Laser Probing of Metastable Atoms and Molecules Deflected by Electron Impact

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A new technique for determining electron impact metastable excitation cross sections which exploits angular deflection of the target is presented. The deflected targets, further excited by laser radiation which is resonant between the metastable state and a high-n Rydberg state, are detected using field ionization. The target deflection angle and flight time determines the associated electron scattering angle, whereas the laser probes individual states of the electron excited manifold. New information over a full range of scattering angles from forward to backward scattering is then obtained. [S0031-9007(99)09364-3]

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The study of electron collision processes has a long tradition in the field of atomic and molecular physics, much of this work involving either excitation [1] or ionization [2] of the target. Determination of the incident and scattered electron momenta has allowed differential cross section measurements for comparison with state-of-the-art quantum mechanical models. The most sophisticated techniques employ coincidence methods to maximize the information obtained about the collision. For ionization, $(e, 2e)$ coincidence studies where the incident, scattered, and ionized electron momenta are determined allow a detailed description of the ionization process to be ascertained. For excitation studies of the target atom or ion, decay radiation from the electron excited state is measured in coincidence with the scattered electron momentum. By measuring the polarization or angular distribution of the radiation a set of atomic collision parameters can be obtained, allowing the alignment and orientation of the target to be determined for comparison with theory [1,3].

By contrast, excitation to a metastable target cannot use conventional coincidence techniques since the target does not decay by dipole radiation [4]. At present no measurements have been made to determine atomic collision parameters for these targets. Since electron impact excitation to metastable atomic and molecular states plays an important role in the energetics of many processes including plasma formation, fluorescent lighting, and astrophysics it is advantageous to understand these excitation mechanisms in detail.

One technique that can be used to determine metastable alignment and orientation parameters is to transfer the information from the metastable target to a higher lying state using resonant laser excitation. Polarization analysis of fluorescence from this laser excited state measured in coincidence with the scattered electron can then be implemented. The upper state radiation carries information about the metastable state which can be extracted by carefully modeling the laser interaction process [5]. This technique has been successfully used to determine a complete set of collision parameters for the 6^1P_1 state of mercury [6,7]; however, it has not as yet been applied to metastable targets. These stepwise electron-laser coincidence experiments are difficult due to the very low coincidence yield, requiring long term stability of the laser and electron spectrometer. To facilitate acceptable statistics in these experiments run times of weeks to months for each scattering angle are required, making the experiments very difficult in practice.

A new experimental technique is presented here which allows the collision process to be studied by considering details of the excited targets themselves. This is achieved by using a pulsed supersonic target beam which has a well defined initial momentum (in contrast to the effusive atomic beams used in conventional experiments). A pulsed electron beam excites and spatially deflects the targets, with the deflection angle being selected using a metastable detector which rotates around the interaction region [8]. Following electron collision both the deflection angle and time of flight of the excited targets is accurately measured. Since the distance from the interaction region to the detector remains constant, this allows the final momentum of the excited targets to be determined.

The equations of motion which govern the final target state momentum $\mathbf{P}_{af}$ are given by [9]

$$\left\{ \begin{array}{l} P_{af} = m_a v_{af} = \sqrt{P_{ei}^2 + P_{ai}^2 + P_{ef}^2 - 2\xi P_{ef} \sin(\theta_e + \Delta)} \\ \theta_a = \tan^{-1}\left(\dfrac{P_{ef} \sin\theta_e - P_{ai}}{P_{ef} \cos\theta_e - P_{ei}} \right) \\ \xi = \sqrt{P_{ei}^2 + P_{ai}^2} \quad \& \quad \Delta = \tan^{-1}\left(\dfrac{P_{ei}}{P_{ai}} \right) \end{array} \right\}, \tag{1}$$

where m_a is the target mass, $\mathbf{v}_{af}$ is the final velocity of the target, $\mathbf{P}_{ai}$ is the initial target momentum, $\mathbf{P}_{ei}$ and $\mathbf{P}_{ef}$ are,

respectively, the initial and final electron momenta, θ_e is the electron scattering angle, and θ_a is the measured target deflection angle. Measurement of the momentum of the deflected targets (i.e., the time of flight and target deflection angle) then allows the associated electron scattering angle to be obtained.

As an example in the present experiments for helium with a 40 eV incident electron beam the 2^1S_0 and 2^3S_1 metastable excited targets deflect through scattering angles from ~4° for forward electron scattering through to ~28° for backward electron scattering. Between these extremes different electron scattering angles produce different target momenta which can be measured using time of flight techniques at a given detector angle. Since a unique relationship exists between the electron scattering angle and momentum transferred to the deflected target [Eq. (1)], it is not necessary to detect the scattered electron to determine the differential cross section. Furthermore, since the excited targets can be measured for all possible electron scattering angles, a complete differential cross section is determined. Until recently conventional methods have prevented measurements in either the fully forward and/or high angle backward scattering geometries; however, a new and alternative technique has recently been developed using a modified electron spectrometer which also allows cross sections in these regions to be determined [10].

Figure 1 shows an example of the metastable target signal as a function of deflection angle and time of flight obtained from the interaction of a pulsed supersonic helium beam with a 40 eV pulsed electron beam crossed perpendicular to the target beam. The electron beam temporal pulse width was 4 μs and the pulse repetition rate was 150 Hz. The first graph shows a single peak obtained at a target detection angle of 4° corresponding to forward electron scattering. As the detection angle is increased two distinct peaks emerge, corresponding to different electron scattering directions. Between target deflection angles of 10° and 24° the peaks are fully resolved. As the detection angle further increases the peaks once more start to merge until at 28° deflection angle only a single peak remains. This corresponds to electron scattering in the backward direction. Since the area under each peak is proportional to the metastable excitation cross section, the differential cross section is obtained over a wide range of scattering angles as shown in Fig. 2.

It should be noted that this measurement involves contributions from both the 2^1S_0 and 2^3S_1 metastable helium states. Further, a contribution arises from cascading into these states from higher lying electron excited states. It is possible to resolve the cascade contribution by applying a coincidence technique between the metastable signal and the scattered electrons using an electron analyzer set to pass only electrons of the correct energy for metastable state excitation. This has a considerable advantage over other coincidence techniques since the electron scattering angle is already known from the detection angle and flight

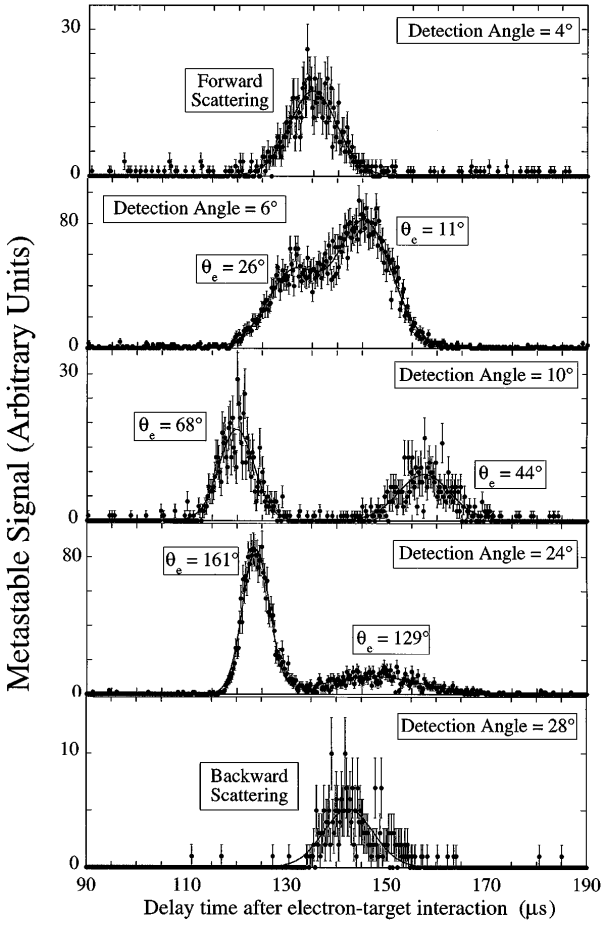

FIG. 1. Deflected metastable helium momentum measurements for a 40 eV incident electron beam. The analyzer set between 4° and 28° detects both 2^1S_0 and 2^3S_1 metastable targets over a complete range of electron scattering directions. The electron scattering angle corresponding to the individual target peaks is shown.

time of the metastable targets. The electron analyzer can be positioned at this angle, the resulting coincidence count between electrons and metastable targets eliminating all cascade contributions to the signal. Experiments implementing these selective coincidence methods are currently being set up in the laboratory.

The new experimental technique presented here will allow ro-vibrationally selective measurements in molecules. This selectivity is impossible within the metastable manifold of a molecule even with the highest resolution electron spectrometers [11] since the levels are very closely spaced energetically. In the experimental technique described here a high resolution tunable laser is used to resonantly excite the deflected targets from a particular metastable state to a high lying Rydberg state [12] and the energy resolution of the incident electron is therefore immaterial. The resolution of the laser is exploited to select individual metastable states from the manifold of electron excited states. In metastable helium, individual contributions of the 2^1S_0 and 2^3S_1 metastable helium states,

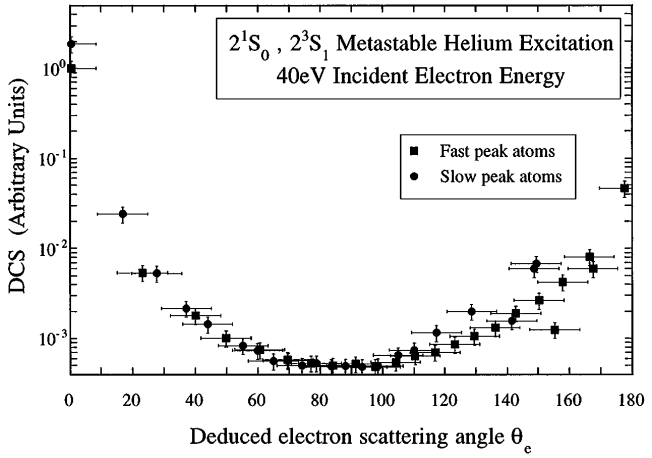

FIG. 2. The extracted differential cross section for excitation to the 2^1S_0 and 2^3S_1 metastable states at 40 eV incident energy determined from momentum deflection measurements, a subset of which is shown in Fig. 1.

separated by 798 meV, can be determined using a high resolution electron gun and analyzer, but here we establish the viability of the laser probing scheme for wider application.

This new experimental method has been successfully implemented using helium as the target prior to conducting experiments on a molecular target. Figure 3 shows the excitation mechanism that has been used. A 40 eV pulsed incident electron beam deflects the helium atoms and excites them to the unresolved 2^1S_0 and 2^3S_1 metastable states as described above. A pulsed laser beam interacts with the targets upstream of the metastable detector, resonantly exciting the 2^1S_0 targets to the 30^1P_1 state using radiation at 313.651 nm. These highly excited targets are ionized using a pulsed electric field which also accelerates the resulting ions towards an apertured ion detector where they are counted. By measuring the ion yield as a function of flight time in the detector the individual contribution to the differential cross section from the 2^1S_0 target excitation is determined.

Figure 4 shows an example of the laser probed 2^1S_0 time of flight ionization signal at a detection angle of 6°. The metastable signal arising from both 2^1S_0 and 2^3S_1 metastable targets is also shown for comparison. The ion signal is measured as a function of flight time by delaying the laser pulse to select different regions of the metastable targets as they pass through the laser beam interaction region inside the detector. The field ionized signal is counted earlier than the metastable signal since the laser interacts with the metastable targets upstream from the metastable detector. This also results in the relative narrowing of the signal which is observed.

The difference in intensity of the metastable signal and ion signal mainly arises due to the efficiency of the laser interaction process, and due to the low repetition rate of the pulsed laser which operates at 20 Hz. The ion signal was measured for 1000 sec at each data point. The most striking difference is seen comparing the fast peak (whose area is proportional to the cross section for an electron scattering angle of 26°), to the slow peak (whose area is proportional to the cross section for an electron scattering angle of 11°). For the combined 2^1S_0 and 2^3S_1 metastable target manifold the differential cross section ratio between these measurements is around 1.7:1, whereas for the individual 2^1S_0 state this ratio is approximately 3:1. This indicates that the cross section varies more rapidly with scattering angle for the singlet state compared to the triplet state. By moving to a different angle and repeating the experiment over the range of detector angles from 4° to 28° a full differential cross section for the 2^1S_0

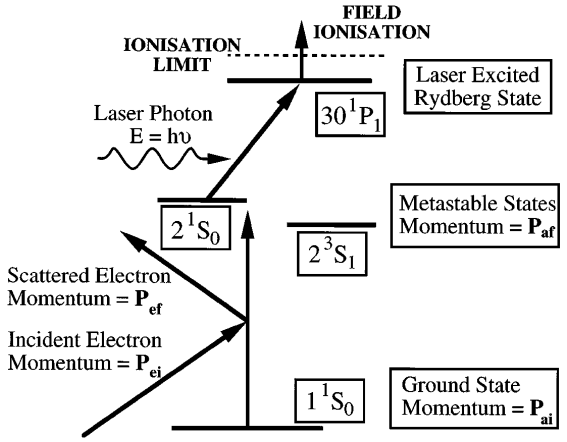

FIG. 3. Resonant laser excitation in helium from the 2^1S_0 metastable state to the 30^1P_1 state. A pulsed electric field ionizes the laser excited targets, the ions being counted as a function of flight time. The laser selects only the 2^1S_0 metastable targets, thereby allowing individual state cross section measurements to be ascertained.

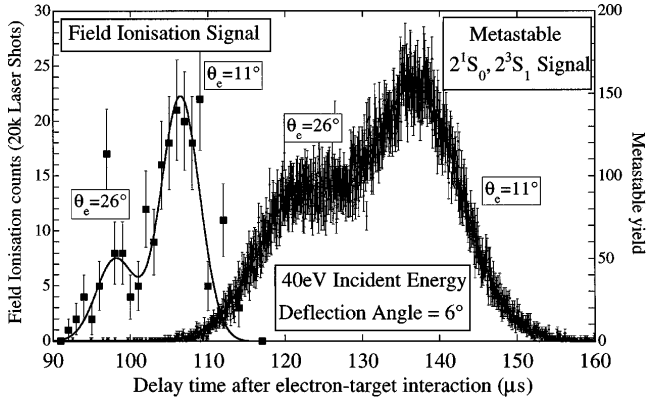

FIG. 4. Time of flight measurements of the 2^1S_0 and 2^3S_1 metastable targets at a detection angle of 6° compared to the field ionized signal obtained from laser excitation of the 2^1S_0 targets. The ion yield occurs at a shorter flight time since the laser interacts upstream from the metastable detector. The difference between the 2^1S_0 target yield and the combined metastable target yield as a function of scattering angle is seen.

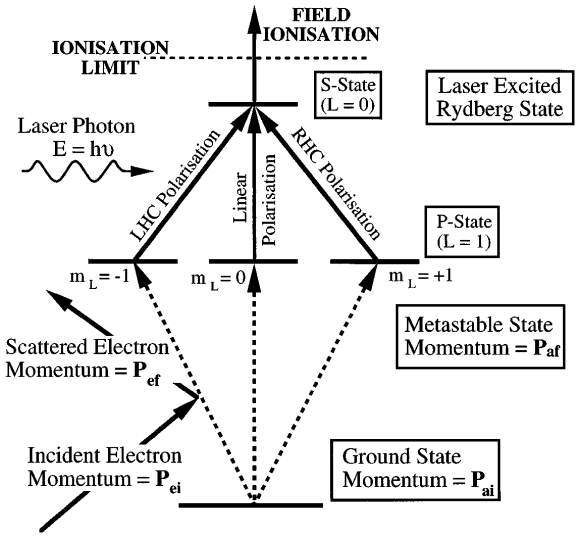

FIG. 5. Resonant laser excitation from a metastable P state ($L = 1$) to a high lying S state ($L = 0$). The laser polarization allows excitation from individual substates, circular polarization exciting from the outer $m_L = \pm 1$ states while linear polarization excites from the $m_L = 0$ state. The target in the upper state is field ionized and the ion yield is measured as a function of laser polarization, detection angle, and flight time to determine individual substate cross sections.

state can be determined. Similarly, the cross section for 2^3S_1 state excitation can be determined by repeating the experiment using laser radiation resonantly tuned between the 2^3S_1 and 30^3P states. These measurements will be conducted when the coincidence techniques have been fully implemented and tested, since cascade contributions can then be eliminated from the signal which is obtained.

The experimental results presented here using helium indicate the viability of this new experimental method. For atoms or molecules which have metastable states with nonzero orbital angular momentum (e.g., for atomic P states with $L = 1$), further information can be obtained about the electron collision process by varying the polarization of the laser beam to excite from different magnetic substates prior to field ionization. By choosing a laser wavelength to excite from the $L = 1$ metastable state to a high Rydberg $L = 0$ state, the polarization of the laser accesses individual substates as shown in Fig. 5. Circularly polarized radiation excites the metastable target from the outer $m_L = \pm 1$ substates, while linear polarization excites only from the $m_L = 0$ substate. Measurement of the field ionization yield as a function of laser polarization, target deflection angle, and time of flight then allows detailed information about these substate excitation cross sections to be determined.

Laser excitation studies are currently in progress to characterize the laser transitions from individual ro-vibrational states in the molecular $c^3\Pi_u H_2$ metastable manifold to higher lying Rydberg states prior to carrying out deflection experiments on this molecule using the techniques described here. This will then allow individual ro-vibrational state electron impact differential cross section measurements to be obtained from a molecular target for the first time.

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