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AUTOIONIZING STATES IN THE ALKALIS*

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INTRODUCTION

Atoms and ions are frequently excited to states which in turn de-excite spontaneously by electron rather than photon emission. This radiationless process, known as autoionization, is energetically possible when an excited state is degenerate in energy with a state in some ionization continuum. The simultaneous excitation of two or more loosely bound electrons (multiple-excitation) or the excitation of a single more tightly bound inner shell electron (core-excitation) are two processes which give rise to discrete or quasi-discrete states that are embedded in a continuum. The rate of autoionization is determined by the magnitude of the matrix element connecting the discrete state with states of the continuum of equal energy. It can be shown that for such matrix elements to be non-zero, the states being mixed must be of the same parity and total angular momentum and moreover, for allowed autoionizing transitions (i.e., transitions induced by the inter-electronic electrostatic or Coulomb interaction) in the L-S coupling approximation, L and S must also be conserved. Forbidden autoionization, which is induced by the magnetic (relativistic) interactions, allows greater freedom on L and S but the rates for such processes (in near neutral systems) are smaller by $\sim \alpha^4$ than those for the allowed process. Slight deviations from the L-S coupling scheme can also cause appreciable transition rates through second-order perturbations due to neighboring levels of the same parity and J but different L and S which themselves have allowed autoionization decay channels. The discrete energy electrons ejected in the autoionization process have kinetic energies equal to the difference in binding energies of the initial state and the

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final state consisting of the residual ion. Under certain circumstances it is also possible for radiative decay processes to compete favorably with autoionization for the de-excitation of core-excited states, this being particularly true, of course, if the state is metastable against autoionization.

The simplest alkali, lithium, can be used as an illustrative example of the types of states involved in the present investigation and the decay processes that are possible. Figure 1 shows a partial energy level diagram of the Li atom in which the region between 55-67 eV has been expanded to show the positions of some core-excited states. Doublet and quartet states associated with core-excited configurations of the type $1s2sn\ell$ ($n \geq 2$) and $1s2pn\ell$ ($n \geq 2$) are shown to lie at least 50 eV above the $(1s^2 2s)^2S$ ground state. Such highly excited states, being adjacent to continuum states of the type $(1s^2)^1S_0 + k^2\ell$, are energetically permitted to autoionize. The selection rules on the allowed autoionization process (in the L-S coupling approximation) permit most doublet states (states of the type $1s2p(^1, ^3P)n\ell^2P$ are an exception) to autoionize rapidly via the Coulomb interaction e.g., $(1s2s^2)^1S_{1/2} \rightarrow (1s^2)^1S_0 + k^2S_{1/2}$. The quartet states are, however, metastable against autoionization but can autoionize via the weaker magnetic interactions. An investigation (using beam-foil excitation) of these metastable autoionizing states in high Z lithiumlike ions has previously been reported by the Oak Ridge group.¹ Radiative (E1) transitions among quartet states in Li have been observed in several sources including beam-foil² and beam-gas³ sources. Wavelength determinations from such experiments have produced important information on the relative spacings of these quartet states. In addition, a recent electron spectroscopic measurement of the $(1s2s2p)^4P$ states energy by Ziem et al.⁴ can be used to establish an accurate, experimentally determined, absolute energy scale. Buchet et al.⁵ have observed a line in the foil-excited spectrum of Li which probably corresponds to the $(1s^2 2p)^2P^0 - (1s2p^2)^2P$ transition permitting a determination of the energy of the upper, core-excited $(1s2p^2)^2P$ state - a metastable autoionizing doublet. Such transitions, which produce the so-called satellite lines, become more probable for high Z ions where they have been observed in laboratory and astrophysical plasmas⁶ as well as in the beam-foil source.⁷ Berry et al.⁸ have also observed several E1 transitions between autoionizing and metastable autoionizing states in Li. These transitions are characterized by anomalously large linewidths due to the short lifetime of the autoionizing state involved in the transition. It appears that the unclassified line at $\lambda 4590$ observed in the work of Berry et al.⁸ probably arises from the transition $1s(2s2p^3p)^2P^0 - (1s2p^2)^2P$. The present work describes binding energy measurements on rapidly decaying autoionizing states in neutral or near neutral alkali systems (Li, Na, Mg^+ , and K) in

which a core electron has been excited from the outermost closed subshell into a higher orbital. Preliminary results for Li and Na have been published.⁹

OTHER INVESTIGATIONS

Autoionizing states have been experimentally investigated by methods involving either the process of excitation (for example, inelastic and elastic electron scattering and photoabsorption techniques or the process of de-excitation (for example, ejected electron spectroscopy of a target or projectile and, under certain circumstances, photon emission spectroscopy). A summary of these methods is shown in Table I. Whereas a great deal of work has been done on autoionizing states in helium and the inert gases, the alkalis until recently have received little attention outside of photoabsorption investigations which, although accurate, suffer from the limitations set by the electric dipole selection rules. Only very recently have particle-impact (electron- and ion-impact) excitation methods been applied to the alkalis in order to complement the photoabsorption studies by exciting optically forbidden states as well as optically allowed ones. For example, besides the present ion-impact (projectile de-excitation) investigation of Li there have been recent studies of the autoionizing states in Li following ion-impact excitation⁴ (target de-excitation), beam-foil excitation¹⁰ and electron-impact excitation¹¹. A similar situation exists in the case of Na where two recent electron-impact studies^{12,13} have produced results which appear to be in very good agreement with the present ion-impact measurements. Similar comparisons can be made for Mg⁺ and K which have been recently studied by electron-impact methods at Freiburg¹⁴ and Southampton¹⁵ respectively. On the theoretical side of the picture considerably less data exists. In the case of Li there exists a definite need for calculations on allowed autoionizing doublet states associated with configurations of the type $1s2s3s$ and $1s2s3d$ for example. A recent calculation by Junker¹⁶ for the energy of the lowest lying doublet, $(1s2s^2)^2S$ appears to be in good agreement with the present measurements. Severe computational difficulties arise for core-excited states in the alkalis heavier than Li due to the rather large-spin-orbit interaction associated with the p-subshell vacancy in these core-excited states. This situation can lead to considerable state mixing and an eventual breakdown of the L-S coupling scheme for some configurations. Despite this, calculations by Weiss¹⁷, who used the Superposition of Configurations (SOC) method, appear to be quite successful for states in Na and Mg⁺. The breakdown of L-S coupling in certain configurations of K is apparent from the work of Martin *et al.*¹⁸. Clearly more theoretical work will be necessary in the future before many of the lines present in particle-impact excited alkali spectra can be positively identified.

Table I

EXPERIMENTAL METHODS OF PRODUCTION AND DECAY OF AUTOIONIZING STATES

EXPERIMENTAL METHOD

Spectroscopic measurements are made on electrons emitted in the autoionizing decay-in-flight of collisionally-excited atoms or ions which themselves form a fast moving, unidirectional beam. Intense and well collimated singly charged alkali ion beams were obtained from the Universities Isotope Separator at Oak Ridge (UNISOR) and then passed at 70 keV through a differentially pumped gaseous target cell ($p \sim 20$ m torr) which served to both charge exchange (if necessary) and collisionally excite the components of the beam. The foil-excitation method could not be used in this work due to the severe degradation in resolution brought about in part by such effects as multiple scattering and energy straggling of the beam in the foil target. In the present arrangement the states are produced and rapidly decay within a small viewing window defined by the entrance slit system of an electron spectrometer. Thus discrete energy electrons emitted at a mean polar angle of 42.3° from a small section (length 0.15 mm, diameter 1.5 mm) of the excited beam were collected and energy analyzed by means of a cylindrical mirror electrostatic analyzer. A schematic of the essential apparatus is shown in Figure 2. The diameters of the inner and outer electrodes of this analyzer are 5.7 cm and 12.2 cm. The spread in the polar angle of acceptance of the analyzer, defined by the size and spacing of the entrance slit system, was restricted in these experiments to 0.09° in order to reduce spectral peak broadening due to kinematic effects, the most important source of resolution loss. This severe collimation in polar angle is, however, partially compensated for by the use of an azimuthal acceptance angle of 120° . The spectra were accumulated by applying a linear voltage ramp signal simultaneously to the outer electrode (the inner electrode was held at ground potential) and to a divider chain connected to the input of a MCA via a linear gate. This gate was opened by the signal pulses from an electron multiplier located behind the spectrometer exit slit. In this configuration the MCA can be gated by pulses from a beam-current integrator. Fringe field correction rings are spaced at logarithmic radial intervals between the inner and outer electrodes of the analyzer. A triple layer of annealed conetic magnetic shielding reduces the ambient field inside the spectrometer to an average value of ~ 3 mG. The whole spectrometer was housed in a vacuum chamber maintained at a residual pressure of $\sim 10^{-8}$ m torr. The spectrometer was calibrated using well established energies of electron-impact excited Auger lines from inert gas sources. Such target lines are, of course, far less broadened by kinematic effects than the projectile lines studied in the actual experiment. Line broadening due to the fast moving projectile beam source results from any source of transverse or longitudinal spread in the beam velocity vector. Because of the finite spread in the polar angle of acceptance, the most important source in the present

Figure 2. Schematic of the apparatus shown the cylindrical mirror analyzer in a plane containing the beam axis.

experiment is due to the transverse spread. Of course, beam collimation should be made comparable to the polar angle spread. Multiple scattering effects in the target will also produce divergence in the beam direction and the use of the gaseous targets as opposed to foil targets in the present work was found necessary to reduce both multiple scattering and energy straggling effects. Even so, the linewidths observed in the Li spectrum, for example, are approximately a factor of four larger than the estimated natural linewidths, thus precluding any lifetime determinations. One consequence of the use of the rather small source volume determined by the stringent polar acceptance angle collimation conditions is that the instrument is effectively more sensitive to the observation of fast decaying states than states with decay times much larger than the "window" defined by the entrance slit system.

RESULTS

Lithium

A typical spectrum of electrons emitted by a 70 keV Li beam

undergoing autoionizing decay-in-flight following collisional excitation in a He gaseous target cell is shown in Figure 3. The spectral features are associated with the allowed autoionizing decay of doublet states in Li involving core-excited configurations of the type $1s2sn\ell$ ($n \geq 2$) and $1s2pn\ell$ ($n \geq 2$). Electron energies are expressed with respect to the rest frame of the emitting atom and the corresponding excitation energies of the core-excited states are obtained by adding the ionization potential (5.39 eV) of Li to the peak energies. Table II shows the present results (excitation energies in eV) along with other particle-impact and photoabsorption measurements. Theoretical estimates, where available, are also shown in the table. In the present work the energies of the optically allowed $1s(2s3p^3p)^2p^0$, $1s(2s2p^1p)^2p^0$ and $(1s2s^3s)3p^2p^0$ states, which have been measured accurately by Ederer et al.¹⁹ using a photoabsorption method, have been used to establish an accurate absolute energy scale. The lowest energy feature of the Li spectrum arises from the autoionizing decay of the optically forbidden doublet, $(1s2s^2)^2S$. The presently measured excitation energy of this state, 56.31 ± 0.03 eV, is in agreement with the recent calculation of Junker¹⁶ of 56.43 eV and in excellent agreement with the ion-impact (target de-excitation) measurement of Ziem et al.⁴ who obtained a value of 56.35 ± 0.05 eV. A beam-foil measurement, under conditions of limited resolution, by Bruch et al.¹⁰ yielded a result of 56.0 ± 0.5 eV. The energy of the optically forbidden $(1s2p^2)^2D$ state is measured to be 61.04 $\pm$ 0.03 eV to be compared with a value of 60.99 ± 0.05 eV obtained by Ziem et al.⁴. A theoretical estimate by Nicolaides²⁰ of 62.0 eV appears to be somewhat high. The other features of the spectrum which are less intense and partially blended are also recorded in Table II and an estimated uncertainty of $\sim \pm 0.1$ eV should be attached to their energy values. Due to the relative closeness of the $(1s2s)^1S$ and $(1s2p)^3P$ limits one might expect considerable mixing between Li configurations involving these parents. Further calculations are required to identify many of the lines observed in the Li spectrum.

Sodium

Figure 4 shows a spectrum of electrons emitted in the autoionizing decay-in-flight of a 70 keV Na beam following collisional excitation in a differentially pumped He gas target cell. The spectral features are associated with the decay of states formed from core-excited configurations of the type $2p^53sn\ell$ ($n \geq 3$) and $2p^53pn\ell$ ($n \geq 3$), in which a vacancy exists in the L shell. Accurate photoabsorption results of Wolff et al.²¹ for the energies of the $(2p^53s^2)^2p^0$ and $(2p^53s^3p)3d$ optically allowed states have been used to establish an accurate absolute energy scale in the present work. Table III shows the present results (estimated uncertainties in most cases ~ 0.1 eV) compared

Figure 3. Electron spectrum from collisionally-excited Li. Energies expressed relative to the rest frame of emitting atom.

to the photoabsorption results of Wolff *et al.*²¹ and theoretical predictions by Weiss.¹⁷ The major new feature of the present work appears to be the presence of lines in the spectrum associated with the optically forbidden core-excited configuration $2p^5 3s 3p$ for which some SOC calculations have been made by Weiss¹⁷ for the quartet states. The present measurements seem to agree quite well with these computed values and in addition to the quartet assignments it appears that we observe doublets associated with this configuration for which theoretical estimates are not presently available. Very recent electron-impact results (as yet unpublished) from both the Freiburg¹² and Southampton¹⁵ groups indicate very good agreement with the present ion-impact measurements. As stated earlier, computational difficulties do arise for the heavier alkalis due to the rather large spin-orbit interaction associated with the vacancies in the p-subshell of these atoms, a situation which can imply considerable state mixing. However Connerade *et al.*²² have pointed out that it appears from their photoabsorption data that L-S coupling may still be substantially valid for many core-excited configurations of the Na atom although these authors do also discuss alternative coupling schemes. The effect of this

Table II. AUTOIONIZING STATES IN LITHIUM

Excitation Energy (eV)		Theory	Assignment
Present	Other Expt.		
56.31	56.35 ^a , 56.2 ^b	56.43 ^c	(1s2s ²)2S
58.91	58.910 ^d , 58.91 ^a	58.96 ^e	1s(2s2p ³ p) ² p°
60.40	60.396 ^d , 60.39 ^a	60.60 ^e	1s(2s2p ¹ p) ² p°
61.04	60.99 ^a	62.0 ^f	(1s2p ²) ² D
61.8	-	-	
62.0	61.99 ^a	-	(1s2s3s) ² S ?
62.42	62.419 ^d , 62.39 ^a	62.46 ^e	(1s2s ³ S)3p ² p°
62.5	-	-	
62.9	62.89 ^a	-	(1s2s3d) ² D ?
63.0	-	-	
63.2	-	-	
63.3	63.356 ^d	63.36 ^e	(1s2s ³ S)4p ² p°
63.4	-	-	
63.6	63.54 ^a	-	(1s2p ²) ² S ?
63.7	-	-	
63.8	63.753 ^d	-	(1s2s ³ S)5p ² p°
64.0	-	-	
64.1	-	-	
64.3	-	-	

^a Reference 4	^c Reference 16	^e Reference 17
^b Reference 10	^d Reference 19	^f Reference 20

Figure 4. Electron spectrum from collisionally-excited Na. Electron energies expressed with respect to rest frame of emitting atom. Inset shows typical calibration spectrum obtained for electron-impact excitation of Kr.

Table III. AUTOIONIZING STATES IN SODIUM

<u>Present</u>	<u>Electron-Impact^a</u>	<u>Photoabsorption^b</u>	<u>Theory^c</u>	<u>Assignment</u>
30.77	30.77	30.768	30.88	$(2p^5 3s^2)^2 p^{\circ}_{3/2}$
31.0	30.93	30.934	31.35	$(2p^5 3s^2)^2 p^{\circ}_{1/2}$
32.67	-	-	32.72	$(2p^5 3s 3p)^4 S$
32.8	32.83	-	-	-
33.08	33.06	-	33.09	$(2p^5 3s 3p)^4 D$
33.2	33.26	-	-	-
33.38	33.57	-	33.32	$(2p^5 3s 3p)^4 P$
33.9	33.99	-	-	-
34.1	-	34.128	-	-
34.4	34.42	34.468	-	-
34.79	-	34.628	-	-
35.6	35.64	35.566, 35.631	35.59	$(2p^5 3s^3 p) 4s^4 P$
35.8	-	35.768, 35.827	35.79	$(2p^5 3s^3 p) 4s^2 P$
35.99	35.98	35.985	35.97	$(2p^5 3s^3 p) 3d^2 P$
36.1	36.21	36.217	36.17	$(2p^5 3s^1 p) 4s^2 P$ or $(2p^5 3s^3 p) 3d$
36.46	36.30	36.394	36.50	$(2p^5 3s^3 p) 4p$ or $(2p^5 3s^3 p) 5d$
36.82	36.86	36.906	-	$(2p^5 3s^3 p) 4p$ or $(2p^5 3s^3 p) 5s$
37.0	-	37.070	36.99	$(2p^5 3s^3 p) 4d$ or $(2p^5 3p^3)^4 P$
37.3	37.23	37.292, 37.273	37.21	$(2p^5 3s^3 p) 5d$ or $(2p^5 3p^2)^4 D$
37.5	37.56	37.497	37.50	$(2p^5 3p^2)^4 S ?$
37.8	-	37.800	-	-
38.1	38.12	-	-	-

^aReference 13^bReference 21^cReference 17

rather strong spin-orbit interaction can be seen in the present spectra in the presence of partially resolved structures caused by the comparatively large fine structures (~ 0.1 - 0.2 eV) and in the presence of quite strong lines arising from the decay of states which are at least partially quartet in character. Such "quartet" states are of course metastable against autoionization in the L-S coupling approximation but become shorter lived in the heavier alkalis due both to mixing with fast decaying doublets of the same parity and J and to increases in the rates for the forbidden autoionization process brought about by the stronger magnetic interactions. Clearly, with the new ion-impact and even higher resolution electron-impact data available, theoretical estimates of the energies of core-excited states in Na and its isoelectronic ions are urgently required.

Single Ionized Magnesium

Part of the electron spectrum obtained following collisional excitation of a 70 keV Mg^+ beam is shown in Figure 5. Many of the spectral features are associated with the autoionizing decay-in-flight of core-excited states of neutral and singly ionized magnesium with configurations of the type $2p^5 3s^2 n\ell$ ($n \geq 3$) and $2p^5 3s n\ell$ ($n \geq 3$) respectively. The Mg^+ spectrum is, of course, similar in structure to that of Na with further increases in fine structure splittings and state mixing effects. The Mg spectrum can, however, be quite complicated due to the fact that for this alkaline-earth system there exists more than one final state for the residual Mg^+ ion. The photoabsorption results of Esteve and Mehlmann²³ for optically allowed transitions have been used in the present work to establish an absolute energy scale. The present ion-impact results for Mg^+ are shown in Table IV (estimated accuracy ~ 0.1 eV) along with photoabsorption values and the results of SOC calculations by Weiss.¹⁷ There is good agreement again between the present results and theory for the three quartets and six doublets associated with the optically forbidden configuration $2p^5 3s 3p$. Very recent, and as yet unpublished, electron-impact measurements by the Freiburg group¹⁴ should provide a good comparison with the present work.

Potassium

Figure 6 shows a spectrum of electrons emitted in the autoionizing decay-in-flight of core-excited states of K following the passage of a 70 keV K^+ beam through a He gas target cell. The spectrum is very rich in lines, most of which are incompletely resolved from one another. Recent work using electron-impact¹⁵ and photoabsorption methods²⁴ should be very valuable in the identification of some of the features in this spectrum. The lines represent the autoionizing decay of states formed from core-excited

Figure 5. Electron spectrum from collisionally-excited Mg and Mg⁺.

Table IV

AUTOIONIZING STATES OF SINGLY IONIZED MAGNESIUM

Excitation Energy (eV)		Theory ^b	Assignment
Present	Other Expt. ^a		
49.90	49.898	50.24	(2p ⁵ 3s ²) 2p _{3/2}
50.1	50.166		(2p ⁵ 3s ²) 2p _{1/2}
53.2	-	53.25	(2p ⁵ 3s3p) ⁴ S
53.9	-	53.95	⁴ D
54.3	-	54.38	⁴ P
54.5	-	54.55	2p ⁵ (3s3p ³ p) ² D
54.7	-	54.74	² P
55.2	-	55.34	² S
56.8	-	57.03	2p ⁵ (3s3p ¹ p) ² D
57.1	-	57.19	² P
57.3	-	57.39	² S
57.8	-	-	
59.3	59.30	-	(2p ⁵ 3s3p) 4s ² p
59.5	-	-	(2p ⁵ 3s3p) 3d ² p
59.8	-	-	(2p ⁵ 3s ¹ p) 4s or (2p ⁵ 3s ³ p) 3d
50.9	60.92	-	(2p ⁵ 3s ³ p) 3d
61.4	61.38	-	(2p ⁵ 3s ¹ p) 3d
61.5	-	-	2p ⁵ 3s4p or (2p ⁵ 3s ³ p) 5s

^aReference 23

^bReference 17

Figure 6. Electron spectrum from collisionally-excited K. Energies expressed in rest frame of emitting atom.

configurations of the type $3p^5 4s n l$, ($n \geq 3$), $3p^5 3d n l$ ($n \geq 3$), $3p^5 4p n l$ ($n \geq 3$), etc. It has been shown by Martin *et al.*¹⁸ that strong state mixing effects have caused considerable departures from the L-S coupling scheme in the case of the $3p^5 4s 3d$ configuration. It appears, however, that the two peaks indicated are associated with the $J = 1/2$ and $3/2$ levels of the $(3p^5 4s^2) 2P^0$ term which have been observed in earlier photoabsorption work by Beutler *et al.*²⁵ and Hudson *et al.*²⁶

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