values for these parameters were obtained. We find that the effective values of G^1 and ζ are obtained by scaling the Hartree-Fock values by 0.866 and 1.319 respectively. We apply these scaling factors to the Hartree-Fock values throughout the isoelectronic sequence. This procedure gives the relative energies of the two terms; to determine the absolute energies, we compare the Hartree-Fock value of n^* with the experimental value for the few cases where the latter is known. For the $^1p^o$ terms of Zr^{4+} , Nb^{5+} , and Mo^{6+} , and the $^3p^o$ term of Mo^{6+} , we find that $n^*_{\text{HF}} - n^* = 0.0365$, 0.0329 , 0.0365 , and 0.0357 respectively. We thus adopt 0.036 as a standard correction, i.e., we take $n^* = n^*_{\text{HF}} - 0.036$. The resulting values of excitation energy computed from Eq. (7) are given in Table 4.

We now consider the various possibilities for decay of the $4s4p^65p$ state. First of all, it should be noted that although this state autoionizes in the

first few members of the isoelectronic sequence, it is bound after Sr^{2+} . Thus, only radiative decay mechanisms need be considered in the cases of greatest interest. Other depopulating mechanisms associated with electron and ion collisions will certainly be important in practice, but their treatment is beyond the scope of the present paper.

There are basically two types of radiative transitions to consider: those in which the 5p electron changes state, and those in which a 4p electron drops into the 4s hole. Two-electron transitions will presumably be much less probable in highly ionized atoms.

Of these two types, the 5p transitions will become more rapid at high ionization stages, since they involve a change in principal quantum number. The electric dipole transitions of this type are $4s4p^65p \rightarrow 4s^24p^6$ and $4s4p^65p \rightarrow 4s4p^64d$. The transition probabilities for these two cases are probably comparable: we have calculated some values using Hartree-Fock wave functions and have obtained, for example, $A(5p-4s) = 1.2 \times 10^{11} \text{ sec}^{-1}$ and $A(5p-4d) = 7.7 \times 10^{10} \text{ sec}^{-1}$ for the singlet states of Cd^{12+} . The 5p-4s transition produces the most energetic radiation, but in the context of Eq. (3) it describes sum-frequency generation. This may turn out to be a feasible method of generating coherent x-radiation (in Cd^{12+} the wavelength would be 6.4nm); but true laser action requires a population inversion, which may be difficult to establish with respect to the ground state. The 5p-4d transition, on the other hand, will be amplified by a population inversion if the lower state is not also resonantly excited. Its wavelength is somewhat longer, being about 14 nm in Cd^{12+} . This transition scheme is not self-terminating, since the $4s4p^64d$ state will decay to $4s^24p^54d$.

The $4s4p^65p - 4s^24p^55p$ transitions occur at yet longer wavelengths, and will have smaller rates than the 5p transitions at high ionization stages.

The oscillator strengths of the 4p-4s transitions decrease slowly along the isoelectronic sequence. We have done a calculation of the rate of the $4s4p^6 - 4s^24p^5$ transition in Cd^{+13} , which should be close to the sum over all final states of the transition rates for Cd^{+12} $4s4p^65p - 4s^24p^55p$. The result obtained in the Hartree-Fock approximation is $A(4p-4s) = 8.1 \times 10^{10} \text{ sec}^{-1}$. This is comparable to the rates of transitions involving the 5p electron. Thus, if pumping is done on a picosecond time scale, radiative decay to the ground state will not much diminish the population of the excited state. Gain could take place on either the 5p-4d or 4p-4s transitions.

The energies of the $4s4p^65p - 4s^24p^55p$ transitions follow the ionic energies of Tables 1, 2, and 3 quite closely. The screening effect of the 5p electron reduces the mean energy of the 4p-4s transitions by less than 0.2 eV throughout the isoelectronic sequence. However, there are nine possible $4s^24p^55p$ final states of the transitions, i.e., all those with $J=0,1,2$, and the associated "satellite" structure can be used to increase the energy of the stabilizing transition. We have evaluated the energies of the $J \leq 2$ states of the $4s^24p^55p$ configuration by using scaled Hartree-Fock values of the Slater-Condon parameters. The configuration has been studied fairly extensively at low ionization stages,^{12,13,22} and we have adopted the scaling factors found for Sr, Y, and Zr. We shall not present detailed results here, since our main purpose is to evaluate the degree to which fine structure can increase the transition energy. The fine structure intervals increase with ionization stage, so the largest effect is found in Cd^{12+} . This case is depicted in Fig. 3, and it is seen that fine structure can increase the transition energy by about 20% above the configuration-average value. We have not made a quantitative estimate of the effect of fine structure in the 5p-4d

transitions, but it is not expected to result in displacements of more than a few eV.

Some features of alternative $4s4p^6n\ell$ states are worth noting. When $n\ell = 5s$, there are no electric dipole-allowed, $\Delta n = -1$ transitions; so, radiative decay should proceed via $4s4p^65s \rightarrow 4s^24p^55s \rightarrow 4s^24p^6$. This might be a preferred scheme for amplifying the $4p-4s$ transition. For $n\ell = 5d$ or $5g$, there will be no electric dipole transitions to the ground state, but there will be $\Delta n = -1$ transitions to $4s4p^54f$ final states. The wavelength of these transitions will decrease rapidly along the isoelectronic sequence.

B. Transitions Involving Double Excitation

If two valence electrons are excited to orbitals with higher values of principal quantum number, energetic radiation can be emitted by a downward transition of one electron. We discuss in particular excitation of $4s^24p^6 - 4s^24p^45s^2$, followed by $4s^24p^45s^2 \rightarrow 4s^24p^65s + \gamma$.

There is presently no photon spectroscopy of these doubly-excited states, since they cannot be reached by single photon absorption by the ground state. There have been some electron-impact excitation studies^{23,24} which have identified the $4s^24p^45s^2$ states in Kr, which are autoionizing features with widths of the order 0.1 eV. There is no data of which we are aware on other members of the Kr isoelectronic sequence. Since there is no basis for extensive comparison of theoretical calculations with experimental data, we have not invested any effort in refining our theoretical treatment beyond the Hartree-Fock level. Thus, all the numerical data presented in this subsection is based on raw Hartree-Fock calculations, which are expected to be in error by at least a few eV. However, we believe that they will correctly reflect systematic trends.

A principal attraction of the $4s^2 4p^4 5s^2$ configuration is that it must become bound at some point in the isoelectronic sequence. This can be concluded on the basis of the hydrogenic limit approached at high ionization stages, in which the binding energy of two 5s electrons ($2/25 Z_{\text{eff}}^2$ Rydberg) exceeds that of one 4p electron ($1/16 Z_{\text{eff}}^2$ Rydberg). When this configuration becomes stable, it should also lie below most other doubly excited configurations, and its predominant decay mechanism should be a radiative transition to $4s^2 4p^5 5s$. Table 4 shows the Hartree-Fock term-averaged excitation energy $4s^2 4p^6 - 4s^2 4p^4 5s^2$ along the isoelectronic sequence. Comparison with Table 1 indicates that crossing below the ionization limit occurs in the vicinity of Mo^{6+} .

The configuration-averaged energy of the 5s-4p transition is shown in Fig. 4. The minimum wavelength attained is 8.9 nm in Cd^{12+} , and this can presumably be further shortened by use of fine structure. If a suitable transition can be found in Cd^{12+} , about 50 KrF photons must be absorbed to excite it.

The results of this section have shown the existence of several candidates for soft x-ray lasing action, though much additional spectroscopic investigation is required, particularly of the doubly-excited states. We now discuss some problems of multiphoton absorption relevant to this scheme.

3. MULTIPHOTON EXCITATION OF IONS

The preceding section has shown that Kr-like ions possess singly and multiply excited states, some of which are bound and some autoionizing, which should provide strong radiative transitions in the region $\approx 10\text{nm}$ to lower-lying excited states. In the scheme being proposed in this paper, we consider multiphoton excitation of these initial states using radiation near 248nm.

Several experiments in recent years have shown that multiphoton ionization, including multiple ionization, can occur with large numbers of absorbed photons with remarkable efficiency. Experiments in Ca^5 and in Xe^{25} for example, have shown evidence of saturation in the production of singly and doubly charged atoms using 1.064μ radiation with processes involving up to 29 photons. The question of interest here involves consideration of these processes with ionized initial systems, and whether it will be equally possible to put large numbers of ions into discrete excited states by multiphoton excitation. Few experiments have been carried out using tunable lasers and even fewer experiments have looked for fluorescence, so many of the concepts discussed here must be considered speculative. Nevertheless, we will see that there are some clear suggestions concerning the most promising way to proceed with experiments.

A major aspect of experiments involving multiphoton excitation in high incident fields is the absence of an adequate theory to explain even the qualitative nature of processes involving the absorption of large numbers of photons. For a simplistic picture, however, one can consider a rate equation approach. This is further justified by the spatial and temporal inhomogeneity of the driving laser field. Consider the case where the only resonances are an N photon resonance between the ground state ψ_1 and the excited state of interest ψ_2 , and an M photon resonance between ψ_2 and the ionization continuum (see Fig. 5).

The excitation rate from ψ_1 to ψ_2 is $R_{12} = \sigma_{12}^{(N)} I^N$, where $\sigma_{12}^{(N)}$ is the N -photon generalized cross section between ψ_1 and ψ_2 , and I is the incident flux density. Similarly, $R_{2c} = \sigma_{2c}^{(M)} I^M$ is the ionization rate out of ψ_2 . In

the case where $R_{12} \gg R_{2C}$, the population N_2 in ψ_2 maximizes at $1/2 N_0$, where N_0 is the initial population in ψ_1 and ψ_2 . If $R_{2C} > R_{12}$, N_2 maximizes at $N_0 \frac{R_{12}}{R_{2C}} \ll 1/2 N_0$. Clearly, it is most efficient to choose a case with $\frac{R_{12}}{R_{2C}}$ as large as possible. Barring anomalies in the generalized cross sections $\sigma_{12}^{(N)}$ and $\sigma_{2C}^{(M)}$, this is generally achieved by making the ratio $\frac{N}{M}$ as small as possible, which in turn results from having ψ_2 as far below ionization as possible relative to the ionization potential of ψ_1 . In the previous section, we saw that because of the scaling laws, the trend as one increases the stage of ionization is to decrease the ratio of $\frac{N}{M}$. For the $4s^2 4p^6 + 4s 4p^6 5p$ transition, measured relative to the $4s 4p^6$ continuum, we find $\frac{N}{M} = 9.9$ for Kr and only 1.2 for Cd^{12+} . Thus, for high ionization species such as Cd^{12+} , we would appear to have the best chance for pumping into intermediate bound states without simultaneously depleting that state by rapid ionization. Of course, the higher stages of ionization require high order processes for excitation, 39 photons in the case of Cd^{12+} , but processes involving as many as 99 photons have been reported in collision-free environments.^{1,26}

A second aspect to consider in multiphoton excitation to discrete levels is the role of resonances in the excitation. In the previous section, the locations of various energy levels were calculated under field-free conditions. In the presence of an intense optical field, the position of the levels will be shifted. Again, accurate calculations for N^{th} order processes are difficult to make. As a measure of the magnitude of the shift, however, we can consider the second order Stark shifts of the ground state by the laser field as corresponding to absorbing and re-emitting one photon. The shift is

given by²⁷

$$\Delta = \sum_j \frac{|\bar{d}_{ja} \cdot \bar{e}_1|^2}{\hbar^2(\omega_{ja} - \omega)}, \quad (8)$$

where the summation is over all intermediate states, $\bar{e}_1$ is the amplitude of the incident field (cgs units), $\hbar\omega_{ja} = E_j - E_a$ is the energy separation of the ground state and the intermediate state j , $\omega < \omega_{ja}$ is the laser frequency, and $d_{ja} = e \int \psi_j^* r \psi_a dr$ is the transition dipole moment. If we take $d = 5 \times 10^{-18}$ esu cm, corresponding to $\langle r \rangle = 10^{-8}$ cm and $\hbar(\omega_{ja} - \omega) = 200$ eV corresponding to Cd^{12+} , we find an optical Stark shift of $\Delta = 1.22$ I, with Δ in units of radians sec^{-1} and I in watts cm^{-2} . Thus, for $I = 10^{16}$ W cm^{-2} the optical Stark shift of the ground state is greater than the photon energy. The energy of the transition also depends on the Stark shift of the upper level and on the shift due to higher order terms, but it is clear that the effect can be comparable to or larger than the energy separating an N photon from an $N+1$ photon process.

During the time evolution of a pulse, and across its spatial extent, the frequency of the multiphoton transition thus undergoes a sufficiently large sweep due to field effects as to effectively destroy the importance of resonances. This variation also gives justification to the use of rate equations in calculating populations. The result is that essentially every system sweeps into and out of resonance, but at the expense of an effective line broadening of the transition with an attendant decrease in optical gain. All systems become approximately equal, but none are as effective as the resonant, unperturbed system. The broadening of the resonance frequency across the sample and consequent decrease in gain vanished as the pump laser is turned off. The entire sample returns to the same resonant frequency,

neglecting plasma broadening and Doppler effects, and the gain across the sample should rise substantially. This "gain switching" may help allow efficient multiphoton pumping during the high intensity part of the pulse, followed by an intense super-radiant pulse as the pump laser turns off.

The time scale on which the pumping and any benefits from gain changes must occur is determined by the lifetime of the excited states. Again, there is an advantage in working with higher stages of ionization. In the case of Kr, Rb⁺, and Sr²⁺, the 4s4p⁶5p levels are above the 4s²4p⁵ continua and subject to autoionization. For Y³⁺ and higher ionization stages, the excited state drops below the continuum threshold, and the lifetimes are determined by radiative decay. Similarly, for Tc⁷⁺ and above, the 4s²4p⁴5s² level lies below the continuum threshold. For radiation, the decay rate is given by

$$A_{ki} = 6.67 \times 10^{15} \lambda^{-2} f_{ki} g_i / g_k \text{ sec}^{-1}, \quad (9)$$

where λ_{ki} , the transition wavelength, is in Å, f_{ki} is the oscillator strength, and g_i and g_k are the degeneracies of the states. For $\lambda = 100\text{Å}$, $f=0.16$, and $g_i=g_k$, we obtain $A_{ki} = 10^{11} \text{ sec}^{-1}$ or $\tau_k=10 \text{ ps}$. Thus, we see that the 100Å region is properly matched to pumping schemes operating in the 1 ps to 10 ps regime.

A final consideration associated with the choice of high ionization stages is related to the concept of core excitation and subsequent loss of an entire electron shell, which has been postulated for the observed high ionization stages under strong laser field (10^{16} - $10^{17} \text{ W cm}^{-2}$) and collision-free conditions. In this model, unspecified coherent processes result in

excitation of the core electrons. This excitation contains sufficient energy to remove much or all of an entire valence shell, thus explaining some of the trends seen in these experiments. As one proceeds to higher stages of ionization, the binding energy of the valence electron rises much faster than the excitation energy, as pointed out before in considering multiphoton ionization and autoionization. Here the benefit is that it becomes much more difficult to drive the system hard enough so that the stored energy is sufficient to remove all of the valence electrons. It would seem much more likely that if there is core excitation, the subsequent relaxation would result in partial ionization at most, with a high likelihood that the remaining valence electrons would be in an excited state.

4. CONCLUSIONS

In summary, it appears that there are major advantages in going to highly ionized systems and short ($\leq 10^{-11}$ sec) pulse excitation to obtain substantial gain in the 100eV-200eV spectral region. The higher optical fields necessary for multiphoton pumping with high ionization systems are accompanied by a greatly reduced ionization rate out of excited states when compared with excitation rates, an elimination of competing autoionization, a relaxation of the resonance condition, possible gain switching, and a reduction in the likelihood of fully stripping the optically active shell by core excitation and subsequent Auger-like ionization. The appropriate time scale for the driving laser appears to be 1 ps to 10 ps.

We have identified candidate transitions in the Kr isoelectronic sequence which may be used to amplify soft X-ray radiation in circumstances where the resonance excitation condition applies (i.e., laser intensities $\leq 10^{15}$ W cm⁻²).

Two obvious avenues are open to extending this approach to shorter wavelengths: utilization of tighter closed-shell isoelectronic ions, i.e., in the He, Ne, Ar, and Ni sequences; and the use of much higher stages of ionization.

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Table 1. Comparison of experimental and scaled Hartree-Fock ionization energies $4s^2 4p^6 - 4s^2 4p^5 \ ^2P_{3/2}$ for the Kr isoelectronic sequence.

<u>Ion</u>	<u>Experimental (cm⁻¹)</u>	<u>Scaled HF (cm⁻¹)</u>
Kr	112915 (a)	112790
Rb ⁺	220072 ± 20 (b)	220131
Sr ²⁺	345900 ± 200 (b)	345961
Y ³⁺	488750 ± 100 (b)	488926
Zr ⁴⁺	648250 ± 150 (b)	648184
Nb ⁵⁺	823150 ± 150 (b)	823172
Mo ⁶⁺	1013550 ± 150 (b)	1013483
Tc ⁷⁺	---	1218817
Ru ⁸⁺	1439000 ± 400 (c)	1438937
Rh ⁹⁺	1673700 ± 400 (c)	1673662
Pd ¹⁰⁺	---	1922838
Ag ¹¹⁺	---	2186343
Cd ¹²⁺	---	2464070

(a) from Ref. 16

(b) from Ref. 9

(c) from Ref. 13

(d) Ref. 13 estimates a value from isoelectronic extrapolation of
1924200 ± 1000

(e) as for (d), estimated 2189500 ± 1000

Table 2. Comparison of experimental and scaled Hartree-Fock values of the $4p^5$ interval. $\zeta = 2/3 (E(J=1/2) - E(J=3/2))$.

Ion	(cm ⁻¹)	
	Experimental	Scaled HF
Br	2457	2466
Kr ⁺	3581	3582
Rb ²⁺	4920	4916
Sr ³⁺	6487	6485
Y ⁴⁺	8307	8308
Zr ⁵⁺	10405	10406
Nb ⁶⁺	12790	12799
Mo ⁷⁺	15518	15510
Tc ⁸⁺	---	18563
Ru ⁹⁺	22037	21980
Rh ¹⁰⁺	25867	25788
Pd ¹¹⁺	30120	30013
Ag ¹²⁺	35168	34681
Cd ¹³⁺	40424	39819

Table 3. Comparison of experimental and scaled Hartree-Fock values of the $4s^2 4p^6 - 4s 4p^6$ ionization energy.

<u>Ion</u>	<u>Experimental (cm⁻¹)</u>	<u>Scaled HF (cm⁻¹)</u>
Kr	221918	221782
Rb+	350108	349908
Sr ²⁺	496405	496425
Y ³⁺	659696	660052
Zr ⁴⁺	839821	840015
Nb ⁵⁺	1035651	1035795
Mo ⁶⁺	1247380	1247027
Tc ⁷⁺	---	1473440
Ru ⁸⁺	---	1714835
Rh ⁹⁺	---	1971049
Pd ¹⁰⁺	---	2241953
Ag ¹¹⁺	---	2527450
Cd ¹²⁺	---	2827454

Table 4. Comparison of experimental and calculated values of the excitation energies $4s^24p^6 - 4s4p^65p$ $^3p^{\circ}_1, ^1p^{\circ}_1$. Entry in the last column indicates whether resonance exists with energy of N photons in the KrF range 248-250 nm.

Ion	Experimental (cm ⁻¹)	Scaled HF (cm ⁻¹)	N
Kr $^3p^{\circ}_1$ $^1p^{\circ}_1$	201005 (a)	201378	5
	201600 (a)	202064	5
Rb+	--	286650	-
	--	288625	-
Sr ²⁺	--	375732	-
	--	379034	-
Y ³⁺	--	469437	-
	--	474131	-
Zr ⁴⁺	--	568003	-
	573776 (b)	574179	-
Nb ⁵⁺	--	671461	-
	679967 (b)	679295	17
Mo ⁶⁺	780390 (b)	779870	-
	789696 (b)	789575	-
Tc ⁷⁺	---	892343	-
	---	905031	-
Ru ⁸⁺	---	1011543	-
	---	1025685	-
Rh ⁹⁺	---	1134796	-
	---	1151573	-
Pd ¹⁰⁺	---	1262951	-
	---	1282740	-
Ag ¹¹⁺	---	1396029	-
	---	1419159	-
Cd ¹²⁺	---	1534025	-
	---	1560857	39

(a) from Ref. 21

(b) from Ref. 10

Table 5. Hartree-Fock excitation energies for $4s^2 4p^6 - 4s^2 4p^4 5s^2$, averaged over all terms of final state configuration.

<u>Ion</u>	<u>Energy (cm⁻¹)</u>
Kr	189800
Rb ⁺	201250
Sr ²⁺	423100
Y ³⁺	553900
Zr ⁴⁺	698200
Nb ⁵⁺	851400
Mo ⁶⁺	1014900
Tc ⁷⁺	1188600
Ru ⁸⁺	1372500
Rh ⁹⁺	1566500
Pd ¹⁰⁺	1770500
Ag ¹¹⁺	1984800
Cd ¹²⁺	2209000

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Figure Captions:

- Fig. 1 Adjusted Hartree-Fock vs. experimental values of the $4p^6-4p^5\ 2p_{3/2}$ ionization energies for the first seven members of the Kr isoelectronic sequence. The adjusted Hartree-Fock values are given by $I_{HF} = E(4p^5) - E(4p^6) - \zeta/2$, where E is the non-relativistic total energy and ζ is obtained from ζ_{HF} via Eq. (5). Sources of experimental energies are listed in Table 1.
- Fig. 2 Hartree-Fock vs. experimental values of the spin-orbit parameter ζ of the ground state of Br-like ions.
- Fig. 3 Energy levels of Br- and Kr- like ions of Cd. The absolute energies of the two ions have been displaced so that the $5p$ states of Cd^{12+} can be seen as "satellites" of the states of Cd^{13+} configurations; 41 eV is the energy separation of the centers of gravity of the Cd^{13+} configurations; 52.7 eV is the maximum energy separation between states of Cd^{12+} .
- Fig. 4 Hartree-Fock values of the wavelength of $4p^45s^2 - 4p^55s$ transitions in the Kr isoelectronic sequence.
- Fig. 5 Schematic N-photon resonant, N + M-photon ionization process, as discussed in Sec. 3.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5

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