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Electron Capture in Very Low Energy Collisions of Multicharged Ions with H and D in Merged Beams

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ABSTRACT: An ion-atom merged-beams technique is being used to measure total absolute electron-capture cross sections for multicharged ions in collisions with H (or D) in the energy range between 0.1 and 1000 eV/amu. Comparison between experiment and theory over such a large energy range constitutes a critical test for both experiment and theory. Total capture cross-section measurements for O^{3+} + H(D) and O^{5+} + H(D) are presented and compared to state selective and differential cross section calculations. Landau-Zener calculations show that for O^{5+} the sharp increase in the measured cross section below 1 eV/amu is partly due to trajectory effects arising from the ion-induced dipole interaction between the reactants.

MASTER

1. INTRODUCTION

With the merged-beams technique it is now possible to measure with a single apparatus total electron capture cross sections for multicharged ions in collisions with H or D over an energy range covering four orders of magnitude: 0.1 eV/amu to over 1000 eV/amu. Comparison of experiment with theory over such a wide energy range provides a stringent test of our understanding of such processes which are fundamental to modelling of both laboratory and astrophysical plasmas. Simple scaling laws (e.g., see Gilbody 1986) which have been used to parametrize the behavior of the total capture cross section in the keV/amu energy range, are not appropriate in this range. At these low energies, theoretical calculations must take into account that the nuclear motion between collision partners is slow compared to the orbital motion of the active electrons of the system. Electrons of the temporary quasi-molecule formed in the collision have sufficient time to adjust to the changing interatomic field as the nuclei approach and separate. State-of-the-art molecular orbital coupled-state calculations are rather complex and, at these low energies, treat the collision dynamics quantum-mechanically.

Comparison between theory and experiment over this large energy range permits evaluation of a number of the theoretical parameters and methods used in these calculations. For example, for the O^{5+} + H(D) system, such comparison (Andersson et al. 1991) at the higher energies permits evaluation of theoretical parameters (e.g., adiabatic potential energies and non-adiabatic coupling matrix elements) which are used to calculate the collision dynamics throughout the whole energy range. As will be illustrated for both O^{3+} and O^{5+}, electron capture for these systems is extremely state-selective at low energies, with the dominant (nl) final state of the multicharged ion changing as a function of energy.

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In addition, there are several aspects of the theoretical calculation of electron capture cross sections whose importance is energy dependent. For example, electron translation effects are of decreasing importance below 1 keV/amu (Gargaud and McCarroll 1985). Calculations have predicted that rotational coupling mechanisms, while important for some systems at energies around 1 keV/amu (Gargaud et al. 1988), are of negligible importance below 10 eV/amu. As collision energies decrease below 1 eV/amu, the ion-induced dipole attraction between the ion and neutral may become strong enough to significantly affect the reactant trajectories. These trajectory effects can lead to an enhancement in the cross section that increases with decreasing collision energy. In addition, at these low collision energies several quantal calculations (Rittby et al. 1984, Shimakura and Kimura 1991) have predicted "orbiting" resonances. These resonances are due to the excitation of ro-vibrational states in the shallow potential well formed by the attractive induced dipole in combination with the repulsive angular momentum barrier.

The ion-atom merged-beams apparatus (Havener et al. 1989) at Oak Ridge National Laboratory has recently been improved (Havener and Phaneuf, in prep.), allowing measurements to access collision energies as low as 0.1 eV/amu. Previous measurements (Havener et al. 1989, Huq et al. 1989, Havener et al. 1991) have been restricted to the 1 to 1000 eV/amu collision energy range. The first measurements to extend significantly below 1 eV/amu were made for the O^{3+} and O^{5+} + D systems and are presented here. Comparison with previous measurements and with state-selective (Gargaud 1987) and differential (Andersson et al. 1991) cross-section calculations illustrates the state selective nature of electron capture and accompanying, potentially large angular scattering of the products. To provide some physical insight into the collision dynamics below 1 eV/amu, Landau-Zener calculations are used to determine the energy dependence of the capture cross section for different reactant trajectories. Resultant estimates of the cross section show that the observed sharp increase in the cross section for O^{5+} + D below 1 eV/amu can be attributed in part to the ion-induced dipole attraction modifying the reactant trajectories. This enhancement in the cross section can be further amplified by considering collisions with H atoms rather than the heavier isotope D.

2. EXPERIMENTAL

The merged-beams method (Havener et al. 1989, and ref. within) is well suited for these measurements. In this technique, beams of neutral atoms and multicharged ions each having energies in the keV range are merged, resulting in a relative velocity of the two beams that can be "tuned" over a very large range. Figure 1 is a simplified schematic of the apparatus. The multicharged ion beam X^{q+} is merged electrostatically with a neutral H or D beam. The merged beams interact in a field-free region for a distance of 47 cm, after which the primary beams are magnetically separated from each other and from the product or "signal" $H(D)^+$ ions. The $X^{(q-1)+}$ product of the reaction is not measured separately, but is collected together with the primary X^{q+} in a large Faraday cup. The neutral beam intensity is measured by secondary-electron emission from a stainless steel plate, and the signal H^+ or D^+ ions are recorded by a channel electron multiplier operated in pulse-counting mode. A 99.98% pure ground-state beam of H or D atoms is produced by passing a 6- to 9-keV beam of H^- or D^- ions through the optical cavity of a 1.06-μm Nd:YAG laser, where up to 600 W of continuous power circulates and typically 0.5% of the negative ions undergo photodetachment. A nearly parallel beam of H(D) atoms is produced having a diameter of 2 to 4 mm FWHM and an equivalent intensity of 10 to 20 (particle) na. The divergence of this beam is typically less than 0.2°. A 50- to 90-keV, 2- to 5-μA beam of X^{q+} ions is produced by the ORNL-ECR source with a typical diameter of 6 to 8 mm FWHM in the merge path and a divergence of less than 0.5°. The finite divergence of the primary beams results in a distribution of merging angles, creating an energy spread of about 0.1 eV/amu at collision energies near 0.1 eV/amu.

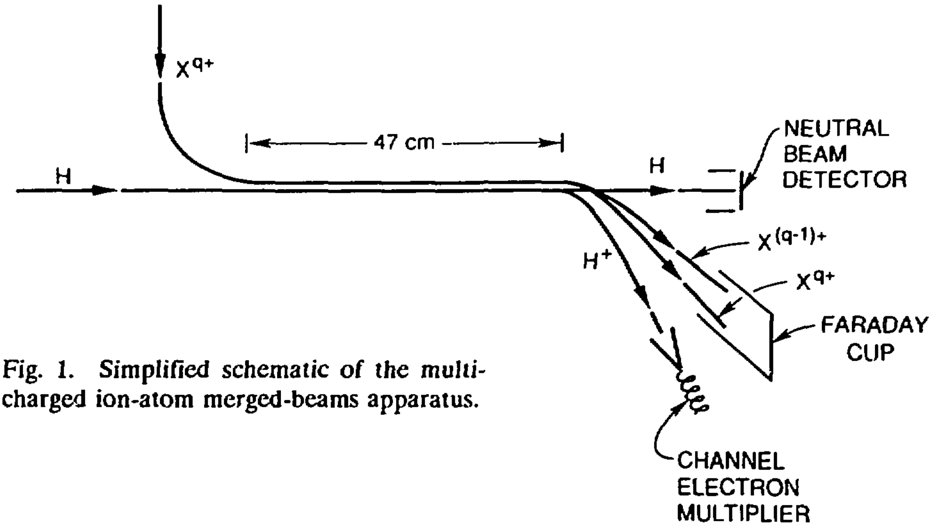


Fig. 1. Simplified schematic of the multi-charged ion-atom merged-beams apparatus.

Electron-capture cross sections are determined absolutely by measuring the rate of $H^+(D^+)$ ions produced by the beam-beam interaction over the merged path. The cross-section value is determined at each velocity from directly measurable parameters which include the signal count rate, intensities of the two beams, and the form factor which is a measure of the spatial overlap of the two beams. The integrated three-dimensional form factor is estimated from two-dimensional measurements of the overlap at three different positions along the merge path.

There have been some recent changes that have led to significant improvements to the technique. These improvements and their benefits are discussed elsewhere (Havener and Phaneuf, in prep.) in more detail, and are only briefly outlined here. While most of the beam-beam signal is generated at the beginning of the merge path where the overlap of the beams is typically better, the background due to H stripping on background gas is created uniformly along the merge-path. For this reason, improvements in vacuum and reduction of the merge-path from 80 to 47 cm has resulted in an increase in the signal-to-noise by at least a factor of 3, thereby extending the range of measurements downward to almost a factor of 10 lower in energy. Reduction of the small excited state component of the H(D) beam formed by stripping of H^-on background gas has resulted in a 99.98% pure ground-state beam. Such beam purities are essential, since even a small residual fraction of excited states can contribute significantly to the beam-beam signal because of the significantly larger cross sections for electron capture from excited atoms by multicharged ions. Enlargement of the physical aperture of the signal collection optics has resulted in a significant increase in the angular collection. Ion-trajectory modelling of the product D^+ trajectories has indicated that for a typical 8 keV D neutral beam, the angular collection of the apparatus was increased from an average angle of 1.8° to a minimum of 2.3°.

3. ANGULAR COLLECTION

An important advantage of the merged-beams technique in low-energy measurements is the potentially large angular collection of the reaction products. The low-energy electron capture collisions under study are exoergic and both products are positively charged, so that significant angular scattering can occur in the center-of-mass frame (Olson and Kimura 1982). However, due to the kinematic frame transformation, this angular scattering is significantly compressed

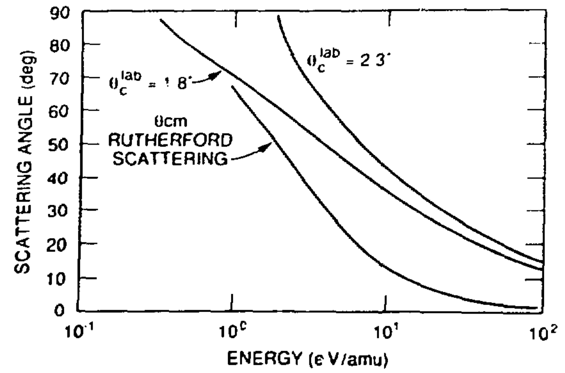


Fig. 2. Angular scattering and collection in the center-of-mass frame for the O^{5+} + (8 keV) D collision system. The angular collection estimates correspond to the original (θ_c^{lab} = 1.8°) and to the present (θ_c^{lab} = 2.3°) apparatus. The angular scattering estimate corresponds to a simple "half-Coulomb" Rutherford scattering calculation (see text).

in the laboratory frame, the frame in which the products are collected. As an example, Fig. 2 shows the angular collection for the O^{5+} + (8 keV) D system in the center-of-mass frame for a lab frame angular collection of 1.8 and 2.3°. Note that with the present apparatus, which has a 2.3° minimum angular collection, all products will be collected below 2 eV/amu for this system. Also shown in Fig. 2 is the Rutherford minimum scattering estimate (Olson and Kimura 1982). Recent detailed differential cross-section calculations (Andersson et al. 1991) predict significantly larger scattering for the O^{5+} + D system, requiring an angular collection in the lab of at least 2.3° for a collision energy between 1 and 10 eV/amu. Further discussion specific to this system is presented in the next section.

4. CROSS-SECTION MEASUREMENTS

The ion-atom merged-beams apparatus has been used to measure total capture cross sections for O^{3+} and O^{5+} + D collisions; these measurements along with other experimental and theoretical results are shown in Figs. 3-5. The error bars shown correspond to an uncertainty in the reproducibility of the measurements estimated with a 90% confidence level. The absolute uncertainty in the measurements corresponds to about 12% and must be added in quadrature to the relative uncertainty. For both the O^{3+} + H(D) and O^{5+} + H(D) collision systems, the merged-beams data join smoothly with other measurements (Phaneuf et al. 1982, Meyer et al. unpublished) at the higher energies based upon ion beam-gas target methods, and verify the normalization methods used for the latter.

For the O^{3+} + H(D) system the present measurements lie significantly below the calculations of Bienstock et al. (1983) which predict a large contribution from capture to the ($1s^22s^22p3s$) and ($1s^22s^22p3p$) configurations at these energies (see Fig. 3). Capture to the 3d state is not expected to contribute significantly in this energy range. Both the old and more recent merged-beams measurements are more consistent with the calculations of Gargaud et al. (1989) which predict a significantly smaller contribution from capture to the 3p state at the lower energies. Both calculations predict that capture to the 3s state dominates at the higher energies (see Fig. 4). This prediction agrees with the state-specific translational spectroscopy measurements of Wilson et al. (1988) which were performed at collision energies between 200 and 700 eV/amu.

Our previous O^{3+} measurements, which could only be extended down to 1 eV/amu, were not able to verify the cross section rise due to the predicted capture to the 3p state at low energies. However, the present merged-beam measurements, which extend down to 0.1 eV/amu, follow closely the predicted energy dependence of the 3p capture cross section. Nevertheless, as can be seen in Fig. 4, there remains a discrepancy between the measurements and theory throughout

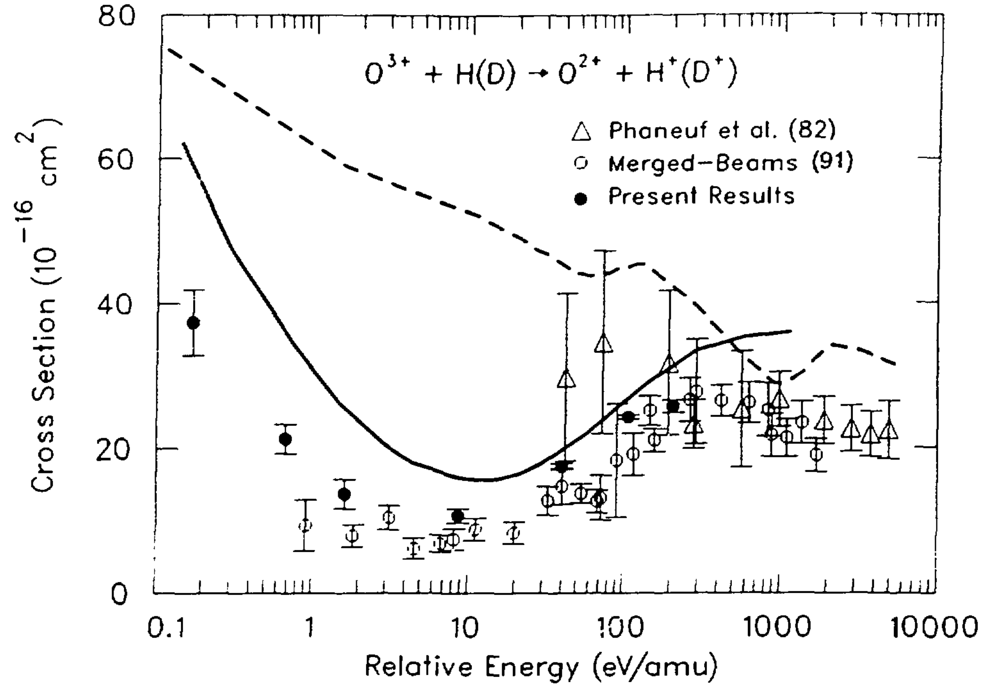


Fig. 3. Comparison of merged-beams data for O^{3+} + H(D) with other measurements (Phaneuf et al. 1982) and theoretical calculations (dashed curve, Bienstock et al. 1983; solid curve, Gargaud et al. 1989).

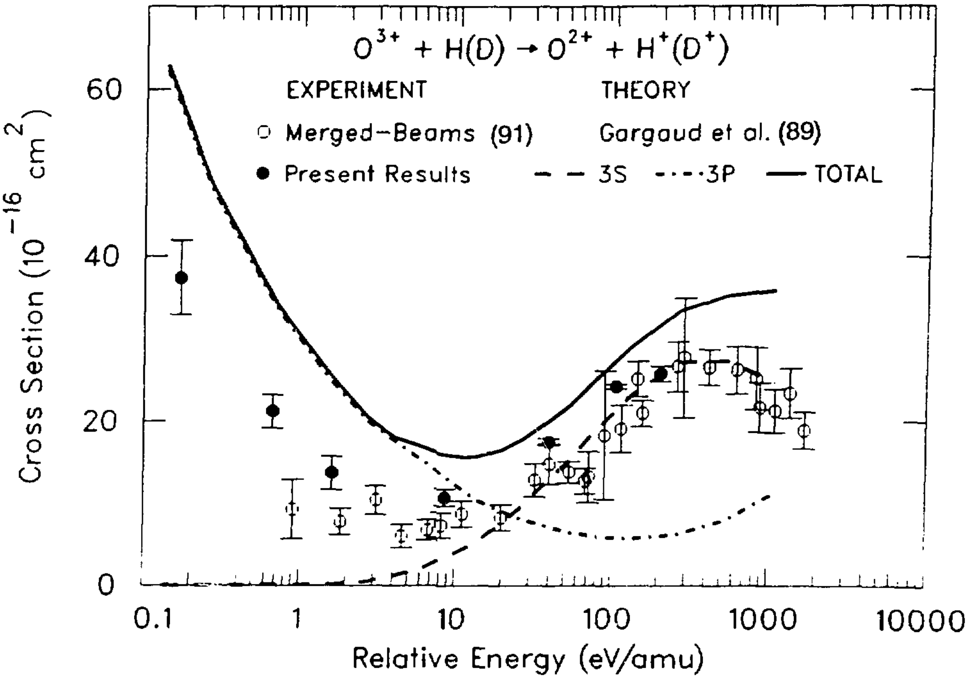


Fig. 4. Comparison of merged-beams data for O^{3+} + H(D) with theoretical calculations (Gargaud et al. 1989) for capture to the 3s and 3p states.

the whole energy range. A possibility exists that the calculations may overestimate the contribution due to capture to the 3p state. However, it should be noted that a finite fraction of the B-like O^{3+} ion beam is in the $(1s^2 2s2p^2)^4P$ metastable state, which may also contribute at least in part to this discrepancy. This fraction has been previously estimated (Phaneuf et al. 1982) to be on the order of 16% for beams from a Penning multicharged ion source.

In Fig. 5, the merged-beam measurements for O^{5+} + H(D) are presented along with other measurements and theoretical calculations (Gargaud 1987, Bottcher unpub.). Unlike the O^{3+} case, the present measurements for O^{5+} show as much as a 30% deviation from previous merged-beams results (Havener et al. 1989) in some energy regions. This discrepancy is now understood as having originated from two sources: the small fraction of excited states in the H(D) beam and the insufficient angular collection of the $H^+(D^+)$ products in the previous apparatus between 1 and 10 eV/amu. It has been observed experimentally that for collision energies less than about 50 eV/amu, the collision-energy-dependent beam-beam signal due to the estimated 0.1% of the H beam which was in excited states can account for a significant fraction of the measured signal. In the first merged-beams measurements, this contribution was not subtracted from the measured signal and therefore resulted in artificially high cross section values below 50 eV/amu.

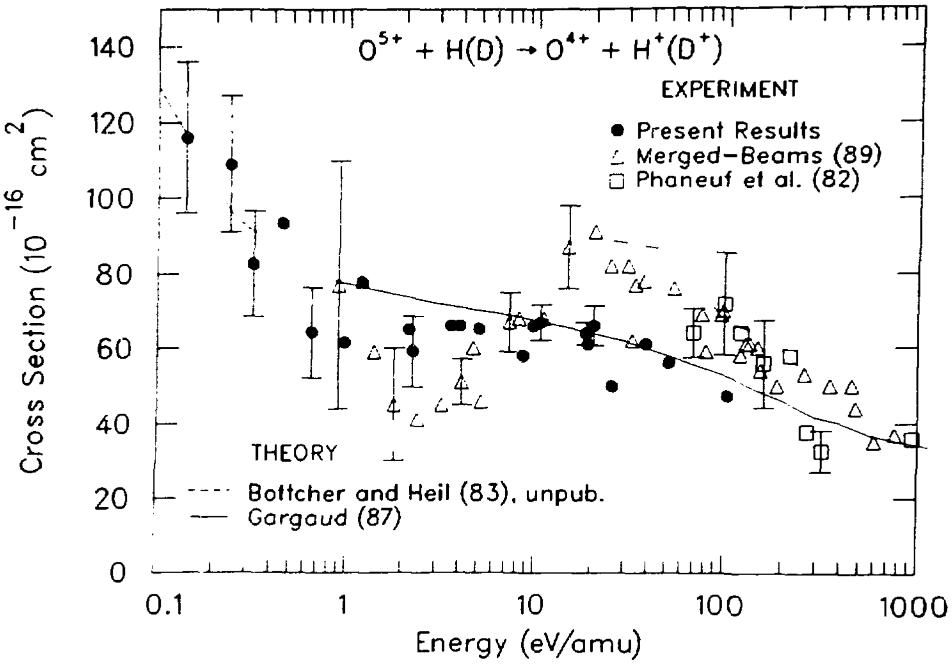
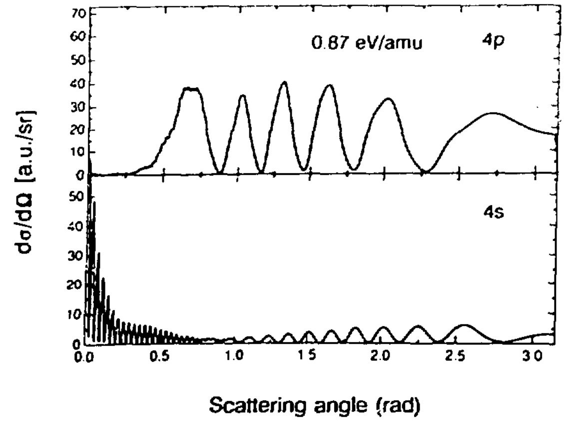


Fig. 5. Comparison of merged-beams data for O^{5+} + H(D) with other measurements and theoretical calculations.

The previous O^{5+} measurements also show a distinct minimum in the cross section between 1 and 10 eV/amu. For this energy range, the calculations predict that the total capture cross section consisted of a decreasing contribution from capture to the 4s state and an approximately equally increasing contribution from 4p capture. As reported recently (Andersson et al. 1991), no reasonable variation of theoretical parameters was successful in bringing theory into agreement at a few eV/amu without destroying the good agreement with the measurements at energies greater than 20 eV/amu. However, differential-cross-section calculations, as shown in Fig. 6, show that while the angular scattering of the 4s capture channel is forwardly peaked, the capture to the 4p state results in large scattering angles in the center-of-mass frame. A detailed

Fig. 6. Differential cross sections (Andersson et al. 1991) for the 4s and 4p electron capture channels in the center-of-mass frame for the O^{5+} + H system at 0.87 eV/amu collision energy.



analysis of the angular collection of the experiment versus angular scattering has been performed by Andersson et al. (1991). The angular collection of the present apparatus which is at least 2.3° in the laboratory frame, is sufficient to guarantee no significant loss of signal. Indeed, as can be seen in Fig. 5, the present O^{5+} measurements agree well with the calculations of Gargaud. The sharp increase in the cross section below 1 eV/amu is attributed to a trajectory-enhanced capture into the 4p state. The degree of this enhancement is estimated using simple Landau-Zener calculations and is discussed in the next section.

5. TRAJECTORY EFFECTS AT LOW ENERGIES

As the ion and neutral approach, a dipole is induced in the neutral atom, causing an attractive force between them. The resultant interaction potential is given by

$$V(r) = -\alpha \, q^2/2r^4 \qquad (1)$$

where α is the polarizability of H, q is the charge of the ion, and r is the internuclear separation. For sufficiently large q and for collision energies below 1 eV/amu the attraction is strong enough to significantly modify the trajectories of the reactants and thereby possibly affect the total capture cross section. Indeed, the simple classical orbiting model (Gioumousis and Stevenson 1958), which uses a straightforward geometrical interpretation of orbits that decay into a reaction sphere, predicts a strong 1/v increase in the cross section at these low energies.

To obtain an estimate of such trajectory effects, Landau-Zener calculations were used to determine the energy dependence of the cross section with and without the ion-induced dipole attraction taken into account. The capture cross section is calculated by integrating the total transition probability, 2p(1-p), over the impact parameter b,

$$\sigma = 2\pi \int_0^{b_{max}} 2p(1-p) \, b db \qquad (2)$$

where p is the Landau-Zener probability given by

$$p = \exp\left(-\frac{2\pi\Delta^2}{v_r\,|H'_{11} - H'_{22}|}\right) \tag{3}$$

Δ is the energy splitting at the avoided crossing, R_c; v_r is the radial component of the collision velocity; and $|H'_{11} - H'_{22}|$ is the difference in slopes of the diabatic potential energy curves at the crossing.

Since the crossings occur at relatively large internuclear separations, the potential energy, V, of the initial state can be approximated by that due to the ion-induced dipole interaction [Eq. (1)] and the final-state potential energy by (q-1)/r (a.u.). Δ is estimated by matching, for a specific state, the maximum in the Landau-Zener cross section to the corresponding maximum in the full quantal calculations. Having obtained Δ by normalization in this way, Landau-Zener calculations for other final states which cross the initial state at different internuclear separations R'_c could be performed by estimating the appropriate energy splitting using an $R_c^2 e^{-R_c}$ scaling (Butler and Dalgarno 1980). The Landau-Zener cross sections could be evaluated with straight-line or with trajectories modified by the ion-induced dipole potential (polarization interaction) by straightforward modifications to H_{11}, b_{max} and v_r. The polarization interaction results in the reactant trajectories with impact parameters larger than R_c being able to access the avoided crossing where capture occurs. In Eq. (2) the integrand thus becomes

$$b_{max} = R_c\sqrt{1 - V/E} \tag{4}$$

where E is the collision energy. Also the radial velocity, v_r, used in Eq. (3), is modified along the incident trajectory and is given by

$$v_r = v_0\sqrt{1 - b^2/r^2} \tag{5}$$

where v_0 is the initial collision velocity.

Figure 7a shows the results for O^{3+} + D. The Landau-Zener parameters were estimated by normalization at the calculated (Gargaud et al. 1989) cross section maximum for capture to 3s. As may be seen in the figure, the energy dependence for capture to 3s and for capture to 3p agree qualitatively with the full quantal calculations of Gargaud et al. (1989) (see Fig. 4). The Landau-Zener estimates are plotted with and without the effect of the polarization interaction. Note that only for capture to 3p at the lowest energies is there any enhancement due to the interaction. Figure 7b shows similar calculations for O^{5+} + D, where the ion-induced polarization attraction is stronger due to the increased charge. For O^{5+}, capture to the 4s shows only a small enhancement at the low energies, while capture to the dominant 4p channel shows a significant enhancement that increases toward lower energies. Capture to the 4d (not shown) shows a similar enhancement but only accounts for less than 20% of the total cross section. At an energy of 0.1 eV, the enhancement in the 4p accounts for 30% of the 4p capture cross section. The sharp increase at low energies in the measured cross section can be interpreted as evidence of trajectory effects due to the ion-induced dipole attraction for this system.

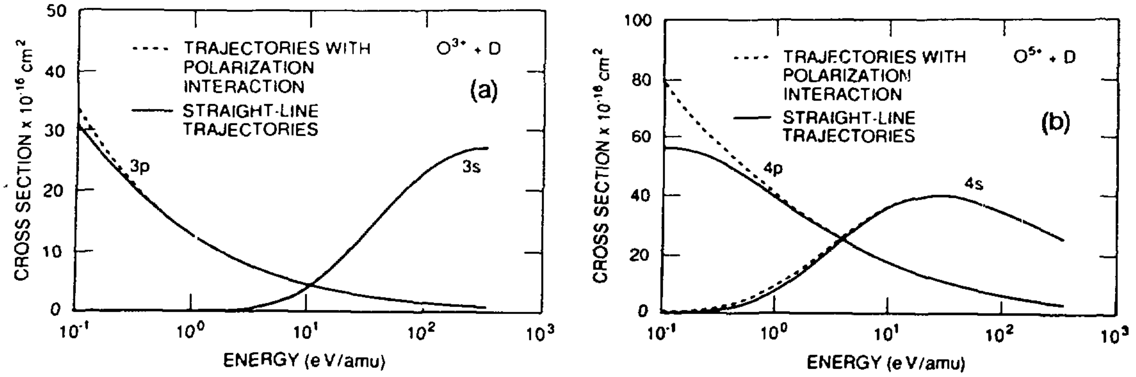


Fig. 7. Landau-Zener calculations for the O^{3+} + D system(a) and O^{5+} + D system (b) for capture to specific states (as labelled). Both straight-line (solid line) and trajectories with the polarization interaction (dashed line) are shown.

Until now we have not made a distinction between measurements with atomic hydrogen or the heavier isotope deuterium. Generally in our past measurements the heavier isotope was used both to decrease the angular scattering after the reaction, and to increase the angular collection of the apparatus in the center-of-mass frame. Detailed angular scattering estimates (Andersson et al. 1991) show that for measurements with a 6-keV-H or 8-keV beam, the angular collection of the present apparatus is sufficient below 1 eV/amu to collect all the signal. To obtain direct evidence for the enhancement in the cross section due to trajectory effects, we are in the process of performing measurements with both H and D below 1 eV/amu for O^{5+}. The expected enhancement for these different systems has been estimated using Landau-Zener calculations for capture to the 4p state and is shown in Fig. 8. In the figure the cross section is calculated for an H or D projectile, with and without trajectory effects. The difference in enhancement at 0.1 eV/amu between H and D is estimated to be on the order of 20 x 10^{-16} cm^{-2}, or 20% of the total cross section with H. The full quantal calculations (Gargaud 1987) for the O^{5+} + H system have also been performed for O^{5+} + D for energies as low as 0.87 eV/amu. At this energy, though, the enhancement amounts only to a few percent in the total capture cross section, in agreement with the Landau-Zener estimate.

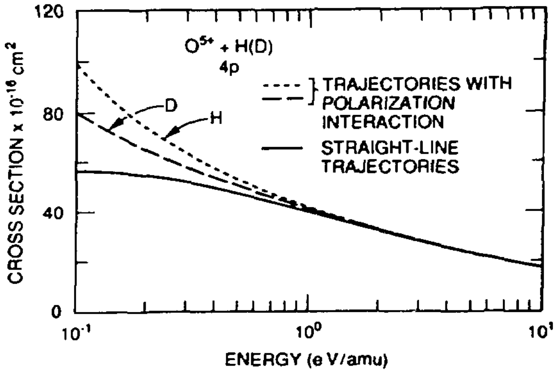


Fig. 8. Landau-Zener 4p cross section calculations for the O^{5+} + H (short-dashed line) and D (long-dashed line) collision systems with trajectories modified by polarization effects. Also shown is the calculation due to straight-line trajectories for H or D (solid line).

6. CONCLUSIONS

The merged-beams method now makes possible absolute total electron-capture cross section measurements in the energy range between 0.1 and 1000 eV/amu for multicharged ions colliding with ground-state H or D atoms. Comparison with state-of-the-art theory over such a large energy range permits a systematic evaluation of both experiment and theory and can provide further insights into collision mechanisms and dynamics. New measurements for O^{3+} have observed the predicted rise in the cross section due to capture to the 3p state below 1 eV/amu. Detailed differential cross section calculations have predicted surprisingly large angular scattering for the O^{5+} system. An upgrade of the merged-beams apparatus to accommodate such scattering has resolved the discrepancy between measurement and theory for this system. Landau-Zener estimates for the cross section indicate that trajectory effects may be significant for the O^{5+} + D system at low energies. Comparison with measurements planned for O^{5+} + H should be capable of directly probing such enhancements.

7. ACKNOWLEDGEMENTS

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