A versatile gas phase coincidence spectrometer for use at the CLS.

A. Padmanabhan (1), P. Thorn (1), T. Reddish (1), C. Ryan (2), L. Zuin (2), M. MacDonald (2)

Principal Contact:

Principal Investigator	T. Reddish University of Windsor reddish@uwindsor.ca (519) 253-3000 Ext 2655

(1) Department of Physics, University of Windsor
(2) Canadian Light Source, Inc.

Introduction

Electron-electron correlation is intrinsic in the formation of chemical bonds and leads to observable effects in atomic and molecular physics. Without an irreducible core of electron correlation every atomic process would be governed by effective one-particle physics. Photodouble ionization (the direct ejection of two or more electrons from the absorption of a single photon) is a direct consequence of these correlation effects.

An instrument to probe these effects through angle-resolved photoelectron coincidence spectroscopy has previously been built [1-3] and is being commissioned on the PGM beamline of the CLS.

Experimental

The spectrometer consists of two toroidal, electrostatic charged particle analyzers, with their associated entrance and exit lenses, detectors, a gas jet and differential pumping - all housed in a stainless steel chamber. The analyzers collect charged particles from the interaction region (overlap of the gas jet and SR beam) emitted orthogonal to the SR beam, as shown in Figure 1.

The analyzers are independent from each other, *i.e.* they are able to detect dissimilar electron energies, with different resolutions or even one - ions and the other - electrons. The advantage of using a toroidal geometry for the analyzer is that allows the emission angle of the charged particles and its energy to be collected simultaneously. The analyzers can be azimuthally rotated about the SR beam, which is 100% linearly polarized.

The instrument is expected to be used in three experimental configurations – (a) angle resolved electron - electron coincidence, (b) angle resolved electron - photoion coincidence, (c) angle integrated threshold electron - photoion coincidence. During this reporting period commissioning began on the first two of these.

Photodouble ionization of helium

Direct photoionization of helium produces two electrons which can share the excess energy between them. The double photoionization threshold is 79 eV, so excitation at 129 eV produces a total excess energy of 50 eV. One analyzer was set to collect a low energy (7.5 eV) electron ejected approximately in line with the electric vector ($\boldsymbol{\varepsilon}$) of the linearly polarized SR beam. The other analyzer collects the high (42.5 eV) energy electron. When two electrons are detected in coincidence, their emission angles are logged.

After post-processing, the data is displayed as figure 2. Each frame shows the angular distribution of the fast electron, θ_2, in coincidence with a slow electron emitted at the θ_1 angle given in the insert ($\Delta\theta_1 = \pm 5°$). The theoretical triple differential cross sections (TDCSs) are the results of Time Dependent Close Coupling (TDCC) [4] calculations from [5]; the observed shapes of the TDCSs are in agreement with theory, and with published results under similar conditions [6]

Photoionization of aligned CO

Single photoionization of CO, may produce CO^+ in its ground state, an excited state, or a dissociative state. In the latter case, as long as the dissociation is rapid, collection of the ion fragment will give the orientation of the CO molecule prior to the ionization event. Here the absorption of a 51.5 eV photon produces CO^+ ($4^2\Sigma^+$) with a photoelectron of 19.5 eV and a photoion (C^+) fragment of ~3.2 eV. One detector is set to collect the photoion and one the photoelectron. When they both register in coincidence the two collection angles are recorded.

Figure 3 shows the photoelectron angular distribution (between 140° and 280°) for the photoelectron producing CO^+ ($4^2\Sigma^+$) for all CO aligned between ±25° with respect to $\boldsymbol{\varepsilon}$. The data set can be processed to better define the CO alignment and further data would complete the angular distribution. Although more work is required, the present data are quite consistent with published results [7], including the observation of the small side lobe.

Discussion

Photoion-photoelectron coincidence studies have only been possible in the last decade, due to the availability of undulator radiation and sophisticated charged particle detection techniques; most studies have been in the area of core ionization. This is the first time the toroidal spectrometer has been used for such experiments and, in conjunction with the PGM beamline, we have an excellent opportunity for state-of-the-art inner valence ionization studies in small molecules.

This report covers the first step in a process to make the instrument described available to the user community. It is hoped to extend the commissioning to threshold photoelectron spectroscopy in coincidence with the photoion in 2010. This instrument will greatly extend the range of gas phase molecular science available. CLS

especially once the variable polarization undulator is available. Software and hardware upgrades are also required.

Conclusion

We have started commissioning a versatile coincidence spectrometer for use at the CLS. It can be used in three different modes for double photoionization, photoelectron angular distributions and threshold photoelectrons.

References

1. Reddish, T.J., Richmond, G., Bagley, G.W., Wightman, J.P. and Cvejanovic, S. 1997. Dual toroidal photo-electron spectrometer for investigating photodouble-ionization in atoms and molecules. Review of Scientific Instruments 68(7), pp. 2685-2692.
2. Wightman, J.P., Cvejanovic, S., and Reddish, T.J. 1998. A procedure for measuring photoelectron angular distributions from gas-phase targets using an angle- and energy-dispersive spectrometer. Journal of Electron Spectroscopy and Related Phenomena 95(2-3), pp 203-209.
3. Slattery, A.E., Wightman, J.P., MacDonald, M.A., Cvejanovic, S. and Reddish, T.J. 2000. Threshold photoelectron studies of Kr and Xe. Journal of Physics B: Atomic, Molecular and Optical Physics 33, pp 4833-4848.
4. Colgan, J., Pindola M.S., and Robicheaux, F., 2007 Physical Review Letters, 98, pp153001-4
5. Colgan, J. Private Communication.
6. Cvejanovic, S., Wightman, J.P., Reddish T.J., Maulbetsch, F., MacDonald, M.A., Kheifets, A.S. and Brayk, I. 2000. Photodouble ionization of helium at an excess energy of 40 eV. Journal of Physics B: Atomic, Molecular and Optical Physics 33, pp 265-283.
7. Hikosaka, Y. and Eland, J.H.D. 2000. Molecular frame photoelectron angular distributions in inner valence photoionisation of CO. Physical Chemistry Chemical Physics 2, pp 4663-4668.

Acknowledgements

T.J. Reddish acknowledges support from NSERC and the University of Windsor.

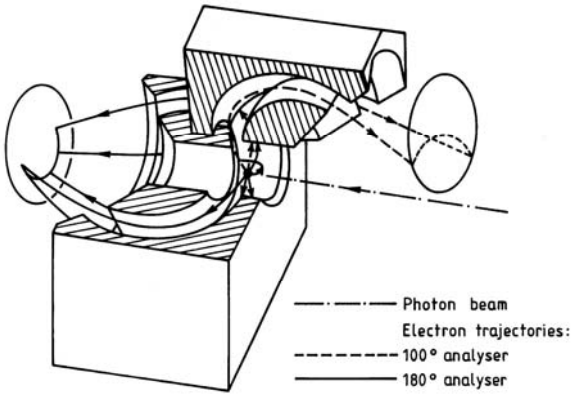

Figure 1: [Reddish_1.jpg] A schematic diagram showing the configuration of the two partial toroids along with lines indicating central trajectories of electrons with as election of emission angles, as discussed in the text. The electron lenses are not shown for reasons of clarity. The mechanical angular acceptances of the two analyzers in the plane orthogonal to the photon beam are 100° and 180°.

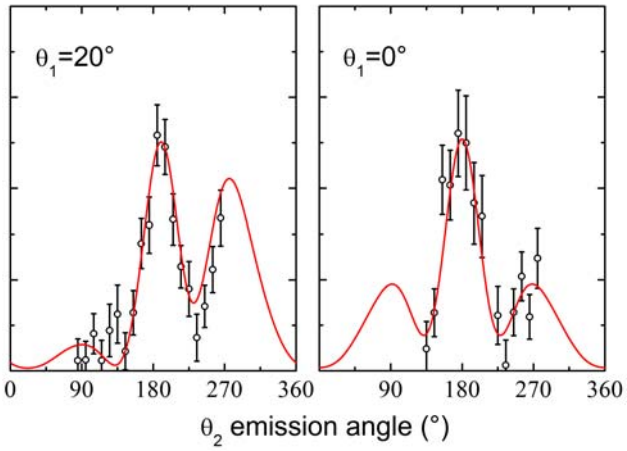

Figure 2: [Reddish_2.jpg] Photoelectron distributions for He double photo-ionization at hv = 129 eV (50 eV excess energy). The inset in each graph shows the collection angle of the 7.5 eV electron, while the graph shows angular distribution of the 42.5 eV electron collected in coincidence. The red curve is from TDCC theory [4,5] and is arbitrarily normalized to the data.

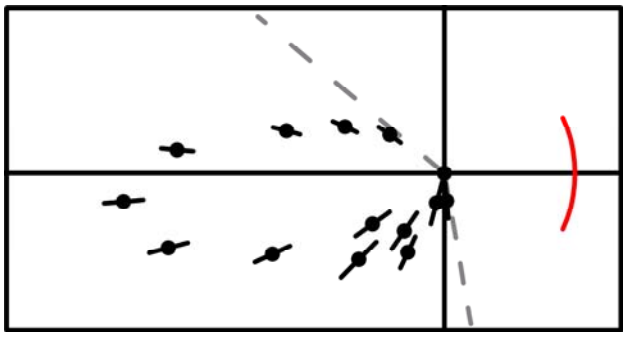

Figure 3: [Reddish_3.jpg] Photoelectron distribution (between the dashed lines) for CO^+ single photoionization at hv = 51.5 eV (19.5 eV electron energy) in coincidence with the C^+ fragment emitted in the range shown by the red line. (The horizontal line corresponds to ε direction). There is a high degree of correlation between the Σ component of ionization and photoelectron's direction.