CONF-901116--15

Electron Capture by Multicharged Ions from Hydrogen Atoms at eV Energies

C. C. Havener, M. P. Nesnidal, M. R. Porter and R. A. Phaneuf

Oak Ridge National Laboratory, Oak Ridge TN, 37831-6372, USA

CONF-901116--15

DE91 001214

To quantitatively study electron capture during collisions of multiply charged ions with neutral atoms at near-thermal energies, keV-energy multicharged ion beams are merged with ground-state beams of H or D atoms of chosen velocity such that collisions in the relative energy range 1-1000 eV/amu result. Recent data for O^{3+}, O^{4+} + H(D) are presented and compared with theoretical predictions. Recently completed modifications to the apparatus are described that will provide a significant improvement in signal-to-background and angular collection. These improvements will allow measurements to be extended to lower energies, where effects due to the ion-induced dipole attraction may be evident.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

"The submitted manuscript has been authored by a contractor of the U.S. Government under contract DE-AC05-84OR21400. Accordingly, the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes."

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

1. Introduction

Only very limited data are available for electron capture by multicharged ions colliding with neutral atoms in the velocity range ($v < 0.1$ a.u.). In such low-velocity collisions, cross sections do not obey any simple scaling law. 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. Calculations in this regime require involved theoretical approaches which often disagree at these low energies. At sufficiently low collision velocities, the attractive force due to the ion-induced dipole or polarization interaction between reactants is strong enough to cause collision trajectories to sample internuclear separations which are smaller than the impact parameter, and may thereby lead to an enhancement of the cross section for ion-neutral reactions. Published merged-beams experimental data from this laboratory for O^{5+} + H(D) collisions[1] are suggestive of such an enhancement, although no such evidence was found in subsequent measurements of N^{3+}, N^{4+}, and N^{5+} ions.[2]. At these low energies, there has been speculation[3,4] that the ion-induced dipole attraction between the reactants may cause orbiting trajectories[5]. A rich resonance structure attributed to the quasi-bound states associated with the classical phenomenon of orbiting trajectories has been predicted in cross section calculations[6] for N^{3+} + H at collision energies below 0.1 eV.

In order to extend these studies of low energy ion-atom collisions, measurements have been performed for O^{3+}, O^{4+} + H(D) and are presented here. Previous measurements[7] above 30 eV/amu for O^{3+} were not able to access the 1-10 eV/amu energy range where factor of three differences in theoretical calculations[8,9] exist. There is particular astrophysical interest[8] in the O^{3+} + H system, since spectral line emission from states populated by charge transfer furnishes direct information on the ionization structure of gaseous nebulae. Recent fully quantal calculations[10]

for O^{4+} + H and Si^{4+} + H investigate the large effect that the nature of the ionic core has on these low-energy collisions.

2. Experimental Technique

Total electron-capture cross sections are measured at these low energies using the merged-beams technique. This technique permits measurements to be made over a wide range of interaction energies, provides a large angular collection of reaction products, and produces independently absolute cross-section measurements. In this technique a fast (30-80 keV) multicharged ion beam is merged electrostatically with a 6-9 keV neutral beam travelling at nearly the same speed, resulting in a relative velocity of the two beams which can be "tuned" over a large range. Figure 1 is a simplified schematic of the ion-atom merged-beams apparatus. Multicharged ion beams (X^{q+}) of necessary intensity are produced by the Oak Ridge National Laboratory Electron Cyclotron Resonance (ORNL-ECR) multicharged ion source. A ground-state beam of H or D atoms is produced by passing a 6-9 keV beam of H^- or D^- ions through the optical cavity of a 1.06-μm Nd:YAG laser. The merged-beams interact for a distance of 80.8 cm. The overlap of the two beams, which are a few mm in diameter and have a mean divergence of less than 0.5°, is measured along the merged path at four positions by horizontal and vertical profile monitors. These profiles are used in a calculation of the beam-beam overlap (form factor). The $H(D)^+$ products of the collision (signal) are magnetically separated from the primary beams and detected by a channel electron multiplier operating in pulse counting mode. The primary neutral H(D) beam is measured by secondary electron emisson from a stainless steel plate. Absolute calibration of this neutral detector is performed in situ. A fast two-beam chopping scheme is used to separate signal from background. The background (typically 10 kHz) is due mainly to stripping of H(D) atoms on residual gas. To keep

this background to a minimum, pressure in the merge-section is maintained as low as possible, typically 2 x 10^{-10} Torr or less during measurements. Since the number of ion-atom collisions is proportional to the relative velocity of the beams, the signal-to-background ratio determined the lowest energies at which measurements could be made. The lower-energy limit is approximately 0.2 eV/amu, and is a result of the finite beam divergence.

An important advantage of the merged-beams technique is the potentially large angular collection of the reaction products. The low energy electron capture collisions under study are exoergic, and, since both products are positively charged, they can undergo significant angular scattering in the center-of-mass frame[11]. An average angular collection in the laboratory frame along the 80.8 cm merge path was determined by detailed trajectory calculations[1] to be 1.8°. Angular scattering in the center-of-mass frame becomes significantly compressed in the laboratory frame where the $H(D)^+$ product ion is collected. This compression increases as collision energies decrease. In fact, for O^{5+} + D measurements performed with a 9 keV D beam, D^+ trajectories with up to 90° scattering in the center-of-mass frame are collected at 1 eV/amu collision energies. However, at collision energies above 1 eV/amu, even though the estimated angular collection was sufficient to collect products scattered within angles predicted by a Rutherford half-collision model[11], it was not sufficient for the larger scattering predicted in recent differential cross section calculations[12]. Further details of the apparatus used for the measurements presented here are given in Ref.[1].

3. Results & Discussion

The data for O^{3+} + H(D) and O^{4+} + H(D) are presented along with other experimental and theoretical results in Figs. 2-4. Error bars shown correspond to an

estimated uncertainty in the reproducibility of the measurements estimated with a 90% confidence limit. The absolute uncertainty in the measurements corresponds to about 12 % [1] and must be added in quadrature to the relative uncertainty. For both the O^{3+} + H(D) and O^{4+} + H(D) collisions, the merged-beams data join smoothly with other measurements 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 system (see Fig. 2), the present measurements lie significantly below the calculations of Bienstock et.al.[9] which predict that capture to the (2p3p) configuration dominates at low energies. The measurements are more consistent with the calculations of Gargaud et al.[8] which predict that capture to the 3p state dominates below 10 eV/amu while capture to the 3s state is important at the higher energies. This prediction agrees with translational spectroscopy measurements by Wilson et al.[13]. As seen in Fig. 3, within the experimental uncertainties, the present measurements are consistent throughout the whole energy range with the calculation including only capture to the 3s state. This suggests that even Gargaud et al.'s calculation may overestimate the 3p contribution by roughly a factor of two. It should be noted that a finite fraction (previously estimated[7] to be on the order of 16% for beams from a Penning multicharged ion source) 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.

In Fig. 4, the merged-beam measurements for O^{4+} + H(D) are compared with calculations of Gargaud et. al.[10] which predict that for this system capture to the 3d state dominates, especially below 200 eV/amu. In these calculations the effect of rotational coupling involving the $3d\sigma$, $3d\pi$, and $3d\delta$ orbitals is considerable and sensitive to the relative energy separations of the different orbitals used in the calculation. In Figure 4, the cross section calculation is presented with and without the effect of

rotational coupling (denoted by a solid and broken line, respectively), illustrating the significant enhancement due to this effect throughout most of the energy range. As may be seen in the Figure, for energies above 200 eV/amu, previous measurements of Phaneuf et al.[7] were somewhat lower than the calculation (which includes the enhancement due to rotational coupling), but suggest agreement at energies below 200 eV/amu. The present merged-beams measurements, though, are significantly lower than the calculation over the whole energy range. As can be seen in the figure, this discrepancy with theory is on the order of the predicted enhancement due to rotational coupling. Of course, this discrepancy could also be caused by an overestimation of the 3d contribution in the calculation.

Previously noted disagreements between measurements[1] and calculations[14] for the O^{5+} + H(D) system have led to calculations[12] of the angular scattering in an attempt to reconcile striking differences between calculations and measurements for this system in the collision energy between 2 and 10 eV/amu. The differential cross section calculations[12] suggest that for capture to some states (e.g., to the 3p state) in this energy range there may be significant angular scattering, somewhat beyond the estimated collection of the apparatus. In response to this, modifications to the apparatus (shortening the merged-path to 50 cm and increasing the diameter of collection lenses and beam pipes) have been made which have resulted in the estimated angular collection to increase from an average of 1.8° to a minimum of 2.3°. This corresponds to a significant increase in the angular collection in the center-of-mass frame and, according to the recent angular scattering calculation[12], should be sufficient to resolve the differences between present theory and experiment. As a result of these modifications together with vacuum improvements in the merged-section, the signal-to-background ratio has been increased by at least a factor of three. With this large increase in sensitivity and angular collection, it should be possible to

extend measurements to lower energies and to perform measurements for ions with higher charge states. It is for such collisions that predicted effects due to the ion-induced dipole attraction may become evident in the relative collision energy range facilitated by the present technique.

4. Summary

Comparison of merged-beams measurements of O^{3+}, O^{4+} + H(D) with theory are providing insight into the low energy electron capture process. The measurements for O^{3+} + H(D) suggest that for this system, electron capture is dominated by capture to the 3s state, more so than predicted by the most recent calculations. The measurements indicate that contributions due to capture to the 3p state may have previously been overestimated. The measurements for O^{4+} + H(D) suggest that recent calculations may overestimate the enhancement due to rotational coupling. Recent modifications to the apparatus will provide a significant increase in angular collection and sensitivity at the lowest energies. It is hoped that with these improvements, new measurements will be possible that will shed light on existing discrepancies between experiment and theory for the O^{5+} + H system, and that the merged-beams technique will extend to lower energies where orbiting resonances are predicted to occur.

Acknolwedgments

This research was supported by the Division of Chemical Sciences, U.S. Department of Energy, under Contract No. DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc. M. P. Nesnidal was sponsored by the Great Lakes Colleges Association/Associated Colleges of the Midwest. M. R. Porter was sponsored by Science Alliance, a state of Tennessee Center of Excellence, through the University of Tennessee.

REFERENCES

[1] C. C. Havener, M. S. Huq, H. F. Krause, P. A. Schulz, and R. A. Phaneuf. *Phys. Rev. A*, 39:1725, 1989.

[2] M. S. Huq, C. C. Havener, and R. A. Phaneuf. *Phys. Rev. A*, 40:1811, 1989.

[3] C. Bottcher and T. G. Heil. *Chem. Phys. Lett.*, 86:506, 1982.

[4] R. E. Olson. *in Electronic and Atomic Collisions, N. Oda and K. Takayanagi, eds.*, 391, 1980.

[5] G. Gioumousis and D. P. Stevenson. *J. Chem. Phys.*, 29:294, 1958.

[6] M. Rittby, N. Elander, E. Brandas, and A. Barany. *J. Phys. B: At. Mol. Phys.*, 17:L677, 1984.

[7] R. A. Phaneuf, I. Alvarez, F. W. Meyer, and D. H. Crandall. *Phys. Rev. A*, 26:1892, 1982.

[8] M. Gargaud, R. McCarroll, and L. Opradolce. *Astron. Astrophys.*, 208:251, 1989.

[9] S. Bienstock, T. G. Heil, and A. Dalgarno. *Phys. Rev. A*, 27:2741, 1983.

[10] M. Gargaud and R. McCarroll. *J. Phys. B: At. Mol. Opt. Phys.*, 21:513, 1988.

[11] R. E. Olson and M. Kimura. *J. Phys. B: At. Mol. Phys.*, 15:4231, 1982.

[12] L. R. Anderson, M. Gargaud, and R. McCarroll. *Abstracts of XVI ICPEAC, New York, New York*, 566, 1989.

[13] S. M. Wilson, R. W. McCullough, and H. B. Gilbody. *J. Phys. B:Atom. Mol. Opt. Phys.*, 21:1027, 1988.

[14] M. Gargaud. *Doctoral Thesis, Universite de Bordeau I*, 1987.

FIGURES

FIG. 1. Simplified schematic of the ion-neutral merged-beams apparatus.

FIG. 2. Comparison of merged-beams data for O^{3+} + H(D) with other measurements (Ref.[7]) and theoretical calculations (Refs.[9,8]). The dashed curve is from Ref.[9] and the solid curve is from Ref.[8].

FIG. 3. Comparison of merged-beams data for O^{3+} + H(D) with theoretical calculations (Ref.[8]) for capture to the 3s and 3p states.

FIG. 4. Comparison of merged-beams data for O^{4+} + H(D) with other measurements (Ref.[7]) and theoretical calculations (Ref.[10]) with (solid curve) and without (broken curve) the enhancement due to rotational coupling.

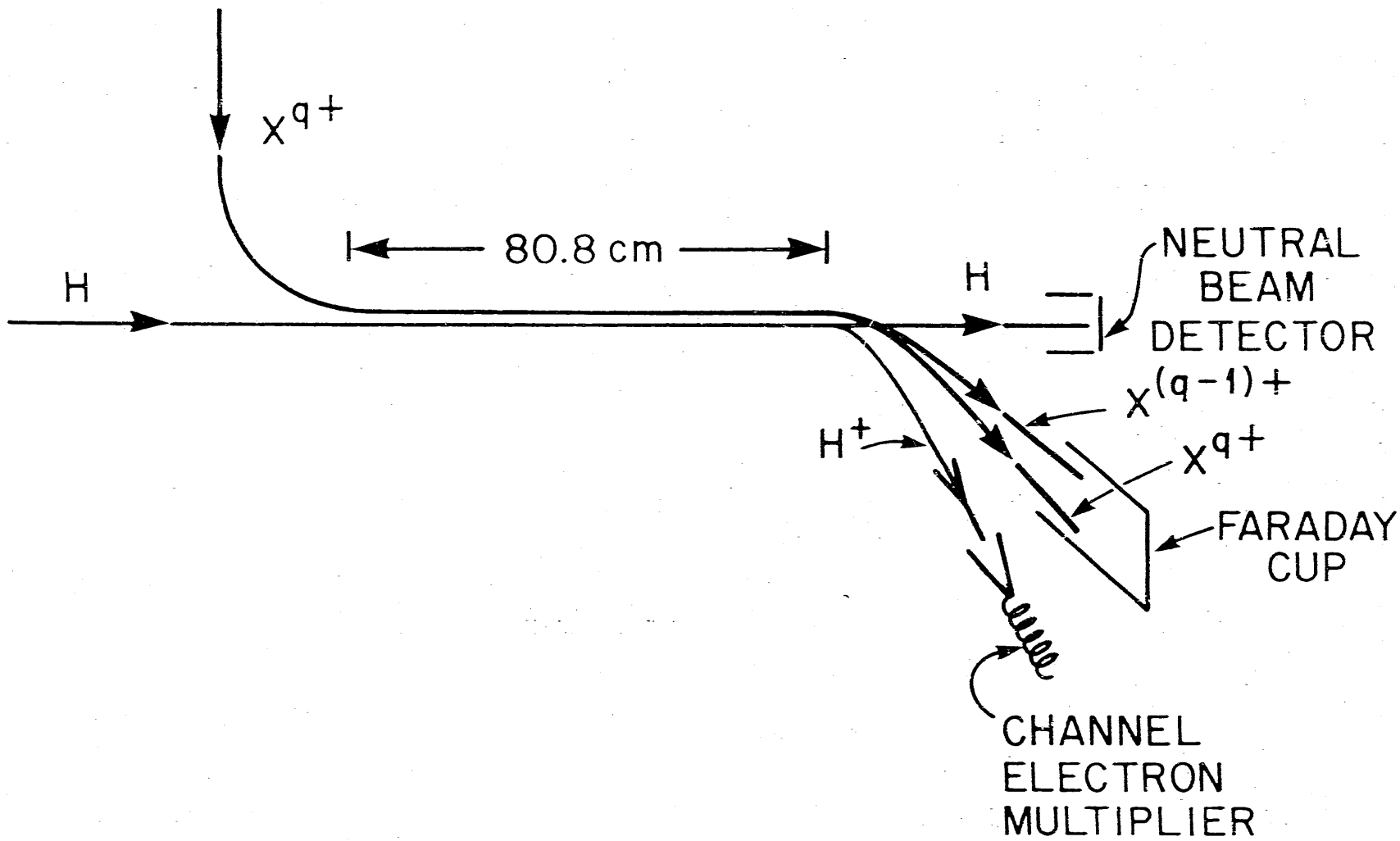
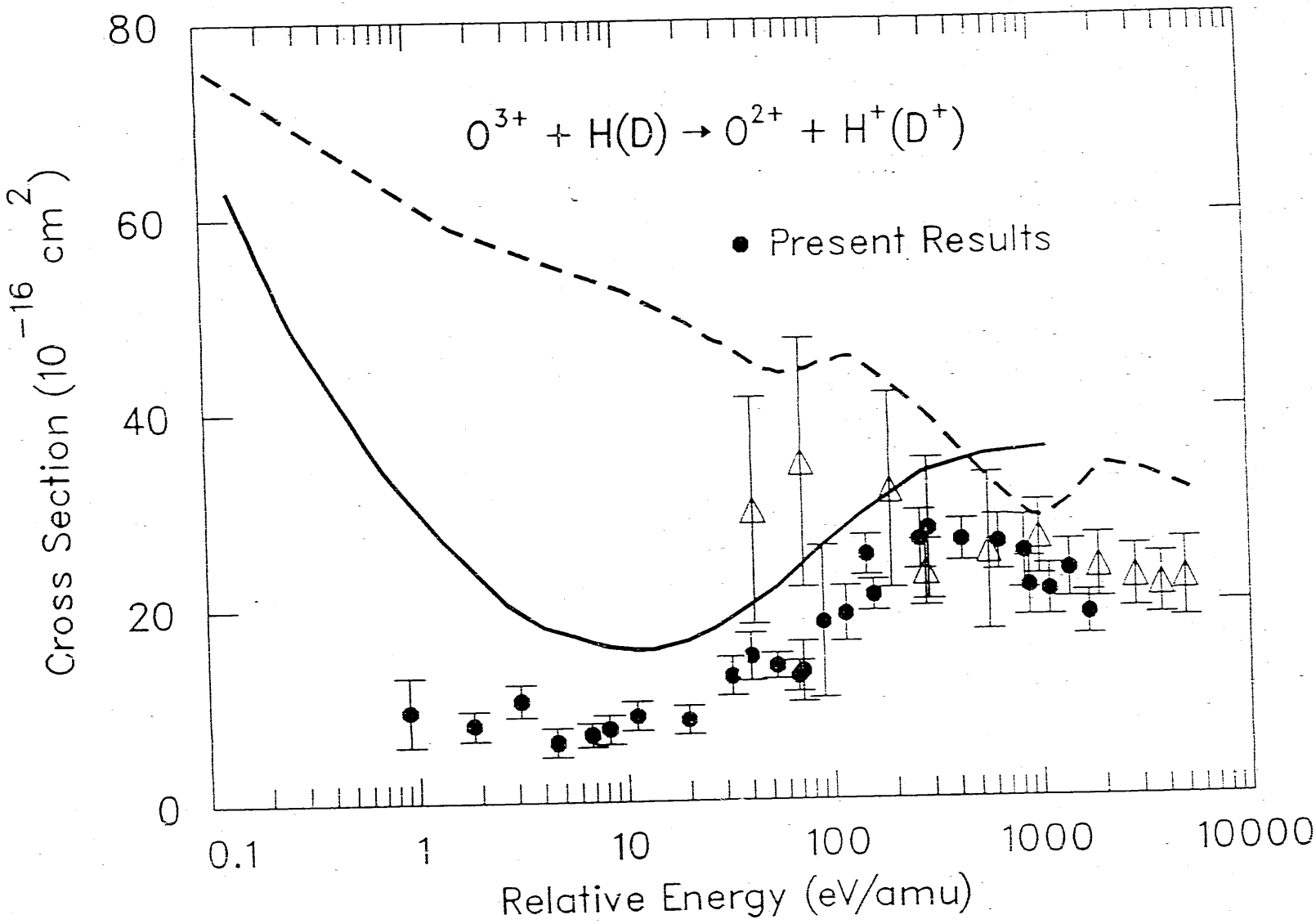
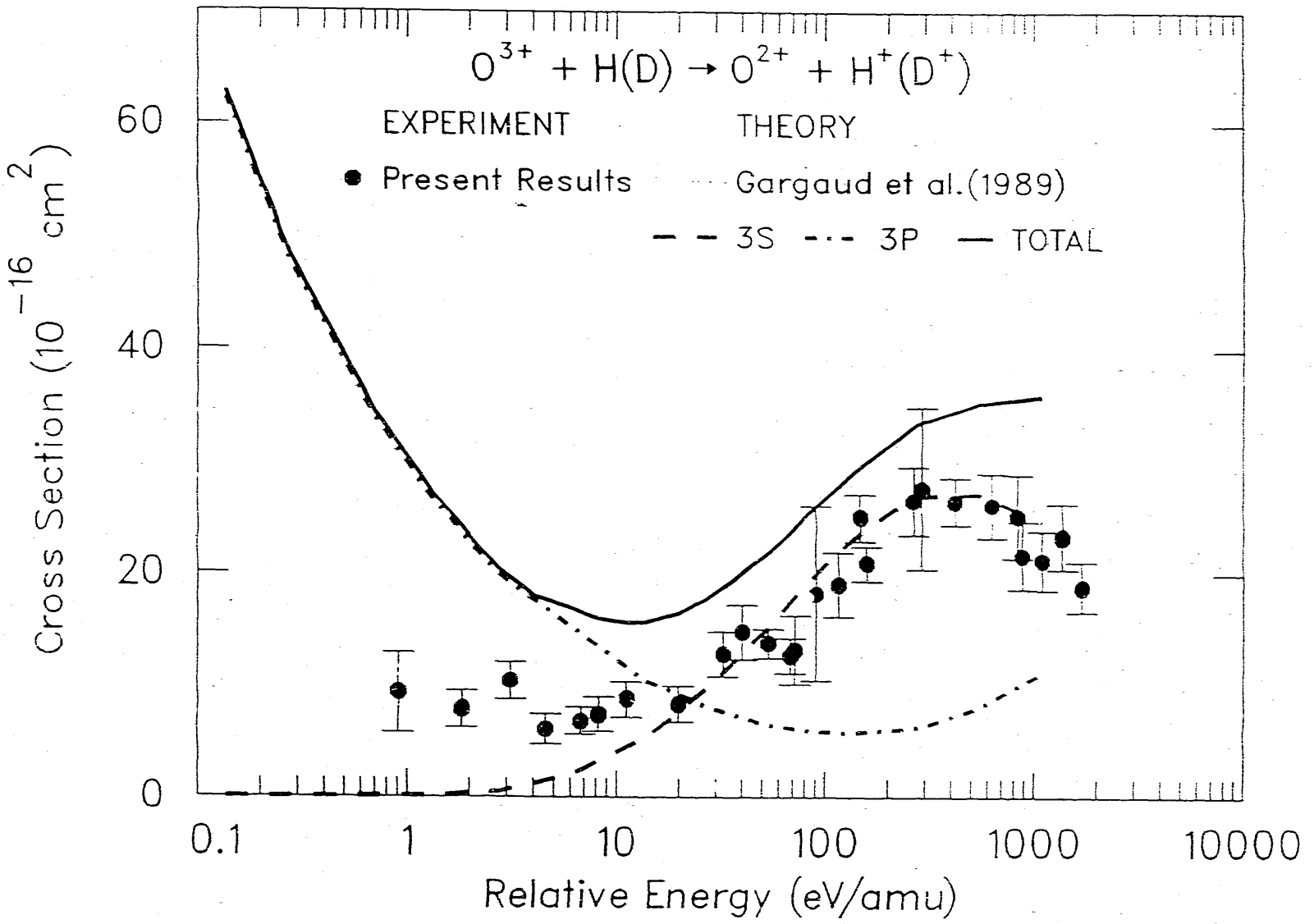
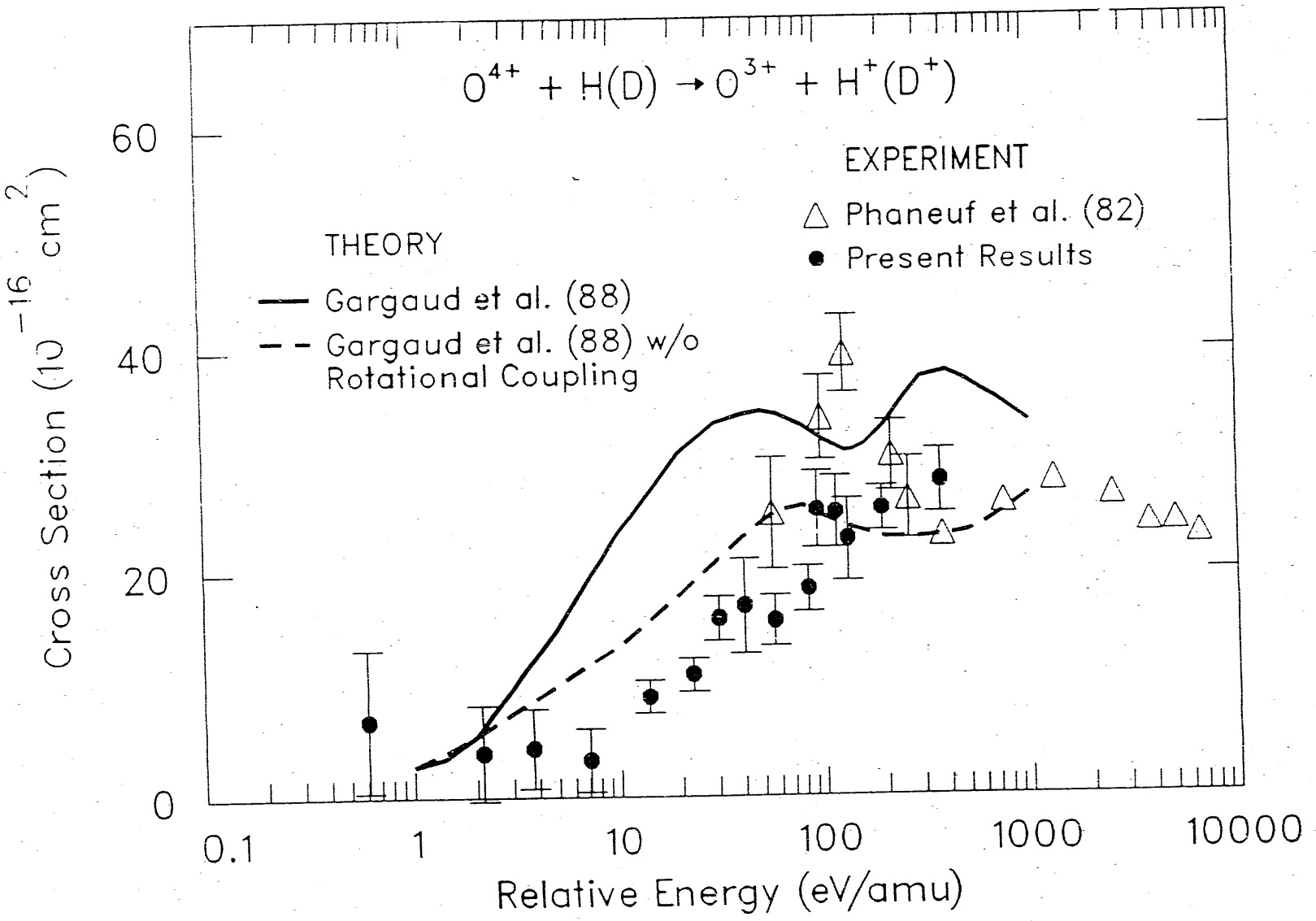

X^{q+}
80.8 cm
H
H
NEUTRAL BEAM DETECTOR
$X^{(q-1)+}$
H^+
X^{q+}
FARADAY CUP
CHANNEL ELECTRON MULTIPLIER

Fig. 1


$O^{3+} + H(D) \rightarrow O^{2+} + H^{+}(D^{+})$
Present Results
Cross Section (10^{-16} cm^{2})
80
60
40
20
0
0.1
1
10
100
1000
10000
Relative Energy (eV/amu)


$O^{3+} + H(D) \rightarrow O^{2+} + H^{+}(D^{+})$
EXPERIMENT
Present Results
THEORY
Gargaud et al.(1989)
3S
3P
TOTAL
Cross Section (10^{-16} cm^{2})
60
40
20
0
0.1
1
10
100
1000
10000
Relative Energy (eV/amu)



$O^{4+} + H(D) \rightarrow O^{3+} + H^{+}(D^{+})$
EXPERIMENT
Phaneuf et al. (82)
Present Results
THEORY
Gargaud et al. (88)
Gargaud et al. (88) w/o Rotational Coupling
Cross Section (10^{-16} cm^2)
0
20
40
60
0.1
1
10
100
1000
10000
Relative Energy (eV/amu)

Fig. 4

-END-

DATE FILMED

11 / 05 / 90