Lifetime Measurements in Multiply Ionized Atoms

Recent advances have made possible spectroscopic and decay rate studies of isoelectronic sequences with charge states ranging from negative ions to very heavy one-electron ions. The highly charged ions constitute a new theoretical regime where interactions which are negligible in neutral atoms can become dominant. Applications in a number of areas require a reliable atomic structure data base for these systems. The existing capabilities for time-resolved lifetime measurements in multiply ionized atoms are reviewed and compared with theoretical results based on *ab initio* methods, as well as with semiempirical approaches.

Key Words: *lifetimes, transition probabilities, relativistic effects, accelerator-based experiments*

INTRODUCTION

During the 1980's, significant progress has been made in the spectroscopy of multiply ionized atoms. Technical advances have made possible the spectroscopic study of systems ranging from negative ions to one-electron uranium, U^{91+}. Spectroscopic classification work has been completed through extremely high stages of ionization for some isoelectronic sequences, and new classes of states have been added to the spectroscopic data for previously studied systems. Furthermore, it is now possible to perform time-integrated studies (to determine excitation energies and relative transition probabilities) as well as time-resolved studies (to measure lifetimes and absolute transition probabilities) for practically any

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ion. These developments have added a new dimension to the field of atomic physics, and provide opportunities and challenges for the 1990's.

These recent trends have been brought about by the development of new light sources and technical advances in the use of traditional light sources. The work has been motivated by urgent needs for atomic data in specific applications. These sources include the use of fast ion beams, tokamak- and laser-produced plasmas, recoil ions from ion–atom collision studies at high energy, electron beam ion sources (EBIS), electron beam ion traps (EBIT), and electron cyclotron resonance ion sources (ECRIS), as well as improved use of more traditional spark and vacuum arc sources.

In some ways highly ionized atomic systems constitute a new theoretical regime, since interactions that are negligible in neutral atoms can dominate in these systems. For high central charge, relativistic and quantum electrodynamic interactions can become large. Similarly, "forbidden" decay processes such as magnetic dipole (M1), electric and magnetic quadrupole (E2 and M2, respectively), and two photon decay, all of which have rates that scale with high powers of Z (being typically proportional to Z^6–Z^{10}), may rival or sometimes even exceed electric dipole (E1) transition rates, the latter scaling as Z ($\Delta n = 0$) or Z^4 ($\Delta n = \pm 1$). To meet these new conditions, calculational methods are undergoing important conceptual and computational refinements which require experimental verification.

In this connection, the accurate specification of wavelength and energy level data does not insure correct predictions of other atomic structure properties, most notably atomic transition probabilities and lifetimes. Measurements of the latter quantities are particularly crucial here, since they provide absolute rate values which can be used to normalize relative transition probabilities obtained by time-integrated techniques. In some cases, such lifetime measurements have even been used to indirectly measure fine structure and Lamb shift energy separations. The accurate specification of the transition probabilities for highly ionized atoms has important applications which must be addressed in the 1990's. One application is in the determination of elemental abundances in the interpretation of astrophysical spectra, an area which is expected to expand rapidly with the deployment of the Hubble Space Tele-

scope as well as future devices of high-resolution stellar spectroscopy in the vacuum-ultraviolet (VUV) and X-ray regions. Moreover, atomic data, including lifetimes and transition probabilities for multiply ionized atoms, are badly needed in the determination of stellar opacities. Another application involves the diagnosis and modeling of controlled fusion plasmas. Here transition probabilities are necessary both to determine concentrations and to predict the rates of dielectronic recombination, radiative cooling, etc. Other areas which require atomic lifetimes are in the development of X-ray lasers and in the interpretation of atomic collision studies.

In this article we shall review current experimental techniques in the context of a number of recent measurements, compare these results with the capabilities of theoretical methods, and consider the roles of experiment, theory, and semiempirical systematization in the development of the comprehensive data base of lifetime information that is needed.

FAST ION BEAM EXCITATION METHODS

Although many new spectroscopic light sources have impacted lifetime studies by providing essential energy level data, they either operate in a time-integrated way or lack the necessary time resolution for lifetime measurements in multiply charged ions. Here the only generally applicable technique is provided by thin foil excitation of a fast ion beam, which frequently permits measurements on the 10^{-12} s scale. This method has been reviewed by several authors in recent years,[1-6] so we shall provide only a brief summary of the general method and mainly stress recent developments through descriptions of specific measurements.

The beam-foil method utilizes an isotopically pure, monoenergetic, collimated beam of ions from a particle accelerator. After further ionization and excitation in a thin foil, the ions emerge into a field-free and collision-free region in which they spontaneously decay. The photons thereby emitted are wavelength analyzed and detected as a function of distance from the foil, which provides the time base. By varying the accelerator energy, virtually any ionization stage can be reached, and the high density within the foil leads to copious production of inner shell vacancies and

multiply excited states. However, the excitation is nonselective, which leads to cascade repopulation of the levels, and the Doppler effects inherent in the source limit the spectral resolution and may result in line blending.

In the early 1970's it was pointed out[7] that the lifetime data reported prior to that time could contain substantial errors. For certain transitions which were easy to treat theoretically, beam-foil data showed poor agreement with the calculated values. The conclusion was that cascading and blending were severe problems in these early measurements, but a number of subsequent experimental developments have greatly improved this situation. Thus, the method of refocussing the spectrometer[8] to compensate for Doppler effects in the light source resulted in a quite favorable spectral resolution. Moreover, the cascade problems were practically solved by means of the so-called ANDC method,[9] discussed below. Parallel to these developments, a number of new experimental methods were introduced which largely eliminated cascading effects. Of these, the beam-laser excitation method[10] is undoubtedly the most efficient. However, while this technique, which involves selective optical excitation, has been successful in the case of transitions in neutral and singly ionized atoms, it has only in exceptional cases (Lamb-shift measurements for one-electron systems) been extended to higher ionization states. Another interesting method[11,12] combines the beam-foil and beam-laser techniques. Here a two-step excitation process is used in which the ionization and nonselective excitation in a carbon foil is periodically followed by selective excitation of a transition in the foil emergent ion, achieved by means of a chopped tunable dye laser. Thus, although the populations are affected by cascades, the cascade contributions vanish from the differences between the populations with laser on and with laser off. After some initial measurements[11] in 1974, this technique has recently been further developed and applied[12] to obtain cascade-free lifetime data for C III and C IV.

ANDC MEASUREMENTS IN HEAVILY CASCADED SYSTEMS

Cascading effects can be reliably corrected for if the energy level scheme and the decays of both the primary level and those re-

populating it can be measured. If the primary level is very long lived relative to those feeding it (e.g., in the case of metastable states), cascading does not pose a problem. Similarly, if the cascading levels have much longer decay times than the primary lifetime, the latter can be determined by curve fitting techniques. However, this simple technique is unreliable if the various lifetimes involved in the decay chain are close to each other. In such a case more refined methods of analysis must be applied.

If there is a limited number of measurable strong cascades, then the ANDC (arbitrarily normalized direct cascade) method[9] permits the lifetime to be extracted. The method is illustrated in Fig. 1, from Reistad *et al.*[13] The primary level is repopulated by a set of (here three) higher levels, and all of their decay curves $I(t)$ are measured under the same conditions, but with arbitrary intensity

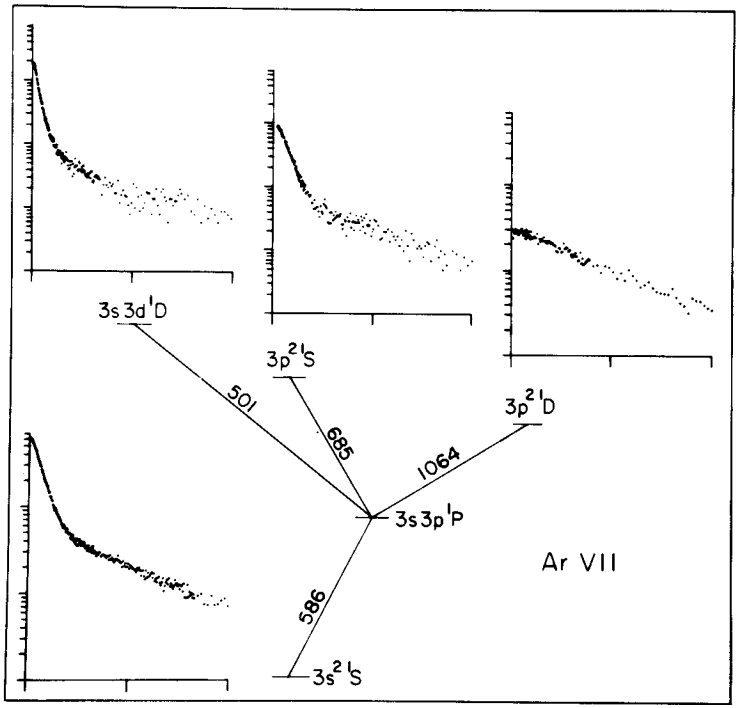


FIGURE 1 Partial Grotrian diagram for the singlet system of Mg-like Ar VII and decay curves of the primary level 3s3p ^{1}P and of the levels directly repopulating it, 3s3d ^{1}D, 3p^2 ^{1}S and 3p^2 ^{1}D, from Reistad *et al.* (Ref. 13).

normalization in any convenient decay branch. These decay curves are then interrelated through the population equation, which can be written in terms of the measured decay curves (denoting the primary by i and its cascade feeders by j) as

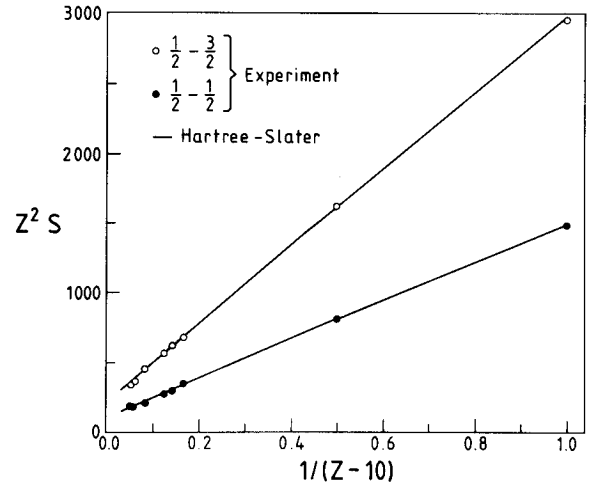
$$dI_i(t)/dt = -I_i(t)/\tau_i + \sum_{j>i} \xi_j I_j(t) \qquad (1)$$

By virtue of the large number of common time coordinates t at which the decay curves are measured, Eq. (1) comprises an overdetermined set of linear equations, from which the parameters τ_i and ξ_j (the desired lifetime and the relative normalization factors) can be determined. Indirect cascading is automatically included within the time dependence of the direct cascades. A very efficient computer program for ANDC analyses, CANDY, is available.[14] In the case shown in Fig. 1 a decomposition of the decay curve of the first excited level, 3s3p ^{1}P in Ar VII, into a sum of exponentials yielded an "apparent" lifetime of 0.195 ± 0.03 ns, a value that strongly deviated from the theoretical result of 0.130 ns. The latter[15] is based on elaborate multiconfiguration Hartree–Fock (MCHF) calculations and should therefore be quite reliable. The problem was solved by the ANDC analysis,[13] however, which resulted in a lifetime of 0.132 ± 0.005 ns, in excellent agreement with theory. While this fact is very satisfying, it is still important to extend the experimental work to other systems so as to explore the merits and shortcomings of various theoretical methods.

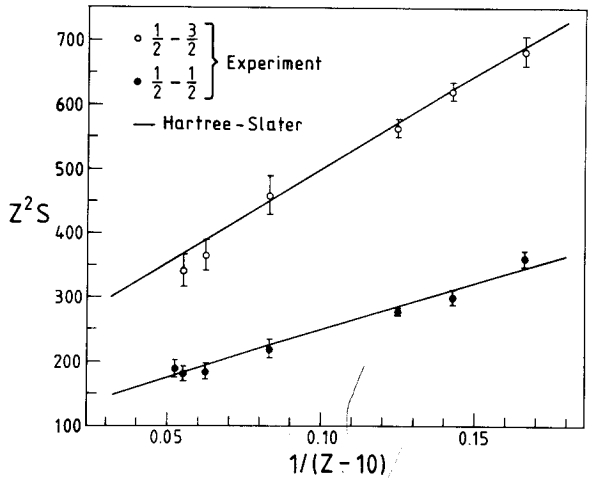
The ns ^{2}S–np ^{2}P resonance transitions in alkali-like isoelectronic sequences provide an experimental case which—because of extensive cascading—is difficult if studied by curve fitting techniques, but is ideally suited to ANDC analysis. Early measurements which utilized only curve fitting methods systematically yielded overestimates of the lifetime, and discrepancies between theory and experiment were generally dismissed as experimental artifacts. While more careful measurements and ANDC analysis[13,16] resulted in much better agreement with theory, a recent experimental study[17] of the 3s ^{2}S–3p ^{2}P resonance doublet in Na-like Ti, Fe, Ni and Cu showed that there existed a small systematic disagreement with the theoretical predictions, based on the relativistic multiconfiguration Dirac–Fock (MCDF) method. This fact was unexpected

because of the relative simplicity of Na-like ions (only one electron outside closed shells). However, the new data,[17] being consistent with the results[13,16] for Na-like S, Cl and Ar, further displayed the same trend as the highly accurate (with quoted uncertainties far smaller than 1%) cascade-free beam-laser measurement[18] for the 3s–3p multiplet in Na I. It was noted some years ago that the latter value differs from an accurate theoretical (MCHF) result[19] by 1.8% which is about 8 times larger than the experimental uncertainty. The possibility of a small systematic error in the experiment was sometimes brought up, but the beam-laser result[18] has recently been corroborated by another very accurate measurement using the pulsed-laser technique.[20]

Whereas there thus existed a small but significant discrepancy between the results from careful measurements and elaborate *ab initio* theoretical calculations, it was demonstrated that semiempirical calculations[21,22] could provide a clue to the problems. The latter theoretical treatment utilized the Hartree–Slater method and incorporated a realistic model potential to account for core polarizability, spin-orbit interaction, and exchange effects. Empirical core polarizabilities and fine structure splittings were used in the calculation to construct the model parameters, and the wavefunctions were forced to yield the empirical energies. Thus the method is not *ab initio*, but it provides excellent predictions over the entire range from the neutral atom to the highest measured stage of ionization. The magnitudes of the individual terms in the model potential indicate that core polarization is the dominant effect that accounts for the lengthening of the lifetimes (in comparison with the *ab initio* data). This suggests that electron correlation has not been adequately included in the MCDF calculations. Recently, Guet *et al.*[23] reported accurate relativistic many-body perturbation calculations for the 3s ^{2}S–3p ^{2}P lines in Na-like ions. The transition probabilities obtained in this way are in very good agreement with semiempirically determined values[22] and the experimental data.[13,16–18,20] To the latter a new, accurate beam laser result[24] for Mg II can now be added. The present situation is illustrated in Figs. 2(a) and 2(b). Here the quantity Z^2S (where S is the line strength in a.u.) is plotted as a function of $1/(Z - 10)$. Such a representation, recommended by Edlén,[25] often results in nearly straight lines which can be reliably used for interpolation



(a)



(b)

FIGURE 2 (a) Graphic representation of transition probabilities for the 3s $^2S_{1/2}$–3p $^2P_{1/2,3/2}$ resonance doublet in the Na I isoelectronic sequence. The quantity Z^2S (where S is the line strength in a.u.) is plotted vs. $(Z - 10)^{-1}$. Here 10 is an estimate of the screening of the nucleus by the passive electrons. The experimental points originate from Refs. 13, 16–18, 20, 24, whereas the lines connect theoretical data, Ref. 22. (b) An enlargement of the high-Z region of Fig. 2(a).

or extrapolation purposes. It is also possible to use it together with a method of "isoelectronic smoothing" to further reduce the statistical errors of the individual measurements.[26]

The transition probabilities in the Ne isoelectronic sequence represent another interesting example of recent progress. The structure of Ne-like ions has been the subject of many studies in recent years. This activity was largely stimulated by observations of laser action in the VUV and soft X-ray regions for highly charged Ne-like ions.[27] In such work inverted populations between levels of the excited $2p^53s$ and sp^53p configurations have been achieved, resulting in stimulated emission at wavelengths as low as 100–200 Å. Until about 10 years ago, detailed knowledge of the energy level structure of Ne-like ions was limited to the first 5–6 ionization stages, whereas now quite substantial progress has been made. Since the decay times of the various $2p^53s$ and sp^53p levels are important parameters in the laser research, several lifetime studies have been undertaken. For example, Westerlind *et al.*[28,29] have made careful lifetime determinations for the $2p^53p$ levels in Ne-like S VII, Cl VIII and Ar IX. Their data show that lifetimes based on ANDC corrected data can differ by as much as 30% from those obtained from the simple decomposition of decay curves into sums of exponentials. In other cases ANDC and multiexponential fits give similar results. This fact further underscores the importance of ANDC analyses—only in this way is it possible to avoid the ambiguities that may enter in the case of less rigorous methods for data analysis. In Fig. 3 some experimental lifetime data, from Westerlind *et al.*[28,29] and Träbert[30] (who studied Ne-like Ti XIII), are displayed. The variation with the nuclear charge Z follows a regular pattern and the experimental results are consistent with recent theoretical values. The $3p\ ^1S_0$ level, which energetically lies far above the other nine 3p-levels, is particularly interesting. Its excitation energy is surprisingly difficult to calculate accurately unless quite elaborate *ab initio* methods[31] are employed. In the X-ray laser experiments this level was populated to a much lesser degree than expected from calculations,[27] which led to many additional experimental and theoretical studies. The experimental and theoretical lifetimes for this level now show good agreement, however.

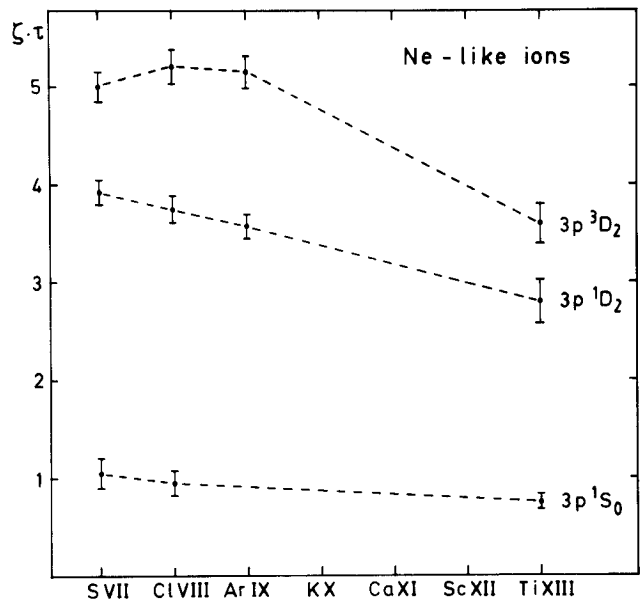


FIGURE 3 Experimental lifetimes (in ns), multiplied by the spectrum number ζ (net charge of the core) for three 3p-levels in Ne-like ions S VII, Cl VIII, Ar IX and Ti XIII (Refs. 28–30).

INTERCOMBINATION LINES IN ALKALINE EARTH-LIKE SPECTRA

Resonance transitions for highly ionized members of alkali-like and alkaline earth-like spectra (i.e., those with one and two valence electrons) become very short lived, with increasing Z in an isoelectronic sequence, and thus increasingly difficult to measure by time of flight methods. In alkaline earth-like spectra, intersystem transitions of the form $ns^2\ {}^1S_0 - nsnp\ {}^3P_1$ are forbidden in pure LS coupling by the $\Delta S = 0$ selection rule, but this breaks down with increasing Z due to the (relativistic) spin-orbit-induced singlet-triplet mixing. In terms of a singlet-triplet mixing angle ϑ which characterizes the normalized amplitudes of the wavefunctions, the resonance transition probability is proportional to $\cos^2\vartheta$ and the intercombination transition probability is proportional to $\sin^2\vartheta$.

Thus, for reasonably high Z, these intercombination transitions become significant and they provide a measurable decay channel between the ns^2 and nsnp configurations even when the decay time of the $ns^2\ ^1S-nsnp\ ^1P$ resonance line is too short to allow reliable measurements.

Collision rates in plasmas are often comparable to intercombination decay rates. Thus, the measurement of intensity ratios between intercombination lines and allowed resonance lines can be a useful diagnostic probe of plasma conditions (electron densities, electron temperatures, etc.). Since the atomic systems of particular relevance to plasma diagnostics are often characterized by significant configuration interaction effects, lifetime measurements are especially valuable. Several examples of such experiments have already been reviewed in this journal.[32,33]

One type of example concerns the $2s3p\ ^3P_J$ levels in the Mg sequence.[36] In both cases the three $J = 0,1,2$ levels all have E1-allowed $\Delta n = 0$ transitions to a $^3S_1$ level, but only the $J = 1$ level has an intercombination channel to the $^1S_0$ ground state due to the spin-orbit induced mixing of the $^1P_1$ and $^3P_1$ states. Thus, through differential measurements of the J-dependent lifetimes of the three ^3P_J levels, it has been possible to determine the intercombination rates. Theoretical calculations of these rates are particularly challenging because they involve intermediate coupling, configuration interaction, relativistic corrections, and can be affected by plunging levels.[37]

Lifetimes corresponding to intercombination transitions of the form $ns^2\ ^1S_0-nsnp\ ^3P_1$ have been measured to very high stages of ionization, as has recently been discussed by Träbert *et al.*[38] These singlet-triplet intercombination transitions are very closely related to the corresponding singlet-singlet resonance transitions $ns^2\ ^1S_0-nsnp\ ^1P_1$, the two differing only through singlet-triplet mixing modifications and wavelength factors (in the single configuration nonrelativistic approximation the transition moments are the same), which can be written as

$$[\tau\lambda^{-3}]_{res}/[\tau\lambda^{-3}]_{int} = \tan^2\vartheta. \qquad (2)$$

It has been shown[39] that, to a good approximation, the value for the mixing angle obtained from lifetime data in Eq. (2) is identical

to that obtained from energy level data alone, which is given by

$$\tan\vartheta = \sqrt{1 + W^2} - \sqrt{W^2},$$
$$W \equiv [E_2 - 3E_1 + 2E_0]/\sqrt{2}[E_2 - E_0], \tag{3}$$

where E_2 and E_0 are the energies of the $J = 2$ and 0 triplet levels and E_1 is the centroid of the $J = 1$ singlet and triplet levels. Thus, if the lifetime of either the intercombination or the resonance line is known, then the lifetime of the other can be reliably predicted using only energy level data. Since the decay times of the intercombination and resonance lines often differ by three to four orders of magnitude, this can permit a conveniently measured lifetime to be used to determine a neighboring lifetime that is either too long or too short to be studied by time of flight methods.

For the metastable $2s2p\ ^3P_1$ level in Be-like ions, which via spin-forbidden transitions de-excites to the $2s^2\ ^1S_0$ ground state, lifetime measurements have been made[40] for a broad range of ionization stages, from Be-like C to Xe, and the experimental data agree with theory. For the corresponding level in the Mg I sequence, $3s3p\ ^3P_1$, the range is narrower, but lifetime measurements include Mg I–Si III and Fe XV–Zn XIX (for details see Ref. 38). Similarly, for the $4s4p\ ^3P_1$ level in the Zn-like ions, lifetime measurements[41] are available only for Kr VII, Mo XIII and Ag XVIII, but some additional work is being undertaken. In all these cases experiment and theory are in satisfactory agreement. An interesting point worth mentioning is that after the identification of the intercombination transition, lifetime measurement and data evaluation are not complicated. Thus, cascading levels play a minor role because their lifetimes are usually too short to significantly affect the decay curve of the intersystem transition.

An interesting recent example concerns alkaline earth-like spectra that have been observed for the 62 electron Sm I isoelectronic sequence. This involves a type of behavior that exists only for highly ionized members of this sequence. At the neutral end of the sequence the ground configuration is $4f^65s^25p^66s^2$. As Z increases the electrons in $n = 5$ and $n = 6$ shells gradually begin to

occupy the half-filled 4f shell until a $4f^{14}5s^2$ ground configuration, and thus a two valence electron structure, is reached. Similarly, the highly stripped Nd-like ions (60 electrons) will assume a $4f^{14}$ closed shell ground state and the Pm sequence (61 electrons) will exhibit an alkali-like spectrum, the ground configuration and the first excited one being $4f^{14}5s$ and $4f^{14}5p$, respectively. The suggestion that these "hyperalkali" and "hyperalkaline earth" systems exist, and that their transitions could lead to large radiation losses in fusion plasmas containing high Z contaminants, was originally made by Curtis and Ellis,[42] and experimental searches for them have been carried out.[43]

The wavelengths and lifetimes for the Pm sequence were predicted by Curtis and Ellis[42] and for the Sm sequence by Curtis.[44] Measurements by Träbert and Heckmann,[43] using the beam-foil method, have supported the wavelength predictions of the resonance transitions in the Pm sequence for Au XIX and for the intercombination lines in the Sm sequence for Os XV, Ir XVI, Pt XVII and Au XVIII. Decay times have been reported for the intercombination lines in the Sm sequence, which are a factor of 2 shorter than predictions made using MCDF methods, but in good agreement with results obtained using Hartree–Fock methods with relativistic corrections. Thus the results again indicate that correlation effects may not be adequately included in existing versions of fully relativistic codes.

TRANSITIONS IN HIGHLY IONIZED ONE- AND TWO-ELECTRON ATOMS

Despite great progress in theoretical investigations of one and two electron atoms, fundamental questions remain, some of which can be addressed by the study of level lifetimes in highly ionized members of the H and He sequences. Successes in predicting energy levels at low Z do not test the perturbative inclusion of interactions which involve high powers of Z, particularly in the calculation of decay rates, which depend sensitively on noncentral interactions.

A recent advance in the measurement of lifetimes in highly ionized atoms involves the efficient use of photon-photon coinci-

dence methods. Earlier attempts to use coincidence techniques in beam-foil applications[45] suffered from the low detection efficiency of the optical photons, leading to long data accumulation times, low statistical accuracy, and high accidental backgrounds. In highly ionized atoms, where emitted photons are in the keV range, the use of Si(Li) detectors can greatly improve the detection efficiency.

In recent measurements of the two-photon decay of the 2s $^2S_{1/2}$ and 1s2s $^1S_0$ states in H-like and He-like nickel, Dunford and coworkers[46] were able not only to require a coincidence between the two simultaneously emitted photons, but also to use pulse height analysis to insure that the sum of the energies of the two photons had the proper value. This eliminated all backgrounds and blends (although it did not eliminate cascades).

In the hydrogen-like system, the 2s $^2S_{1/2}$ level decays to the 1s $^2S_{1/2}$ ground state either by emission of two E1 photons (the frequency distribution of which is continuous but the sum equals the 1s–2s energy difference) or a single M1 photon. In the helium-like system the $J = 0 \rightarrow J = 0$ transition (to the $1s^2$ $^1S_0$ ground term) is strictly forbidden to single photon decay, and the 1s2s $^1S_0$ level decays only by two photon emission. The theoretical calculation of the helium-like two photon decay is much more complicated than the hydrogen-like case because of the need to account for two-electron correlations. The values obtained in the Ni experiment[46] were 217.1 ± 1.8 and 156.1 ± 1.6 ps for the hydrogen-like and helium-like decays, which compare with theoretical values[47] of 215.45 and 154.3 ps. A few years earlier Marrus *et al.*[48] reported a beam-foil measurement of the 1s2s $^1S_0$ lifetime for He-like Kr^{34+} which showed the importance of relativistic effects on the decay rate.

The lifetime of the 1s2p $^3P_0$ level in helium-like systems undergoes a number of interesting changes as a function of Z and A. For any He-like ion there is an allowed (E1) decay channel to the lowest triplet term 1s2s $^3S_1$. However, another interesting decay mode is possible when the nuclear spin I differs from 0 (i.e., for odd A nuclei). Here the total angular momentum is not J but $F = J + I$. The $F = I$ states of the 1s2p $^3P_0$ and $^3P_1$ states mix and E1 transitions from 1s2p ^3P ($J = 0$, $F = I$) to $1s^2$ ^1S ($J = 0$, $F = I$) become possible. This "hyperfine quenching" of the $^3P_0$ lifetime is strongly sensitive to A, being particularly significant

above $Z = 20$ where odd A nuclei may have appreciable nuclear magnetic dipole moments. This has been demonstrated in a number of cases, as discussed in Refs. 2 and 49. The experimental values for the rate of the hyperfine-induced decay, as obtained from 1s2p $^3P_0$ lifetime measurements, are in good agreement with theory. At very high Z, the combined corrections for electron self-energy, vacuum polarization and nuclear size reduce the splitting between the 1s2s $^3S_1$ and 1s2p $^3P_0$ levels by more than 20%, which significantly affects the E1 decay rate of the 1s2p $^3P_0$ level. At the same time, the two-photon decay (one E1 photon and one M1 photon) of the 1s2p $^3P_0$ level to the $1s^2$ $^1S_0$ ground state also becomes significant. These facts cause the Z dependence of the lifetime of the 1s2p $^3P_0$ lifetime to be a very useful probe of various interactions.

The sensitivity of the 1s2p $^3P_0$ lifetime to the Lamb shift was exploited by Munger and Gould,[50] who were able to determine the $^3P_0$ lifetime in helium-like uranium. From a detailed comparison with theoretical predictions, an experimental value for the one-electron Lamb shift was extracted.

Another application involved the lifetime measurement by Marrus *et al.*[51] of the hyperfine quenched lifetime of the 1s2p $^3P_0$ level in Ag^{45+}. Here a near level crossing between the $^3P_0$ and $^3P_1$ levels occurs, and the $^3P_0$ lifetime is sensitive to that separation. Thus, in this case, a fine structure splitting is inferred by a lifetime measurement.

MULTIPLY EXCITED LEVELS

Some years ago Andersen and Mannervik[52] in this journal reviewed the spectroscopy of multiply excited levels in three electron systems. More recently a very thorough review of doubly excited states has been written by Mannervik.[53] Such states, which are abundantly populated in ion-gas or ion-foil collisions, have several interesting decay channels. For instance, the quartet levels 1s2snl and 1s2pnl ^4L in Li-like ions are normally not able to autoionize into the $1s^2\varepsilon l$ doublet continuum via the Coulomb mechanism (because of the $\Delta S = 0$ selection rule), but they can decay to lower quartet states by E1 radiative transitions. However, spin-forbidden

E1 transitions to the $1s^2nl\ ^2L$ doublet system as well as spin-orbit induced autoionization are sometimes possible. The decay rate of a given level is then the sum of several transition probabilities which scale differently with Z. Such decay modes are shown in Fig. 4 for the two lowest quartet terms, $1s2s2p\ ^4P$ and $1s2p^2\ ^4P$ in Li-like ions. (This graph is similar to the one shown by Charalambidis *et al.*[54]) For low values of Z the fine-structure levels of the upper term, $1s2p^2\ ^4P$, predominantly decay to the metastable term $1s2s2p\ ^4P$ by means of E1, $\Delta S = 0$ transitions, and their lifetimes are practically independent of J. As Z increases, mixing with doubly excited doublet terms, e.g., $1s2p^2\ ^2P$ and ^2D, as well as spin-orbit induced autoionization grow in importance. These effects being strongly J-dependent, the result is that the three levels of $1s2p^2\ ^4P$ will have quite different total decay probabilities.[55,56] Careful lifetime measurements thus provide valuable checks of modern theories used in describing these non-relativistic and relativistic decay processes. A selection of experimental results,[57] for Li-like B III–Ne VIII, is shown in Fig. 5, where the data are compared

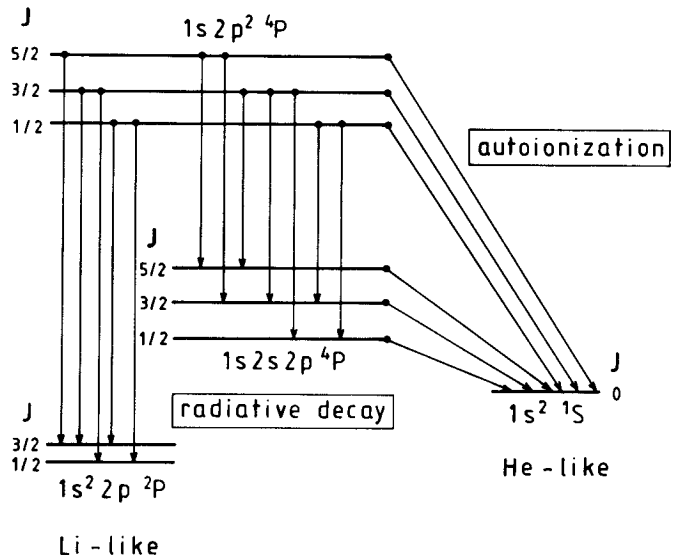


FIGURE 4 Partial Grotrian diagram showing possible decay channels of the low-lying quartet levels in Li-like ions. The energy axis is not to scale.

with the theoretical results of Davis and Chung,[58] obtained by means of the saddle-point complex-rotation method. The difference between the lifetimes for the $J = 1/2$ and 3/2 states is not large, whereas the $J = 5/2$ state has a more rapid decay because of mixing with the $1s2p^2\ ^2D_{5/2}$ level. For completeness it should be mentioned that the three fine-structure levels of the $1s2s2p\ ^4P$ also have interesting decay modes, for instance, spin-orbit and spin-spin induced autoionization as well as forbidden radiative decay.[53,54] Experimental and theoretical material is available on several ions.

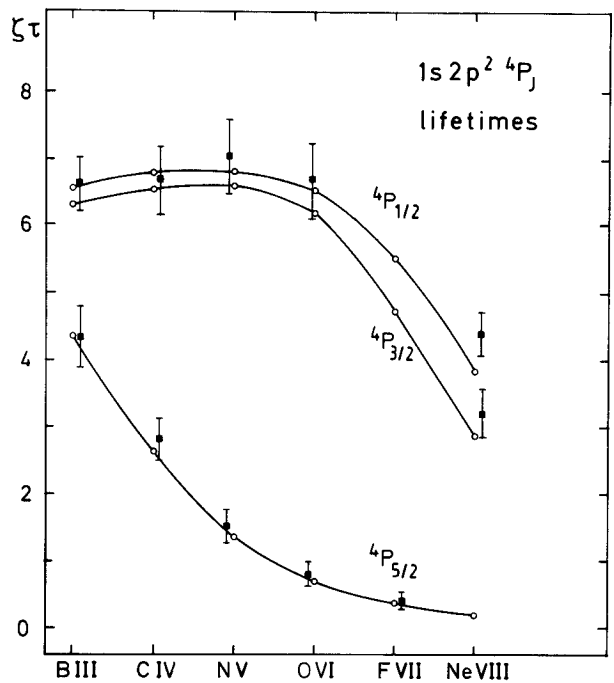


FIGURE 5 Experimental and theoretical lifetimes of the $1s2p^2\ ^4P$ levels in Li-like B III–Ne VIII. Only a selection of all experimental points is shown. (Note that for lower values of Z it is usually not easy to resolve the $J = 1/2$ and $J = 3/2$ levels.) Reference 57 contains all experimental data (except for some early, less reliable results). The theoretical points (circles) were taken from Ref. 58.

In recent years interest has been focussed on doubly excited levels of Na I and Na-like ions. The lowest configurations now are $2p^5 3s3p$, $2p^5 3p^2$, $2p^5 3s3d$ and $2p^5 3s4s$, which have doublet as well as quartet states. The latter have been discussed as candidates for laser action in the extreme UV.[59] Although the spectra are somewhat more complicated than those of the Li-like ions, there exist many similarities. Being energetically above the $2p^6$ limit, the doubly excited levels can autoionize into the $2p^6 \varepsilon l\ ^2L$ continuum. This is the principal decay mode for the $2p^5 nln'l'$ doublet levels which therefore have very short lifetimes (the rates of Coulomb autoionization are typically $10^{13}-10^{15}\ sec^{-1}$). As in the case of Li-like ions, the $\Delta S = 0$ rule precludes this rapid autoionization for the quartet levels. Besides decaying by photon emission to lower quartets, these levels can also de-excite into the "normal" doublet system $2p^6 nl\ ^2L$ by spin-forbidden E1 transitions, or alternatively, autoionize via the spin-orbit or spin-spin interactions. Experimental lifetimes[60] have been reported for Na I and Mg II and these results are generally in good agreement with theory.[61] The work has now been extended to Na-like Cl VII and preliminary results are available for 7 levels.[62] In this spectrum cascading appears to be fairly serious, which shows that many higher-lying doubly excited levels are being populated by the beam-foil excitation process. It is now a challenge to identify these levels and determine their excitation energies and decay probabilities.

CONCLUDING REMARKS

It is sometimes observed that the field of atomic lifetime measurements appears to have waned (at least relative to the intense but sometimes unreliable activity that occurred 15–20 years ago). Although fewer laboratories are engaged in these measurements today, it would be erroneous to conclude that such experiments are of diminished importance, or that theoretical calculations could generally supplant the need for these measurements.

In the present article we have selected a number of cases which should demonstrate that lifetime measurements are still capable of producing very valuable and interesting information. One important and frequently overlooked fact is that the precision of modern ion-beam lifetime studies is much higher now than that

attainable in the 1960's and 1970's. As a consequence of this, quite detailed investigations of the merits and limitations of various theoretical methods can be made, e.g., by comparing recent theoretical and experimental results. As examples of this we have discussed the Na-like and Ne-like ions. Difficulties are often encountered in performing *ab initio* calculations for transitions of low probability such as the intercombination and forbidden lines. Here, a wealth of experimental data also have appeared in recent years. The same holds for levels in extremely highly stripped ions for which "exotic" decay channels are possible because of relativistic, nuclear structure and QED effects. Here lifetime measurements, followed by comparisons of the data with theoretical predictions, provide unique information concerning quantities of fundamental interest. In the case of multiply excited levels it is possible to explore in detail the Z-dependence of a number of nonrelativistic and relativistic decay modes and the coupling of discrete states with continua. The possibility of varying Z (isoelectronic studies) is a particularly attractive factor in accelerator-based experiments.

As a final remark, we must acknowledge that lifetime measurements are frequently difficult and time-consuming. When large blocks of data are needed, for instance, in connection with the opacity program for stellar envelopes,[63] calculations using supercomputers are the only realistic way to proceed. However, even here selected measurements are well worth undertaking so as to provide a firm test of the theoretical uncertainties.

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