

SOME CURRENT PROBLEMS IN BEAM-FOIL SPECTROSCOPY

L.J. CURTIS

Department of Physics and Astronomy, University of Toledo, Toledo, Ohio 43606, USA.

The use of tilted foils to produce alignment and orientation excitation asymmetries in fast ion beams is reviewed. These asymmetries, their observed dependence on various experimental parameters (ion energy, foil tilt angle, observation geometry, etc.) and their utilisation for the study of atomic and nuclear structure properties are discussed. In addition, measurements are described that utilise differential decay curve measurements to suppress cascade effects and to determine decay rates into forbidden or inhibited radiative and autoionising intercombination channels.

1. Introduction

In this review I shall attempt to summarise briefly the current status of two specific areas of research employing beam-foil excitation methods: the first is the production, study and utilisation of atomic alignment and orientation, and the second involves the use of differential lifetime measurements to determine decay rates into forbidden or inhibited channels. The two areas are quite different, but both occur by virtue of the presence of dynamical asymmetries among closely spaced energy levels – in excitation for the former and in deexcitation for the latter.

2. Alignment and orientation in beam-foil spectroscopy

Since this Conference occurs just over one decade after the first tilted-foil orientation measurements were made in Toledo [1], it is particularly fitting to review its development. Excitation asymmetries in the population of magnetic substates are essential to many of the techniques used in beam-foil work, such as determinations of fine and hyperfine structure, g factors, etc. by quantum beat and level crossing methods. The production of these asymmetries can also provide a probe of particle-solid and particle-surface interactions, short-range interionic interactions, and other collisional properties.

It has long been known that alignment (quadrupolar anisotropies) can be produced about the beam axis. Although systematic studies are far from exhaustive, alignment generally decreases for heavier atoms and higher stages of ionisation. Since the production of highly ionised atoms is one of the strongest features of beam-foil excitation, there was a clear need for an experimental parameter other than beam energy to control the production of excitation anisotropies. The use

of foil tilt can provide such a parameter [2].

A rotation of the foil about an axis perpendicular to the beam introduces a pseudovector and (despite the large microscopic surface irregularities inherent to the foil) permits the possibility of circularly polarised light. We searched for circular polarisation in an experiment at the University of Toledo 400 keV Van de Graaff accelerator in December 1973. To our great surprise, our results were positive. Since then a large number of studies have been made in attempts to understand the origin of this circular polarisation and to learn to optimise and utilise its production.

2.1. The excitation process

The phenomenological parametrisation of the problem has been described many times (cf ref. [3]). The state of polarisation of the emitted light is represented by the relative Stokes parameters M/I (the linear polarisation), C/I (related to the tilt angle of the polarisation ellipse) and S/I (the circular polarisation). The angular geometry can be removed by reformulation in terms of four spherical tensors, such as the Fano-Macek parameters A_0^c , A_1^c , A_2^c and O_1^c (cf ref. [3]) which determine some or all of the coherent excitation amplitudes of the atom if reexpressed as elements of the density matrix in the magnetic substate representation.

Some typical experimental results [4] are presented in fig. 1. This shows a contour plot over both beam energy and foil tilt angle for the $2s\ ^1S-3p\ ^1P$ transition in He I for the three relative Stokes parameters. Several trends appear. Notice that M/I (a) is oscillatory in energy, passing through maxima at approximately 150 and 700 keV (for sufficiently high ion energy it passes through zero and becomes negative [5]); and (b) decreases from positive through zero to negative values as a function of tilt angle. Similarly, notice that S/I (a) is positive, corresponding to an electron orbit turning in a

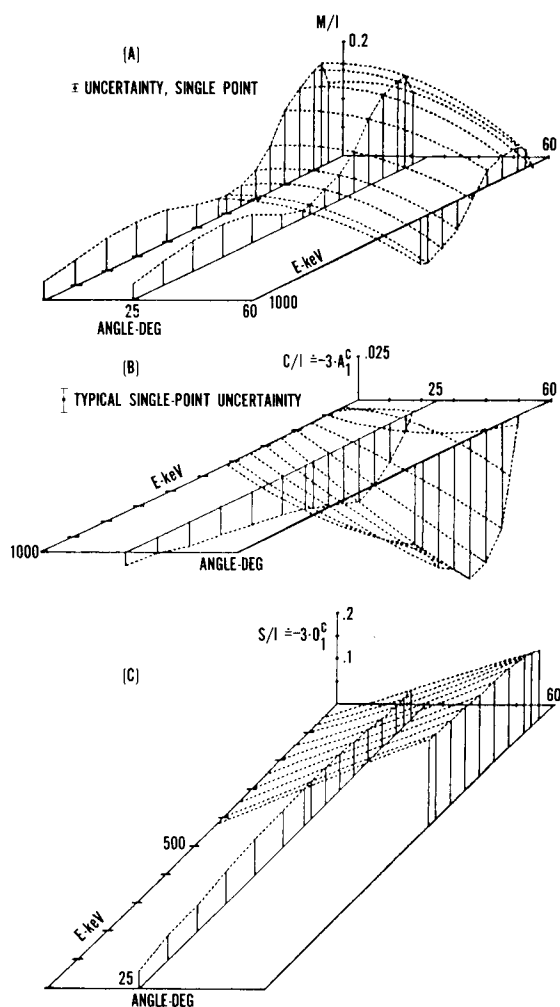


Fig. 1. Relative Stokes parameters for the He I 5015 Å $2s\ ^1S-3p\ ^1P$ transition as a function of beam energy and foil tilt angle [4].

direction opposite to that through which the foil is rotated, as a surface electron pickup model would predict; and (b) increases approximately linearly with increasing tilt angle (breakdowns in this linearity at large angles have been suggested [6], but the direction seems to be experiment-specific). Fig. 1 was drawn [4] in 1980, and the range of measurements has been extended since that time (e.g. ref. [5]), but the qualitative trends are unchanged. A number of interesting theoretical models for have been proposed (cf ref. [3]) invoking e.g. interactions with the foil bulk, the foil surface, surface fields and electron pickup, but most models contain too many free parameters to be definitively tested at present.

The viewing geometry normally chosen is a zenith angle $\theta = 90^\circ$ to the beam direction (the z axis) and two azimuth angles, $\phi = 0^\circ$ (the x axis) and 90° (the y axis)

within and perpendicular to the ($x-z$) plane formed by the beam and the normal to the foil surface. The two azimuths permit the separate determination of A_0^c and A_2^c , since

$$(M/I)_x \sim A_0^c + A_2^c; \quad (C/I)_y \sim A_1^c$$

$$(M/I)_y \sim A_0^c - A_2^c; \quad (S/I)_y \sim O_1^c.$$

This has recently led to an interesting result. When the light was viewed from the x axis it was found by Brooks et al. [7] that M/I is independent of foil tilt for the $3p\ ^3P$ level in He I. Dehaes and Carmeliet [8] have recently shown that this is also true for the $3p\ ^1P$ and $3d\ ^1D$ levels in the same spectrum, and have shown further that a model based on a strong electric field perpendicular to the foil predicts that the sum $A_0^c + A_2^c$ is independent of foil tilt angle whereas the difference $A_0^c - A_2^c$ falls off as a constant minus a secant-squared term.

Although the data of the type described above are regular and reproducible, the interactions are complicated and multifaceted. It is possible that some underlying symmetries will be found, but it is also possible that the results are system-specific, and not easily generalised. One of the major strengths of beam-foil spectroscopy lies in the study of highly ionised and highly and multiply excited atoms, and it is possible to utilise the empirical production of excitation asymmetries without inquiring more deeply into its origins.

2.2. Applications to atomic structure measurements

I have selected a few examples to illustrate the potential of the method for atomic structure measurement, especially with systems inaccessible to other methods.

For example, it has been demonstrated that doubly excited states can be aligned and oriented. Gaupp et al. [9] have measured quantum beats in the Li I $1s2s2p\ ^4P^o-1s2p^2\ ^4P$ transition using straight foils and Brooks and Pinnington [10] have observed quantum beats in the He I $2s2p\ ^3P^o-2p3d\ ^3D$ transition implying $S/I = -40\%$ for large foil tilt angles. In another area, alignment has been used by Gay and Berry [11,12] and Groeneveld et al. [13] as a tool to study the breakup of molecules. By observing the destruction of alignment in HeH^+ ions as a function of foil thickness, Gay and Berry [12] concluded that the range of the molecular influence is approximately 30 a.u.

Some success has been obtained in the production of orientation in highly ionised systems through the use of tilted foil excitation. In $^{14}\text{N IV}$ quantum beat studies of hyperfine structure have been made by Silver et al. [14] using straight foils. For the same system Berry et al. [15] have used tilted foils to observe beats between the $F=0$ and $F=1$ hyperfine levels, which are forbidden by excitation with untilted foils. Although a search [16] for

tilted foil produced circular polarisation yielded $S/I = 0$ for states in O VII and O VIII at 36 MeV at the Uppsala tandem, a similar study [17] at 4 MeV at the Lund tandem produced states of O VI with $S/I = 30\%$. The production of oriented nuclei through the transfer of atomic orientation produced by a tilted foil or a stack of tilted foils has now become a standard technique in nuclear physics (e.g. refs. [18–20]). The promising use of IBSIGI to produce substantial nuclear orientation that can subsequently be retransferred to the electrons via the hyperfine interaction and observed after passage through a straight foil is discussed by Andr  elsewhere in these Proceedings.

Alignment and orientation can also be used for the suppression of cascade effects in decay curve measurements. If excitation asymmetries occur primarily by direct excitation at the foil and there is little cascade induced alignment (CIA) or cascade induced orientation (CIO), repopulation effects can be made to cancel through appropriately normalised differential measurements [21]. In a straight foil measurement in Li II Berry et al. [22] have found that cascade tails cancelled from the efficiency-corrected difference between decay curves measured in light polarised parallel to and perpendicular to the beam axis. In a tilted foil measurement in O VI Huld et al. [17] have found indications of a similar cancellation between decay curves measured in right and left circularly polarised light. Although the ANDC method has been found to account very successfully for cascading in systems with a known level scheme and only a few strong cascades, many more systems may exhibit excitation anisotropies than satisfy the ANDC criteria.

3. Differential measurements of transition probabilities of forbidden or inhibited decay channels

Although the use of excitation asymmetries described above has not yet achieved widespread use, other types of differential decay curve measurements that can also accomplish cascade suppression have recently been applied. One class of studies has been made possible by improved wavelength resolution in lifetime measurements, and involves differences between decay curves within a fine structure multiplet due to differing decay channels. The isoelectronic behaviour of such transitions can be interesting theoretically, because the rates usually consist of several contributions with different Z dependences.

3.1. Radiative intercombination channels to the ground state

Spin intercombination transitions have recently received much experimental and theoretical study and the

subject will be treated in a forthcoming review by Martinson and Ellis [23]. The $\Delta S = 0$ selection rule on E1 transitions is rigorously valid only for pure (nonrelativistic) LS coupling and its validity is markedly diminished along an isoelectronic sequence. A number of measurements of spin-forbidden transition probabilities for metastable levels belonging to ground state complexes have been performed, but another interesting type of measurement has received much recent attention. This concerns the determination of transition probabilities of spin-intercombination decay channels for terms that are not within the ground complex and are not metastable, but exhibit decay asymmetries among fine structure levels.

3.1.1. The $2s3p\ ^3P_j^o$ levels in the beryllium sequence

The $2s3p\ ^3P^o$ term in the Be sequence provides an excellent example of this type of system. It has E1-allowed transitions to the $2s3s\ ^3S$ term for all three fine structure levels, and the $J = 1$ level acquires an E1 transition channel to the ground state $2s^2\ ^1S$ as spin mixing causes the $\Delta S = 0$ selection rule to break down. Owing to its large excitation energy relative to ground and the small energy difference between the $2s3p\ ^1P_1^o$ and $^3P_1^o$ levels, this intercombination channel becomes competitive with the intraspin channels at moderate stages of ionisation. Relative decay curve measurements of the $J = 2, 1, 0$ levels were performed for N IV, O V and F VI by Engstr m et al. [24] and for Ne VII by Hardis et al. [25]. Semilogarithmic plots of the three decay curves for the Ne VII case are shown in fig. 2. The difference between the reciprocal lifetimes of the $J = 0, 2$ and the $J = 1$ curves is essentially equal to the intercombination rate. This rate can be obtained directly from the time derivative of the logarithmic intensity ratio of the $J = 2$ and $J = 1$ decay curves, shown in fig. 3. These measurements have motivated a number of theoretical studies [26–28], and Ellis [27] has cited this

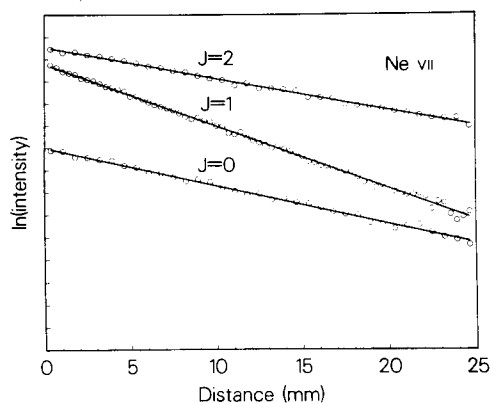


Fig. 2. Semilogarithmic plot of the fine structure decay curves of the $2s3p\ ^3P_j^o$ levels in Ne VII [25].

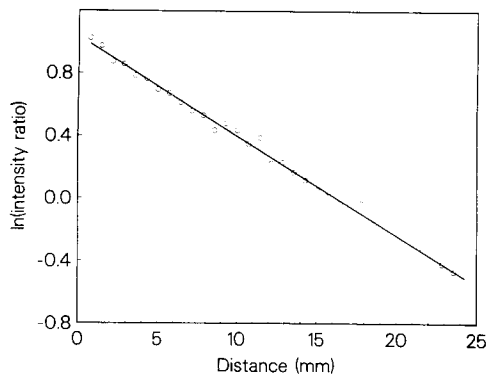


Fig. 3. Semilogarithmic plot of the ratio of the intensities of the $J=2$ and $J=1$ decay curves shown in fig. 2.

lifetime difference as a useful test of theoretical methods, since the system is simple enough to be tractable, yet the calculation must include configuration interaction, intermediate coupling and relativistic effects. An isoelectronic plot of available measurements and calculations is given in fig. 4.

3.1.2. The $3s4p\ ^3P_j^o$ levels in the magnesium sequence

The situation becomes even more interesting if we move to the homologous term in the Mg sequence. Since, unlike the Be case, the ground complex here contains d electrons, a large number of "plunging levels" cross the $3s4p$ levels along the isoelectronic sequence. Unfortunately, until recently few spectroscopic analyses that included more than the ground complex for this sequence were available, but lifetime and classification studies have now been constructively combined.

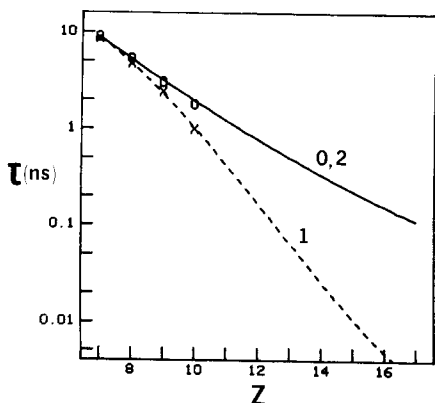


Fig. 4. Lifetime versus atomic number for the $2s3p\ ^3P_j$ levels in the Be isoelectronic sequence. Theoretical values [26,27] are represented by a solid line for $J=0$ and 2, and by a dashed line for $J=1$. Experimental measurements [24,25] are represented by (O) for $J=0$ and 2 and by (x) for $J=1$.

The laboratory at Lund University is unique in its capability and tradition of doing high resolution classical spectroscopy side-by-side with beam-foil measurements, and their recent measurements in this sequence provide a breakthrough. Complete and comprehensive spectroscopic analyses have been completed at Lund for P IV [29], S V [30] and Cl VI [31]. The Cl VI work (by Jupén and Fremberg [31]) is especially noteworthy because it was done in large part using high wavelength resolution beam-foil measurements.

Lifetime studies were motivated when sliding spark spectroscopic measurements revealed that intercombination lines from $3s4p\ ^3P_1$ to all lower singlet levels were anomalously strong in S V. Beam-foil decay curve measurements were subsequently made at Lund by Reistad et al. [32] which provide useful insights into the system. The lifetime extractions were accomplished using the ANDC technique with the programme CANDY [33] incorporating the important $3s4d\ ^3D$ cascade into the analysis. Unlike the case for the Be sequence, there is no measureable J -dependence in the decay curves for the five times ionised member, Cl VI, but a striking difference occurs for S V. The spectroscopic analysis shows that this arises from a crossing of the $3s4p\ ^3P$ configuration by the plunging $3p3d\ ^3P$ term, shown in fig. 5, which strongly mixes the singlet and triplet. Calculations made by Reistad et al. [32] and by Godefroid [34] confirm these results.

It would be interesting to extend the studies further along this sequence to see if a smooth isoelectronic variation of the $J=2$ and $J=1$ lifetimes, similar to that in the Be sequence, occurs. For members of this sequence a few charge states higher the wavelengths would fall within the range of position sensitive detectors. This could permit the intensity ratios of the fine structure decay curves to be registered directly and simultaneously, greatly reducing the uncertainties introduced by experimental drifts over the long accumulation times required to detect these small differences. Homologous measurements of the $4s5p\ ^3P_j^o$ in the zinc sequence would also be interesting. A necessary prerequisite for these lifetime measurements is a comprehensive spectroscopic analysis, and this indicates the tasks which confront beam-foil spectroscopy.

3.2. Intercombination autoionising channels to the continuum

Multiply excited states sometimes provide a selective decay channel to the continuum that is similar to the decay channel to the ground state provided by singly excited spin intercombination transitions. The lithium-like quartets exemplify this since they represent the simplest case of doubly excited states that differ in spin multiplicity from the singly excited states. They are metastable both to Coulomb autoionisation and to tran-

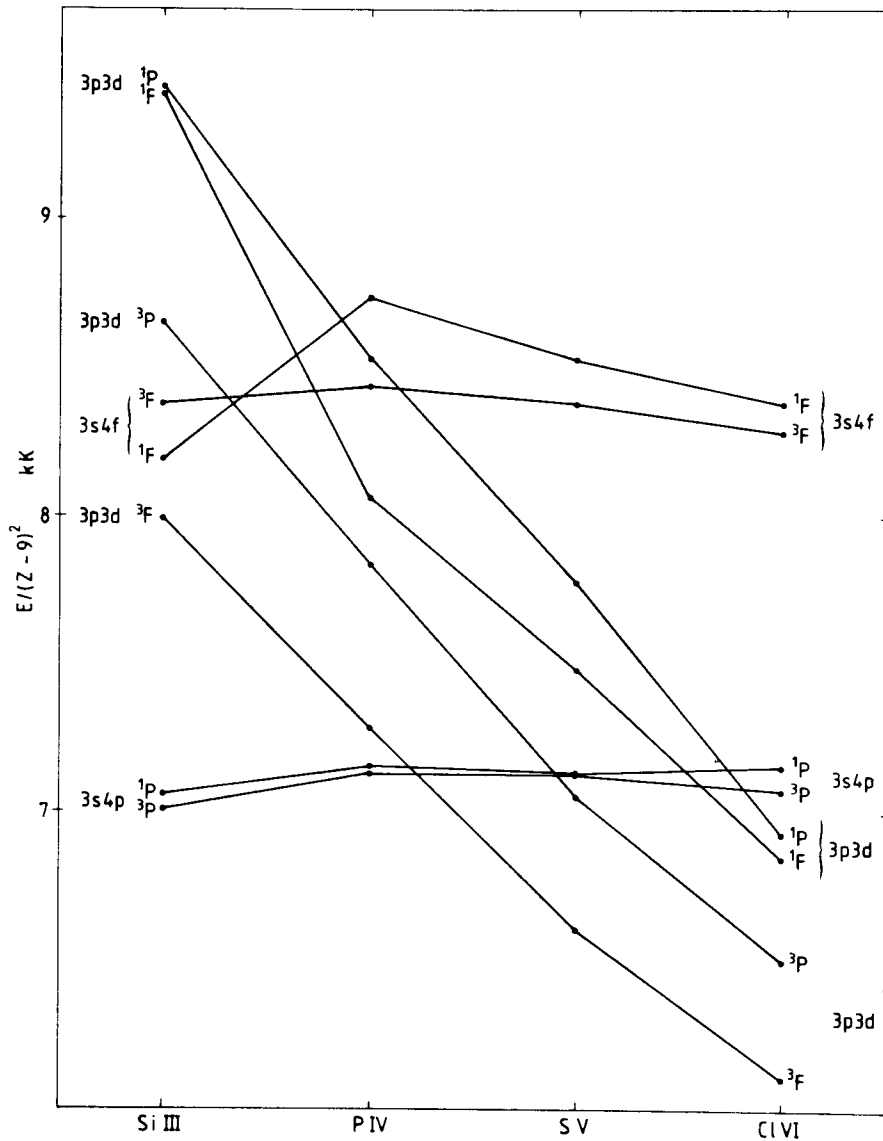


Fig. 5. Isoelectric study of some of the low configurations of the Mg sequence from Si III to Cl VI [32].

sitions to singly excited states, allowing E1 transitions among doubly excited states to compete favourably with other deexcitation modes and to significantly affect X-ray fluorescence yields.

The $1s2p^2\ ^4P$ term provides an autoionising analogue to the radiative intercombination channels described in section 3.1, since it undergoes transitions to the lower lying $1s2s2p\ ^4P^o$ inner-shell doubly excited state. The lifetimes of the fine structure components thus exhibit a "differential metastability" in which a weak mixing of the $1s2p^2\ ^4P_{5/2}$ level with the rapidly autoionising $^2D_{5/2}$ level of the same configuration causes a substantial shortening in its lifetime relative to those of the $^4P_{3/2}$ and $^4P_{1/2}$.

Recent spectroscopic measurements of the fine structure wavelengths [35–40] and fine structure lifetimes [35,38–42] have stimulated theoretical interest [43–46] in this system. These studies require reasonably good spectroscopic resolution, which can be obtained through the technique of spectrometer refocusing to a moving light source. As an example, one spectral scan from our Ne VIII [40] study is shown in fig. 6. All seven fine structure components can be discerned, as well as the $n = 5-7$ hydrogenic transition in lithium-like Ne VIII. The centers for all but the $\frac{1}{2}-\frac{1}{2}$ and $\frac{5}{2}-\frac{5}{2}$ were determined to within $\pm 0.05\ \text{\AA}$, specifying the four fine structure intervals of the upper and lower states. The results agree with the calculations of Chung [45] to within about $\pm 0.2\ \text{\AA}$.

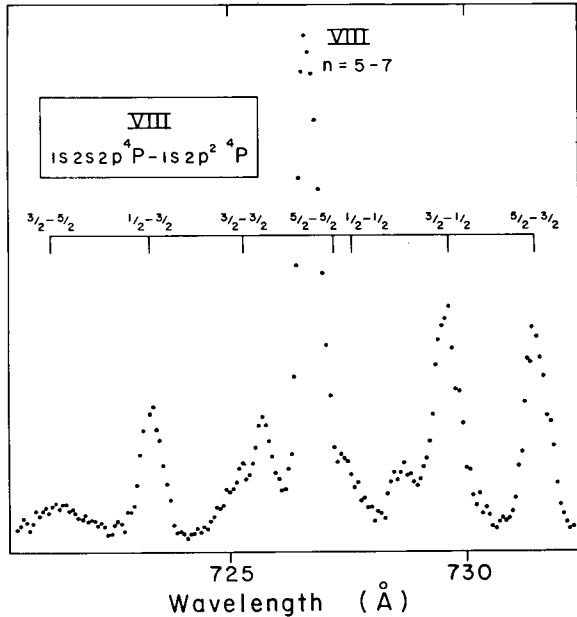


Fig. 6. Spectrum of foil-excited 7 MeV neon showing the resolved fine structure of the multiplet connecting the two lowest quartet terms and the quasihydrogen-like $n = 5-7$ manifold in lithium-like Ne VIII [40].

In addition to our measurements [40] of the lifetimes in Ne VIII, recent lifetime measurements have been made in F VI at Lund [38], in Mg X and Al XI at Bochum [42] and in Al XI at Lyon [39]. The existing base of lifetime data for the $1s2p^2\ ^4P_j$ levels is shown in fig. 7 and compared with the theoretical calculations of Chen et al. [43]. These calculations include relativistic intermediate coupling with configuration interaction, and are in much closer agreement with experiment than were earlier nonrelativistic calculations [46]. The difference between the reciprocal lifetime of the $J = \frac{5}{2}$

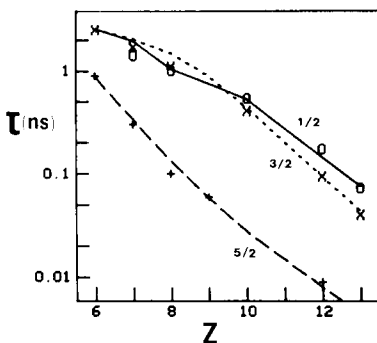


Fig. 7. Lifetime versus nuclear charge for the fine structure components of the $1s2p^2\ ^4P_j$ decay. Experimental points [35, 38-42] are denoted by (○) for $J = \frac{1}{2}$; (×) for $J = \frac{3}{2}$; and (+) for $J = \frac{5}{2}$. Theoretical curves [43] are solid for $J = \frac{1}{2}$; dotted for $J = \frac{3}{2}$; and dashed for $J = \frac{5}{2}$.

level and that of either the $J = \frac{3}{2}$ or the $J = \frac{1}{2}$ levels is essentially a measure of the Auger decay rate of the $J = \frac{5}{2}$, which (as in the radiative case of fig. 3) is given directly by the time derivative of the logarithmic ratio of the $J = \frac{5}{2}$ and $J = \frac{3}{2}$ decay curves.

Beam-foil methods are well suited to differential decay curve measurements of this type since they utilise the unique excitation and decay properties of this source and are not seriously affected by the cascading.

I am grateful to J.C. Dehaes, E. Träbert and N. Reistad for providing me with their results prior to publication and to H.G. Berry, D.G. Ellis, I. Martinson, E.H. Pinnington and R.M. Schectman for helpful suggestions. This work was partially supported by the US Department of Energy, Division of Chemical Sciences, under contract number DE AS05 80ER10676.

References

- [1] H.G. Berry, L.J. Curtis, D.G. Ellis and R.M. Schectman, Phys. Rev. Lett. 32 (1974) 751.
- [2] D.G. Ellis, J. Opt. Soc. Am. 63 (1973) 1232.
- [3] H.G. Berry and M. Hass, Ann. Rev. Nucl. Part. Sci. 32 (1982) 1.
- [4] R.M. Schectman, R.D. Hight, S.T. Chen, L.J. Curtis, H.G. Berry, T.J. Gay and R. DeSerio, Phys. Rev. A22 (1980) 1591.
- [5] T.J. Gay, H.G. Berry, R. DeSerio, H.P. Garnir, R.M. Schectman, N. Schaffel, R.D. Hight and D.J. Burns, Phys. Rev. A23 (1981) 1745.
- [6] J.C. Dehaes, M. Schenkel and J. DeVooght, Nucl. Instr. and Meth. 202 (1982) 235.
- [7] R.L. Brooks, H.G. Berry and E.H. Pinnington, Phys. Rev. A25 (1982) 2545.
- [8] J.C. Dehaes and J. Carmeliet, J. Phys. B 17 (1984) 3029.
- [9] A. Gaupp, M. Dufay and J.L. Subtil, J. Phys. B 9 (1976) 2365.
- [10] R.L. Brooks and E.H. Pinnington, Phys. Rev. A22 (1980) 529.
- [11] T.J. Gay and H.G. Berry, J. Phys. B 13 (1980) L199.
- [12] T.J. Gay, H.G. Berry and R. DeSerio, Phys. Rev. A23 (1981) 1761.
- [13] K.O. Groeneveld, G. Astner, S. Hultberg, S. Mannervik and P.S. Ramanujam, J. Phys. B 13 (1980) L205.
- [14] J.D. Silver, J. Desesquelles and M.L. Gaillard, J. Phys. B 8 (1975) L219.
- [15] H.G. Berry, L.J. Curtis, D.G. Ellis and R.M. Schectman, Phys. Rev. Lett. 35 (1975) 274.
- [16] L.J. Curtis, R. Hallin, J. Lindskog, J. Pihl and H.G. Berry, Phys. Lett. A60 (1977) 297.
- [17] S. Hultdt, L.J. Curtis, B. Denne, L. Engström, K. Ishii and I. Martinson, Phys. Lett. A66 (1978) 103.
- [18] C. Broude, E. Dafni, A. Gelberg, M.B. Goldberg, G. Goldring, M. Hass, O.C. Kistner and A. Zemel, Phys. Lett. B 105 (1981) 119.
- [19] E. Dafni, G. Goldring, M. Hass, O.C. Kistner, Y. Niv and A. Zemel, Phys. Rev. C25 (1982) 1525.

- [20] Y. Nojiri and B.I. Deutch, *Phys. Rev. Lett.* 51 (1983) 180.
- [21] L.J. Curtis, R.M. Schectman, J.L. Kohl, D.A. Chojnacki and D.R. Shoffstall, *Nucl. Instr. and Meth.* 90 (1970) 207.
- [22] H.G. Berry, L.J. Curtis and J.L. Subtil, *J. Opt. Soc. Am.* 62 (1972), 772
- [23] I. Martinson and D.G. Ellis, *Comments Atomic Molecular Physics* 16 (1985) 21.
- [24] L. Engström, B. Denne, S. Hultdt, J.O. Ekberg, L.J. Curtis, E. Veje and I. Martinson, *Physica Scripta* 20 (1979) 88.
- [25] J.E. Hardis, L.J. Curtis, P.S. Ramanujam, A.E. Livingston and R.L. Brooks, *Phys. Rev. A* 27 (1983) 257.
- [26] A. Hibbert, *J. Phys. B* 12 (1979) L661
- [27] D.G. Ellis, *Phys. Rev. A* 28 (1983) 1223.
- [28] A. Weiss and Y.-K. Kim, to be published.
- [29] P.O. Zetterberg and C.E. Magnusson, *Physica Scripta* 15 (1977) 189.
- [30] I. Joelsson, P.O. Zetterberg and C.E. Magnusson, *Physica Scripta* 23 (1981) 1087.
- [31] C. Jupén and J. Fremberg, to be published.
- [32] N. Reistad, T. Brage, J.O. Ekberg and L. Engström, *Physica Scripta* 30 (1984) 249.
- [33] L. Engström, *Nucl. Instr. and Meth.* 202 (1982) 369.
- [34] M. Godefroid, personal communication.
- [35] A.E. Livingston and H.G. Berry, *Phys. Rev. A* 17 (1978) 1966.
- [36] E. Träbert, H. Hellmann, P.H. Heckmann, S. Bashkin, H.A. Klein and J.D. Silver, *Phys. Lett. A* 93 (1982) 76.
- [37] E. Träbert, H. Hellmann and P.H. Heckmann, *Z. Phys.* A313 (1983) 373.
- [38] I. Martinson, B. Denne, J.O. Ekberg, L. Engström, S. Hultdt, C. Jupén, U. Litzén, S. Mannervik and A. Triguerios, *Physica Scripta* 27 (1983) 201.
- [39] J.P. Buchet, M.C. Buchet-Poulizac, A. Denis, J. Desesquelles, M. Druetta, S. Martin, J.P. Grandin, D. Hennescart, X. Husson and D. Lecler, *J. de Phys. Lett.* 45 (1984) L-361.
- [40] A.E. Livingston, J.E. Hardis, L.J. Curtis, R.L. Brooks and H.G. Berry, *Phys. Rev. A* 30 (1984) 2089.
- [41] J.R. Mowat, K.W. Jones and B.M. Johnson, *Phys. Rev. A* 20 (1979) 1972.
- [42] J.H. Blank, P.H. Heckmann and E. Träbert, *Jahresberichte, Dynamitron Tandem Laboratorium, Bochum* (1983) p. 57.
- [43] M.H. Chen, B. Crasemann and H. Mark, *Phys. Rev. A* 26 (1982) 1441.
- [44] J. Hata and I.P. Grant, *J. Phys. B* 16 (1983) 915.
- [45] K.T. Chung, *Phys. Rev. A* 29 (1984) 682.
- [46] T.W. Tunnell and C.P. Bhalla, *Phys. Lett. A* 67 (1978) 119.