40 years of Atomic Physics using Synchrotron Radiation

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Abstract Experiments in atomic physics using several synchrotron radiation sources worldwide over the last 40 years, and in which I have been personally involved, are described.

1 Introduction

It would be impossible for me to give a worldwide account of SR work in atomic physics, so in this paper I shall focus on some of the experiments in which I was involved and which marked my career in SR research. My story began in 1970, when I joined Geoff Marr's VUV Spectroscopy group at Reading University as a post-doctoral research assistant. I began to work closely with Keith Codling; at that time the main interest was in setting up a beamline on the Daresbury 5 GeV electron synchrotron to carry out photoionization measurements on gases. My task was to design and have built a grazing incidence monochromator with emphasis on order sorting ability, Throughout my career my primary interest and ability has been in the construction of optical instruments and experimental apparatus, with a view to putting them to use in making absolute measurements as well as measurements of electronic spectra.

2 Absolute photoionization cross section measurements of atoms: the heat pipe

The aim here was to measure the photoionization cross sections of metal atoms, and for this a vapour cell was required. The heat pipe was chosen, a device which contains the metal vapour by an inert buffer gas at each end, an internal mesh being used to circulate the condensed metal within the containment region. The technique avoids having to use vapour pressure curves, which are in many cases accurate to only a factor of two or worse, to determine the metal vapour density. By measuring the absorption difference between the cell with just buffer gas in it and the cell when hot, the absorption path length and the buffer gas pressure, the absolute cross section can be obtained. At this point it became clear that in the spectral region where we were working, 50 – 400Å, the helium buffer gas cross section was not known with sufficient accuracy. So we embarked on a series of measurements to determine this, expanding it to cover other rare gases.

The cell worked best at high temperatures, because that defined the length of the absorbing column better, and we noted that David Ederer and Caroline Mehlman were already using the same device to make measurements on lithium. Our work focused on sodium (1), which worked reasonably well, and cadmium for which it worked less well. Our measurements had an uncertainty of ± 25%, and for both these two atoms these remain, to my knowledge, the only absolute photoionization cross section measurements available in this spectral region.

3 Photoelectron spectroscopy of the rare gases

With the much greater photon intensity provided by synchrotron radiation compared to helium continuum or discharge sources, it became possible to make a continuous series of

measurements of the partial cross sections and electron angular distributions on the inner shells of the rare gases. In this way electron-electron correlations could be highlighted, and the effect on the 5s and 5p electrons of xenon in the region of the 4d ionisation was seen, clearly reflected in the 5s and 5p cross sections and the electron angular distribution data (2), the latter being completely at variance with calculations at the time. Notably, a calculation using the RPAE method, by Miron Amusia and his student Vadim Ivanov (3) before they had seen the experimental data, was in excellent agreement with experiment.

4 Absolute cross section measurements on atomic ions

After the closure of the 5 GeV electron synchrotron in 1977, work on photoelectron spectroscopy (PES) of atoms and molecules ceased at Daresbury until the SRS came into operation during the early 1980s. During this period of shutdown I spent a year as SURF fellow at NBS, a year which proved to be highly enjoyable as well as very productive. Most of the research carried out during that period was on PES of molecules, focussing on shape resonant phenomena, so is not appropriate to this paper. However some atomic work was undertaken, on the β-values and branching ratios in the regions of the $3s3p^64p$ and $5s5p^66p$ $^1P_1$ resonances in argon and xenon (4). The sensitivity of the β-parameter to the resonances was evident; it was notable that the minima in β and the absorption cross section occur at different wavelengths

My interest in atomic work continued with measurements on the alkaline earths, in this case strontium, during time spent at the Photon Factory in Japan. Using a time-of-flight spectrometer, the single and double ionization spectra of the neutral atom were measured. But these were relative measurements; an entirely new kind of experiment in photoionization, brought to Daresbury by Ken Dolder and Barry Peart from the University of Newcastle in the UK, led the way to making absolute photoionization cross section measurements on atomic ions. They applied the technique of merged beam spectroscopy, which they had developed for beams of electrons and ions in their own laboratory, to make measurements on singly charged atomic ions. The absolute cross section can be readily calculated from this experiment; full details are given in reference 5. Initial results were not encouraging, since the photon flux from the 5-metre normal incidence MacPherson monochromator was much lower than expected. There was a hint of a high cross section around 580Å in the Ba$^+$ spectrum, but this result was already known from trial experiments using a helium lamp at the university in Newcastle.

It remained to find out the cause of the low photon flux from the monochromator, and this turned out to be due to some confusion over the direction of the blaze angle in the diffraction grating in the monochromator. Once this was corrected by rotating the grating in its mount, the flux increased 100-fold to 10^{12} photons/sec/Å. This, together with an improved method of aligning the photon beam to the ion beam, transformed the situation. The first results, on the Ba$^+$ ion, were staggering (5). We watched amazed as the signal at the main peak of the resonances in the 5p – 5d resonance region kept climbing, with a peak cross section of 3000Mb, ie 3 X 10^{-15} cm^2 and an oscillator strength of 2.8 for the four peaks seen. Further measurements on Ca$^+$ and Sr$^+$ followed in the region of their p-d resonances.

These data encouraged a great deal of theory activity, including a rare excursion on my part into theoretical work with Vadim Ivanov to try and identify the structure seen in the whole of the calcium spectrum in the photon energy region from 28 – 43eV. A later more complex calculation (6) confirmed and enlarged upon our earlier theoretical analysis.

At Daresbury there was no instrumentation available to continue such measurements at higher photon energies. A collaboration was begun with the University of Aarhus, using the grazing incidence monochromator originally used at Daresbury on the old NINA accelerator. An extensive programme of measurements on atomic ions was carried out on ASTRID using the same merged beam technique; details are contained in two reviews (7,8). I highlight here the interesting case of Cr^+, where it eventually turned out that theory was right and experiment wrong. The Cr^+ spectrum had been measured using the dual laser plasma technique in a collaboration between NIST and Dublin City University in both theory and experiment (9). It appeared that there was a remarkable difference between the Cr and Cr^+ spectra in the region of the 3p-3d and 3p-4d resonances, which theory could not reproduce. It was later realised that the experiment was probably affected by the presence of metastable states (10), a worrying problem with all ion spectroscopy measurements, and one which cannot always be solved. It was some years later before the experiment could be repeated using the merged beam technique at ASTRID (11). This allowed better control of metastable contamination in the case of Cr^+, due to the type of ion source used, and agreement with theory was much improved.

5 The "complete" experiment and double photoionization of metal atoms

In the last experiments to be presented in this paper, the aim initially was to measure all the photoionization parameters in the one experiment. This had already been done a while ago by Ulrich Heinzmann and his colleagues by measuring the spin polarisation of the ejected electron and its angular distribution; a different approach was adopted in this work.

The atom chosen was calcium, photoexcited in the 3p-3d resonance (12). One decay channel for this resonance leaves the Ca^+ ion in an excited 4p state after ejecting an electron. The ion then decays to the ground state emitting a photon, and by measuring the polarisation of the photon in coincidence with the photoejected electron, and in a separate measurement measuring the angular distribution of the electron, it is possible to determine the ratio of the dipole amplitudes of the two ejected photoelectron channels, and the phase difference between them. Absolute values of the dipole amplitudes could not be calculated since the total photoionization cross section of calcium was not known.

This experiment evolved into measurements of double photoionization of metal atoms, where the two ejected electrons were detected in coincidence and as a function of the angle between them, a triply differential cross section (TDCS) measurement. Calcium and strontium were chosen; by this time we had become experienced in producing vapour beams of these metals, and had developed an oven in which the light beam entered along the axis of the oven by means of a capillary light guide to maximise the interaction between the photons and the metal vapour. The two 127° analysers had to be protected from the vapour beam by trapping the vapour on a liquid nitrogen cooled shield. At the Daresbury SRS the photon flux was not sufficient to work off resonance, so we worked in the regions of the 3p-3d and 4p-4d resonances. Data were taken at 31.41eV, the peak of the 3p-3d resonance in Ca, for unequal energy sharing of the two photoejected electrons (13). In this case one of the analysers was fixed at 0°, ie along the main E-vector component of the incoming radiation. With the rotatable analyser at ± 90°, a value of 0.6 was measured for the photon polarisation, confirming that LS-coupling was valid at this photon energy. There were obvious differences between the two energy sharing cases. Secondary peaks were evident in one set of data, and also noticeable was the non-zero value of the TDCS when the two electrons were ejected in

directly opposite directions, a breakdown of the symmetry rule which does not occur when the two electrons have equal energies.

Data were also taken for equal energy sharing of the two electrons. In this case the symmetry rule was observed, ie the TDCS was zero for the two electrons ejected at 180° to each other. The results were also different when the photon energy was changed to another position in the region of the 3p-3d resonance at a photon energy of 31.59eV where there is another peak in the photoionization spectrum. The peaks became broader although the symmetry rule was not broken. It is well known that in fact this resonance has many components, incompletely resolved in this experiment, and showed that the results obtained were definitely resonance dependent.

For this reason a further experiment was attempted at an off-resonance photon energy for calcium using the Trieste SR source ELETTRA, where an undulator beamline was available. Since the photoionization cross section was approximately 2000 times lower off-resonance, this was an extremely difficult experiment, even with the much improved intensity from the undulator beam-line. The fixed detector was placed as before aligned to the E-vector of the incoming beam, and the experiment was carried out for equal energy sharing of the two ejected electrons. The results (14) showed a marked difference from the corresponding case for helium (15), with the main peak occurring at a relative angle of 60° between the two analysers and being much narrower than the helium case

6 Acknowledgments

7 References

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Footnote

This is a short report; an expanded version, complete with diagrams and a full set of references, is available by contacting the author using the e-mail address given above.