Recent Progress in Atomic-structure Investigations with Fast-ion Beams

The excitation of fast-ion beams in solid or gaseous targets yields important information about the atomic structure of highly ionized species. Recent developments in this area of atomic physics are reviewed, with the emphasis on accurate determinations of energy-level structure and decay times for multiply ionized atoms.

Introduction

Beams of heavy ions from particle accelerators can be successfully used for atomic-physics experiments. The most common technique of this kind, beam–foil spectroscopy (BFS), was introduced in the early 1960s by Kay[1] and Bashkin.[2] It has gradually developed into a standard method in atomic physics. The quantities which can be studied by BFS include excitation energies in atoms and ions, fine and hyperfine structure, radiative lifetimes and sometimes quantum-electrodynamic (QED) effects such as Lamb shifts. In a typical BFS experiment the fast mono-energetic ions are sent through a thin (~500 Å) carbon foil, located in an evacuated target chamber. When emerging from the foil, the ions are in excited states which decay spontaneously by photon (or electron) emission. The BFS method thus provides a light source for atomic-spectroscopy experiments. Since the velocity of the excited ions is accurately known, measurements of spectral-line spatial decay lengths yield lifetimes of excited states in atoms and ions. This was the earliest and most obvious application of the BFS method. Although it was later found to possess some disadvantages, it is the only presently available approach by which atomic lifetimes in highly ionized systems can be determined. Much interest has recently been focused on such data, largely because of their relevance to the impurity problems in Tokamak discharges.[3–5]

Some recent progress in BFS is reviewed in this Comment. For detailed discussions of new results, we refer to the proceedings of the latest beam–foil conference[6] and to the thorough review by Andrä.[7]

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As a spectroscopic light source, the foil-excited ion beam has several interesting properties. Beams of practically any element can be obtained from modern heavy-ion accelerators, and very many ionization and excitation states are available (nonselective excitation) in BFS. The ion-foil excitation processes favor the population of multiply excited or inner-shell excited configurations in the fast ions. Also, high-lying Rydberg states (with high n and l quantum numbers) are strongly excited at ion energies in the MeV range. The wavelength accuracy of beam-foil measurements has some inherent limitations, caused by Doppler effects of the moving ions. However, this Doppler dispersion possesses an angular dependence which can be exploited to narrow and enhance the lines. Optical "refocusing" methods have thus been developed[8-10] and beam-foil spectra of high quality can now be routinely obtained. Livingston *et al.*[11] have recently obtained 0.5-0.8 Å linewidths in the region 600-900 Å and they determined the wavelengths of the 1s2s $^3S_1$-1s2p $^3P_{02}$ transitions in He-like Si, S and Cl with relative errors as low as 10^{-4}. For He-like Si, similar work has been reported by Armour *et al.*[12] The high accuracies of these two BFS investigations made it possible to determine the QED corrections (which here amount to 0.4-0.7% of the transition energies) with uncertainties as low as 3%. The effects of QED for high-Z systems can thus be quite accurately studied by means of BFS methods.

In an investigation of the 1s2s2p ^4P-1s2p^2 ^4P multiplet in Li-like C IV, N V and O VI, Livingston and Berry[13] obtained 0.3Å linewidths and this resolution made it possible to determine the fine structure of these doubly excited terms very accurately. Using relativistic multiconfiguration Hartree-Fock wave func-

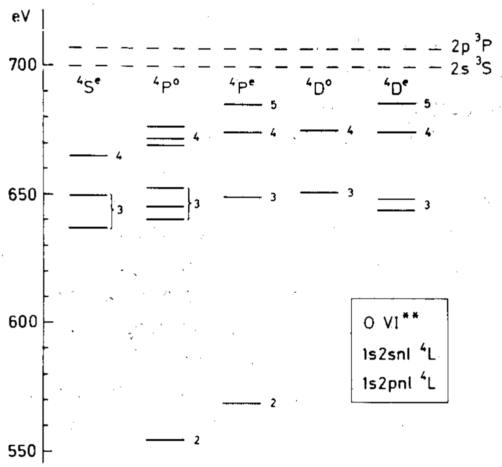

FIGURE 1. Energy-level diagram of the 1s2snl and 1s2pnl ^{4}L levels in O VI (according to Hannebauer *et al.*[17]). These levels lie more than 400 eV above the O VII ground state, and they converge to the 2s and 2p levels of this ion.

tions, Cheng *et al.*[14] subsequently calculated these fine-structure separations. The importance of the Breit interaction was thereby clearly demonstrated.

The 1s2s*nl* and 1s2p*nl* ^{4}L levels in Li-like systems are strongly populated in beam–foil experiments, and thus they have been investigated by photon or electron spectroscopy for Li I–Ar XVI.[15,16] These quartet levels have high excitation energies (they lie far above the $1s^2\ ^1S$ ionization limit) but – because of the $\Delta S = 0$ selection rule – they cannot autoionize into the $1s^2 \epsilon l$ doublet continuum via the Coulomb repulsion process. Figure 1 shows the energy-level diagram for the quartets in O VI, based on the beam–foil work of Hannebauer *et al.*[17] The levels indicated have been connected with many transitions in the range 100-200 Å. There is considerable configuration interaction, e.g., the odd ^{4}P terms can arise from 1s2s*n*p, 1s2p*n*s and 1s2p*n*d configurations. Note that the lowest quartet term, 1s2s2p $^4P^o$, lies more than 400 eV above the O VII ground state! Autoionization via the spin–orbit, spin–other-orbit or spin–spin mechanisms starts to play an important role when Z increases. The 1s2s2p $^4P_{5/2}$ state autoionizes via the spin–spin process. (For high Z, the M2 decay to the $1s^2 2s\ ^2S$ ground state must also be considered.) Lifetime measurements by electron spectroscopy have provided experimental data for many ions up to Ar XVI. A systematic deviation from theoretical calculations has been noted here.[16] However, a recent experimental reinvestigation[18] for Ar XVI produced results in good agreement with theoretical data.[19] Additional work, preferably extended to higher Z values, is clearly desirable. The high excitation energy and metastability of the 1s2s2p ^{4}P term makes it an interesting candidate for laser processes. Harris[20] has thus proposed a technique whereby a 207-Å laser can be constructed.

The high cross sections for populating high-lying nonpenetrating states in beam–foil experiments facilitate accurate determinations of transition wavelengths for the hydrogen-like parts of the spectra.[21] (It should be noted that such lines are absent or weak in most other light sources.) The wavelengths for these transitions can be predicted by a simple model in which an outer electron orbits a polarizable core. However, the experimental data are also useful for the determination of dipole and quadrupole polarizabilities of the core, as well as of ionization energies. The dependence of lifetimes on core polarizability has been investigated recently,[22] and predictions which could be tested by suitable beam–foil experiments have been made.

Lifetime Studies

The improved spectral resolution, as noted above, has been most useful for lifetime determinations. Livingston and Berry[13] were thus able to measure the lifetimes of the $1s2p^2\ ^4P_J$ levels individually and they found that the $J = 5/2$ state had a lifetime which was an order of magnitude shorter than those of the $J = 3/2$ and $J = 1/2$ levels, presumably because of mixing with the $1s2p^2\ ^2D_{5/2}$ state, which rapidly autoionizes to the doublet continuum. Similarly, Engström *et al.*[23] have obtained 1-Å linewidths in the near ultraviolet in a study of triplet P levels of four-electron systems in nitrogen, oxygen and fluorine, which enabled them to measure separately the $1s^2 2s3p\ ^3P_J$ fine-structure levels. They found that the $J = 1$ states had slightly shorter lifetimes than the $J = 2$ and $J = 0$ states, in a manner which varied systematically with the nuclear charge. They attributed this difference to the spin-forbidden $2s^2\ ^1S$–$2s3p\ ^3P_1$ transition, and therefore were able to deduce the transition probability for the intercombination line from dipole-allowed lifetime measurements. The ability to study such effects isoelectronically demonstrates one of the strengths of the beam–foil method.

The development of beam-detection windows with very narrow spatial resolution is another important improvement. In the extreme ultraviolet region To and Drouin[24] have developed a near-beam auxiliary-entrance-slit modification for a grazing-incidence monochromator which permits viewing of a beam segment as short as 80 μm. For the x-ray region, Varghese *et al.*[25] have utilized a near-beam source slit to view a beam segment as short as 20 μm. By these methods picosecond lifetimes are now being reported with uncertainties as low as 5%. At present this seems to be the lower limit for direct lifetime measurements, but shorter lifetimes have been measured indirectly by nonproportional x-ray yields[26] and by natural linewidth studies.[27]

Great advances have also been made in the statistical accuracy and reproducibility of beam–foil-measured decay curves. In the very early work, problems due to foil degradation and beam fluctuations were sometimes severe, but today these problems have been largely eliminated through improved instrumentation. The decay curves are obtained by stepwise translation of the foil along the beam axis with a machine screw of low backlash and high positional accuracy (often to within ±0.05 mm), usually driven by a stepping motor which is shaft encoded to provide gate pulses to the light-detection and multiscaling apparatus. The foil advance is often gated by some monitoring system so as to account for small beam fluctuations and foil-aging effects. (For example, the foil may be advanced after a fixed number of counts are accumulated from a light monitor at a fixed distance from the foil, with dark count corrections made for the dwell time.) Through the use of computer-automated equipment the decay curves are retraced quickly and repetitively to average over short-term fluctuations. Successive runs can be compared for consistency in buffer storage units. The speed of the foil-emergent beam can be continuously monitored by an electrostatic analyzer or by a Doppler-shift measurement.[28] Many of these refinements were developed primarily to make possible the observation of fast, low-amplitude quantum beats,[7] but this increased instrumental sophistication has had a dramatic effect on the reliability of beam–foil lifetime measurements.

Much attention has been devoted to the problem of cascade repopulation in beam–foil measurements. Because of the nonselective nature of the excitation process, the decay curves consist of a sum of exponential contributions, one for the primary level and one for each level which cascades (directly or indirectly) into it. This problem is not unique to beam–foil excitation, but occurs in nearly all atomic-lifetime measuring techniques which have any degree of generality in their application. However, other techniques are often beset with so many additional problems (pressure dependences, sample purity, interionic fields, etc.) that cascading is not so prominently mentioned as it is in beam–foil applications. Unlike the analogous case in nuclear physics, the various sequential decay processes in atomic systems often have lifetimes of the same order of magnitude, which can be difficult to separate if the exponential term corresponding to the level of interest is not dominant. In most cases this is not a serious problem, and reliable lifetimes can be extracted by multiexponential fitting techniques. In cases where this is a problem, reliable alternative analysis techniques are available,[29] but these require combined measurements of all important cascade-related levels, greatly complicating the experimental requirements. The measurements must therefore involve a judgment as to whether the state studied can or cannot be extracted by multi-exponential fitting. This is not normally a difficult decision, and can be made on the basis of the apparent complexity of the decay curve. However, decay curves can occasionally appear to be deceptively simple. In order to identify troublesome cascade situations, much effort has been devoted to the simulation of beam–foil decay curves,[30,31] using theoretical lifetimes and assumed population models. These efforts have shown that probably the most difficult of all cascading problems occur for the lowest-lying $\Delta n = 0$ resonance transitions in the highly ionized members of the alkali and alkaline-earth sequences, which are affected much too severely by cascade repopulation to permit lifetime extraction by exponential-curve-fitting techniques. This does not, however, preclude their reliable determination by beam–foil methods, if appropriate analysis techniques are employed. For the Kr VIII 4s–4p transition, a case judged to be very difficult by these simulation criteria,[31] recent beam–foil measurements[32,33] using the "ANDC method" have extracted lifetimes which agree to within 5% with theoretical estimates (which should be expected to be reliable for this single-valence-electron system), superceding earlier curve-fitting results which overestimated the lifetimes by 40%. The ANDC method[29] requires the measurement of the decay curves of the primary level and all important cascade-coupled levels, which are analyzed through correlations inherent in the population differential equation. Thus the results indicate that the theoretical estimates are reliable for the highly ionized single-valence-electron spectra (e.g., the Na and Cu isoelectronic sequences) and that the ANDC technique does extract reliable lifetimes even when severe cascading problems are present. Thus the ANDC method can, with confidence, be applied to two-valence-electron spectra (e.g., the Mg and Zn isoelectronic sequences). Here, theoretical lifetime calculations are much more difficult, and great reliance must be placed on experimental determinations. Since the lifetimes of these highly ionized one- and two-valence-electron systems have important applications in fusion-reactor

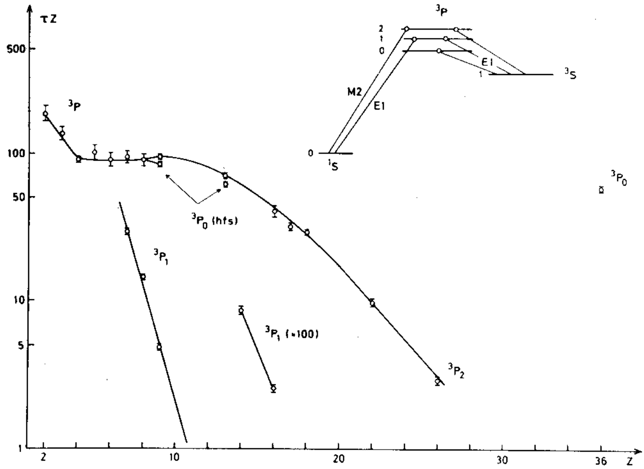

FIGURE 2. Lifetimes (multiplied by Z) of the 1s2p $^3P_{0,1,2}$ levels in the He I isoelectronic sequence. All data shown here originate from beam-foil experiments. Note the effects of the spin-forbidden decay mode on the $^3P_1$ lifetime and the shortening of the $^3P_2$ lifetime because of the M2 decay channel. For systems with nonzero nuclear spin, the $^3P_0$ lifetime may also be shortened.

development,[3-5] the use of ANDC analyses of beam-foil measurements can be expected to provide particularly valuable information in the near future.

One somewhat puzzling result has come out of the simulation analysis described above – the presence of very pronounced long tails on the experimental decay curves, which are not present in simulated decay curves unless use is made of population models that have a preponderance of excitation in states of very high principal quantum numbers.[34] Various alternative population mechanisms have been suggested,[35] but the origin of these very long tails is not presently understood.

A good illustration of the importance of isoelectronic studies is shown in Figure 2, which displays the lifetimes (multiplied by the nuclear charge Z) for the 1s2p $^3P_{0,1,2}$ levels in the He I isoelectronic sequence. For low Z, all three ^{3}P levels have similar lifetimes, determined by the 1s2s ^{3}S–1s2p ^{3}P transition probability. With increasing Z, the $^3P_1$ lifetime is drastically shortened, because of the intercombination transition to the 1s^2 ^{1}S ground state. A similar shortening of the $^3P_2$ lifetime (caused by M2 transitions) takes place when Z increases further. This was first shown by Marrus and Schmieder.[36] (A fine review of forbidden transitions in one- and two-electron spectra has been written by Marrus and Mohr,[37] where references to most of the beam-foil data shown in Figure 2 can be found.) For systems with nonzero nuclear spin I, the $^3P_0$ lifetime also undergoes shortening, because of hyperfine-induced E1 transitions to the ground state. In highly ionized atoms this effect was first studied by Gould *et al.*[38] In later beam-foil experiments, Engström *et al.*[39] and Denne *et al.*[40] have determined the probability for this hyperfine-induced decay in F VIII ($I = 1/2$) and Al XII ($I = 5/2$). The data confirm the calculations of Mohr,[41] based on perturbation theory.

About 10 years ago, there was a good deal of excitement about some accurate beam-foil lifetimes for Fe I[42] which, together with other modern *f*-value determinations, proved that the previously assumed Fe abundance in the solar photosphere was wrong by an order of magnitude.[43] Such work has been continued both for the iron-group elements and for other systems, and much data of relevance to solar-abundance problems have been obtained in recent years. The beam-foil studies have also been extended to the rare-earth ions, the solar abundances of which present interesting problems.[44] In a typical experiment, Andersen *et al.*[45] determined the lifetimes of several excited levels in singly ionized La, Ce, Pr, Nd, Sm, Yb and Lu. These data showed that previously assumed solar abundances of these elements had to be drastically revised whereas agreement between solar and meteoritic abundances improved substantially.

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