Electron impact ionization and excitation studies of laser prepared atomic targets

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Abstract. A review of current work at Manchester is given, with emphasis on low energy electron impact ionization & excitation experiments from laser-prepared targets. Three different methods are considered: super-elastic scattering from targets prepared by laser radiation in an optical enhancement cavity, (e,2e) studies of laser-aligned atoms, and electron impact ionization studies from laser cooled atoms in a new type of atom trap – the AC-MOT. The status of each experimental programme is detailed, and new data presented. Future directions and techniques are then discussed.

1. Introduction
The study of collisions between electrons, atoms and molecules has a long history. Two types of collision may occur. In elastic scattering the energy of the colliding particles remains unchanged, however their momenta may alter. Inelastic collisions occur when both energy and momentum of the particles change. In this case, the target may be excited, fragmentation may occur, the target may be ionized or a combination of these processes may result. Unlike the incident electron, the target has structure, and so different types of interaction may occur. These range from Coulombic interactions between the incident electron, target electrons and nucleus, through exchange processes, polarization of the target and post collisional interactions between scattered and ejected particles. In the low to intermediate energy regime studied in Manchester all processes are important, and so theoretical models in this region are complex and challenging. Nevertheless, close coupling models and distorted wave theories are proving successful for a number of different targets.

In the experiments described here, inelastic collisions leading to excitation and ionization are considered for atomic targets. Excitation is studied using super-elastic scattering, which can be considered as the ‘time-reversal’ of electron photon coincidence experiments. Ionization is described for two different targets: those pre-aligned using coherent laser excitation, and those cooled and trapped using laser and magnetic fields. In the former case, Mg is excited to the 3P1 state prior to (e,2e) measurements in a coplanar geometry. In the latter, potassium is cooled and trapped in a new type of atom trap (the AC-MOT) before studying ionization as a function of electron energy. These combined laser and electron experiments are difficult, as they require simultaneous and precise control of both the electron spectrometer and laser system. Such experiments have become feasible with the advent of newer lasers whose wavelength can be fixed for long periods using electronic feedback.

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A new direction for these laser-based experiments is being developed at Manchester, using optical enhancement and optical storage cavities to control the laser beam at the interaction region. This new method will greatly expand the number of targets that can be studied, and will allow the efficiency of pulsed laser experiments to be significantly increased. These new techniques are described below.

2. Super-elastic scattering studies.

Electron impact excitation studies of atoms first used coincidence techniques, as pioneered by the Stirling group [1]. This was followed soon after with the development of the super-elastic technique by Hertel and co-workers [2]. Present studies almost exclusively adopt super-elastic methods, as they provide data of much greater precision, and the atomic collision parameters (ACP’s) describing the ‘shape’ of the target can be determined over a much wider range of angles. Indeed, new techniques recently developed in Manchester adopted a Magnetic Angle Changer inside the spectrometer that allowed the ACP’s \((P_{\text{lin}}, \gamma, L_{\perp})\) to be determined over all angles for the first time [3,4].

Although very powerful, the super-elastic technique is restricted by the range of laser wavelengths and intensities available using present day continuous wave (CW) lasers. The experiments are almost exclusively limited to alkali and alkali-earth targets (although Yb has also been studied [5]), with excitation being from S to P-states. It is clearly advantageous to expand this range of targets, and to consider collisions to states of higher angular momentum so as to further test quantum theories.

Figure 1 shows the experimental setup. A laser beam is directed into the interaction region to promote atoms to an excited state. An incident electron from the gun may then de-excite a target, and so will emerge with higher energy (super-elastic scattering). The scattering probability through an angle \(\theta\) then depends upon the ‘shape’ of the laser-excited charge cloud prior to the collision. By changing this shape, the scattering probability is determined. The de-excitation probability is directly related to the excitation probability through time-reversal, and by carefully considering the effects of the laser on the atom. The derived excitation probability is then related to the atomic collision parameters [2-5].

In practice, measurements are obtained by varying the polarization of the incident laser radiation. To determine the alignment parameters \((P_{\text{lin}}, \gamma)\), a linearly polarized laser beam is used, the direction of the polarization vector being rotated around the scattering plane. Early experiments measured the time-inverse Stokes parameters to determine \(P_{\text{lin}} \& \gamma\), however this is less accurate than modern methods that fit the ACP’s directly to the data. \(L_{\perp}\) is determined using circularly polarized laser radiation. Full details of these methods can be found in the references at the end of this paper.

Figure 2 shows an example of results recently obtained from a calcium target, with the incident electron energy being \(\sim 10\) eV [6]. The results are compared to different theories [7-10]. The energy of the experiment has been corrected by \(\sim 2\) eV to allow for effects of surface charging in the interaction region, as noted in [6]. The CCC theory is in closest agreement with the data at this energy, although the R-matrix with B-splines calculation (BSR) also agrees well. The distorted wave model (which is accurate at higher energies) does not reproduce the data in this low energy regime.
Figure 2. Comparison of experimental data to theory for an incident electron energy ~10eV. Calculations include a convergent close coupling theory (CCC) [7], R-matrix calculations (RM [8], BSR [9]) and relativistic distorted wave calculation (RDW) [10].

2.1. Introduction of an optical enhancement cavity to super-elastic measurements.

The super-elastic technique produces accurate data over a wide range of energies and angles, and so provides the most stringent test of current theories, as shown above. However, the method requires laser radiation of high spectral purity and high intensity (typically greater than 100mW/mm²) to efficiently excite the atoms prior to the collision. This radiation is produced by tuneable CW lasers, which operate in the visible and near infra-red regions of the EM spectrum. Frequency doubling methods have recently become available to increases the wavelength range of CW lasers down to ~215nm, however the power available from these systems is low (typically <10mW at 215nm).

![Figure 3](image)
Figure 3. The optical enhancement cavity surrounding the interaction region.

To overcome these restrictions a new experiment is under development which uses an optical enhancement cavity surrounding the interaction volume, as shown in figure 3. An enhancement cavity is basically a photon ‘storage’ device that rapidly builds up radiation between the mirrors until a steady state is reached. This steady state is governed by losses inside the cavity, and the reflectivity of the mirrors. For a simple cavity, the maximum intensity occurs inside the cavity when the transmission of the input coupler $T_I$ matches losses $L$ inside the cavity (assuming all other mirrors have ~100% reflectivity). For scattering experiments inside the cavity, these losses arise due to the atoms spontaneously emitting light from the cavity. Calculations show ~2% of the laser light is absorbed and spontaneously re-emitted by a well collimated atomic beam [11], and so by choosing an input coupler with a reflection ~98%, maximum power is delivered into the cavity. Under these conditions the power enhancement $G_p$ inside the cavity is given by $G_p = L^{-1} = 50$. For a 10mW incident laser beam, this equates to ~500mW inside the cavity. Super-elastic experiments then become possible at UV wavelengths for the first time.
The enhancement cavity has now been installed inside the spectrometer to carry out these new experiments. The mirror spacing is controlled using high-speed electronics to ensure the cavity remains on-resonance with the laser beam, and the laser wavelength is set by monitoring fluorescence from the atomic beam. Measurements are underway from calcium to check the performance of the cavity against existing data, after which experiments will be performed in the UV for the first time. New collision data will then be obtained from targets including Cu, Ag, Mg and Zn.

3. \((e,2e)\) studies from laser excited and aligned atoms

The most accurate studies of electron impact ionization are carried out using the \((e,2e)\) coincidence technique. In these experiments an incident electron of momentum \(k_0\) scatters from the target, producing ionization. Two electrons emerge, and are detected in coincidence so as to determine their momenta \(k_1, k_2\). Since the only restrictions on the scattering process are conservation of energy and momentum, the electrons may emerge at any angle with respect to the incident beam direction. The \((e,2e)\) technique determines the probability that the two outgoing electrons are correlated, and relates this to a three fold differential cross section \(TDCS(k_0, k_1, k_2)\).

Since electrons may emerge at any angle, it is necessary to use a configuration that allows different kinematical conditions to be studied. The most common geometry adopted is coplanar, where the incident, scattered and ejected electrons all occupy a plane. This geometry usually produces the highest ionization probability, although not always [12]. For studies of non-coplanar interactions, it is necessary to design a more complex apparatus. To study these interactions the \((e,2e)\) spectrometer in Manchester adopts a configuration where the electron detectors define a ‘detection plane’, while the electron gun moves in and out of this plane, as shown in figure 4. Studies can then be conducted from coplanar geometry (\(\psi = 0^\circ\)) to the perpendicular plane (\(\psi = 90^\circ\)). A common point exists for all \(\psi\) when \(\xi_1 = \xi_2 = 90^\circ\), allowing the measured TDCS to be inter-related over all angles. A large body of data has now been obtained for a wide range of targets, providing a robust test of current theory.

Whereas theory and experiment compare well for lighter atoms, the comparison is often poor for molecules, particularly at low to intermediate energies. A key difficulty lies in the problem of making a true comparison: at present it is impossible to take measurements from \textit{pre-aligned} molecules (although some data exists for targets that fragment after ionization, revealing the alignment direction), and so theory must integrate the calculation over all possible orientations of the target. This is computationally very intensive, and so approximations are made for all but the simplest target (\(H_2\)). This may explain why comparisons between experiment and theory are poor at present. There is clearly a need to develop experiments that provide accurate results for aligned targets. As a step towards this goal, measurements were recently carried out from excited Mg atoms that were pre-aligned using a laser field. In this case it is the \textit{electron charge density} that is aligned with the laser beam, the nucleus of the target remaining at rest with respect to the scattering geometry. Such experiments can therefore be thought of as a pre-cursor to more challenging experiments in the future, where the distributed nuclei of a molecular target will be pre-aligned prior to ionization.

![Figure 4. The Manchester \((e,2e)\) geometry, where the incident electron makes an angle \(\psi\) with respect to the detection geometry. The outgoing electrons emerge at angles \(\xi_1, \xi_2\) in the detection plane. A common point exists when \(\xi_1 = \xi_2 = 90^\circ\).](image-url)
In these atomic experiments the laser accurately controls the alignment direction of the Mg electron density, in the same way as for the super-elastic studies described above. The laser excites the Mg atoms from the $3^1S_0$ ground state to the $3^1P_1$ state using radiation at ~285nm. Ionization then occurs from the laser-excited atoms, the electrons emerging with the required energy as selected by the analysers. The detected electrons are then time-correlated to produce a four-fold differential cross section $Q_{DCS}(\mathbf{k}_0, \mathbf{k}_A, \mathbf{k}_1, \mathbf{k}_2)$ that also depends upon the alignment direction of the target $\mathbf{k}_A$.

The first experiments to make these difficult measurements were recently carried out in a coplanar asymmetric geometry, with outgoing electrons selected to have 20eV energy. In this case $\psi = 0^\circ$, $\xi_1 = 30^\circ$ and $\xi_2$ was varied from $\xi_2 = 35^\circ$ to 120°. Measurements were taken from the $3^1S_0$ ground state of Mg and from the laser-aligned $3^1P_1$ excited state, with alignment angles $\varepsilon = 0^\circ$ ($\mathbf{k}_A$ in the detection plane), $\varepsilon = 45^\circ$ and $\varepsilon = 90^\circ$ ($\mathbf{k}_A$ perpendicular to the detection plane). The data were inter-related through the relative density of excited targets compared to atoms in the ground state [12].

![Diagram of experimental setup](image)

**Figure 5.** QDCS measurements from laser-aligned Mg atoms compared to the TDCS from Mg in the ground state. The results are on a common scale, and are normalised to unity at the peak of the TDCS.

Figure 5 shows the results from these experiments. The QDCS peak for $\varepsilon = 0^\circ$ is seen to be larger than the TDCS, although the overall shape of the cross section is similar. As the alignment angle emerges out of the plane, the QDCS steadily decreases, until at $\varepsilon = 90^\circ$ the peak is ~50% of that for the TDCS. In this configuration the QDCS peak has also moved to smaller scattering angles, and there is evidence of a dip in the cross section around $\xi_2 = 50^\circ$. This type of feature is often seen when ionizing a p-electron (eg in the heavier noble gases), and is attributed to the momentum of the p-electron being zero at the origin. It is interesting to note that for the ejected electron to emerge in the scattering plane with $\varepsilon = 90^\circ$, multiple scattering from the core must occur. No theory yet exists to compare with these data, and it will be interesting to see how successful different models will be.

Experiments wishing to *pre-align* a molecule prior to the collision are very challenging. Adiabatic alignment of molecules is possible using large laser fields (as found in high power pulsed lasers), however the alignment rapidly relaxes once the field is removed. Experiments studying alignment in molecules have hence considered the interaction only when the laser pulse is on, which is typically ~5ns at a repetition rate of 10Hz (ie the molecules are aligned only 0.000005% of the time). It is impossible to carry out precise (e,2e) measurements from aligned molecules with such a small
alignment time, since (e,2e) experiments require many hours of operation to measure the QDCS, even with a high density of targets in the interaction region.

It should be possible to significantly increase this interaction time by again employing an optical cavity around the interaction region. This requires careful cavity design so that a high power laser pulse could enter the cavity, pass through the interaction region many times with low loss, then exit the cavity to be regenerated prior to re-injection. Figure 6 shows such a possible arrangement. For this to be successful the cavity must obey the usual rules of stability (i.e. $0 \leq g_1 g_2 \leq 1$; $g_{1,2} = 1 - L/R_{1,2}$ where $L$ is the optical path between the mirrors whose radius of curvature are $R_1, R_2$), the mirrors must have very high reflectivity (>99.999% is possible) and the gain from the laser medium must just balance losses due to reflection inside and outside the cavity. Since the molecules are not in resonance with the laser beam, there will be negligible losses from the alignment interaction (in contrast to the enhancement cavity described above, where this interaction provides the dominant loss mechanism).

In principle, such an arrangement will extend the interaction time of the high power pulse with the atoms by several orders of magnitude, making (e,2e) experiments possible. We are developing these experiments in Manchester, as well as through collaboration with Nick Martin’s group in Kentucky.

**Figure 6.** Example of an optical storage cavity proposed for (e,2e) experiments from laser-aligned molecules. The laser beam circulates many times through the interaction region before being passed by the Pockel switch into a regenerative gain medium, which compensates for losses in the cavity. The laser pulse is then re-injected into the cavity to continue molecular alignment. In this example the laser passes 36 times through the interaction region before regeneration, with a loss ~4% from the mirrors.

4. **Electron collision studies from cold atoms – the AC-MOT**

The direction of atomic physics research in recent years has moved significantly into the study of cold atoms. Much of this work studies the properties of Bose Einstein Condensates and the interactions that occur in these ultra-cold quantum systems. In the field of collision physics the use of cold targets also has advantages (eg by exploiting the momentum exchanged to the cold target during the collision). There has been very few studies in this area however due to the complexity of the experiments, and due to problems that occur when the fields from the magneto-optic atom trap (MOT) influence the momenta of charged particles. This is particularly critical in the study of low energy electron collisions, as they are extremely sensitive to the magnetic $\mathbf{B}$-fields generated by the MOT.

To eliminate these problems a new type of atom trap was invented in Manchester, the AC-MOT [14]. This trap has the same efficiency as a conventional DC-MOT, and can cool the atoms to the same temperature. The key difference is that the $\mathbf{B}$-fields generated by the AC-MOT can be switched off ~300 times faster than for the DC-MOT, making low energy electron scattering experiments possible.

In a conventional MOT, the field is generated by a constant current passing through a set of coils to produce an anti-Helmholtz magnetic field in the interaction region. Six orthogonal circularly polarized laser beams illuminate the atoms to produce an optical molasses, which reduces their velocity and so cools them. The combination of $\mathbf{B}$-field gradient and laser polarization results in an additional restoring force that directs the cooling atoms back to the trap centre, where they accumulate. The lowest temperature of the atoms in the trap is then dictated by their recoil from a single photon.
When the current is quickly switched off in a DC-MOT, eddy currents are induced in conducting components within the spectrometer. These currents produce additional $B$-fields that decay with a lifetime set by the inductance and resistance of the components. Studies show the typical time for these fields to reduce to $<5\text{mG}$ (where electron scattering experiments can be performed) is $\sim6-10\text{ms}$. In this time a significant number of the trapped atoms escape, and the trap density rapidly reduces. An additional problem arises due to the low cycling rate required due to this delay, which is $\sim50-80\text{Hz}$. Both problems mean electron scattering from trapped atoms has not been widely adopted.

These problems are eliminated in the AC-MOT. The key differences compared to a conventional MOT are that an alternating sinusoidal $B$-field is generated in the AC-MOT, and the laser polarization is switched from $\sigma^+$ to $\sigma^-$ at the same rate as the field. In this way, atoms in the trapping region still experience a time varying restoring force which is directed towards the centre of the trap. The laser beams again produce a ‘molasses’ force which reduces the atomic velocity to the photon recoil limit. A high density of trapped atoms rapidly accumulates in the same way as for a conventional MOT. By switching the AC-current off when the current reaches zero, the effects of induced eddy currents are eliminated. Since there is then no induced $B$-field, the electron beam can be injected into the trap almost immediately (in practice a delay $\sim18\mu\text{s}$ is required due to the slew rate of the amplifier driving the coils). Since the cold atoms are virtually stationary during this time, no losses occur from the trap during the electron interaction (apart from those due to collisions with the background gas, which are always present). Electron scattering experiments can hence be performed at a repetition rate $\sim300-500$ times faster than is possible with a conventional MOT.

Figure 7 shows the methodology used for electron impact ionization experiments from laser-trapped potassium atoms in Manchester. The trap was pre-loaded for 1000ms to accumulate a high density of atoms which cooled to the recoil limit ($\sim250\text{µK}$). The AC-field was then switched off, and the electron spectrometer switched on for 200$\mu\text{s}$, corresponding to a single cycle of the 5kHz field. The AC-MOT was then switched back on for 400$\mu\text{s}$ to re-establish the trap. This cycle was repeated for several seconds before the trap was reloaded, and the cycle recommenced. Electron impact ionization experiments were performed from $\sim2\text{eV}$ to 70$\text{eV}$ incident energy, the resulting ions being detected in a time of flight detector. Figure 8 shows the results of these studies, which demonstrate both single and double ionization of the potassium targets. The shape of the associated curves is directly related to the total cross section for single and double ionization of potassium as a function of incident energy. These are the first comprehensive electron collision results from a cold atom ensemble, proving the success of the AC-MOT for these studies. These cold targets can be exploited in new experiments, including COLTRIMS and other experiments that measure the recoil of the ion to establish the cross section. A further advantage occurs for laser-based studies, since the Doppler profile of the cold atom ensemble is virtually eliminated. This allows efficient excitation to higher lying $(n,l)$ states using CW lasers. New experiments are under preparation in Manchester which will exploit these advances.
5. Conclusions
This paper has discussed three new experiments that combine laser excitation with electron collisions. Electron impact excitation has been considered using super-elastic techniques, and a new experiment using an optical enhancement cavity detailed. Ionization studies have been described both from laser-excited targets, and for targets cooled and trapped in a new type of atom trap. All three experiments promise to progress the field of electron collisions into new directions, with the powerful combination of laser and electron interactions allowing new data to be obtained. Several experiments in Manchester are moving towards collision (and trapping) studies inside optical cavities, and two examples have been given to show the advantages of this new technique. Enhancement cavities allow low power lasers to efficiently excite the atoms under study, opening up access to many new targets. These cavities can also be used in atom cooling and trapping experiments, thereby reducing the cost of the laser systems that are required. It should further be possible to use storage cavities with pulsed lasers, to increase the laser-interaction time for (e,2e) studies of aligned molecules. This is one possible method which may solve this very difficult problem in the future.

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6. References
[1] Eminyan M, MacAdam K, Slevin J and Kleinpoppen H 1973 Phys. Rev. Lett. 31 576
[2] Hertel I V and Stoll W 1974 J. Phys. B 7 570
[3] Hussey M J, Murray A J, MacGillivray W R and King G C 2008 J. Phys. B 41 055202
[4] Murray A J, MacGillivray W R and Hussey M 2008 Phys. Rev. A 77 013409
[5] Li Y and Zetner P W 1994 J. Phys. B 27 L293
[6] Knight-Percival A, Jhumka S, Hussey M J and Murray A J 2011 J. Phys. B 44 105203
[7] Fursa D V and Bray I 2008 J. Phys. B 41 145206
[8] Kawazoe S, Kai T, Chauhan R K, Srivastava R and Nakazaki S 2006 J. Phys. B 39 493
[9] Zatsarinny O, Bardchat K, Bandurina L and Gedeon S 2007 J. Phys. B 40 4023
[10] Chauhan R K, Srivastava R and Stauffer A D 2005 J. Phys. B 38 2385
[11] Murray A J, Hussey M J and Needham M 2006 Meas. Sci. Tech. 17 3094
[12] Colgan J et al 2008 Phys. Rev. Lett. 101 233201
[13] Nixon K L and Murray A J 2011 Phys. Rev. Lett. 106 123201
[14] Harvey M and Murray A J 2008 Phys. Rev. Lett. 101 173201