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Theoretical simulation of the decay of the $4s4p\ ^1P_1^o$ level in Kr VII following beam-foil excitation

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The decay of the $4s4p\ ^1P_1^o$ level in Kr VII following beam-foil excitation has been simulated using theoretical initial-state populations and calculated transition probabilities. It was found that the decay curve was substantially influenced by cascading from higher levels and that such cascades dominate the decay in the region beyond three times the lifetime of the $4s4p\ ^1P_1^o$ level. The simulation is in good agreement with an experimental decay curve at short decay times, but is unable to reproduce the long-time behavior of the experimental data. However, recent spectroscopic evidence indicates that line blending is responsible for this aspect. Complications due to core-excited states and other complex cascade mechanisms are discussed.

I. INTRODUCTION

An accurate understanding of the radiative properties of highly ionized heavy atoms is needed for the diagnostics and modeling of high-temperature plasmas occurring both astrophysically and in plasma devices used for magnetic fusion research.[1,2] In particular, the resonance lines of ions with one or two electrons outside closed shells are of great interest since they constitute prominent spectral features which are readily identifiable in laboratory plasmas.[3] A review of the available oscillator strength data for the $\Delta n = 0$ resonance transitions of two such isoelectronic sequences, the Cu and Zn sequences, has shown that systematic discrepancies exist between the best available theoretical and experimental data.[4] The discrepancies are of the order of 30% for the Cu sequence and increase to more than 100% for the higher end of the Zn sequence, with the experimental data almost always yielding much lower oscillator strengths than theory. Practically all experimental data have been obtained with the beam-foil spectroscopy method,[5] while the principal theoretical data are from multiconfiguration self-consistent-field approaches or the relativistic random-phase approximation, which include electron correlation to various degrees. On the basis of a simulation of experimental beam-foil decay data, two of the present authors (S. M. Y. and W.L.W.) proposed[6] that cascading effects may be responsible for this discrepancy. Such cascading, which is always present in beam-foil experiments, due to the nonselective excitation mechanism, is difficult to treat adequately. The decay curves of several Cu-like ions have been simulated by us in detail and compared with available experimental decay curves. It was concluded that the usual analysis of the time-of-flight decay of the foil-excited beam in terms of a multiexponential fit was inadequate for the resolution of a true many-exponential decay encountered in the typical beam-foil experiment. The substantial population of highly excited high-angular-momentum states was shown to lead to a complex masking of the true primary lifetime even when some statistical indicators of cascading, such as the replenishment ratio, did not point to a serious problem. A confirmation of these ideas based on new experimental observations has recently been made by Livingston *et al.*,[7] who performed an ANDC (arbitrarily normalized decay curve) analysis of the $4s$-$4p$ resonance line of copperlike krypton (Kr VIII).[7] This method takes into account some of the complex cascades present in the decay and will lead to a more accurate primary lifetime than would be obtained from exponential fitting. The lifetime extracted by Livingston *et al.* is in excellent agreement with our earlier analysis as well as the best theoretical results.

The zinc sequence represents a much more complex case of cascading than the one-electron Cu sequence because of the presence of (a) multiply excited bound states, such as $3d^{10}4p^2$ and $3d^{10}4p4d$, as well as (b) core-excited bound states, such as $3d^94s4p^2$. Both of these decay directly into the $3d^{10}4s4p$ state.

Since our previous study was performed before an experimental decay curve on a Zn-like ion was available to aid us in establishing the contributions of these states, we refrained from a detailed examination of the Zn-sequence case. Recently, an experimental decay curve for the Zn-like ion Kr VII has become available, thereby making the present simulation study feasible. Also, renewed interest by several beam-foil groups in measuring cascade-free lifetimes in the Zn sequence has provided increased motivation to investigate from a theoretical standpoint what cascade effects might be expected.

II. THEORETICAL SIMULATION OF THE $4s4p\,^1P$ DECAY IN Kr VII

The techniques used in the simulation of beam-foil decay curves have been described by two of us in detail elsewhere.[6] Briefly, one constructs a hierarchy of exponential decay equations which describe the cascading of highly excited electrons into the primary state whose lifetime is being determined. The key parameters on which such a simulation is based are the lifetimes of the cascading states and their initial population distribution following foil excitation. Thus fairly reliable lifetime data, from calculations or other non-beam-foil sources, and a realistic model for the distribution of initial populations must be known.

The principal cascade routes into the $4s4p\,^1P^o$ level are illustrated in Fig. 1. Analogous to our work in the Cu sequence, we have chosen the initial population distribution as

$$N_{nl}(t=0)=\frac{(2l+1)}{(n^*_{nl})^3}\,, \tag{1}$$

where n^*_{nl} is the effective quantum number[6] and l is the angular momentum. Clearly such a simple model should not be accurate for core-excited or multiply excited states, particularly when equivalent electrons are involved. It has been retained for these cases, however, both from considerations of consistency and since additional simulations assuming much larger populations in multiply excited states than those based on n^* showed that their effects on the extracted lifetime are small.

The transition probability data were chosen as follows: For the primary level the multiconfiguration Hartree-Fock oscillator strength[8] value of $f=1.61$ was converted to the transition probability $A=104\times10^8\ \text{sec}^{-1}$. The $4p^2$ and $4s4d$ level lifetimes were derived from the superposition-of-configuration calculations of Weiss.[9] Transitions from the core-excited $3d^94s4p^2$ state are satellitic to those from the Ni-like $3d^94p$ state, and based on previous studies,[9] these levels are expected to have roughly the same lifetime. We have therefore taken as an approximation to the $3d^94s4p^2$ lifetime the $3d^94p\,^1P^o$ lifetime data of Cowan.[10] The $4s4f$ lifetime was computed with Hartree-Fock orbitals with approximations for configuration mixing. The remainder of the "yrast" lifetimes (see Ref. 6) were obtained from Coulomb approximation data which are expected to be reasonably accurate for

TABLE I. Initial population ratios and cascade lifetimes used in the Kr VII $4s4p\,^1P^o$ decay simulation. The theoretical primary lifetime is 0.096 ns.

Level	τ(ns)	$N_n(0)/N_{4s4p}(0)$ Model I, II	Model III
$4s4d$	0.032	1.02	1.20
$4s4f$	0.093	0.86	1.20
$4s5g$	0.086	0.53	0.94
$4s6h$	0.25	0.37	0.80
$4s7i$	0.57	0.28	0.69
$4s8k$	1.1	0.21	0.61
$4s9l$	2.2	0.17	0.55
$4s10m$	4.3	0.14	0.50
$4s11n$	8.3	0.12	0.45
$4s12o$	9.3	0.097	0.42
$4p^2\,^1D$	1.3	1.36	1.45
$4p^2\,^1S$	0.024	0.25	0.27

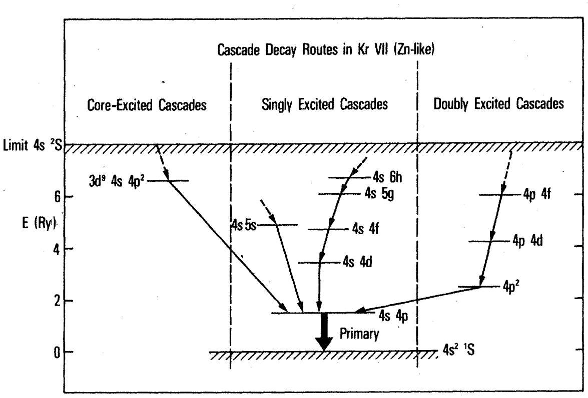

FIG. 1. Cascade decay routes in Kr VII. Core-excited, doubly excited, and lower singly excited level positions were derived from the superposition-of-configuration calculations of Weiss (Ref. 9). High-lying singly excited levels were computed in the quantum-defect approximation.

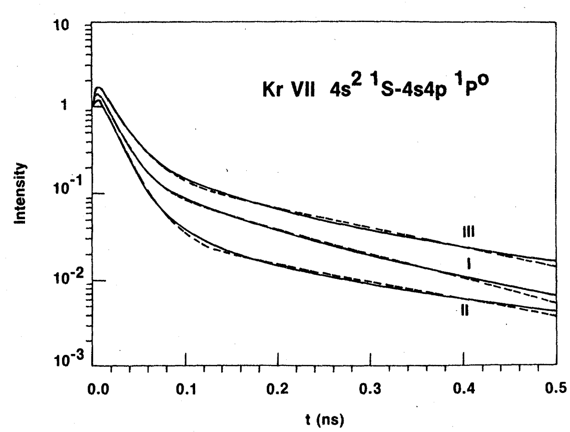

FIG. 2. Theoretical simulations of the decay of the $4s4p\,^1P_1^o$ level in Kr VII following foil excitation. The solid curves correspond to the models described in the text [model I: yrast cascades, plus cascades from $4p^2\,^1D$ and $4p^2\,^1S$ states, with $(2l+1)/(n^*)^3$ initial population; model II: yrast cascades only with $(2l+1)/(n^*)^3$ initial population; model III: both types of cascades, but with $(2l+1)/(n^*)^2$ initial population]. The dotted curves are two-exponential fits to the full simulations.

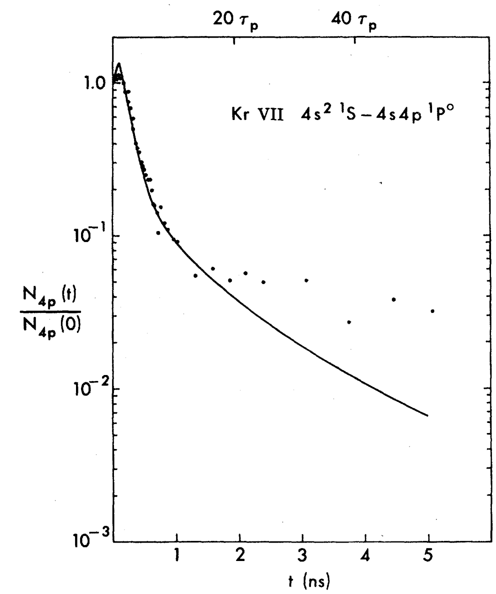

FIG. 3. Comparison of the simulated decay curve using model I [primary plus yrast plus $4p^2$ assuming a population distribution according to $(2l+1)/(n^*)^3$] to experimental beam-foil data. The background count was taken as the beam-off dark count. The time axis of the experimental data was shifted to obtain the best fit to the simulated curve.

the high-angular-momentum states employed. The initial population ratios and lifetimes used in the present simulation are given in Table I.

Several theoretical simulations of the decay of the $4s4p\ {}^1P$ level following foil excitation are illustrated in Fig. 2. Curve I includes yrast cascades ($4p \leftarrow 4d \leftarrow 4f \leftarrow 5g \leftarrow 6h \leftarrow \cdots$) up to $n=12$ as well as cascades from the doubly excited $4p^2\ {}^1D$ and $4p^2\ {}^1S$ states assuming an initial population distribution given by Eq. (1). Curve II includes only yrast cascades. Curve III includes both types of cascades but assumes a population distribution of $(2l+1)/(n^*)^2$. The dotted curves are the best two-exponential fits (see below) to each of the simulated curves.

Of key importance to this simulation study is the availability of an experimental decay curve with which various simulated curves can be compared. The present decay curve for the 585-Å resonance line of Kr VII was generated by a 1.5-MeV Kr^+ beam from a Van de Graaff accelerator. (Details of the experiment may be found in Ref. 16.) The post-foil beam velocity of 1.86 mm/ns, combined with the spatial resolution along the beam axis, yielded a time window of 0.08 ns. Data points near the beginning of the curve have a spacing of 0.027 ns, gradually increasing to steps of 6.75 ns at the shallow tail of the decay.

A constant background of 13 counts per channel was subtracted from the data points. This was obtained by summing the average dark count rate with an estimate of the additional small scattered light contribution which would be expected at large distances from the foil. The last six data points, which span a time of flight of some 27 ns, show an essentially flat behavior with an average of 17 counts per point. Other background rates were also attempted (16 and 25 counts per point), but the same conclusions were reached in all three cases.

The data were normalized to the simulated curve at $t=0.146$ ns. For comparison, the simulated decay curve giving the best fit with experiment, i.e., the curve comprising primary plus yrast plus $4p^2$ cascades (curve I of Fig. 2), has been included in Fig. 3. The absolute number of counts in the peak is 1018. It is seen to be in good agreement with the experimental data for short decay times, but is unable to reproduce the long-time behavior. However, the long "tail" is very likely due to line blending. A recent spectroscopic investigation of

TABLE II. Comparison of experimental, simulated, and theoretical lifetimes (in ns) for the $4s4p\ {}^1P^o$ level of Kr VII. Both the experimental and simulated lifetimes were obtained from a two-exponential fit to the corresponding decay curves. Numbers in parentheses indicate the cascade lifetime.

Beam-Foil Data	Simulation Model I	Simulation Model II	Simulation Model III	Theory
0.19 ± 0.02[a]	0.19	0.19	0.23	0.096[d]
	(1.5)	(2.2)	(5.2)	
0.19 ± 0.02[b]				
0.163 ± 0.006[c]				
(0.95 ± 0.08)				

[a]Reference 12.
[b]Reference 13.
[c]Reference 14.
[d]Reference 8.

the Cu I isoelectronic sequence using many members of this sequence yields for the $4f$-$5g$ transition an interpolated value of 585.6 ± 0.4 Å in Kr VIII.[11] This wavelength coincides within the experimental resolution of about 1 Å with that of the Kr VII resonance line being studied (which is at 585.3 Å); hence, the measured decay curve will also contain components due to this Kr VIII line. Such yrast transitions are known to appear strongly in beam-foil spectra and are usually associated with slow decays. The long tail in the decay curve is expected to exhibit the entire yrast chain arising from levels above $5g$.

It is worth noting that curve III of Fig. 2, i.e., the primary plus yrast plus $4p^2$ cascades with a $(2l+1)/(n^*)^2$ population distribution, would be significantly above the experimental points, indicating that this model substantially overpopulates the cascade states with respect to the primary. However, in all cases the effect of the cascades is to substantially alter the decay curve from the case where only the $4s4p\,^1P^o$ level is populated.

A cascade analysis similar to one which is often applied to experimental data was performed on the theoretical curves. Two-exponential fits assuming variable coefficients and exponents were made to each simulation curve with the results given in Table II and by the dotted curves in Fig. 2. In cases I and II the primary lifetimes extracted from the fit are in excellent agreement with recently measured beam-foil lifetimes, but not with the theoretical value actually used in the construction of the simulation, which is shorter by about a factor of 2. Thus the many highly excited cascades effectively mask the true primary decay to such an extent that the customary multiexponential fit to the decay curve is inadequate to recover the true value. (Note the extremely close agreement of the many-exponential simulation and the two-exponential fits illustrated in Fig. 2.) This is similar to the case of the $4p\,^2P^o$ state in Cu-like ions discussed in Ref. 6. As in the Cu sequence, an ANDC (Ref. 7) (arbitrarily normalized decay curve) analysis is expected to produce an accurate lifetime, since cascading through the $4s4d$ state represents the dominant decay channel.

III. DISCUSSION

There are four principal classes of cascade levels which may contribute to the decay of the $4s4p\,^1P_1^o$ level in most ions of the Zn sequence: Rydberg, yrast, multiply excited closed-core, and core-excited levels. An examination of the lifetimes of the low-lying Rydberg states for Kr VII indicates that they will give rise to "growing in" cascades which affect only the initial part of the decay. There are appreciable uncertainties in the transition probabilities associated with these cascades, and in the Kr VII simulation they were omitted. It is of interest to note that in our previous study[6] of the Kr VIII $4p\,^2P^o$ decay we also found that such cascades had very little effect on the extracted lifetime.

The yrast states constituted the most important class of cascades in the Kr VIII simulation and Fig. 2 illustrates that they also dominate the decay in Kr VII. Recent studies of the initial populations of beam-foil excited ions indicate that the population model given by Eq. (1) is at least qualitatively correct.[15] Note that the $4s4f\,^1F^o$ lifetime, 0.093 ns, is very close to the primary lifetime of 0.096 ns.

Multiply excited states are potentially important cascade sources; moreover, each multiply excited state has its own yrast feeder chain which further complicates the decay scheme. The length of this chain is uncertain in that at some point the configuration $3d^{10}nlml'$ becomes autoionizing, but for Kr VII one might expect that at least the $4p4f \rightarrow 4p4d \rightarrow 4p^2 \rightarrow 4s4p$ decay sequency will exist. Since no specific data on the initial populations of such multiply excited states in a foil-excited beam are available and the transition probabilities are quite uncertain, only the $4p^2$-$4s4p$ cascades were included in the present simulation. In order to test the sensitivity of the simulation to the population of the $4p^2$ state, several curves were constructed with different $N_{4p^2}(0)$; the net effect on the extracted lifetime was very small although the magnitude of the decay curve varied considerably. Note the good agreement between the lifetime extracted from curve I of Fig. 2, which includes cascading from $4p^2$ states, and the value extracted from curve II, which omits them.

Core-excited states are even more difficult to evaluate as potential cascades in beam-foil lifetime experiments. In light ions there is evidence that substantial numbers of core-excited states are produced[16]; even larger effects should be observed for heavy ions which lose most of their outer electrons during passage through the foil and must "rebuild" these electron shells upon their exit. The large energy differences involved in core-repopulating transitions, however, as well as expected large line strengths should result in extremely short lifetimes. Cowan's value[10] of $\tau = 0.013$ ns for the $3d^9 4p\,^1P$ level in Kr IX suggests that the satellite $3d^9 4s4p^2$-$3d^{10}4s4p$ in Kr VII is also short compared to the primary lifetime of 0.096 ns. Thus, even if core-excited cascades are heavily populated, their effect on the decay curve would only be at very short decay times, and would probably not seriously influence the determination of the primary lifetimes of non-core-excited states.

There is another mechanism by which multiply excited and core-excited states may affect the long-time behavior of the decay curve, that is, by the "selective dumping" of populations of multiply excited or core-excited states into long-lived yrast or Rydberg states. Examples of this type of indirect cascades are $3d^{10}4p4d \rightarrow 3d^{10}4s4d$ and $3d^9 4s4p4f \rightarrow 3d^{10}4s4f$. Even if the branching ratios for such transitions were small, large initial populations in these states could significantly affect the final decay curve.

In view of all these uncertainties, principally in the modeling of the population distributions, it seems most appropriate to rely on a comparison with an actual beam-foil decay curve as the main test for the simulation. There are, however, some potential difficulties with the interpretation of such comparisons, mainly associated with the experimental techniques used to record the decay. One of the most serious of these is the ambiguity of the "background" count in a beam-foil experiment. Often the background is given as the "beam-off" or "dark count". This may not reflect the actual background of the recorded decay, however, which is the detected signal with the beam on but with the spectrometer positioned slightly off resonance. It is also not clear whether this background count is constant over the length of the measured decay curve, since each point in the curve corresponds to a different position of the spectrometer-detector system relative to the foil and other potential scattering sources. Since the influence of cascades on the primary decay is to produce a long-lived tail at low intensity it is important to be able to distinguish the actual signal from the background.

IV. SUMMARY

We have simulated the decay of the $4s4p\,{}^1P^o$ level of Kr VII by using theoretical estimates for the initial population distribution and lifetimes. In contrast to our previous studies of Cu-like ions, we found that very extensive cascading from several different *types* of excited states (e.g., doubly excited, core-excited) may be present in the decay curve of a Zn-like ion produced in a foil-excited beam. Nevertheless, it appears that the long-time behavior of the curve is again dominated by the $4snl$ yrast chain of states. The quality of the simulation at large distances from the foil is difficult to assess due to the low experimental count in this region and the ambiguity in the background count present. Line blending is also a potential source of difficulty, especially in complex spectra, as has been seen in the present study. Large differences persist between the cascade-dominated decay curve and the primary decay for any reasonable choice of assumptions. As was the case in our simulation of the $4p\,{}^2P^o$ level in Kr VIII, we found that a two-exponential fit to the Kr VII decay resulted in a primary lifetime in good agreement with the quoted experimental values, but not with the theoretical value actually used in the construction of the simulation. Only when a more sophisticated analytical technique, such as the ANDC method, is applied will the extracted lifetime agree with the original value.

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