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Lifetimes of the metastable autoionizing $(1s\,2s\,2p)\,{}^4P_{5/2}$ states of lithiumlike Al^{10+} and Si^{11+} ions: Comparisons with theory over the isoelectronic sequence $Z = 8$–18*

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Lifetimes of the lowest, most metastable, autoionizing quartet state $(1s\,2s\,2p)\,{}^4P_{5/2}$ of the lithiumlike ions Al^{10+} and Si^{11+} have been measured by the electron spectrometric, time-of-flight technique and are found to be 2.9 ± 0.2 and 2.1 ± 0.1 nsec, respectively. These results can be compared to very recent Dirac-Hartree-Fock lifetime calculations of Cheng, Lin, and Johnson which give 2.58 and 1.84 nsec, respectively. Comparison of our experimental results for the sequence $Z = 8$, 9, 13, 14, 16, 17, and 18 with the new theoretical calculations are now possible with the result that the measured lifetimes tend to be systematically larger than those predicted from theory, after proper allowance for an $M2$ radiation-decay channel is made. Possible reasons for these systematic differences are mentioned.

I. INTRODUCTION

The lifetimes and energies of the lowest lying quartet states in three-electron systems have been investigated extensively both experimentally[1] and theoretically[2] because they are metastable against Coulomb autoionization [as well as against radiative decay, in the case of the lowest quartet state $(1s2s2p)\,{}^4P^0_{5/2}$]. These states autoionize only through the spin-orbit and spin-spin interactions in three-electron systems (for $J = \frac{5}{2}$, spin-orbit matrix elements also vanish). Hence their study has provided a sensitive test of the spin-orbit and spin-spin interactions in three-electron systems, including the effects of electron correlation. Comparison of experiments (the present experiments in particular) with theoretical predictions shows that accuracy in wave functions for the three-electron system is extremely critical for predicting level lifetimes.

Interest in the quartet states of three-electron systems stems historically from the early observations[3] of the existence of a bound state associated with the three-electron helium ion, He^-. Wu[4] suggested that the $(1s2s2p)\,{}^4P^0_{5/2}$ state of He^- would be bound, and since it was expected to be also metastable against both autoionization and radiation, it accounted for the observations. Subsequent studies such as those by Wu and Shen,[5] Holøien and Midtal,[6] Garcia and Mack,[7] Holøien and Geltman,[6] and Junker and Bardsley[8] were directed at the study of those highly excited states which were metastable against autoionization. Herzberg and Moore[9] suggested that lines that had been observed[10] in the lithium optical spectrum and which failed to fall into the normal classification scheme for singly excited configurations were in fact due to radiative transitions between doubly or core-excited states of lithium that were metastable against autoionization. Recently these types of transitions have been studied[11] optically in other three-electron ions using the foil-excitation method. Satellite lines on the long-wavelength side of the resonance lines of two-electron ions were first observed by Edlén and Tyrén[12] who interpreted them as arising from radiative transitions between doubly or core-excited states and the normal singly excited three-electron ion states. Similar optical lines have also more recently been observed in both laboratory plasma[13] and solar sources.[14]

Feldman and Novick[15] experimentally studied the forbidden autoionizing decay of metastable autoionizing states in several of the alkali atoms. The binding energy of the $(1s2s2p)\,{}^4P^0_{5/2}$ level in lithium was determined and its lifetime measured by observing the change in the charge states of an electron-impact excited lithium beam following autoionization in flight. Blau, Novick, and Weinflash[16] used a similar method to measure the lifetime of the corresponding state in He^-. The $(1s2s2p)\,{}^4P^0_{5/2}$-state lifetime was studied in other three-electron ions ($Z = 4$–8) by Dmitriev *et al.*,[17] who also used a time-of-flight technique to track the charge change in a fast, foil-excited, accelerator beam. Manson,[18] Balashov *et al.*,[19] and most recently Cheng, Lin, and Johnson[20] have calculated the lifetime of this lowest-lying quartet state in several ions of the lithium sequence. The present experimental method is similar to that used by Dmitriev *et al.* with the important exception that our method includes energy analysis of the emitted autoionization electrons which we believe represents a considerable advance over the previous time-of-flight methods. In fact, the lifetimes of Dmitriev *et al.* showed a residual dependence on

beam velocity, which of course corresponded to a systematic error of unknown origin in their experiments. We are nonetheless indebted to these earlier experimenters for being the first to demonstrate the possibility of measuring Auger lifetimes by applying time-of-flight methods to fast beams of excited heavy ions.

II. EXPERIMENTAL METHOD

Our electron spectrometric time-of-flight technique is basically unchanged from that used in our previous lifetime experiments[1] on other ions in this isoelectronic sequence. The technique has been described most completely in two of our previous papers[21] and only basic procedures and certain significant changes need to be noted here. Beams of 15.7-MeV Al ions and 19.2-MeV Si ions of ~20-nA intensity were obtained, using the new universal negative-ion source at the Oak Ridge Tandem Accelerator. The ions were stripped and excited in movable, ~25 μg/cm^2 carbon foils prior to entering the paraxial viewing region of our axially mounted cylindrical-mirror electron-energy analyzer. Electrons emitted at polar angles of ~40.3 ± 0.4 deg by spontaneously decaying ions as they passed through the viewing region were dispersed in an approximately axial plane by the radial electrostatic field between the grounded inner and negatively charged outer analyzer plates. An axially mounted channel electron multiplier detected the energy-selected electrons passing through a symmetrically located exit slit. The spectrum was scanned by applying a ~1-kHz linear ramp voltage to the outer plate, a portion of which voltage was routed to a multichannel analyzer through a linear gate opened by electron multiplier counts. Thus a spectrum of electrons versus a scale proportional to analyzer voltage (and hence electron energy) was obtained.

The usual procedure of studying the decay in flight of energy-selected Auger electrons as a function of time after excitation was adopted. Subsequent to locating the peaks in the Auger electron spectrum at about 1.13 keV and 1.30 keV for Al^{10+} and Si^{11+}, respectively (both energies are expressed in the rest frame of the emitting ion), the spectrometer exit slits were widened to improve "flat-topping" of the well-resolved peaks and to improve counting statistics. For electrons of sharply defined energy, use of an exit slit wider than the entrance slit results in a more constant (flat-topped) detector response versus analyzer voltage near a spectral peak than for symmetric slit widths, a practice useful in reducing point scatter in our lifetime experiments. Small, beam-dependent background corrections were made by detuning the spectrometer voltage slightly, while beam-independent background corrections were made by measurements during intervals when the beam was deflected to a remote beam stop. As usual, the energy resolution of the spectrometer was insufficient to spectrally separate the $J = \frac{1}{2}$ and $\frac{3}{2}$ decays from the desired $J = \frac{5}{2}$ decay, but this could be accomplished by taking data accumulated at sufficient distances downstream ($\gtrsim$1 cm) so that the $J = \frac{5}{2}$ state was temporarily resolved.

III. RESULTS

The decays in flight of the $J = \frac{5}{2}$ component of the $(1s2s2p)\ ^4P^0$ state in Al^{10+} and Si^{11+} were followed out to 2.3 and 3.0 decay lengths, respectively, and were observed to correspond to single exponentials within the limits set by systematic and statistical error considerations, just as in the case of the similar curves we have obtained for other ions in this isoelectronic sequence.[1] The energy-loss corrected beam velocities for Al^{10+} and Si^{11+} were 1.04×10^9 cm/sec and 1.14×10^9 cm/sec, respectively, and led to corresponding lifetime values of 2.9 ± 0.2 nsec and 2.1 ± 0.1 nsec. The errors given are estimates of combined systematic and counting statistical errors and are comparable to similar estimates for our previous experiments. The principal systematic error arises from point scatter in the decay curves stemming from beam steering effects.[21] Data-point normalization is provided by the ratio of observed electron counts to the integrated beam current measured in a Faraday cup situated at the center of the electron spectrometer, and slight variations in beam trajectory as it passes through the spectrometer-viewing region cause jitter in this ratio. The jitter is believed to be the principal cause of error, and accounts for the bulk of the errors quoted above.

IV. DISCUSSION AND COMPARISONS WITH THEORY

The Al^{10+} and Si^{11+} metastable state lifetimes are of course intermediate between those of O^{5+} and Ar^{15+}, which are the extreme cases we have studied previously. Because of the rapid onset[22] of an $M2$ radiative decay channel whose rate scales as ~Z^8, however, these cases are actually somewhat more useful as a test of the Z dependence of Auger rate calculations than are the more mixed cases of S^{13+}, Cl^{14+}, and Ar^{15+}. There are two different aspects of such calculations to be tested: (i) the importance of relativistic effects and (ii) the importance of electron correlation effects. The very recent theoretical work of Cheng, Lin, and Johnson[20] has provided a sufficiently detailed calculation for the first time.

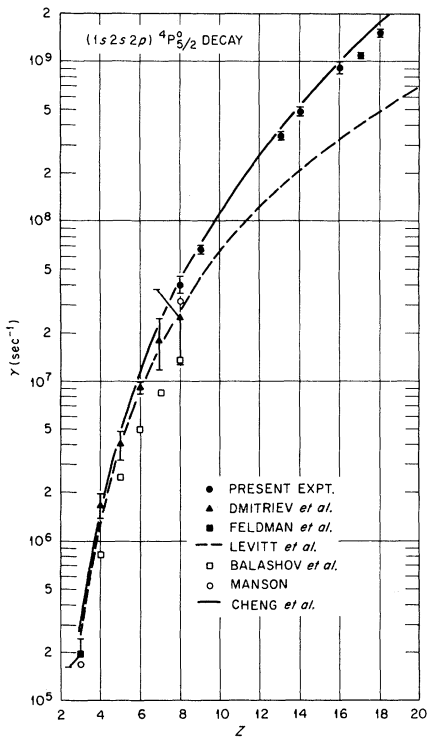

FIG. 1. Decay rate (inverse mean life) of the $(1s2s2p)^4P_{5/2}$ state of lithiumlike ions vs Z, allowing for both Auger and $M2$ decay channels.

We are dealing here with one of the most fundamental examples of core-excited states—the $(1s2s2p)\ ^4P_{5/2}$ state of lithiumlike ions. Because of the requirements that both J and parity be conserved in autoionizing transitions to the final state $(1s^2)\ ^1S_0+k$, only $^2F_{5/2}$ states are possible for the emitted electron, requiring $\Delta S=1$, $\Delta L=2$. These changes rule out Coulomb, spin-orbit and spin-other-orbit autoionization processes, but the decay can be induced by the tensor part of the spin-spin interaction.[2] Hence systematic measurements of the mean life of this state against autoionization in ions of the lithium sequence measures directly the size of the tensor part of the electronic spin-spin interaction in three-electron systems.

The questions of the effects of electron correlation and of relativistic and other higher-order effects for high-Z ions arise. For example, it has been demonstrated that the autoionization branching ratio is reduced by the competing forbidden ($M2$) radiative transition $(1s^22s)\ ^2S_{1/2}$-$(1s2s2p)\ ^4P_{5/2}$. Such radiation (x ray) has been observed for both S^{13+} and Cl^{14+} ions,[22] and this decay channel yields lifetimes in excellent agreement with our previously published electron spectrometric results.[1]

The detailed autoionization and $M2$-rate calculations for the $(1s2s2p)\ ^4P_{5/2}$ state in ions from $Z=3$–26 made very recently by Cheng, Lin and Johnson[20] permit a meaningful comparison of our experimental rates and theory for the first time. Figure 1 presents a variety of experimental and theoretical results including our new and previously published measurements. (Table I shows a more detailed comparison of some experimental and theoretical values, together with branching ratios which also take into account the new results of Ref. 20.) Results from lower-Z ions, based on ion-charge-changing methods, obtained by a group at Columbia University (Novick *et al.*[1]) and at the Moscow State University (Dmitriev *et al.*[1]) are also given, as are earlier theoretical results of Manson[18] and of Balashov *et al.*[19] The dashed curve gives an extrapolated semiempirical fit $\alpha(Z-\sigma)^3$ to the low-Z data proposed by Levitt *et al.*,[1] who considered only the autoionizing channel, where σ is a suitable screening constant. Even allowing for the neglect of the $M2$ channel, however, it is clear that the recent calculations of Cheng *et al.*[20]

TABLE I. Lifetimes of the $(1s2s2p)\ ^4P_{5/2}$ state in some three-electron ions.

Ion	Lifetime (nsec) Expt.	Theory	Autoionization branching ratio	Lifetime ratio expt./theory
O^{5+}	25±3[a], 40±20[b]	23.1,[c] 31[d], 75[e]	0.993[c]	1.08±0.13
F^{6+}	15±1[a]	13.5[c]	0.988[c]	1.11±0.07
Al^{10+}	2.9±0.2[f]	2.58[c]	0.947[c]	1.12±0.08
Si^{11+}	2.1±0.1[f]	1.84[c]	0.929[c]	1.14±0.05
S^{13+}	1.1±0.1[g]			
	1.1±0.2[g]	0.993[c]	0.883[c]	1.11±0.10
Cl^{14+}	0.91±0.04[h]			
	0.95±0.20[g]	0.743[c]	0.855[c]	1.12±0.05
Ar^{15+}	0.66±0.04[h]	0.563[c]	0.823[c]	1.17±0.07

[a]Reference 21.
[b]Reference 17.
[c]Reference 20.
[d]Reference 18.
[e]Reference 19.
[f]This work.
[g]Reference 22; Reference 1.
[h]Reference 26.

represent a marked improvement over the semiempirical extrapolation, and yield a Z dependence nearer $(Z-\sigma)^4$ than $(Z-\sigma)^3$. It is also obvious that the most recent theoretical results systematically overestimate the experimentally measured decay rates for the case $Z \geq 5$, while giving significantly better results for $Z=8$ than the calculation of Manson[18] or of Balashov *et al.*[19]

The calculation of Balashov *et al.* employed simple screened Coulomb functions for the bound electrons. This approach is not expected to yield particularly accurate results. Manson used a single-configuration, separable, single-orbital type, variational wave function and a purely nonrelativistic Hamiltonian in his calculations. Cheng *et al.*[20] used Dirac-Hartree-Fock wave functions and a relativistic Hamiltonian. It is the opinion of these authors that the remaining discrepancy between their theory and our experiments may be due to the neglect of initial-state electron correlation effects as well as distortion by the outgoing, final-state F-wave continuum electron. Though such correlation effects ought to be most significant at low Z, it appears that the lifetime measurements are sufficiently precise to permit sensitive measurements of such effects for ions as heavy as Ar^{15+}.

The last column of Table I shows the remarkably constant ratio of the present experimental to theoretical lifetimes.[20] In a plot of this ratio versus Z, it would be possible to represent the ratio as a constant of ~1.17 within the quoted error bars. The mean ratio for the cases cited is 1.14, and the standard deviation from this mean is ±0.05. Hence it is possible to speak of a systematic difference between theoretical and experimental decay rates of $14\% \pm 5\%$.

The possibility of undiscovered systematic errors of this magnitude of course arises. However, this error would have to have remarkably constant-ratio properties in order to prevail for different experimental configurations ranging from those involving 2.5-MeV oxygen beams from the Oak Ridge Tandem Accelerator to those involving 80-MeV argon beams from the Oak Ridge Isochronous Cyclotron, and involving different target thicknesses, different laboratory decay lengths, and different energy-loss corrections. Cascades, which often apparently lengthen beam-foil lifetime measurements, would also have to have remarkably coincidental behavior in order to lengthen the experimental lifetimes in the same ratio for such a wide variety of atomic systems and beam velocities. We also note that in experiments using very similar techniques in which the decay rates for $M1$ radiation in the $2\,^3S_1$ states of heliumlike ions have been measured,[23] the experimental lifetimes are systematically *smaller* than the presumably well-founded theoretical values.[24] Hence we look to the calculations as the origin of the remaining difference between theory and experiment.

The various calculated $M1$ rates[20,24] for the $2\,^3S_1$ states of the heliumlike ions essentially agree. The $M2$ rates[20,24] (except for small radial integral differences) for the $2\,^3P_2$ heliumlike and $^4P_{5/2}$ lithiumlike states also agree well with each other. Cheng *et al.*[25] believe that their fuller accounting of relativistic effects neither resolves the tendency for the calculated $2\,^3S_1$ $M1$ radiative lifetimes to exceed experimental ones nor the tendency for the calculated $^4P_{5/2}$ Auger lifetimes to underestimate the present experimental ones.

The question is thus raised about whether remaining differences between theoretical and experimental Auger transition probabilities can be accounted for by correcting the Dirac-Hartree-Fock wave functions to allow for correlation effects. These corrections would presumably include both correlation among the three electrons initially present and distortion of the final atomic wave function because of the finite overlap of the outgoing-F-electron wave function with that of the residual heliumlike ion. A complete calculation of this type has apparently not yet been made. It is plausible that such correlation effects should decrease with Z because of the increasing dominance of nucleus-electron interactions and reduction in final-state wave function overlap. It therefore seems peculiar that the ratio between experimental and theoretical lifetimes stays so constant, not only on experimental grounds but on theoretical ones as well.

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