Photoionisation of atoms and molecules using synchrotron radiation at Daresbury

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The Madden and Codling publications of the early 1960's demonstrated convincingly the relevance of sources of synchrotron radiation (SR) to the study of photoionisation phenomena in atoms and molecules [1, 2]. Within a relatively short period of time SR facilities sprang up in France, Germany, Italy, Japan and the U.K.

Ditchburn and Marr, at Reading, appreciated the potential of SR and in 1966 a grant of £73,750 was awarded to them by the Science Research Council (SRC) to develop an SR facility in the UK. The initial idea was to use the East Kilbride 100MeV linear accelerator, coupled with a superconducting magnet, but access to the experimental area would have been problematic and so interest turned to the Glasgow University 300MeV electron synchrotron. In October 1966, Codling took a standard lamp, calibrated by the National Physical Lab., to Glasgow to determine the beam current.

This led, in due course, to NPL becoming interested in SR as a standard of flux in the extreme ultraviolet (XUV). McWhirter (Culham Lab.) and his Glasgow colleagues had recently coupled a spectrograph to the synchrotron and seen radiation down to about 3nm. The Glasgow synchrotron thus became the first U.K. SR Facility, with users from Glasgow (Stewart), Culham (McWhirter), NPL (Key) and Reading.

Quite independently and unknown to the Reading Atomic and Molecular (ATMOL) group, Ian Munro and Scott Hamilton, at Manchester University Physics Dept., took measurements at the Daresbury Laboratory's (DL) 5 GeV synchrotron, NINA, in 1967. They showed that NINA was a potent source of XUV and x-radiation and by early 1969 had submitted a proposal to SRC to establish an SR Facility (the SRF) at Daresbury. The proposal was successful and ultimately the SRF consisted of 2 beam lines and 6 experimental stations, the SR operation being *parasiti c.*

In 1968 Peter Mitchell (Uni. N.S.W., Sydney) joined Reading and was the first physicist to couple SR with the technique of photoelectron spectroscopy (PES). Whilst building his 'retarding potential' photoelectron analyser, he helped to align and test a 'zero deviation', 3-metre grazing incidence monochromator, designed and built at Reading [3]. In 1970 the monochromator was coupled to the Glasgow synchrotron to provide the XUV continuum to allow measurements on electron angular distributions in the rare gases [4]. Mervyn Lynch (Perth) joined the group and used the set-up at Glasgow to make the first measurement of partial photoionisation cross sections, in this case of the 3s electron in argon [5].

Reading Experiments at the SRF

The Reading group was ready to transfer their experiments to the SRF in the early 1970's, but before any experiments could be contemplated, it was necessary to choose the most appropriate normal and grazing incidence monochromators to be attached to the beam lines. One of the big problems in working with high energy synchrotrons such as NINA is that a substantial amount of 'second order', *unwanted* radiation is reflected from a diffraction grating when it is situated in grazing incidence geometry. John West, who took up a PDRA position at Reading in 1972, oversaw the construction of a monochromator to minimise this second order contribution [6]. John subsequently moved to Daresbury and was highly involved in the design, testing, and setting up on line, of many of the XUV monochromators used at the SRF.

The mid-to-late 1970's was a golden period for Reading at the SRF. The improvement in flux, compared to that of Glasgow, could be seen in the data taken by Houlgate et al [7] on the angular distribution and partial cross sections for the 3s and 3p electrons in argon, see **figure 1**. Pam Woodruff measured the 4d, 5s and 5p partial cross sections in xenon and showed the importance of inter-shell correlation effects [8]. John Hamley measured the photoionisation cross section of sodium in the region above the 2p threshold using a heat pipe, the first and only time metal vapour
absolute
cross sections have been obtain above 25eV, the ionisation limit of helium (the buffer gas used to contain the metal vapour) [9]. Steve Shannon measured the partial photoionisation cross sections of cadmium and mercury. This was the first time that an SR source had been coupled with PES to study metal vapours [10].

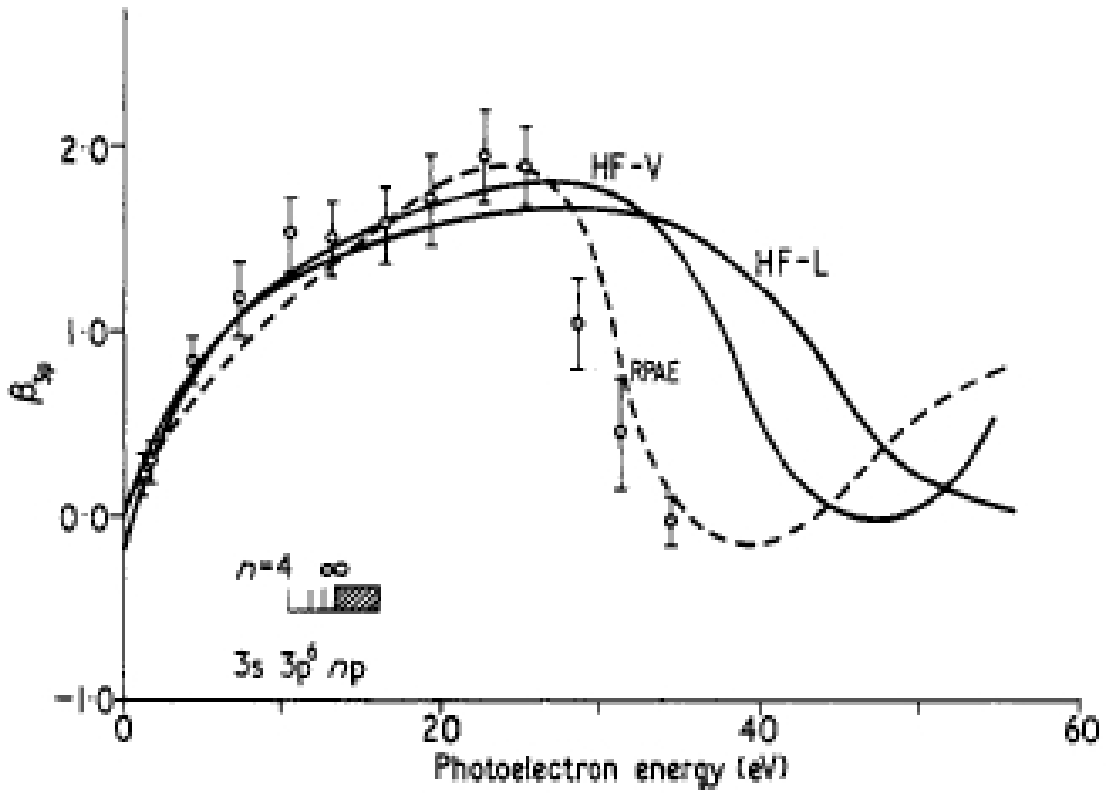


Figure 1 The asymmetry parameter β for the 3p electron of argon; see ref [7] for details.

John Morton and Robin Holmes, with the help of Don McCoy (Adelaide), measured angular distributions of N_2 and CO as a function of photon energy [11] and David Holland determined accurate absolute 'partial' cross sections for multiple ionisation of the rare gases from threshold to 280eV [12], for example the Ar $^{2+}$ partial cross sections, see **figure 2**. John West was co-author on most of the above publications, reflecting his efforts in setting up the SRF monochromators, keeping them in working order and helping during the data-taking process. Somehow he also found time to take his own data. For example, he made the most accurate measurement to date of the rare gas photoabsorption cross sections in the XUV [13].

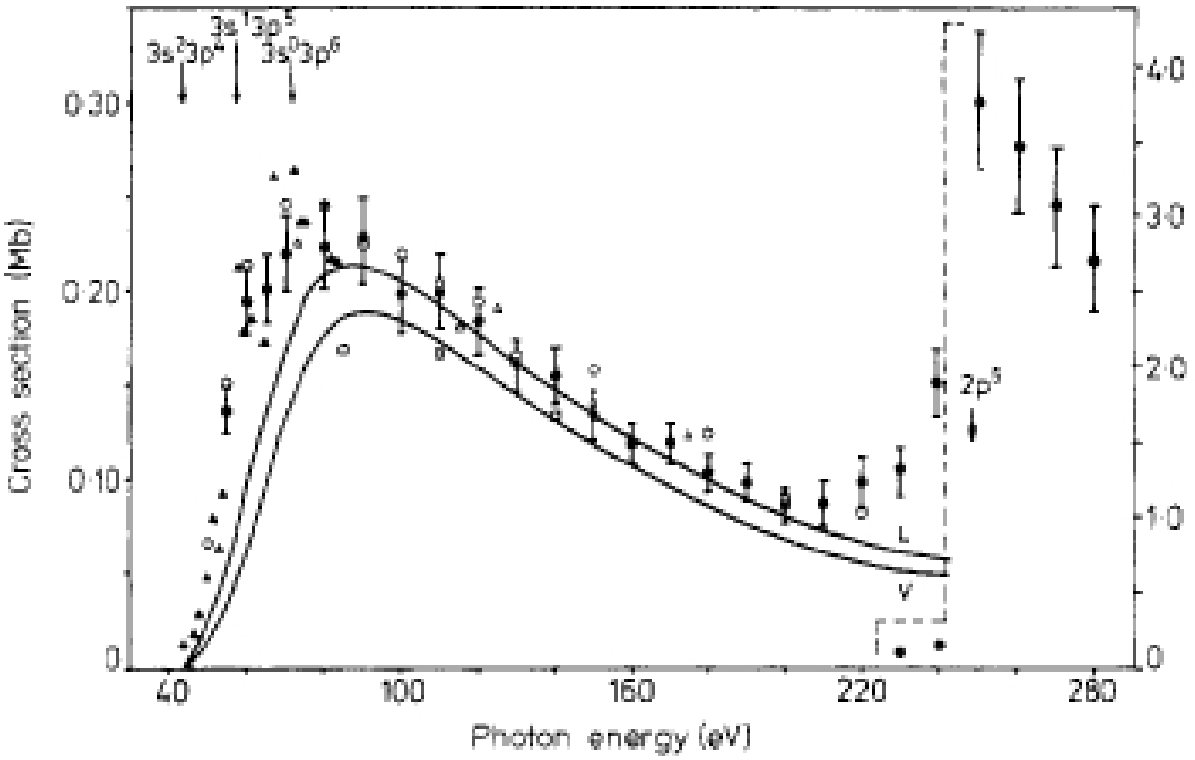


Figure 2 The Ar^{2+} absolute partial cross section. Theoretical curves: L (dipole length) and V (dipole velocity). See ref [12] for details.

Reading Experiments at the SRS

In 1972 the SRC decided that NINA would close in 1977. After a meeting in Reading in 1974 to discuss an SRC panel report: ‘A plan for future research with SR based on a dedicated storage ring’ [14], it was decided that a new accelerator would be built, the world’s first machine *dedicated* to SR research. The Synchrotron Radiation Source (SRS), a 2GeV, 1amp storage ring, was opened in 1980. Unfortunately, Reading did not gain access to the SRS until 1983. Geoff Marr therefore negotiated the use of the Bonn synchrotron for those students who still required data for their PhDs. Reading provided the optics for two new beam lines and Ralph Chamberlain and Geoff Abburrow machined the required stainless steel components. They also made a number of trips to Bonn, taking with them the experiments of Robin Holmes, David Taylor and David Holland.

In 1981 Geoff Marr was appointed to the Chair of Natural Philosophy at Aberdeen. Fortunately for Reading, he was replaced by Leszek Frasinski, who joined the ATMOL group in 1983. There followed another golden period for the group. Leszek and Kevin Randall used a threshold photoelectron-photoion coincidence (TPEPICO) apparatus to study the predissociation of the $c^4\Sigma_u^-$ state of O_2^+ [15]. They coupled a high aperture threshold electron analyser with the ‘single bunch’ mode of operation of the storage ring (0.2ns pulses every 320ns), to allow time-of-flight (TOF) techniques to be employed for the electrons. It was possible, therefore, to obtain an ion energy spectrum in coincidence with ‘threshold’ electrons, with energies less than 50meV.

In 1986 Frasinski introduced two completely new triple coincidence experiments to the field of molecular photoionisation [16]. The results of both were presented in the form of three-dimensional triple coincidence maps. In the first, a photoelectron-photoion-photoion coincidence (PEPIPICO) experiment, the TOF of two ions from the double ionisation of SF_6 (for example SF^+ and F^+) were displayed, see **figure**

3.

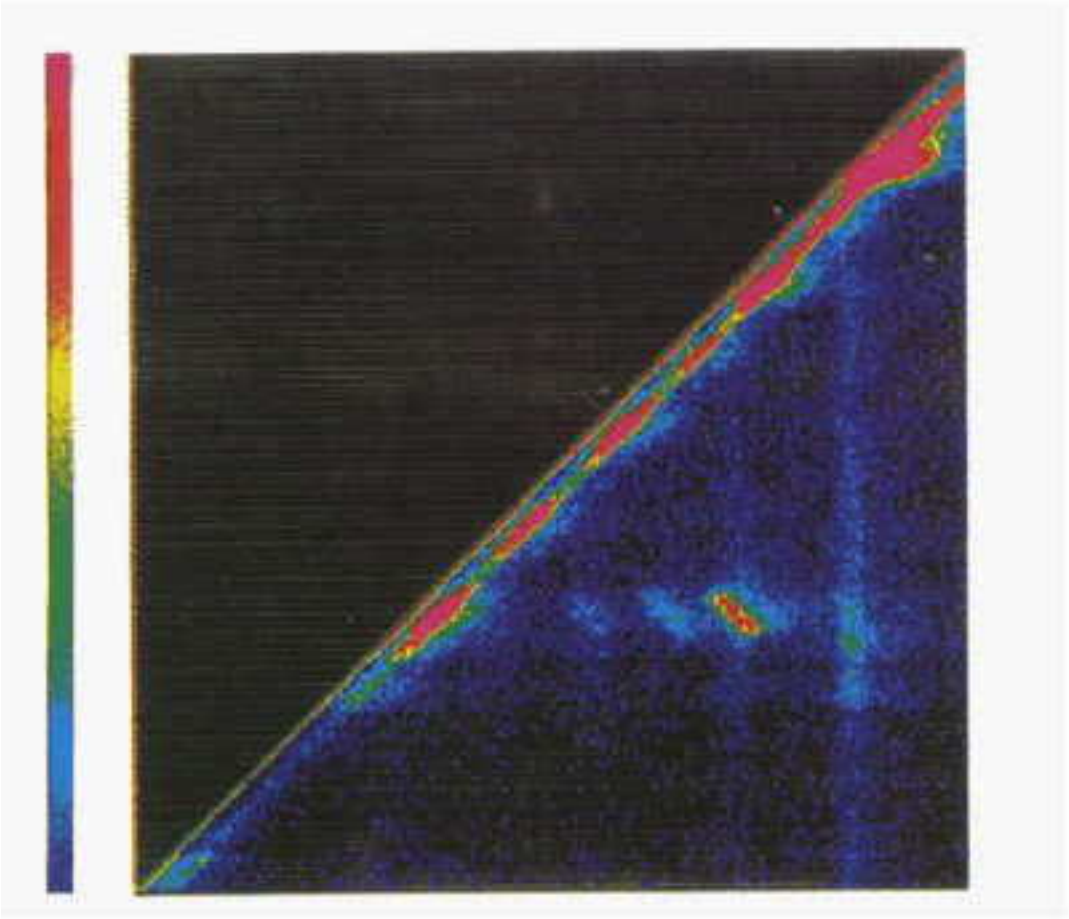


Figure 3 Ion-ion coincidences in the double ionisation of SF_6 at 220 Å (56.4 ev). On the intensity scale, dark blue denotes 1 count, magenta ≥ 30 counts. On the right hand side of the figure, the coincidence features are:

1

, $SF_5^+ + F^+$;

2

, SF_3

+

+ F

+

;

3

, SF

2

+

+ F

+

;

4

, SF

+

+ F

+

.

In the second, using the single-bunch operation of the SRS, the photoelectron and O^+ ion TOFs from the predissociation of O

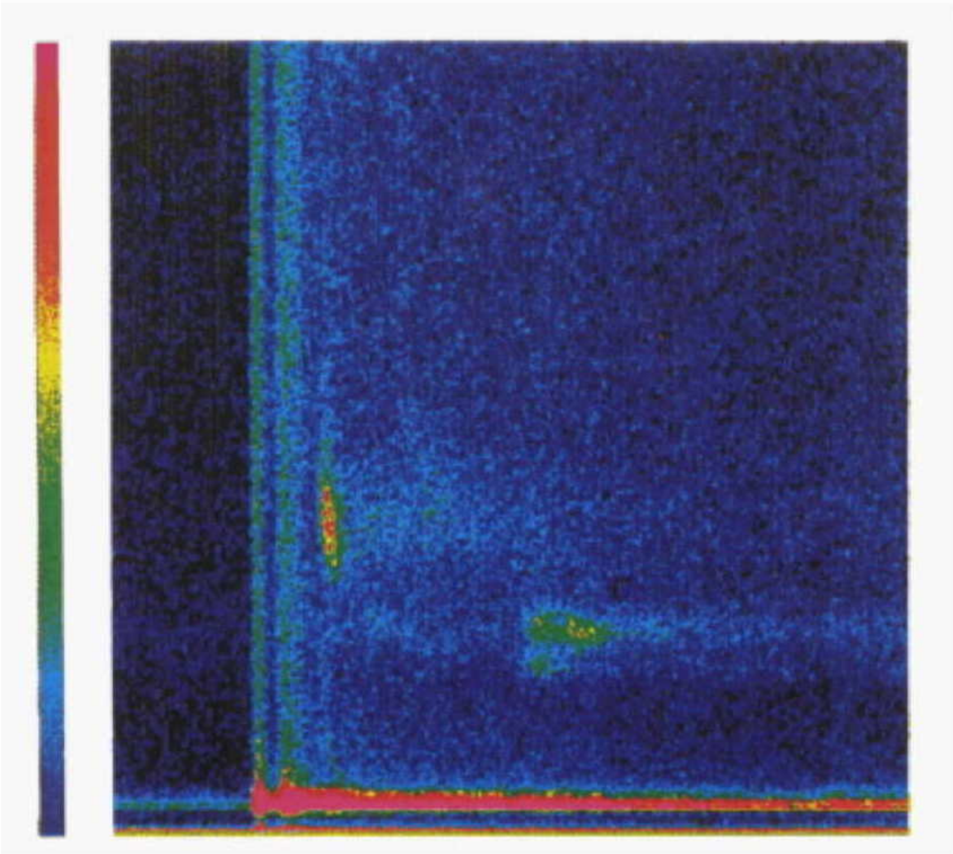
2

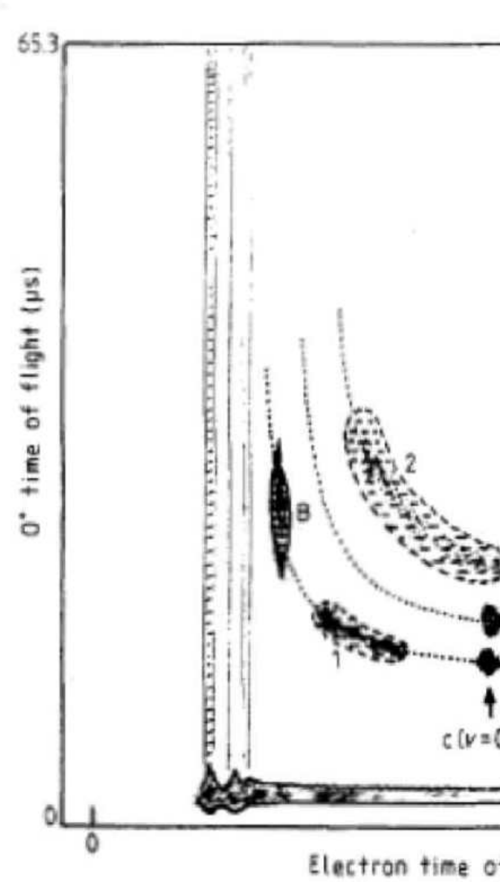
+

were measured in coincidence. This experiment provided a visual insight into the details of dissociative photoionisation that had not been available previously. For example, groups of electrons from two different continua could be distinguished, see

figure

4.




65.3
O+ time of flight (μs)
B
1
2
0
0
Electron time o