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# SEMIEMPIRICAL STUDIES OF ATOMIC STRUCTURE: Progress Report 1 July 1991 - 1 October 1993

L. J. Curtis  
Department of Physics and Astronomy  
McMaster Hall  
University of Toledo  
2801 W. Bancroft St.  
Toledo, Ohio 43606  
tel: (419) 537-2341  
fax: (419) 537-2723  
email: ljc@utphysa.phya.utoledo.edu

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### *Invited Review Article*

- A1. L.J. Curtis and I. Martinson, "Lifetime Measurements in Multiply Ionized Atoms," *Comments in Atomic and Molecular Physics* **24**, 213-33 (1990). [DOE/ER/13958-16]

### *Fast Ion Beam Measurements*

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- A8. K.W. Kukla, A.E. Livingston, F.G. Serpa, A.S. Zacarias, H.G. Berry, R.W. Dunford, E. Kanter, S. Cheng, J. Suleiman, L.J. Curtis, and E. Träbert, "Spectroscopy of Highly-Ionized Atoms Using Position-Sensitive Detection," in *The Physics of Highly-Charged Ions*, AIP Conf. Proceed. No. 274, 398-401 (1993). [DOE/ER/13958-41]
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- A10. R.W. Dunford, H.G. Berry, D.A. Church, T.P. Dineen, M. Hass, C.J. Liu, N. Berrah-Mansour, R.C. Pardo, M.L.A. Raphaelian, L. Young, B.J. Zabransky, and L.J. Curtis, "Precision Spectroscopic Measurements in Few-Electron Ions," *Z. Physik D* **21**, S13-16 (1991). [DOE/ER/13958-26]
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*Calculations*

- A21. C.E. Theodosiou, L.J. Curtis, and M. El-Mekki, "Calculations of  $2p$  Lifetimes in the Li Sequence," *Phys. Rev. A* **44**, 7144-57 (1991). [DOE/ER/13958-23]
- A22. L.J. Curtis, "Cancellation Effects in Alkalilike Doublets," *Can. J. Phys.* **69**, 668-70 (1991). [DOE/ER/13958-21]
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*Accelerator Studies*

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## I. SCOPE OF INVESTIGATIONS

The atomic structure and properties of highly-ionized many-electron systems are studied through the combined use of sensitive semiempirical data systematizations, precision experimental measurements, and specialized theoretical computations. Measurements are made primarily through the use of fast ion beam excitation methods, which are combined with available data from laser- and tokamak-produced plasmas, astrophysical sources, and conventional light sources. The experimental studies are supported and interpreted through large scale *ab initio* calculations. Large blocks of data are systematized and parametrized along isoelectronic, homologous, isoionic, Rydberg, and yrast series.

The use of spectroscopic line intensities as diagnostics for determining thermonuclear plasma temperatures and densities requires laboratory observation and analysis of the spectra of these systems, and computer modeling of both astrophysical and laboratory plasmas requires a comprehensive knowledge of the atomic structure parameters for highly ionized complex systems. Spectroscopic classifications require accuracies exceeding parts per million, which cannot be achieved by standard *ab initio* methods for these highly relativistic many electron systems. Semiempirical systematizations of high precision data can provide these accuracies, and an increasingly large base of such data is becoming available. This program seeks to acquire, systematize, and parametrize large blocks of data to expose and exploit predictive empirical regularities. Trends revealed are tested through well chosen experimental measurements, and *ab initio* methods are used to gain quantitative understanding of the empirical regularities.

Spectroscopic techniques now permit measurements of unprecedented precision for highly ionized, highly excited and highly complex systems. This has pressed the use of quantum defect analyses, core polarizations, and screening parameter formulations into new regimes, and has revealed both new systematic trends and the breakdown of regularities present for lower ionicities. Laser techniques, fast-ion-beam excitation, tokamak-produced plasmas, and advances in the detectability of astrophysical light sources have provided access to high Rydberg states and to multiply excited states, brought about MHz accuracies in the specification of differential energy levels, and yielded spectra for atoms ionized through one electron uranium. Through the acqui-

sition and systematization of empirical data, methods have been developed as part of this project that parametrize and precisely predict excitation energies, transition wavelengths, transition probabilities, level lifetimes, ionization potentials, core polarizabilities, and core penetrabilities for large classes of atomic systems. Furthermore, this precision is sharpened with the subsequent measurements that are motivated by the predictions themselves.

## II. SIGNIFICANT RESULTS

During the thirteen and one-half years since it began, work done on this project has been reported in 81 published papers [1-81]. Of these, 26 [56-81] were produced during the three-year funding period covered by this Report. These papers provide the best context for a description of the significant results of the program, and a set of reprint copies of these 26 publications is attached as Appendix A (henceforth designated as Papers [A1]-[A26]). The project has proceeded through a number of separate but complementary investigations, which can be broken down into specific topics. To facilitate the description, the appended papers have been placed in subjective groupings, rather than in the chronological order given in the bibliography. Thus the 26 latest publications are arranged in the following categories: Invited Review Article [Paper A1]; Fast-Ion Beam Lifetime Measurements [Papers A2-A14]; Data-Based Semiempirical Systematizations [Papers A15-A20]; Calculations [Papers A21-A23]; and Accelerator Studies [Papers A24-A26]. The latest results obtained in this project will be described below within the framework provided by these categories.

### A. Invited Review Article

A comprehensive summary of many of the types of studies carried out in this program is presented in Paper [A1], which is a review prepared in collaboration with Professor Indrek Martinson at the invitation of the Editorial Board of the Journal *Comments on Atomic and Molecular Physics*. This review deals with recent developments in the study of decay rates in systems ranging from negative ions to very heavy one-electron ions. It describes how studies of atomic structure are being extended into highly ionized regimes in which relativistic and quantum electrodynamic interactions and effects due

to the finite size of the nucleus can dominate the dynamical structure of the ions. The existing capabilities for time-resolved lifetime measurements are reviewed and compared with theoretical results based on *ab initio* methods, as well as with semiempirical approaches. This article contains a useful overview of our program, and provides an ideal context for the descriptions which will be given below.

## B. Fast Ion Beam Lifetime Measurements

A large number of atomic lifetime studies have been conducted during this three year period, using a variety of particle accelerators. The facilities used include: (1) the 330 kV Heavy Ion Accelerator at the University of Toledo; (2) the 3 MV Tandem Pelletron Accelerator at the University of Lund in Sweden; (3) the ATLAS Tandem Linear Accelerator System at Argonne National Laboratory; (4) the K1200 Cyclotron at the National Superconducting Cyclotron Laboratory at Michigan State University; (5) the Tandem Van de Graaff Accelerator at the University of Notre Dame; and (6) the 4 MV Dynamitron Accelerator at the University of Bochum in Germany. Studies have been made for elements as light as carbon and as heavy as gold, with ionization degrees ranging from neutral to 35 times ionized, and for systems with as few as one and as many as 78 electrons. Short descriptions of some of these studies will be given below.

### 1. Lifetime Measurements at 100-330 keV

- *Lifetimes in the Platinum Sequence*

The lifetimes of levels of ions in the platinum isoelectronic sequence are needed for a number of applications, but the reliability of theoretical methods for the specification of this 78 electron system is untested. The lowest configuration of odd parity is  $5d^9 6p$ , which contains twelve levels with  $J$  ranging from 0 to 4, but only those with  $J=1$  have E1-allowed transitions to the  $5d^{10} {}^1S_0$  ground state. In  $LS$  notation these would be the  ${}^1P_1$ ,  ${}^3P_1$ , and  ${}^3D_1$  levels, but they are heavily influenced by intermediate coupling and configuration interaction with, *e.g.*,  $5d^8 6s 6p$ . We have performed experimental studies of these systems for Hg III [Paper A2] and Au II [Paper A3] using the 330 kV Heavy Ion Accelerator at the University of Toledo, and have also supported

these measurements with *ab initio* calculations utilizing the Multiconfiguration Dirac-Fock (MCDF) package GRASP and the program suite of Cowan, run in the relativistic Hartree-Fock (HXR) mode.

One of the first projects of the Goddard High Resolution Spectrograph (GHRS) on board the Hubble Space Telescope (HST) was the registration of a high resolution spectrum of a chemically peculiar star,  $\chi$  Lupi, which has a high abundance of mercury. This analysis verified the earlier finding of an isotope anomaly in the star's atmosphere, in which mercury is present in the form of  $^{204}\text{Hg}$  to 99%, whereas this isotope is only a minor contributor (7%) in the solar system. The study was based on the resonance line of Hg II at 1942 Å. A possible explanation for this isotope anomaly involves a delicate balance between radiative pressure and gravitation, described by a diffusion model developed by Michaud [Phys. Scr. **36**, 112 (1987)]. Determinations of the abundance and isotopic composition of neutral and doubly ionized Hg thus constitute a critical test of this model, and observations of the spectral regions containing Hg I and Hg III lines have been performed with the HST. These spectra permit the determination of isotopic compositions to within a few percent if the relevant atomic data (energy levels, wavelengths, oscillator strengths, *etc*) are available. Such data exist for Hg I and Hg II, but information is fragmentary for Hg III, particularly for transition probabilities. The wavelength interval accessible with the HST at the highest spectral resolution has a lower limit of about 1700 Å, so the Hg III lines observed with the HST appear at around 1740 Å.

With one exception (an electron-excitation measurement of lifetimes of some  $5d^86s^2$  levels), no experimental or theoretical lifetimes or *f*-values had been reported for Hg III prior to our work. We measured lifetimes for three  $5d^96p$  levels and one  $5d^86s6p$  level, and performed calculations using both GRASP and the Cowan suite. As described in Paper [A2], our measurements indicated some specific discrepancies between experimental and theoretical lifetimes, but we generally found that the theoretical values were reliable to within about 15%. While this accuracy may suffice for the analysis of HST spectra of peculiar stars, improvements in the approximations needed in theoretically treating this complex many-electron system must be made if higher accuracy is required.

As discussed above, the GHRS aboard the HST has been used to monitor the region around 1740 Å, where the Hg III lines appear. This region also con-

tains the strongest 6s-6p transition ( $5d^9 6s \ ^3D_3 - 5d^9 6p \ ^3F_4$  in LS notation) in Au II, isoelectronic to Hg III. The wavelength of this transition was recently determined to be 1740.476 Å using the VUV Fourier Transform Spectrometer at the University of Lund. This value agrees with the wavelength measured with the HST to within 2-3 mÅ, and represents a 20-fold improvement over the previously accepted value of 1740.52 Å. This line is also very prominent in the spectrum of  $\chi$  Lupi. Analyses of stellar data, using recent unpublished calculations of gf-values for Au II, yield an overabundance of Au (relative to the solar value) of about four orders of magnitude. This is comparable to the abundance pattern found for the elements mercury and platinum.

Various explanations of these high abundances are now being investigated through systematic studies of the heavy elements. In such work, accurate experimental data (level energies, wavelengths, oscillator strengths, lifetimes, hyperfine structure separations, isotope shifts, *etc.*) are required. Although the knowledge of energy levels can be considered satisfactory (although the important  $5d^8 6s 6p$  configuration is practically unknown), no data on lifetimes or oscillator strengths for Au II had appeared in the literature prior to our measurements, which are described in Paper [A3].

The measurements were performed as in the Hg III studies, and calculations were performed using the Cowan program. To improve the agreement between experimental and calculated energies, least-squares fits of the energy parameters to the observed level values were performed. The results indicated that the theoretical lifetimes of the  $5d^9 6p$  levels are typically 30% shorter than the experimental values. A number of possible explanations for this discrepancy were investigated. There are uncertainties in the theoretical predictions because the  $5d^8 6s 6p$  levels are poorly known, and their configuration interaction with the  $5d^9 6p$  levels could not be accurately included. There are also uncertainties in the experimental measurements introduced by possible line blends and cascades, but our measurement of the lifetime of the  $6p \ ^2P_{1/2}$  level in Au I resulted in very good agreement with accurate laser data, which provides confidence in our the experimental data for Au II. In view of the fact that the present calculations for Au I deviate from these experimental values by about 30%, similar inaccuracies in the theoretical values may occur for the case of Au II. Thus, in Paper [A3] we concluded that the method used to calculate the f-values for Au II has an inherent uncertainty close to 30%. More accurate theoretical values could be obtained

by including the interaction with the 5d electrons, but this would require substantial computational effort.

- *Lifetimes in Neutral Carbon*

Recently, emission studies of the gf values have been made for C I by Goldbach and coworkers [Astron. Astrophys. **181**, 203 (1987); **221**, 155 (1989)]. These studies used a wall stabilized arc light source to record high resolution spectra of C I in the region 1200-2000 Å. The relative oscillator strengths were then normalized, using the best available data, which consisted of beam-foil lifetime data from the late 1960's and early 1970's. However, the beam foil method has undergone a number of significant improvements in recent years, resulting in higher sensitivity, better wavelength resolution, and improved methods for accounting for the contributions of cascade repopulation. In order to confirm the normalization of these new gf values, we undertook a new beam-foil study of C I [Paper A4] in the VUV region, using the Toledo Heavy Ion Accelerator.

Lifetimes were measured for six multiplets in the triplet system and five in the singlet system. For the strongest VUV multiplets our results generally confirmed previous measurements, but for some of the weaker lines our results provided substantial corrections to the earlier experimental data. In some cases our measurements provide the first experimental lifetimes. The accuracy of our measurements was sufficient to test the reliability of theoretical methods that have been applied to this system, and support the calculations of Nussbaumer and Storey [Astron. Astrophys. **140**, 383 (1984)] and of Luo and Pradhan [J. Phys. B**22**, 3377 (1989)]. Our results are also in agreement with values deduced from the emission measurements of Goldbach and coworkers.

- *Lifetimes of Resonance Transitions in the Na Sequence*

Alkalilike systems, consisting essentially of one electron outside closed shells, have long been assumed to be theoretically calculable with high accuracy. However, small discrepancies between theory and experiment exist for the Li I 2s-2p and Na I 3s-3p resonance transition probabilities. A series of cascade-free beam-laser and pulsed-laser measurements in Li I and Na I have

yielded lifetimes that are consistently longer than those predicted by sophisticated *ab initio* calculations by amounts just prescribed by the accuracies (typically 1% or better) of the measurements. Similar discrepancies with theory exist for the  $\text{Mg}^+$  lifetime, measured in a beam-laser experiment, and for multiply charged ions in the Na sequence, measured by beam-foil spectroscopy and the ANDC technique.

There is a gap between the laser data for Na and Mg and the data for higher ionic charges that have been obtained by ANDC analyses. To bridge this gap, we performed measurements [Paper A5] of the lifetimes of the 3p levels for Na-like Si IV using ANDC analysis. Two other studies of this ion had been reported earlier, both of which used only multiexponential curve fitting methods to determine the 3p lifetimes. We were able to improve on the accuracy of the decay curve data, taking advantage of significant advances in ion source, accelerator, and spectroscopic detection technology in the two decades since the earlier studies.

The results of our analysis indicated that these lifetimes cannot be correctly extracted from the decay curves by curve fitting techniques. When we applied curve fitting methods to our data, the effective parameters obtained agreed with earlier reported lifetimes of the 3p levels, indicating that the new and old measured decay curves were consistent. However, when the 3p-3d and 3p-4s cascade decay curves were incorporated into the analysis via the ANDC method, the values for the 3p lifetimes decreased significantly, agreeing very well with the semiempirical CAHS calculations which we reported earlier [43], and with the recent *ab initio* calculations of Guet *et al.* [Phys. Lett. A **143**, 384 (1990)].

## 2. Multiplexed Decay Curve Measurements

We have utilized position sensitive detection of VUV emission from excited ion beams to obtain multiplexed registration of decay curves, reported in Papers [A6-A8]. This method provides simultaneous detection over a range of wavelengths, yielding a landscape decay curve containing intensity as a function both of wavelength and of decay time. This multiplexed registration greatly increases the efficiency of data collection and provides a more precise determination of the relative lifetimes. Because of the simultaneous measurement, there is a cancellation of the small systematic effects



(beam fluctuations, degradation of foil quality, *etc.*) that limit the accuracy of separately measured relative decay curves.

This type of multiplexed decay curve measurement offers new possibilities for improved accuracy in lifetime measurements. As one example, it could permit sensitive measurements of the relative lifetimes of fine structure components where configuration interaction or intermediate coupling selectively couples a particular J level to produce an additional intercombination or autoionization channel. As another example, it permits long lived metastable decays to be identified and studied efficiently. It also makes possible the real-time application of the ANDC correlated analysis of cascade repopulation for a system if the primary and cascade transitions are within the same wavelength range.

Our measurements were made using a position sensitive detector (PSD) consisting of a 1 inch wide windowless channeled electron multiplier array in the chevron configuration, coupled with a resistive anode position sensor. The position decoding of the photon induced pulses was obtained by comparison of the divided pulse charges appearing at the two ends of the anode.

#### • *Relative Lifetime Measurements of Alkalilike Resonance Transitions*

As one example of the application of this technique, we used fast silicon ions to make precision measurements of the J dependence of the lifetimes of the 2s-2p multiplet in Li-like Si XII [Paper A6]. The experiment was carried out at the Notre Dame Tandem Accelerator in collaboration with A.E. Livingston of Notre Dame and H.G. Berry of ANL. The 42 MeV foil-excited Si beam was viewed with a 1 m normal incidence monochromator, with the exit slit replaced by the PSD. An example of the multiplexed spectrum of Si in the region  $\lambda 1000-1040 \text{ \AA}$  is shown in Fig.1 of Paper [A6]. The two strongest lines are the 2s-2p fine structure resonance transitions (in second order), and the other features arise from the Si XII  $n=5-6$  and Si XI  $n=6-7$  Rydberg transition complexes.

When reduced to eliminate  $\lambda$  dependences, the measured line strength ratio was  $S(1/2-3/2) : S(1/2-1/2) = 1.979(44)$ , which can be compared with *ab initio* calculations of 2.007 (MBPT) and 2.005 (MCDP), and a semiempirical calculation of 2.007 (CAHS).

- *Spin Forbidden Intercombination Lines*

Measurements of both the wavelengths and lifetimes provide a useful probe of spin system intercombination transitions in ions with several electrons outside of closed shells. Since these rates scale as a higher power of  $Z$  than the allowed transitions, the intersystem lifetimes become measurable at ionicities for which many of the allowed transitions become too short for reliable time-of-flight determination. In the section on Data-Based Methods below, we shall discuss how these intersystem rates can be combined with spectroscopic information to produce reliable predictions for allowed rates, based on intermediate coupling considerations. Thus, an entire isoelectronic sequence can sometimes be specified by measuring lifetimes for allowed transitions in few times ionized members, and intercombination lifetimes in highly ionized members.

Beam-foil spectroscopy is presently the only method which allows measurement of long-lived levels of highly charged ions. We have used this method to study intercombination transitions in Mg-like, Al-like, and Si-like bromine in an experiment [Paper A7] carried out at the Argonne ATLAS accelerator. Here a PSD was also used, and a sample multiplexed spectrum is shown in Fig. 1 of Paper [A7]. The spectra contain decays with very different time constants, the most notable in the delayed spectra being the intercombination lines in Mg- and Al-like ions of Br. For calibration purposes, a number of wavelength and lifetime measurements were also made for Na-like bromine.

For the Mg sequence we studied the  $3s^2\ ^1S_0 - 3s3p\ ^3P_1$  transition, and verified trends observed earlier at lower  $Z$  that had indicated agreement with theoretical predictions of Huang and Johnson [Nucl. Instr. Meth. B9, 502 (1985)], and with the predictions which we made in Paper [A15]. Our measurements yielded a lifetime value 1.86(15) ns. For the Al sequence we studied the  $3s^23p\ ^2P_{3/2} - 3s3p^2\ ^4P_J$  transitions. Here the previously available data base was sparse, there being few experimental lifetime values, and none for the longest-lived quartet term,  $3s3p^2\ ^4P_{3/2}$ . Here our measurements yielded wavelengths 255.6(3), 287.4(15), and 260.1(3) Å, and lifetimes 1.9(2), 12(5), and 2.05(10) ns for the  $J=1/2, 3/2$  and  $5/2$   $^4P_J$  levels. For the Si sequence we studied the  $3s^23p^2\ ^3P_{3/2} - 3s3p^3\ ^5S$  transitions. Here our measurements can also check the anticipated result that, with increasing  $Z$ , the intercombination multiplet will be quenched in favor of the  $3s^23p3d$  level decays. Here

our measurements yielded a wavelength 284.7(1) Å and a lifetime 3(1) ns.

Our measurements confirm a number of predictions obtained by multi-configuration relativistic random phase approximation (MCRP) and by MCHF schemes, and represent a considerable extension of available lifetime results to higher nuclear charge.

### 3. Lifetimes in the Ne Sequence

Because of the strong radiative decay of the  $2p^6$ - $2p^53s$  transitions in the Ne isoelectronic sequence, it is possible to create a population inversion between the  $2p^53s$  and  $2p^53p$  levels which can produce XUV laser amplification action. Modeling studies have demonstrated that amplification is optimal near  $Z=36$ , leading to applications in Se and Y plasmas. The lifetimes of these levels are too short for time-of-flight measurements at  $Z=36$ , but we have developed methods that permit reliable semiempirical extrapolation if precise results at lower  $Z$  are available. Together with collaborators at the University of Lund, we have performed precision measurements of the  $2p^6$ - $2p^53s$  -  $2p^53p$  transitions in S VII and Cl VIII [Paper A9].

These studies were made possible by the existence of a uniquely designed beam-foil chamber at the University of Lund which views a very short segment of beam with a grazing incidence monochromator, permitting the precision measurement of lifetimes as short as few ps for wavelengths well below 100 Å. Fig.1 of Paper [A9] shows sample decay curves obtained for the resonance and intercombination lines in S VII. These measurements yielded lifetimes accurate to 3-4%, as compared to earlier measurements which had uncertainties  $\cong 25\%$ . In addition to the curve fits, it was also possible to perform ANDC correlated decay analyses with the primary cascade channel  $2p^53s$ - $2p^53p$  (a proposed laser transition).

The section on Data-Based Methods below describes a semiempirical reduction of transition probabilities for resonance and intercombination lines using spin-mixing reduced charge-scaled line strengths. This can be used to combine our S VII and Cl VIII data with earlier measurements in Ne I and Na II to obtain a predictive semiempirical parametrization. As will be described in the proposal for the next funding period, this produces a linearization that can be extrapolated to make predictions for *e.g.*, Se ( $Z=34$ ) and other ions with lifetimes too short for reliable measurement. While this extrapolation is

still somewhat speculative, existing *ab initio* calculations vary by factors of 2, and a semiempirical approach that is reliable to the 3-4% levels of accuracy provided by our measurements would be valuable for modeling purposes.

#### 4. Lifetimes for the H and He Sequences

The increasing availability of intense beams of high  $Z$  few-electron ions at heavy ion accelerators provides an opportunity for testing relativistic corrections to energy levels and lifetimes of highly charged ions. Few electron systems, particularly hydrogenlike and heliumlike ions, provide an important testing ground for our understanding of atomic structure. In the one electron system, there are no calculational uncertainties, so experiments in these systems provide tests of the underlying theory of Quantum Electrodynamics (QED). Current interest is in the question of whether there is a breakdown of QED in highly-charged one electron atoms. The theory of the two-electron system is not as well understood. Here, one has the additional problem of treating correlations between the two electrons, and a complete Hamiltonian for the system does not exist. The importance of the heliumlike systems is that they are the simplest atoms in which the electron-electron correlations must be taken into account. A review of these methods is presented in Paper [A10].

- *Branching Ratio for the M1 Decay of  $2\ ^2S_{1/2}$  in One-Electron Krypton*

One of the most fundamental lifetimes in one electron ions is that of the  $2\ ^2S_{1/2}$  level. This level decays to the  $1\ ^1S_{1/2}$  ground state either by two-photon emission or by single-photon M1 emission. The two-photon decay rate dominates at low  $Z$ . In fact, the M1 matrix element vanishes if nonrelativistic wavefunctions are used. The operator acts only on the angular parts of the wavefunctions, and the radial parts are separately orthogonal. The M1 decay is therefore inherently a relativistic phenomenon, and the branching ratio for this decay increases with  $Z$  and becomes equal to the two photon decay rate at  $Z=41$ . We have measured [Papers A11, A12] the lifetime of the  $2\ ^2S_{1/2}$  level in H-like Kr, and have made the first determination of the branching ratio for the M1 decay of the  $2\ ^2S_{1/2}$  level of a one-electron ion.

The lifetime of the  $2\ ^2S_{1/2}$  level in hydrogenlike ions has been measured to a precision of about 1% in  $\text{He}^+$ ,  $\text{Ar}^{17+}$ , and  $\text{Ni}^{27+}$ , and to lower precision

in  $O^{7+}$ ,  $F^{8+}$ , and  $S^{15+}$ . The Ar experiment was the first to be sensitive to the contribution to the total decay rate due to the M1 amplitude, and the Ni measurement was the first to be sensitive to the relativistic corrections to the two photon amplitude. Our lifetime measurement yields 36.8(1.4) ps, in good agreement with the theoretical value of 37.008 ps. Our measured branching ratio is 0.351(11), and is in fair agreement with the theoretical value of 0.3642. The corresponding M1 decay rate is  $\Lambda_{M1}=9.54(47)\times 10^9 \text{ s}^{-1}$  and the two photon decay rate is  $\Lambda_{2E1}=1.764(74)\times 10^{10} \text{ s}^{-1}$ , in agreement with the theoretical values  $\Lambda_{M1}=9.844\times 10^9 \text{ s}^{-1}$ , and  $\Lambda_{2E1}=1.718\times 10^{10} \text{ s}^{-1}$ .

- *Hyperfine Quenching in He-like Nickel*

One of the most interesting phenomena in the theory of highly forbidden transitions is the effect of hyperfine quenching, whereby mixing by the hyperfine interaction can significantly alter the lifetimes of the levels. Single photon transitions connecting  $^1S_0$ - $^3P_0$  levels are absolutely forbidden in atoms with a spinless nucleus. When the nuclear spin is nonzero the nuclear magnetic moment mixes electron states of different J, and this decay channel is opened. This hyperfine quenching alters the lifetime of the  $^3P_0$  level from its spin free value. We have reported [Paper A13] the clearest demonstration of this effect to date by direct comparison of the lifetimes of the  $2\ ^3P_0$  level in the heliumlike isotopes  $^{61}\text{Ni}^{26+}$  and  $^{58}\text{Ni}^{26+}$ .

We directly observed the effect of hyperfine quenching by comparing the lifetimes of forbidden decays in these two isotopes. The isotope  $^{58}\text{Ni}$  has nuclear spin  $I=0$  and is not affected by hyperfine quenching, whereas  $^{61}\text{Ni}$  has  $I=3/2$  and hyperfine effects are important to understanding the lifetimes of its excited states. By assuming theoretical values for the lifetimes of the levels and hyperfine matrix elements, our measurements could also be used to specify the  $2\ ^3P_0$ - $2\ ^3P_1$  level splitting, or to confirm the theory of the unquenched lifetimes and hyperfine mixing matrix elements assuming the energy splittings are known. We have also shown that the comparison of decay curves for two different isotopes is a powerful means for studying or eliminating experimental problems such as blended transitions or cascade effects. The quenched lifetime of the  $2\ ^3P_0$  level in  $^{61}\text{Ni}^{26+}$  was found to be 470(50) ps. From this we have deduced the  $^3P_0$ - $^3P_1$  energy splitting to be 2.33(15) eV. We have also measured the lifetime of the  $2\ ^3P_2$  level to be 70(3) ps. This level is only slightly affected by hyperfine quenching.

- *Two Photon Decay Rate of the  $2^1S_0$  state in He-like Bromine*

The  $2^1S_0$  level in heliumlike ions is strictly forbidden to decay to the  $1^1S_0$  ground state by the emission of a single photon because of the requirement that angular momentum be conserved in the decay. This level decays by two photon emission, which is second order in the coupling of the radiation field to the electrons, so this decay is highly suppressed. The emitted photons have a continuous energy distribution, but the sum of the energies of the two photons adds up to the transition energy. The requirement of a coincidence between the two detected photons with the proper sum energy provides a signature to distinguish background counts and allows separation of one- and two-electron decays. We have used the coincidence detection technique to study this decay in He-like bromine [Paper A14]. This system is particularly sensitive to relativistic corrections, and our measurement clarifies earlier apparent discrepancies between experiment and theory.

The lifetime of the  $2^1S_0$  level has been measured to about 1% in  $Kr^{34+}$  and  $Ni^{26+}$ , and to lower precision in He,  $Li^+$ , and  $Ar^{16+}$ . The  $Kr^{34+}$  experiment [Marrus *et al.*, Phys. Rev. Lett. **56**, 1683 (1986)] was the first to be sensitive to the relativistic corrections to the decay rate, but was not a coincidence measurement. Our own  $Ni^{26+}$  measurement [49] was also sensitive to the relativistic corrections and was the first to determine the decay rate using the coincidence technique. However, the relativistic corrections are smaller in Ni than in Br or Kr. The  $Ni^{26+}$  and the  $Kr^{34+}$  measurements yielded lifetimes that were slightly longer than those theoretically predicted. The theoretical formalism for two photon decay was developed by Goeppert-Mayer already in 1931, and has been subsequently studied by many authors. Drake [Phys. Rev. A **34**, 2871 (1986)] has calculated the nonrelativistic decay rates and estimated the relativistic corrections for all two electron ions up to  $Z=92$ . For Br the relativistic corrections are significant, with a nonrelativistic result 37.972 ps, with the inclusion of relativistic corrections yielding 39.63 ps. We obtained a value 39.32(29) ps, in good agreement with the theoretical value.

### C. Data-Based Semiempirical Formulations

The high precision requirements of atomic spectroscopy have traditionally been met by empirical exposition and predictive systematization of measured data. Empirical approaches are necessitated by the high precision that can

be obtained in optical measurements, which often exceeds the capabilities of existing *ab initio* theoretical methods. Empirical regularities are often exploited for predictive guidance in and conceptual interpretation of experimental measurements, as well as to provide insights into the fundamental physical interactions. A number of different empirical methods have been utilized, some of which will be described below.

## 1. Mixing Angle Formulations of Lifetime Measurements

In studies of  $ns^2$ - $nsn'$ p and  $np^6$ - $np^5n$ 's transitions in alkaline-earthlike and inert-gaslike isoelectronic sequences we have developed an empirical data reduction that treats measured transition probabilities for the resonance and intercombination lines as conjugate quantities. The reduction is formulated through a singlet-triplet mixing angle  $\theta$  deduced from measured spectroscopic energy level data, and yields an effective generalized line strength that has a regular and slowly varying isoelectronic behavior. This regularity has been observed to persist even in the presence of strong perturbations, and it can be linearized through simple screening parametrizations that permit quantitative interpolation, extrapolation and smoothing.

In this approach the line strengths obtained from measured transition probabilities of the resonance and intercombination lines  $S(\text{Res})$  and  $S(\text{Int})$  are converted to values reduced for spin mixing effects by dividing by the appropriate empirical mixing angle quantity to obtain  $S_r(\text{Res}) \equiv S(\text{Res})/\cos^2\theta$  and  $S_r(\text{Int}) \equiv S(\text{Int})/\sin^2\theta$ . In some cases it has been found that  $S_r(\text{Res}) \cong S_r(\text{Int}) \cong S(\text{Res})+S(\text{Int})$  and the total effective line strength  $S(\text{Tot})$  can be defined as the sum of the line strengths of the resonance and intercombination transitions. The combined use of resonance and intercombination lines to specify a generalized line strength permits data over a wide range of charge states to be merged. Time-of-flight methods are limited to lifetimes that produce a measurable decay length, and often the intercombination transitions are unmeasurably long-lived at low  $Z$  while the resonance transitions are unmeasurable short-lived at high  $Z$ . Using our own lifetime measurements as well as those of other groups, predictive isoelectronic studies have been completed for a number of systems. In all cases the reduced line strength has been found to be a nearly linear function of the reciprocal screened charge, permitting predictive interpolations, extrapolations, and smoothing.

- *Alkaline Earthlike Intrashell  $ns^2$ - $nsnp$  Transitions*

We applied these methods to use spectroscopic energy level data to systematize lifetime data for the strong low lying intrashell resonance and intercombination lines of the  $2s^2$ - $2s2p$  and  $3s^2$ - $3s3p$  transitions in the Be and Mg isoelectronic sequences [Paper A15] and the  $4s^2$ - $4s4p$  transitions in the Zn isoelectronic sequences [Papers A16 and A17]. The work on the Zn sequence is particularly noteworthy, since our most recent study [Paper A17] revealed errors in both the calculations and measurements of the intercombination lifetimes, which had caused deceptive agreement. Hibbert and Bailie [Phys. Scri. **45**, 565 (1992)] recently suggested that earlier theoretical calculations had overestimated these lifetimes because of a failure to include the effects of core polarization, and indicated a procedure for correcting these predictions. Träbert and coworkers have subsequently made new lifetime measurements of these intercombination transitions for several ions, and demonstrated that the earlier measurements had overestimated the lifetimes because of a failure to adequately account for the effects of cascade repopulation. (A faulty experimental assumption had been made that, because these spin-forbidden transitions are longer-lived than their close-lying cascade feeders, their repopulation would be less significant than in spin-allowed cases.) The correction of these errors has interesting implications when examined within the mixing angle formulation.

In our first mixing angle reduction [Paper A16] of the Zn sequence, the lifetime data then available had indicated that the values of  $S_r(\text{Res})$  and  $S_r(\text{Int})$  were both linear in  $1/(Z-28)$ , but that each had its own characteristic slope and intercept (*cf.* Fig.2 of Paper [A17]). When the revised data for the intercombination lifetimes were included and subjected to this same reduction, a shift occurred which moved the  $S_r(\text{Int})$  values onto the same straight line as  $S_r(\text{Res})$ . Fig.4 of Paper [A17] presents this plot, and demonstrates that, for the  $4s^2$ - $4s4p$  transitions in the Zn sequence,  $S_r(\text{Res})$  and  $S_r(\text{Int})$  are equivalent measures of  $S(\text{Tot})$ .

- *Alkaline Earthlike Intershell  $ns^2$ - $nsn'p$  Transitions*

We have also applied this semiempirical formulation to a study of the resonance and intercombination lines of the  $2s^2$ - $2s3p$  and  $3s^2$ - $3s4p$  transitions in the Be and Mg isoelectronic sequences [Paper A18]. This extension



to intershell transitions provides a stringent test for the method, since it introduces complications from plunging configuration interaction, transition integral cancellations, and multiple exit channels, that were not present in the case of the intrashell transitions. These transitions provide a particularly suitable example, because of a novel experimental technique that we developed earlier ([Engström *et al*, Phys. Scri. **20**, 88 (1979) and [19]). This permits the determination of intercombination transition probabilities through differential decay curve measurements that exploit the J dependence of the exit channels of the triplet levels.

Using our own measurements for the  $2s^2\ ^1S_0 - 2s3p\ ^3P_1$  transition probabilities in Be-like N IV, O V, F VI, and Ne VII, we have obtained a linearizing formulation of the intercombination transition which also predicts the lifetime of the resonance transition with high reliability. A plot of this reduction is shown in Fig.2 of Paper [A18]. When plotted versus  $1/(Z-3)$ , the linear trend of  $S_r(\text{Int})$  obtained from experimental measurements coincides with the linear trend of  $S_r(\text{Res})$  obtained from theoretical calculations [Nussbaumer, Astron. Astrophys. **16**, 77 (1972)]. To within experimental uncertainties, the reduced line strengths for the two transitions here again coincide.

We have also studied [Paper A18] the homologous example of the  $3s^2-3s4p$  levels in the Mg sequence using available experimental measurement and theoretical calculations for S V and Cl VI. In this case the  $3p3d$  configuration complicates the situation by introducing plunging configuration interaction and additional intercombination channels. This produces heavy configuration interaction in S V which is not present in Cl VI. However, the singlet-triplet mixing angle reduction is found to remove this local effect, indicating that the primary effect of configuration interaction (CI) is to enhance the single configuration singlet-triplet mixing, and that its affect on the dipole transition integral is long range and smooth. Thus it was possible to characterize the CI as indirect rather than direct, because the isoelectronic irregularities in the reduced transition probability data could be completely removed by the use the singlet-triplet mixing angle. These mixing amplitudes contain the CI information, since they are obtained from the spectroscopic data as effective values for pseudo single configuration Slater parameters.

#### • *Inert Gaslike $np^6 - np^5n$ 's Transitions*

Similar analyses have been made of the  $2p^6-2p^53s$  and  $3p^6-3p^54s$  resonance

and intercombination lines in the Ne and Ar sequences, which are described in Paper [A20].

As described in the section on Lifetime Measurements above, we have recently performed precision (3-4%) measurements of the resonance and intercombination lifetimes in Ne-like S VII and Cl VIII [Paper A9]. Fig.2 of Paper [A20] plots  $S_r(\text{Res})$  and  $S_r(\text{Int})$  vs  $1/(Z-9)$  for these ions, together with similar reductions of existing measurements of Ne I and Na II. Here the linearity of the trend of each quantity is clear, although the slope and intercepts of the resonance and intercombination data are not the same. As will be described in our Research Proposal for the next cycle, we plan to remeasure and extend the lifetime data near the neutral end of this sequence to verify and sharpen these trends. In this manner, it should be possible to make precise predictions for the lifetimes of high Z members of this sequence, which have lifetimes too short for determination by time-of-flight methods, and too long for determination by line shape studies.

The resonance and intercombination transitions in the Ar sequence provide a similar example, but with an interesting additional feature. As shown in Fig.3 of Paper [A20], the plunging  $3p^5 3d \ ^1P_1$  level causes a strong local enhancement (50%) in the empirical singlet-triplet mixing in Sc IV, in a manner very similar to the S V case in the Mg sequence. We have made studies using theoretical values for the line strengths that indicate that, while both the energy levels and the lifetime are strongly perturbed in the vicinity of the level crossing, the values of  $S(\text{Tot})$  are isoelectronically smooth. Here lifetime measurements would test the validity of the assumption that CI is indirect, perturbing the lifetimes and the energy levels (and hence the mixing angle) but not the transition matrix.

- *Empirical Mixing Amplitude Formulation for  $np^3$  Systems*

The singlet-triplet mixing angles described above pertain to systems in which only two levels are mixed by intermediate coupling. The method can be extended to systems of arbitrary complexity, although this requires the diagonalization of a matrix larger than  $2 \times 2$ . For the  $3 \times 3$  matrix which occurs for an  $np^3$  configuration, specification of the doublet-quartet mixing coefficients requires the solution of a cubic secular equation

$$\lambda^3 + a_2 \lambda^2 + a_1 \lambda + a_0 = 0 .$$

Although the explicit solutions for the energy eigenvalues  $E_i$  as a function of the coefficients  $a_i$  (which involve Slater and spin-orbit integrals) are complicated and cumbersome, the inverse solution is quite simple, given by

$$a_2 = -E_1 - E_2 - E_3 ; a_1 = E_1 E_2 + E_2 E_3 + E_3 E_1 ; a_0 = -E_1 E_2 E_3$$

where the energy eigenvalues are spectroscopically measured quantities. The empirical eigenvectors generated in this way can be used to reduce measured transition probabilities to effective transition integrals or vice versa. The method was applied to make specific calculations for the N and P sequences in Paper [A19].

## 2. Development of Comprehensive Semiempirical Data Bases

Our use of the CAHS method (*cf* Calculations Section below) for the semiempirical computation of transition probabilities requires an accurate and comprehensive data base for excitation energies and ionization potentials. We have therefore sought to interpolate, extrapolate, and smooth the data bases for the Li and Be isoelectronic sequences. This required a critical evaluation of available measurements and calculations, followed by semiempirical studies utilizing quantum defect, Ritz parameter, and screening parameter systematizations, supplemented where necessary by specialized *ab initio* calculations.

- *Regular and Irregular Doublet Intervals*

For the Li sequence it was possible to make very accurate interpolations and extrapolations of the regular and irregular doublet intervals ( $2p\ ^2P_{1/2} - 2p\ ^2P_{3/2}$  and  $2s\ ^2S_{1/2} - 2p\ ^2P_{1/2}$ ). This was done by mapping the residues between experimental values and the RMBPT calculations of Johnson *et al* [Phys. Rev. A **37**, 2764 (1988); **41**, 1698 (1990)] (which included all but QED corrections) into the effective screened charge that would yield the same value for the corresponding Lamb shift in a hydrogenic atom. Our earlier use [47] of this procedure for  $26 \leq Z \leq 42$  yielded predictions which were subsequently verified to high accuracies by tokamak [Denne-Hinnov, Phys. Rev. A **45**, 2135 (1992)] and Bevalac [Schweppe *et al*, Phys. Rev. Lett. **66**, 1434 (1991)] measurements. The results of this study were extended through  $Z \leq 92$  and reported in Paper [A21].

- *Ionization Potentials*

Ionization potentials were obtained by a procedure that is best illustrated for the Be sequence, in a study reported in Paper [A20]. Here the mapping model was constructed of three parts: a fully screened Balmer energy parametrized by a quantum defect; a weakly screened Sommerfeld relativistic energy; and a weakly screened QED energy. For the Sommerfeld and QED energies, hydrogenlike formulae were used with undefected quantum numbers and an effective screening of one electron charge. The available data were first reduced by the Sommerfeld and QED energies, then mapped into the effective quantum defect. An exposition of these data, plotted vs  $1/(Z-1.9)$ , is given in Fig.4 of Paper [A20]. The measured data clearly exhibit a high degree of linearity. To test the high  $Z$  persistence of this linearity, the plot is supplemented by our own MCDF calculations for selected ions in the range  $36 \leq Z \leq 92$ . The importance of the Sommerfeld and QED corrections is demonstrated by the dashed line, which indicates how the same MCDF calculations appear if reduced to an effective quantum defect without first removing the Sommerfeld and QED energies. Thus the apparent linearity is associated with the Balmer energy (which samples the wave function weighted by  $1/r$ , in contrast to the Sommerfeld and QED energies, which sample the wave function weighted as  $1/r^3$ ). Predictions for the ionization potentials for the entire Be sequence are presented in Table II of Paper [A20].

## D. Calculations

### 1. CAHS Studies

In a series of studies ([43], [45], Paper [A20], Paper [A21]) we have shown that experimental measurements of energy level data can be used to make reliable semiempirical predictions of transition probabilities using the Coulomb Approximation with a Hartree-Slater model core (CAHS). This method involves the integration of the Schrödinger equation with a realistic model potential, constrained to yield the experimental binding energy. The model potential includes the effective central field seen by the single active electron, the spin-orbit interaction, and the core polarization potential (as specified by the dipole polarizability). The method is formulated to minimize or eliminate the use of adjustable parameters, so that a unique

prediction for the transition probability is made entirely from the measured inputs, which are the energy levels, ionization potentials, and core polarizabilities. As described in the previous section, the applicability of the method can be broadened by the use of other semiempirical studies that interpolate, extrapolate, and smooth these experimental inputs. Recent applications of this method to alkalilike isoelectronic sequences have produced important insights through comparisons to precision experimental measurements and to *ab initio* calculations.

As was discussed in the section on Lifetime Measurements, systematic 1% discrepancies between high precision experimental measurements and sophisticated *ab initio* calculations for the  $\Delta n = 0$  resonance transitions in alkalilike systems is a current source of concern. As is described in Paper [A20], there now exists a substantial set of cascade-free lifetime measurements made using state-selective laser excitation, which provides lifetimes accurate to 1% or better for a large variety of neutral alkali and singly ionized alkalilike system. These also include ions such as  $\text{Ca}^+$ ,  $\text{Sr}^+$  and  $\text{Ba}^+$ , for which the lifetime involves a branched decay to  $ns$  and  $(n-1)d$  levels. A summary of these data is presented in Table I of Paper [A20], together with a sampling of *ab initio* calculations. These show a tendency for the *ab initio* calculations to yield lifetimes that are about 1% shorter than the measurements, which are typically of 1% precision. While this trend would not be statistically significant for any single measurement, the systematic trend of the overall sample is unmistakable. Similar 1% discrepancies between experiment and theory have also been reported for highly ionized members of the Na isoelectronic sequence.

Table I of Paper [A20] also includes a series of computations that we have made using the CAHS algorithm. Our semiempirical computations agree very well with experiment, and thus possess the same discrepancies with *ab initio* theory as do the measurements. The CAHS calculations include core polarizability in two different ways: it is contained in the single particle wave functions through the model potential from which they are generated from the energy levels; it is also contained in the electric dipole transition moment, given by  $\mathbf{d} = e\mathbf{r} - \alpha_d\mathbf{r}/r^3$ , which coherently combines the moments of both the outer electron and the inner core. Tests of the CAHS calculations indicate that the trial exclusion of the core polarizability from the construction of the wave functions has an insignificant effect on the computed lifetimes,

but a trial setting of  $\alpha_d = 0$  shortens the computed lifetimes by an amount approximately corresponding to the discrepancy between experiment and *ab initio* theory.

We have reported [Paper A21] lifetime predictions for the 2p levels in the Li isoelectronic sequence for  $Z=3-92$  using the CAHS algorithm. This extends our earlier studies of the Na [43] and Cu [45] sequences. As was the case for our earlier CAHS calculations, the lifetimes obtained for the Li sequence agree with all critically selected measurements, and in particular, they agree with the often criticized cascade-free, laser-selective-excited measurement of Gaupp *et al.* [Phys. Rev. A **26**, 3351 (1982)].

## 2. Cancellation Spectroscopy

Atomic transition moments sometimes exhibit cancellation effects that can cause a normally strong transition to be weak or totally absent in certain observed spectra. One class of cancellation effects can occur within a single-electron radial integral as a result of differential phase shifts in the wave functions due to core polarization and penetration. We have developed methods ([Curtis & Ellis, J. Phys. B **11**, L543 (1978)], [2], [8]) to systematically study these radial integral cancellations and to predict their occurrence by simple graphical methods. An example is shown in Fig.2 of Paper [A22], which compares the effective quantum numbers for the 6s-7p transitions in the Au isoelectronic sequence to the locus of cancellation nodes.

Heavy cancellation effects in a specific transition probability do not usually lead to a large anomaly in the lifetime of the upper level, since these cancellations occur only for  $\Delta n \neq 0$  transitions (which possess multiple decay branches), and f sum rules tend to redistribute the missing oscillator strength among other exit channels. However, a recent lifetime measurement by Pinnington and coworkers [Can J. Phys. **69**, 594 (1991)] exhibited an anomaly which we have explained [Paper A22] in terms of such a cancellation. They observed that for the 7p term in P IV the  $J=1/2$  level has a lifetime six times longer than that of the  $J=3/2$ . We have shown that this result is a consequence of a nearly complete cancellation in the 6s-7p radial transition moment for the  $J=1/2$  level (*cf.* Fig.1 of Paper [A22]), coupled with fortuitous  $\lambda^3$  factors which suppress the decay to 7s, 6d, and 6s. As will be described in our Research Proposal for the next cycle, we plan to trace this

trend by lifetime measurements for other members of this sequence.

### 3. Hydrogenic Formulations

In a series of papers produced earlier as part of this project ([7], [21], [36]) classical expressions were developed for the hydrogenic expectation value  $\langle r^p \rangle$ , the radial coordinate  $r$  raised to an arbitrary power  $p$ . The formulation involves a Legendre polynomial with an argument formed according to a simple prescription, and can be very useful in characterizing polarization and penetration contributions to high Rydberg states. It was shown that the exact quantum mechanical expression can be obtained directly from the corresponding classical formula by a simple mnemonic, if a set of rational fraction coefficients is known. In these earlier studies, it was necessary to evaluate these coefficients by comparison between the classical and quantum mechanical expressions, so the classical method could not be used to generate values for which quantum mechanical calculations had not yet been performed. We have recently obtained [Paper A23] a closed form expression for these coefficients, and this formulation can now be used as a mnemonic by which the correct quantum mechanical expression can be generated quickly and accurately for any arbitrary power  $p$ .

This method for specifying these coefficients was suggested by results in a recent paper by Drake and Swainson [Phys. Rev. A **42**, 1123 (1990)] which presented a general method for obtaining the corresponding quantum mechanical expression for this expectation value. This quantum mechanical formulation involved the development of a recursion relation that also has semiclassical meaning, and from which this crucial set of coefficients can be sequentially generated. By combining this recursion formula with the semiclassical formulation, the correct quantum mechanical expressions can now be obtained [Paper A23] from the classical formula by a simple substitutional prescription for the angular momentum operators. This method of embedding the quantum mechanical dependences directly into the classical formulation offers some conceptual advantages, for example, by providing a smooth transition between the predictions of quantum mechanical and classical Hamilton-Jacobi perturbation theories.

## **E. Accelerator Studies**

Several technical papers have recently been published describing various aspects of our program of fast ion beam studies.

### **1. Studies of Failure Times of Beam Bombarded Foils**

In our studies of the spectra and lifetimes of foil excited atomic states, degradation of the foil can cause many problems. Signal normalization is frequently altered by foil breakage. Changes in the properties of the foil can distort the shape of the decay curve. If the foil lifetime is less than the time needed to measure intensities at a variety of decay times, this can preclude the decay curve measurement entirely. Thus the experimental parameters (beam current and profile, beam energy, foil thickness, *etc.*) must be carefully chosen. The eventual failure of stripper foils is also a limitation in tandem accelerator technology. In order to better understand the factors that influence these foil failures, and to attempt to find ways to increase the useful time before failure, we have undertaken a study of these processes.

The response of arc-evaporated carbon foils to ion-bombardment is well-known and the lifetime is limited by various processes. During irradiation, carbon stripper foils thicken in the irradiated area. The thickening is due both to radiation damage and to carbon build-up from the cracking of hydrocarbon vapor within the vacuum system by the ion beam. Correlated to the thickening, a shrinkage of the foils occurs during ion irradiation. Radial stress lines appear at an early stage around the beam spot as the foil contracts, while the foil surface at the beam spot itself assumes a mirror-like appearance. Gradually the stress lines become more prominent and accumulated mechanical stresses eventually cause the foil to rupture. The rupture usually takes place near the edge of the foil, outside the bombardment area.

We have reported studies undertaken to improve the useful lifetime of carbon foils in Paper [A24]. Several approaches were attempted to overcome the shrinkage. This is attributed to the rearrangement of the atoms in the amorphous foil. An improvement by a factor of 3 to 4 in foil lifetime has been obtained by providing a sufficient slackening in the foil tension at the production stage. In this way an increased amount of shrinkage is possible before the foil becomes ruptured. Measured enhancement factors appear to be relatively insensitive to the beam parameters such as energy, mass, atomic



number, current density, *etc.* No obvious dependences on foil thickness have been observed.

The carbon foils had surface densities in the range  $2\text{--}5\text{ }\mu\text{g}/\text{cm}^2$ . They were self-supporting and were mounted in both a standard and a tension-slackened manner. The intensity of the light emitted by the foil-emergent beam was measured as a function of bombardment time. The measured lifetime before beam failure for standard foils was found to agree well with a semiempirical model proposed by Auble and Galbraith, and the slackened foils exhibited usable lifetimes 3-4 times longer than normal foils. The properties of ordinary and slackened foils were compared under realistic conditions. It was concluded that the slackened foils are very well suited for studies of atomic spectra and meanlives of excited states using beam foil excitation methods. These measurements are now being extended though the production and testing of ethylene-cracked carbon foils.

## **2. The Toledo Heavy Ion Accelerator Facility**

The essential features and capabilities of the Toledo Heavy Ion Accelerator (THIA) facility are described in a Paper [A25]. The facility consists of a 30 keV Danfysik Isotope Separator with post acceleration of 0-300 kV. The ion source used is a Danfysik Model 911A, based on the hollow cathode design developed by Sidenius. An electrostatic switchyard is used to direct the beam into one of three beamlines. Most of our studies are performed on the right beam line, where a beam foil chamber is installed which contains a mechanism for holding and translating foils parallel to the beam line. The chamber is equipped with an Acton 1m normal incidence VUV monochromator. In addition to the beam-foil station, beam lines are also available for scattering and ion implantation studies.

Automated on-line data acquisition and experimental control of the THIA facility are achieved by use of the computer program ACCEL. This program possesses a number of interesting features and is described and documented in Paper [A26]. ACCEL is written in Pascal, and runs on an IBM PC in conjunction with a Keithley Series 500 Data Acquisition System for data collection in fast ion beam excitation studies. In addition to photon counting, foil positioning and wavelength scanning can be programmed. Collected data are displayed graphically, printed as collected, and stored on disk for

subsequent analysis. The program allows flexible control and is designed to allow new functions to be added easily.

### **III. COMPLIANCE WITH CONTRACT STIPULATIONS**

#### **A. Travel:**

##### **1. Overseas**

Two trips to Sweden to perform experiments at the University of Lund were made during the three year period of the grant, which were described and budgeted in the original proposal. In both cases it was possible to schedule the trips to coincide with conferences and meetings in Stockholm to which I was invited (and supported) by Swedish sources. Therefore it was only necessary to cover local expenses and travel from Stockholm to Lund with grant funds.

In July 1992 I was an invited participant at the Nobel Symposium on Highly Ionized Atoms held in Saltsjöbaden Sweden (near Stockholm). The Symposium was arranged by Professor Reinhold Schuch and Dr. Anders B  r  ny of the Manne Siegbahn Institute in Stockholm, and was attended by 34 invited scientists from Sweden, Denmark, USA, France, Germany, England, Japan, and Hungary.

During the period 16-17 February 1993, I was an invited participant in an Editorial Conference of the International Scientific journal *Physica Scripta* (I am the North American Editor for Atomic, Molecular, and Optical Physics) held at the Royal Swedish Academy of Sciences in Stockholm. In addition to the meetings to set and steer the editorial policies of the Journal, the Editors were honored at a Banquet at the Historic Astronomical Observatory in Stockholm.

##### **2. Domestic**

During this period I was also supported by the Grant for attendance at three meetings of the Division of Atomic, Molecular, and Optical Physics (DAMOP) of the American Physical Society. The meetings took place in

Washington DC (22-25 May 1991), Chicago Illinois (19-22 May 1992), and Reno Nevada (17-19 May 1993). Support was also provided to attend the three DOE/BES Atomic Physics Workshops, held at Kansas State University in Manhattan, Kansas (1991), Cornell University in Ithaca, New York (1992), and the University of Virginia in Charlottesville, Virginia (1993). Numerous working visits were also made to Argonne National Laboratory, as well as to the University of Notre Dame.

## **B. Reprints of Publications:**

Six reprint copies of publications resulting from this project have been submitted through the Oak Ridge Operations Office as they become available.

## **C. Time and Effort Reports:**

Time and Effort Reports have been submitted to the University of Toledo Grants Accountant which indicate that 25% of the principal investigator's full time activity is released and charged to the University of Toledo for cost sharing of the project during the 9 month academic year. During the 2 month summers 100% of the principal investigator's full time activity is charged to the grant. For the graduate assistant, 50% of his full time activity during the 9 month academic year, and 100% during the 2.5 summer months are devoted to this project and charged to the grant.

## **D. Support of Students**

The grant has provided various levels of support to two Graduate Students, Steven T. Maniak and Richard E. Irving, and two undergraduates, John Arney and Richard Christ. Steven Maniak received his MS in December 1992 and passed his PhD Comprehensive Examination in May 1992. He has been supported by this Grant through a Research Assistantship, travel to other research laboratories, travel to conferences and workshops, and equipment purchases useful in his research. Richard Irving, John Arney, and Richard Christ have also used equipment purchased under this Grant in the pursuit of their research.

## IV. BIBLIOGRAPHY OF PUBLICATIONS

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## DOE INTERNAL REPORTS

1. Progress Report: 1 July 1988 - 1 April 1989
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3. Ref..43 above.
4. Ref. 44 above
5. Ref. 45 above
6. Ref. 46 above.
7. Ref. 47 above.
8. Ref. 48 above.
9. Ref. 49 above.
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19. Ref. 59 above.
20. Ref. 60 above.
21. Ref. 61 above.
22. Ref. 62 above.
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27. Ref. 67 above.
- 28a. Progress Report: 1 Sept. 1990 - 1 Mar. 1992.

- 28b. Ref. 68 above.
29. Ref. 69 above.
30. Progress Report: 1 Mar.1992 - 15 Dec.1992.
31. Ref. 72 above.
32. Ref. 73 above.
33. Ref. 74 above
34. Ref. 75 above.
35. Ref. 76 above.
36. Ref. 77 above.
37. Ref. 81 above.
38. Ref. 70 above.
39. Ref. 71 above.
40. Ref. 78 above.
41. Ref. 80 above.
42. Ref. 79 above.
43. Progress Report: 1 July 1991 - 15 December 1993

*Reprints + Preprints removed*





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