Combining laser and electron interactions – current experiments and future possibilities in excitation and ionization studies

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Abstract. In this brief review a number of different experimental techniques that combine the advantages of laser and electron interactions with atoms and molecules will be described. These processes range from super-elastic electron scattering from laser excited atoms so as to ascertain the electron collision parameters over all scattering geometries, through new electron interactions with laser cooled and trapped atoms, to the process of electron impact ionization of laser-aligned targets. Examples from work carried out in Manchester are given, and new types of measurements that may be possible in the future are described.

1. Introduction

Studying electron interactions with atomic and molecular targets has a long and distinguished history from the time Franck and Hertz first studied excitation of mercury to confirm the quantum nature of the atom, as predicted by Bohr. Since then scattering experiments have markedly improved in sophistication, and the corresponding quantum models describing these complex interactions now accurately predict much of the data. Studies include the processes of excitation, where the target is left in an excited state following the collision, through to interactions where the target is ionized. The most sophisticated measurements adopt coincidence techniques to correlate the reaction products, allowing the most accurate comparison to quantum models. In the case of excitation the scattered electron is measured in coincidence with an emitted photon to determine the collision parameters [1], whereas for ionization, (e,2e) studies determine the probability of ionization as a function of the scattered and ejected electron momenta [2].

The interaction of laser radiation with atomic and molecular targets usually adopts a tuneable laser source that can resonantly excite transitions within the target. High power pulsed lasers may also directly ionize the target, however these type of reactions are not considered here. For resonant excitation either a pulsed or continuous wave (CW) laser may be used, the limited wavelength range accessible from these lasers usually confining the number of targets that can be directly excited to only a few. By contrast, combining electron and laser interactions opens up a far wider range of targets for study, and further allows the unique properties of the laser and electron interaction to be exploited simultaneously. It is these types of experiments that are discussed here, using the examples of super-elastic scattering from a laser prepared target, electron impact ionization from laser cooled atoms, and ionization from laser aligned targets in CW and pulsed laser beams.
2. Super-elastic electron scattering from laser prepared atoms.

In this type of study a high resolution polarized single mode laser (1) coherently and resonantly excites an atomic ensemble from the ground state to an excited state, as shown in figure 1. The excited targets are then de-excited using an incident electron beam (2), so that the reaction products carry away from the interaction both the excess energy given to the atom by the laser beam, and the excited sub-state information that is imparted to the atom by the coherent excitation process. This information is determined at a given electron scattering angle as a de-excitation probability (3), which depends on the polarization and direction of the exciting laser beam. The super-elastic scattering process can hence be considered as a time reversal of the electron-photon coincidence technique. By contrast, the super-elastic process accumulates data many thousands of times faster, since the laser is always input in the same direction and the probability of excitation is high.

The data from super-elastic scattering measurements is usually given as a set of ‘natural frame’ parameters [1] that precisely define the electron excited atom. These are the angular momentum transferred to the atom during the collision \( L \), the alignment parameter \( P_{\text{lin}} \) describing the ‘length’ to ‘width’ of the charge cloud and \( \gamma \), which describes the alignment angle as in figure 1. These parameters relate directly to the scattering amplitudes for excitation that are predicted theoretically.

![Figure 1](image1.png)

**Figure 1.** The super-elastic geometry showing the direction of incident laser and electron beams. The electron detector measures electrons which have gained energy from the simultaneous laser and electron interaction.

**Figure 2.** The collision parameters \( L \), \( P_{\text{lin}} \) & \( \gamma \) for excitation of calcium at 55eV incident energy, as a function of scattering angle. These data were taken using a MAC device inside the super-elastic spectrometer.

Figure 2 shows an example of these measurements for a calcium target taken at an equivalent coincidence incident energy of 55eV, compared to the theoretical prediction of Stauffer and colleagues [3] who used a relativistic distorted wave calculation (r-DWA) to derive the scattering amplitudes. Theory and data are in impressive agreement at this energy. It is also noted that the data have been taken at scattering angles up to and beyond 180 degrees, which was possible using a Magnetic Angle Changer (MAC) in the spectrometer [4-6]. These experiments were the first to adopt this new
technique in these laser-based studies, and are the first to fully ascertain the collision parameters over all geometries. Agreement between theory and experiment becomes less favourable as the incident energy is lowered, and at energies below ~20eV the r-DWA calculation no longer performs well [7]. At these energies it then becomes necessary to adopt different theoretical methods [8], and it is in this low energy regime where current experiments are being conducted.

3. Collisions with cold atoms – the AC-MOT.

One of the key advances in laser-atom interactions over the last 20 years has been in the field of atom cooling and trapping, which adopts near resonant CW laser fields to interact with atoms in a magnetic field gradient [9]. The direct-current Magneto-Optical Trap (DC-MOT) invented by Chu and co-workers [10] has found wide use in areas ranging from metrology through to fundamental studies of Bose Einstein Condensation (BEC). Although DC-MOT’s have been used in selected high energy ion collision studies (eg MOTRIMS experiments [11]), they have not been adopted in electron collision studies. For this type of collision the main limitation of the DC-MOT arises due to the B-fields from the DC-MOT, including induced fields generated by eddy currents in the spectrometer which occur when the DC-MOT current is switched on and off.

A new type of atom trap has recently been invented in Manchester which uses an alternating current to produce the required B-field, and which simultaneously switches the polarization state of the trapping lasers at the same rate [12]. This AC-MOT has proven to be extremely effective, and will allow many new types of collision studies to be conducted in the near future. The AC-MOT has the same trapping performance as a conventional DC-MOT, but the generated B-fields can be switched off more than 300 times faster than for the conventional MOT. This breakthrough means many different collision studies can now be routinely carried out with cold targets, so as to exploit the low Doppler profile and low momentum that these targets possess.

As an example of the enhanced speed of the AC-MOT, experiments were conducted which measured current from a low energy electron beam provided by an electron gun, where the electron beam passed through the centre of the atom trap. The trap was initially switched on with a field gradient in the trapping region of 10G/cm (as required to trap potassium atoms in the experiment). This field resulted in a large deflection of the electron beam, so that no current was observed on a Faraday cup located on the opposite side of the trapping region to the gun. The current in the MOT coils was then switched off in ~20μs, and the Faraday cup current measured as a function of time.

Figure 3 compares the results for the DC-MOT with those for the AC-MOT. At the electron energies used here (~20eV), B-fields greater than ~5mG were found to deflect the electron beam out of the Faraday cup. These measurements therefore provide a very sensitive test of the influence of the magnetic fields generated by the different types of trap when used for electron collision studies.

![Figure 3](image_url)

**Figure 3.** Faraday cup current measured as a function of time for (a) the DC-MOT and (b) the AC-MOT, where the current through the MOT coils was switched off at t = 0sec.
Figure 3(a) shows that experiments with trapped atoms in a conventional DC-MOT would need to wait \(~6000\mu s\) before the \(B\)-field due to induced eddy currents reduce to \(<5\text{mG}\), at which time electron beam collision experiments could be performed. This severely constrains the experiments, as many trapped atoms will have diffused out of the trapping region in this time, and the pulsed repetition rate of these experiments must be kept low (~80Hz). By contrast, figure 3(b) shows for the AC-MOT, the \(B\)-fields decay to \(<5\text{mG}\) within 20\(\mu s\). In this case, eddy currents generated in the spectrometer are effectively eliminated by adopting an equal number of positive and negative cycles in the AC-MOT drive current. In a time of 20\(\mu s\) the trapped atoms are effectively frozen in space, and so diffusion from the trapping region is negligible. Further, since the AC-current can be driven at greater than 6kHz, pulsed collision experiments from these trapped atoms can be carried out many times faster.

Figure 4. Time of flight spectrum for ionization of cold trapped potassium using electron impact.

Figure 4 shows an example of collision data from cold potassium atoms (\(T\sim300\mu K\)) in the AC-MOT, where a time of flight detector was installed to measure the total ionization cross section as a function of incident electron energy. These are the first comprehensive results from electron collisions with cold atoms. The spectrum shows the cross section for \(K^+\) and \(K^{++}\) formation, the incident electron energy ranging from 2eV to 70eV. In these experiments the laser field was on while the collision occurred, and so there is an additional contribution to the cross section from excited potassium. Adoption of the AC-MOT will allow many new experiments to be conducted. As an example, since the Doppler profile of the trapped atoms is effectively eliminated, it becomes possible to carry out super-elastic scattering experiments from targets with high principle quantum numbers. This will allow collision theories to be tested for excitation to highly excited states, with a precision impossible to obtain using conventional methods. Further, the well defined momentum of trapped atoms will allow target deflection techniques that measure cross sections to be more widely adopted. The AC-MOT provides much higher precision than is possible using conventional COLTRIMS and other types of deflection techniques, since the uncertainty in the initial momentum of the target that limits the resolution in these experiments is markedly reduced.

4. Ionization from laser-aligned targets.

The final topic discussed here is that of electron impact ionization, as measured by the \((e,2e)\) technique. These experiments determine the angular correlation between the electron scattered from an ionized target and the electron that is ejected. Since the electrons can emerge from the interaction at any angle, the ionization probability may be measured over the full scattering geometry for a range of incident energies and outgoing electron momenta. As such, experiments usually measure a subset of this five-dimensional space. Often a coplanar geometry is chosen (where the incident, scattered and
ejected electron momenta all lie in a plane), however it is also possible to measure the ionization probability in non-coplanar geometries. The (e,2e) spectrometer in Manchester allows measurements from a coplanar geometry through to the perpendicular plane (where the incident electron momentum is orthogonal to the scattering plane), and so provides a stringent test of current ionization theory. Recent experiments in Manchester have been conducted over a wide range of scattering geometries where the target is either a diatomic molecule (such as H₂ or N₂) [13,14] or tri-atomic (such as H₂O or CO₂) [15,16]. These experiments paralleled significant theoretical developments which consider the collision using a non-spherical basis, or which adopt time dependent close coupling techniques. These theoretical methods predict the ionization probability as a function of the molecular alignment with respect to the scattering plane, or can average the results over all possible target orientations to compare to conventional experiments that do not select the alignment angle. These theories have recently proven to be very successful when compared to new experimental data being produced [17].

Of key significance in these calculations is the marked change in the cross section predicted for different molecular alignment angles. A few experiments have been conducted to test these theories, by adopting an energy scheme that ensures the molecular ion is left in a repulsive state following the interaction [18]. In this case Coulomb repulsion between the nuclei drives the molecule apart, and it is assumed the final ion direction defines the alignment direction of the target prior to interaction. These experiments are very difficult, and have a low yield and low energy resolution due to the steep gradient of the repulsive molecular energy curve. As such the data uncertainties are high. Experimental techniques exist to partially align a polar molecule (e.g. using a hexapole field prior to the interaction region [19]), however these techniques are not applicable to molecules such as H₂, for which current collision theories are proving successful. It is hence worthwhile considering whether other techniques are possible to control the alignment of these targets.

A technique that has proven successful in recent experiments adopts a non-resonant high power pulsed laser beam to adiabatically align supersonically cooled molecules along the direction of the laser polarization [20]. In these experiments the laser field induces a dipole moment in the molecule, the molecule then re-orienting so as to minimise the interaction energy in the field. By supersonically cooling the target ensemble prior to this interaction, the rotational energy of the molecules is minimised, allowing the coupling of the laser field to the molecule to be maximized. These experiments clearly demonstrate that the molecules are aligned in the field, as predicted.

Unfortunately this alignment only occurs when the laser illuminates the target, and for all experiments carried out so far the temporal width of the laser pulse is only ~5ns, with a repetition rate of 10Hz. The molecules are hence only aligned for 50ns in each second, and so carrying out electron scattering coincidence experiments from these laser aligned targets is currently unrealistic.

**Figure 5.** Design of a multi-pass high resolution laser cavity to extend the laser-molecule interaction time.

A new method is suggested here, as shown in figure 5. In this case an incident high power laser pulse is optically switched into a very high reflectivity cavity using a Pockels cell. A multi-pass cell (MPC) incorporated within the cavity ensures the laser pulse passes through the interaction region many times
before the laser beam leaves the MPC, after which it is passed to a regenerative laser amplifier. This amplifier is constructed to add gain to the laser pulse so as to balance losses in the cavity. The regenerated laser pulse is then reflected back into the MPC (and hence through the molecules to align them), and returns to the Pockels cell which has been switched to ensure the laser pulse remains inside the cavity. By choosing the gain of the laser medium to balance cavity losses, it will then be possible to extend the time of alignment of the molecules by several orders of magnitude. Collision based experiments from laser-aligned molecules should then be possible.

A new pulsed (e,2e) spectrometer has been constructed in Manchester to commence preliminary studies in this area. The new spectrometer uses a pulsed supersonic jet to deliver rotationally cold molecules to the interaction region, and angular correlation measurements have now commenced from these targets. The experiments will initially look for differences between cold and hot molecules, by comparing (e,2e) results from the supersonic jet and from an effusive beam at room temperature. An optical multi-pass alignment cavity will then be designed and tested, to determine the feasibility of the proposed technique. Parallel experiments will also be conducted from laser aligned excited atoms in the existing (e,2e) spectrometer, to ascertain the effects of ionization from these targets. This will allow the new theoretical developments to be further refined and directly tested.

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5. References

[1] Andersen N, Gallagher J W and Hertel I 1988 Phys. Rep. 165 1
[2] McCarthy I E and Weigold E 1991 Rep. Prog. Phys. 54 789
[3] Chauhan R K, Srivastava R and Stauffer A D 2005 J. Phys. B. 38 2385
[4] Hussey M J, Murray A J, MacGillivray W R and King G C 2007 Phys. Rev. Lett. 99 133202
[5] Murray A J, MacGillivray W R and Hussey M J 2008 Phys. Rev. A 77 013409
[6] Hussey M J, Murray A J, MacGillivray W R and King G C 2007 J. Phys B 41 055202
[7] Murray A J and Cvejanovic D 2003 J. Phys B 36 4889
[8] Zatsarinny O, Barschat K, Bandurina L and Gedeon S 2007 J. Phys. B 40 4023
[9] Metcalf H J and van der Straten P 1999 Laser cooling and trapping (Springer: New York)
[10] Raab E L, Prentiss M, Cable A, Chu S and Pritchard D E 1987 Phys. Rev. Lett. 59 2631
[11] DePaola B D, Morgenstern R and Andersen N 2008 Adv. At. Mol. Opt. Phys. 55 139
[12] Harvey M and Murray A J 2008 Phys. Rev. Lett. 101 173201
[13] Murray A J 2005 J. Phys. B 38 1999
[14] Murray A J, Hussey M J, Gao J and Madison D H 2006 J. Phys. B 39 3945
[15] Kaiser C, Spieker D, Gao J, Hussey M J, Murray A J and Madison D H 2007 J. Phys. B 40 2563
[16] Hussey M J and Murray A J 2005 J. Phys. B 38 2965
[17] Colgan J, Pidzola M S, Robicheaux F, Kaiser C, Murray A J and Madison D H 2008 Phys. Rev. Lett. 101 233201
[18] Takahashi M, Watanabe N, Khajuria Y, Udagawa Y and Eland J H D 2005 Phys. Rev. Lett. 94 213202
[19] Baugh D A, Kim D Y, Cho A, Pipes L C, Petteway J C and Fuglesang C D 1994 Chem. Phys. Lett. 219 207
[20] Stapelfeldt H and Seideman T 2003 Rev. Mod. Phys. 75 543