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Double Auger processes in the electron-impact ionization of lithiumlike ions

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Contributions to the electron-impact ionization of Li-like ions due to resonant recombination followed by double Auger emission are calculated using first- and second-order perturbation theory. The double Auger rate is found to be quite sensitive to the choice of both continuum phase and excited orbital basis set. In particular we find that resonant recombination to the $1s2s^22p\ ^3P$ term of O^{4+}, followed by double Auger emission to the $1s^2\ ^1S$ term of O^{6+}, has a cross section ranging from 6.4×10^{-22} to 4.9×10^{-21} cm^2 depending on how the double Auger rate is calculated. The maximum represents 0.65% of the total ionization cross section for O^{5+} at the resonance energy of 436.3 eV. Thus a noise level of 1.0%, which is probably a lower limit of current crossed-beam experiments, will mask the contribution in O^{5+}. Similar results are found for other Li-like ions in the first-row Periodic Table elements.

In recent years our understanding of electron-impact ionization processes in atomic ions of the Li isoelectronic sequence has been enhanced by a number of experimental measurements[1–5] and theoretical calculations.[6–8] For these simple ions, the total cross section is dominated by the direct ionization of a $2s$ subshell electron. Indirect contributions to the single-ionization cross section can be made by the following excitation-autoionization processes:

$$e^- + [A(1s^22s)]^{(q+1)+} \rightarrow [A(1s2snl)] \hookrightarrow [A(1s^2)] \,, \tag{1}$$

where q is the charge of the atomic system. For B^{2+} and O^{5+} there is excellent agreement between six-term close-coupling calculations for excitations to states of the $1s2s^2$ and $1s2s2p$ configurations and electron-ion crossed-beam experiments.[5]

It has been speculated theoretically[5,9] that additional indirect contributions to the single-ionization cross section for Li-like ions can be made by the following resonant-recombination auto-double-ionization processes:

$$e^- + [A(1s^22s)]^{(q+1)+} \rightarrow [A(1s2s2p2l)] \hookrightarrow [A(1s^2)] \,, \tag{2}$$

The energies of the $1s2s2p2l$ configurations are less than the energies of the $1s2s^2$ and $1s2s2p$ configurations, which are energetically the lowest autoionizing configurations for Li-like ions. An energy-level diagram for the relevant oxygen ions is shown in Fig. 1. Contributions to the single-ionization cross section of O^{5+} can be made only if the $1s2s2p2l$ configurations decay by double Auger emission. Since the multiplet structure of the $1s2s2p^2$ configuration is relatively complicated, we focus our attention in this paper on the simpler $1s2s^22p$ configuration. As shown later, the resonant-recombination cross section for the ^3P term of the $1s2s^22p$ configuration is much larger than that for the ^1P term. Thus in the following paragraphs we first calculate the double Auger rate for the $1s2s^22p\ ^3P$ term of several Li-like ions and then determine the additional

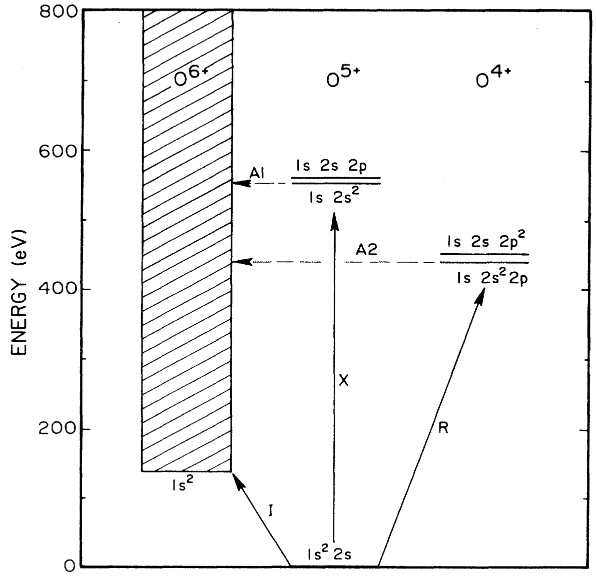

FIG. 1. Energy-level diagram for autoionizing configurations of both O^{5+} and O^{4+}. All energies are relative to the $1s^22s$ configuration of O^{5+}. Cross-hatched area represents the O^{6+} continuum.

contribution to the total ionization cross section made by the rather exotic process of Eq. (2).

Evidence for a double Auger process does exist in photon and projectile induced Auger spectroscopy.[10] From a theoretical perspective the double Auger process is strictly an electron-electron correlation effect. Although calculations have been made using the shakeoff concept,[10] the first many-body perturbation theory calculation was done by Simons and Kelly[11] for the $2s^2 2p$ decay in neutral Li.

Many-body perturbation theory may be used to calculate either the single or double Auger rates. The single Auger rate in lowest-order perturbation theory (in atomic units) is given by

$$A_1 = \frac{4}{k_\beta} \, | \langle \Psi_\beta^+ | V | \Psi_\alpha \rangle |^2 , \tag{3}$$

where $|\Psi_\alpha\rangle$ is the initial autoionizing state of an ion with charge q and $|\Psi_\beta^+\rangle$ is the final state of an ion with charge $q+1$ plus a continuum electron. The total energy $E = \varepsilon_\alpha$ for the initial state and $E = \varepsilon_\beta^+ + k_\beta^2/2$ for the final state. The perturbation V contains both one- and two-electron terms and the continuum normalization is chosen as one times a sine function.

The double Auger rate in lowest-order perturbation theory is given by

$$A_2 = \frac{8}{\pi} \int_0^{k_{\max}} \frac{dk_{\gamma 1}}{k_{\gamma 2}} \times \left| \sum_\beta \sum\!\!\!\!\!\!\!\int_{k_\beta} \frac{\langle \Psi_\gamma^{2+} | V | \Psi_\beta^+ \rangle \langle \Psi_\beta^+ | V | \Psi_\alpha \rangle}{\varepsilon_\alpha - \varepsilon_\beta^+ - k_\beta^2/2} \right|^2 , \tag{4}$$

where $|\Psi_\gamma^{2+}\rangle$ is the final state of an ion with charge $q+2$ plus two continuum electrons. A proper description of $|\Psi_\gamma^{2+}\rangle$ involves the solution of the full three-body problem in the asymptotic region.[12] We employ the standard approximation to the asymptotic region which ensures that the potentials which the electrons experience at infinity are spherically symmetric. In this approximation the phase between the direct and exchange matrix elements is lost and one must make some rather arbitrary choice for this phase. The total energy $E = \varepsilon_\gamma^{2+} + E_{\max}$, where $E_{\max} = k_{\gamma 1}^2/2 + k_{\gamma 2}^2/2$. To avoid a double counting of the continuum states, we set $k_{\max} = \sqrt{E_{\max}}$, which corresponds to an upper limit in an integration over the energy of $E_{\max}/2$. Thus $k_{\gamma 1} \le k_{\gamma 2}$ for all terms in the integrand of Eq. (4).

In this paper we limit our calculations to the first- and second-order perturbation theory expressions of Eqs. (3) and (4), except for the inclusion of certain higher-order effects through the use of a multiconfiguration initial state $|\Psi_\alpha\rangle$. The sums over intermediate states $|\Psi_\beta^+\rangle$ found in Eq. (4) for the double Auger rate include a sum over $(q+1)$ ion states and a sum over a complete set of bound and continuum states of the remaining electron. The symbol $\sum\!\!\!\!\!\!\!\int_k = \sum_n + (2/\pi)\int dk$. A problem common to all perturbation theory calculations is the selection of the excited orbital basis set for which the expansion is most rapidly convergent. By employing two different basis sets, both of which are calculated in a Hartree-Fock V^{N-2} potential, a rough idea of the influence of higher-order effects may be obtained.

To proceed further we focus our attention on the decay of the $1s2s^2 2p\ {}^3P$ term of O^{4+} by double Auger emission. The initial state is assumed to be a configuration-interaction state given by

$$|\Psi_\alpha\rangle = |(a\,|1s2s^2 2p\rangle + b\,|1s2p^3\rangle)\,{}^3P\rangle , \tag{5}$$

where the radial orbitals and mixing coefficients are obtained using the Hartree-Fock bound-state wave-function code developed by Fischer.[13] For O^{4+} the mixing coefficients were found to be $a = 0.97$ and $b = 0.25$.

The complete set of remaining bound and continuum radial orbitals were calculated in the frozen-core Hartree-Fock approximation using two different procedures. A "nonorthogonal" basis set consisted of excited radial orbitals calculated in the Hartree-Fock $1s^2$ potential. An "orthogonal" basis set consisted of excited radial orbitals calculated in the Hartree-Fock $1s^2$ potential with additional projection operators[14,15] to ensure orthogonality with the initial state core orbitals. In this particular case, orthogonality with the $1s$ and $2s$ orbitals is only partially satisfied for s-wave excited orbitals since off-diagonal Lagrange multipliers appear in the original initial state calculation.

The effect of the choice of basis set can best be illustrated by consideration of the partial double Auger rate to the final state $|\Psi_\gamma^{2+}\rangle = |1s^2 k_1 s k_2 p\ {}^3P\rangle$ through the intermediate state $|\Psi_\beta^+\rangle = |1s^2 2skp\ {}^3P\rangle$ from the initial state of Eq. (5). Additional intermediate states, discussed in the next paragraph, are added to the sum inside the absolute value squared. Here our choice of phase between the direct and exchange terms in the two-electron continuum state corresponds to that found from treating the continuum as a discrete set of bound quantum states. For want of a better description we will call this the "natural" phase approximation. Other choices for the direct-exchange term phase are discussed in the following paragraphs. The angular algebra reduction of Eq. (4) leads to a partial rate given by

$$A_2(k_1 s, k_2 p) = \frac{8}{\pi} \int_0^{k_{\max}} \frac{dk_1}{k_2} \left| a \sum\!\!\!\!\!\!\!\int_{kp} \frac{R^0(k_1 s, k_2 p; 2s, kp) R^0(1s, kp; 2s, 2p)}{\Delta\varepsilon - k^2/2} - \frac{a}{3} \sum\!\!\!\!\!\!\!\int_{kp} \frac{R^1(k_2 p, k_1 s; 2s, kp) R^0(1s, kp; 2s, 2p)}{\Delta\varepsilon - k^2/2} + T(k_1 s, k_2 p) \right|^2 , \tag{6}$$

where $\Delta\varepsilon=\varepsilon_\alpha-\varepsilon_\beta^+=436.3$ eV for O^{5+}. For a nonorthogonal basis set the factors $T(k_1s,k_2p)$ arise from the overlap terms in $\langle\Psi_\beta^+ \mid V \mid \Psi_\alpha\rangle$ and are given by

$$\begin{aligned}T(k_1s,k_2p)=&-a \sum\!\!\!\!\!\!\int_{kp} \frac{R^0(k_1s,k_2p;2s,kp)R^0(1s,2p;2s,2p)I(2p,kp)}{\Delta\varepsilon-k^2/2}\\ &-b\sqrt{\tfrac{2}{3}} \sum\!\!\!\!\!\!\int_{kp} \frac{R^0(k_1s,k_2p;2s,kp)R^1(1s,2s;2p,2p)I(2p,kp)}{\Delta\varepsilon-k^2/2}\\ &+\frac{a}{3} \sum\!\!\!\!\!\!\int_{kp} \frac{R^1(k_2p,k_1s;2s,kp)R^0(1s,2p;2s,2p)I(2p,kp)}{\Delta\varepsilon-k^2/2}\\ &+b\sqrt{\tfrac{2}{27}} \sum\!\!\!\!\!\!\int_{kp} \frac{R^1(k_2p,k_1s;2s,kp)R^1(1s,2s;2p,2p)I(2p,kp)}{\Delta\varepsilon-k^2/2}\ .\end{aligned} \quad (7)$$

For an orthogonal basis set the factors $T(k_1s,k_2p)$ arise from one-electron matrix elements as well as two-electron matrix elements involving the $1s$ core electrons within $\langle\Psi_\gamma^{2+} \mid V \mid \Psi_\beta^+\rangle$ and are given by

$$\begin{aligned}T(k_1s,k_2p)=a[&R^0(k_1s,1s;2s,1s)-\tfrac{1}{2}R^0(k_1s,1s;1s,2s)-R^0(k_1s,2s;2s,2s)-R^0(k_1s,2p;2s,2p)\\ &+\tfrac{1}{6}R^1(k_1s,2p;2p,2s)]R^0(1s,k_2p;2s,2p)/(\Delta\varepsilon-k_2^2/2)\ .\end{aligned} \quad (8)$$

In Eqs. (6)–(8) R^λ is the usual Slater radial integral and I is a radial overlap integral. The intermediate state sums are evaluated by summing over a finite number of bound states and then integrating over continuum states using a finite number of points in regions of important momenta. The vanishing energy denominators D are replaced by $\lim_{\eta\to 0}(D+i\eta)^{-1}=\mathrm{P}D^{-1}-i\pi\delta(D)$, where P indicates principal-value integration.

Additional intermediate states $\mid \Psi_\beta^+\rangle$ were included in the calculation for the partial double Auger rate $A_2(k_1s,k_2p)$. Of equal magnitude to those terms in Eq. (6) arising from $\mid 1s^22skp\ {}^3P\rangle$ are those terms coming from $\mid 1s^22pks\ {}^3P\rangle$ with $\Delta\varepsilon=424.2$ eV. Also included are the intermediate states $\mid 1s^22pkd\ {}^3P\rangle$ which only contribute through configuration interaction in the initial state. Further intermediate states, such as $\mid 1s2lk'l'k''l''\ {}^3P\rangle$, were not included since in the calculation by Simons and Kelly[11] for the Li $2s^22p\ {}^2P$ double Auger rate the analogous states were found to make relatively small contributions.

Contributions to the partial double Auger rates for the $1s2s^22p\ {}^3P$ term of O^{4+} are given in Table I. The results are generated using the natural phase approximation and an orthogonal basis set of excited orbitals. The first row of Table I labeled $k_1s\ k_2p$ provides the magnitudes of various contributions to this particular partial double Auger rate. Contributions from imaginary two-electron matrix elements, listed in column 2, include the imagi-

TABLE I. Partial double Auger rates for the $1s2s^22p\ {}^3P$ term of O^{4+} using a natural phase approximation and an orthogonal basis set of excited orbitals.

Final continuum waves	Contribution from imaginary two-electron matrix elements (10^{10} s^{-1})	Contribution from real two-electron matrix elements (10^{10} s^{-1})	Contribution from real one-electron matrix elements (10^{10} s^{-1})	Total contribution from real matrix elements (10^{10} s^{-1})	Total partial rate (10^{10} s^{-1})
$k_1s\ k_2p$	6.04	3.61	11.59	28.05	34.09
$k_1p\ k_2s$	9.03	4.06	22.02	44.96	53.99
$k_1p\ k_2d$	6.41	1.17	1.01	4.35	10.76
$k_1d\ k_2p$	9.23	0.28	0.35	1.18	10.41
$k_1d\ k_2f$	0.31	0.50	0.00	0.50	0.81
$k_1f\ k_2d$	0.10	2.04	0.00	2.04	2.14
$k_1f\ k_2g$	0.11	0.09	0.00	0.09	0.20
$k_1g\ k_2f$	0.15	0.05	0.00	0.05	0.20
$k_1g\ k_2h$	0.00	0.01	0.00	0.01	0.01
$k_1h\ k_2g$	0.01	0.01	0.00	0.01	0.02
Total	31.39	11.82	34.96	81.24	112.63

nary $\delta(D)$ part of the first two terms on the right-hand side of Eq. (6), as well as the same suite of contributions coming from other intermediate states. Contributions from real two-electron matrix elements, listed in column 3, include the real PD^{-1} part of the first two terms on the right-hand side of Eq. (6), as well as the same type of contributions coming from other intermediate states. Contributions from real one-electron matrix elements, listed in column 4, include the terms in Eq. (8) for $T(k_1s,k_2p)$, as well as similar contributions from the intermediate state $|1s^22pks\,^3P\rangle$. The total contribution from real matrix elements, listed in column 5, is not the sum of columns 3 and 4, but the sum of the square of the amplitudes which are used to calculate those results. Finally the total partial rate, listed in column 6, is the sum of the imaginary contribution in column 2 and the real contribution in column 5. As seen in Table I, convergence of the partial-wave expansion for the double continuum state is rapid. The double Auger rate for the $1s2s^22p\,^3P$ term of O^{4+} is found to be 1.13×10^{12} sec^{-1}.

Double Auger rates for the $1s2s^22p\,^3P$ term in the Be isoelectronic sequence are given in Table II for various phase and basis-set choices. The minimum phase approximation is found by squaring the difference between the absolute values of the direct and exchange amplitudes and corresponds to maximum destructive interference. The maximum phase approximation is found by squaring the sum of the absolute values of the direct and exchange amplitudes and corresponds to maximum constructive interference. The range of results found in each row of Table II is surprisingly large. Although the proper choice of phase remains a long-standing problem, it appears that differences in this essentially second-order perturbation theory calculation due to the choice of basis set may reflect the influence of unincluded higher-order effects.

Resonant-recombination auto-double-ionization cross sections in the Li isoelectronic sequence are given in Table III for recombination through the $1s2s^22p\,^3P$ term. The resonance energies and single Auger rates found in Table III are calculated using the atomic structure code of Cowan[16] based on the initial state of Eq. (5). The resonant-recombination cross section, σ_{RR}, found in column 5 of Table III, by the principle of detailed balance, can be written in terms of the single Auger rate as

$$\sigma_{RR}=\frac{2\pi^2}{k^2\Delta}\left[\frac{G_F}{2G_I}\right]A_1(1s2s^22p\,^3P\rightarrow1s^22skp\,^3P)\ , \tag{9}$$

where $G_F=9$ is the statistical weight of the final term in the recombination process, $G_I=2$ is the statistical weight of the initial term of the $(q+1)$ ion, and an energy bin width $\Delta=2.0$ eV is a good estimate for the energy resolution found in a typical crossed-beam experiment. For O^{5+} the resonant-recombination cross section for the $1s2s^22p\,^3P$ term is 5.29×10^{-19} cm^2, while by contrast the recombination cross section to the $1s2s^22p\,^1P$ term, which we have ignored, is only 4.68×10^{-22} cm^2. Cancellation between the direct and exchange amplitudes accounts for the unusually small single Auger rate found for the $1s2s^22p\,^1P$ term. Other ions in the isoelectronic sequence show the same behavior in regard to the cross-section ratio for the two multiplets. The double Auger rates, found in columns 6 and 7 of Table III, are the minimum and maximum rates taken from the calculations summarized in Table II. The resonant-recombination auto-double-ionization cross sections, σ_{RRAD}, found in columns 10 and 11 of Table III, are given by

$$\sigma_{RRAD}=\sigma_{RR}B\ , \tag{10}$$

where the branching ratios, B, are calculated by dividing the double Auger rates by the sum of the single and double Auger rates. We ignore the radiative decay of the $1s2s^22p\,^3P$ term because it is negligibly small.

In conclusion we find that the value of the resonant-recombination auto-double-ionization cross section involving the $1s2s^22p\,^3P$ term decreases as one moves to higher ionization stages in the Li isoelectronic sequence. Of interest in the light of previous speculations[5] of features in the cross section for O^{5+} is that our calculated maximum cross section is 4.9×10^{-21} cm^2 and this corresponds to a ratio of σ_{RRAD} to the background $2s$ direct ionization cross section of 0.65%. This is consistent with the most recent experiment,[17] which appears to be limited by a noise level of at best 1%, and is not able to detect the double Auger induced contribution in O^{5+}. We also find that the ratio of the maximum σ_{RRAD} to the background increases only slightly as one goes to higher ionization stages in the Li isoelectronic

TABLE II. Double Auger rates for the $1s2s^22p\,^3P$ term in the Be isoelectronic sequence.

Ion	Natural phase and orthogonal basis set (10^{12} s^{-1})	Minimum phase and orthogonal basis set (10^{12} s^{-1})	Maximum phase and orthogonal basis set (10^{12} s^{-1})	Natural phase and nonorthogonal basis set (10^{12} s^{-1})	Minimum phase and nonorthogonal basis set (10^{12} s^{-1})	Maximum phase and nonorthogonal basis set (10^{12} s^{-1})
Be	5.19	3.34	5.26	3.22	1.58	3.29
B^+	3.28	1.94	3.33	1.84	0.754	1.88
C^{2+}	2.17	1.21	2.21	1.12	0.392	1.15
N^{3+}	1.55	0.832	1.58	0.775	0.255	0.800
O^{4+}	1.13	0.579	1.15	0.527	0.151	0.547
F^{5+}	0.865	0.432	0.883	0.405	0.111	0.421
Ne^{6+}	0.689	0.335	0.703	0.312	0.081	0.325

TABLE III. Resonant-recombination auto-double-ionization cross sections in the Li isoelectronic sequence for recombination through the $1s2s^22p\ ^3P$ term.

Ion	Resonance energy	Single Auger rate to $1s^22s$ (10^{13} sec^{-1})	Single Auger rate to $1s^22p$ (10^{13} sec^{-1})	σ_{RR} (10^{-19} cm^2)	Range of double Auger rates (10^{12} sec^{-1})		Range of branching ratios		Range of σ_{RRAD} (10^{-19} cm^2)		Maximum % of direct ionization cross section
					min	max	min	max	min	max	
Be^+	105.1	1.42	3.51	7.50	1.58	5.26	0.0311	0.0964	0.233	0.723	0.19
B^{2+}	167.5	2.37	5.19	7.87	0.75	3.33	0.0098	0.0422	0.077	0.332	0.46
C^{3+}	243.8	3.11	6.48	7.10	0.39	2.21	0.0041	0.0225	0.029	0.160	0.64
N^{4+}	333.1	3.69	7.48	6.17	0.26	1.58	0.0023	0.0139	0.014	0.086	0.60
O^{5+}	436.3	4.15	8.25	5.29	0.15	1.15	0.0012	0.0092	0.006	0.049	0.65
F^{6+}	553.2	4.51	8.85	4.54	0.11	0.88	0.0008	0.0065	0.004	0.030	0.75
Ne^{7+}	683.9	4.81	9.31	3.92	0.08	0.70	0.0006	0.0049	0.002	0.019	0.92

sequence. Current experiments would thus appear to gain little by changing charge state. In addition, although correlation effects are generally stronger in heavier atomic systems, the general increase in the number of ionization channels available may tend to mask specific σ_{RRDA} processes. We do hope, however, that direct measurements of double Auger processes will be attempted in the future, since they will certainly shed light on some of the theoretical problems found in calculating pure electron-electron correlation phenomena.

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