Ionization of Aligned Targets by Low Energy Electron Impact

The aims of this project were:

- *To implement new experiments to study ionization of spatially aligned atoms and molecules.*
- *To deliver accurate experimental data for evaluation of the most sophisticated scattering models being developed by theoretical collaborators.*
- *To study ionization from aligned atoms for the first time, thereby providing a bridge between atomic and molecular collision theories.*
- *To produce data for the simplest molecule (H_2) to determine key quantum mechanical processes for ionization by low energy electrons.*
- *To measure ionization of polyatomic molecules of relevance to environmental, medical and technological applications so as to aid in the development of new ionization theories for these complex systems.*

Introduction

The dynamics of electron interactions with atoms and molecules is a rich field for experimental and theoretical exploration. These studies are of both fundamental and practical significance. From a fundamental viewpoint, sophisticated quantum theories must be rigorously tested by experiments that provide high precision data. In this way, models can be extended and refined for application in many areas, with confidence that the results are accurate. From a practical viewpoint, low energy electron impact ionization of atomic and molecular targets plays a key role in many areas, since this energy regime is where the ionization probability is highest. These areas include ionization in the Earth's upper atmosphere, stellar and planetary physics, lighting and laser technologies, as well as studies of secondary ionization in cancer therapy to name a few. *By developing rigorously tested theories that define these interactions, it becomes possible to predict how they occur*. The most detailed description of ionization is provided by the (e,2e) technique. This uses an incident electron of known momentum to collide with and eject an electron from the target. The scattering geometry for an (e,2e) event is shown in Figure 1. The two resulting out-going electrons, *i.e.*, the incident electron scattered in the collision, and the ionized target electron that is ejected, are detected in time coincidence. This ensures the measured events are from a single collision. The momenta of the out-going electrons are measured, so that the collision kinematics are fully determined. *The (e,2e) technique therefore provides the most comprehensive data against which different models can be evaluated.*

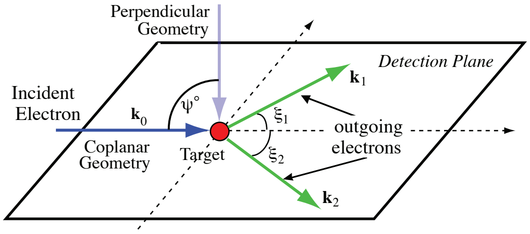

Figure 1: Schematic showing the scattering variables in an (e,2e) collision. The incident electron of momentum $\mathbf{k}_0$ impinges on a target and two out-going electrons (momenta $\mathbf{k}_1$ and $\mathbf{k}_2$) are measured over a range of angles ($25° < \xi < 135°$) in the detection plane. The figure also shows the geometries accessible in Manchester, from coplanar where all three electrons are in the same plane ($\psi = 0°$, solid blue arrow) to the perpendicular geometry ($\psi = 90°$, light blue arrow).

Experimental Methodology

In the research conducted here, three new experiments were undertaken, as described below.

1. (e,2e) from Aligned Atoms

In these experiments ionization from *laser-excited* magnesium atoms was measured, to reveal the three-dimensional character of the scattering dynamics [1]. In these experiments the symmetry of the collision is *reduced* by controlling the direction of the target electron charge-cloud prior to ionization. Magnesium was excited to the 3P state using a laser of high spectral purity and coherence. By controlling the laser beam parameters, the excited target charge cloud was spatially aligned with respect to the scattering geometry. An example of this process is shown in figure 2, which depicts the angular part of the excited charge-cloud under the influence of linearly polarized light. In this example, the target electron is aligned either parallel or perpendicular to the direction of the incident electron, by varying the polarization vector (ε) of the laser beam. The figure shows how the anisotropy of the collision can be varied. The measured changes to the ionization cross-section are shown in figure 3. These pioneering experiments have prompted development of new theories that are considering this alignment with respect to the collision geometry. *These experiments hence 'bridge' between atomic and molecular collision theory, since both must include alignment of the target.*

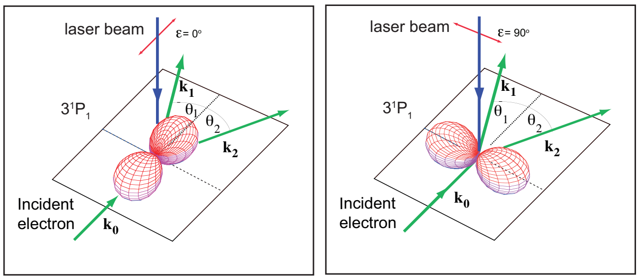

Figure 2: Alignment of the electron charge cloud within an excited target atom using incident laser radiation on resonance with the atomic transition.

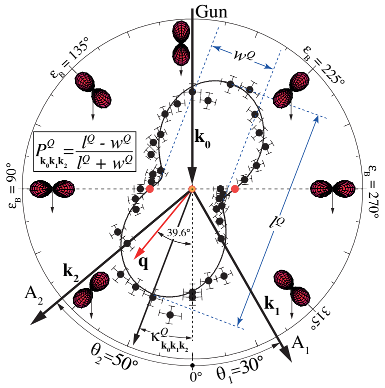

Figure 3: Measurements showing variation of the ionisaiton with alignment of the charge cloud.

2. (e,2e) from H_2 molecules

TDCC calculations [2] predict that electron impact ionization of H_2 should be sensitive to the molecular alignment, particularly in the perpendicular plane. One of the aims of this work was hence to attempt to study ionization of aligned H_2 to test these predictions. The experiments attempted to measure the molecular alignment post-collision, by detecting the direction of the ion resulting from fragmentation of the H_2^+ ion. This requires a *triple* coincidence measurement between the two out-going electrons and the ion fragment. A precursor to these very difficult experiments was carried out where H^+ ions from fragmentation of the ground H_2^+ ion were detected. The fragments are only a small percentage (~1.5%) of the total ion yield, and so were isolated from the thermal H_2^+ ions by energy selection. Measurements of the angular distribution of these ions were obtained at several electron scattering angles in a *double* coincidence experiment. The coincidence yield from these measurements was found to be extremely low, and so a decision was taken not to progress with the more complex *triple* coincidence measurements, since data could not have been obtained with sufficient statistical accuracy.

3. (e,2e) Ionization from Polyatomic Molecules

It is important to measure ionization from polyatomic targets, since these are ubiquitous in nature. As noted above, it is not possible at present to measure ionization from *aligned* molecules, and it is also not possible to execute precise calculations of these collisions at the present time. To progress the field, the third goal of this research was hence to obtain data from polyatomic targets that were randomly oriented in space. Collision theories then have to integrate their calculations over all directions to compare to the data. Only one group (Don Madison, USA) has attempted to calculate ionization of polyatomic targets, using a variation on their Distorted Wave Born theory (M3DW). Their model spherically averages the molecular wavefunction prior to the collision, to attempt to eliminate the difficulties presented by alignment of the molecules. This technique has however proven to have very mixed success. One of the key aims of the work in Manchester was hence to provide guidance to theory. Further, it is expected that the experimental data will stimulate other groups to consider these complex reactions.

Measurements of the *highly symmetric* molecule SF_6 have hence been made, to add to the growing data from polyatomic systems. SF_6 allows the accuracy of the spherical averaging method to be assessed, since the ionization cross-section should be far less sensitive to alignment from this target. New M3DW calculations are currently been undertaken by colleagues in the USA to study these processes.

A further study of the ionization of C_2H_6, a molecule with two *heavy atoms*, has also been conducted to assess the importance of the nuclear frame over the orbital symmetry. These measurements complement results from a series of iso-electronic molecules (H_2O, NH_3 and CH_4) previously studied in Manchester, where each molecule consisted of *one* heavy atom bound to H atoms. In the data obtained from these targets, the molecular frame appears to contribute little to the cross section. By contrast, since C_2H_6 has *two* heavy atoms, the effect of the nuclear charge distribution over two centres is being studied. These data are also now being considered by theoretical colleagues in the USA.

Summary

The experiments carried out during this fellowship have allowed the fundamental nature of electron ionizing collisions to be characterized. All were carried out at low energies, where the ionization probability is highest. The new data from aligned magnesium is a pioneering study that can be extended to other kinematics and geometries, as well as to other targets. Indeed, significant interest from theorists has already been expressed in this area. All experiments carried out in this research programme have strengthened our collaborations with leading theorists throughout the world, and have invoked the development of new models for the ionization of aligned targets. This heralds a new type of study of electron impact ionization for the future, and is expected to lead to further understanding of these important collision processes.

References

1. KL Nixon and AJ Murray (2013) *PRL*, accepted
2. J Colgan, MS Pindzola, F Robicheaux, C Kaiser, AJ Murray and DH Madison (2008) *PRL* **101** 233201