THEORETICAL ATOMIC COLLISION PHYSICS

Final Report

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FINAL REPORT

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"Theoretical Atomic Collision Physics"

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BACKGROUND

This is the final report of research activities supported by the most recent grant to the theoretical atomic collision physics program at Rice University. This group has received funding from the Department of Energy and its predecessors -- in recent years the DOE Office of Basic Energy Sciences, Division of Chemical Sciences -- for over two decades. (Please see the "Background and Introduction" of our latest proposal for a brief description of our earlier research.) The Principal Investigator, many graduate and undergraduate students, postdoctoral research associates and collaborators have benefited from this generous support, and in return, it is hoped that the knowledge gained from the research has been of value to energy research as well as advancing the understanding of atomic collision physics. The Rice group has been fortunate to work with many scientists from other institutions. Dr. Mineo Kimura (Argonne National Laboratory, at the time this research was carried out) held an adjunct faculty appointment at Rice and was a particularly important collaborator.

For this most recent 3-year grant, the focus of the research has been the study of excitation and charge-transfer in atom-atom and ion-atom collisions. Emphasis has been placed on low-velocity collision processes involving initially excited atoms, including "low Rydberg" atoms.

Two particular objectives of this research were to examine the dependence of state-changing collision cross sections and other observables on the orientation and alignment of the initial excited orbitals and to look for "intracollisional interference" effects, speculated to arise from spatially separated interactions during a collision involving a highly excited atom. A number of radiative and non-radiative charge transfer studies involving ground-state atoms and ions were continued from the previous grant period. Because of several unanticipated personnel changes, including the departure

of the original Principal Investigator for a position in government, we were unable to carry out some of the proposed research, including the studies of collisions involving negative ions and electron-attaching atoms and the development of new theoretical techniques for handling the "strong coupling" regime. These remain challenging and fundamentally important research topics.

RESEARCH HIGHLIGHTS

Since more detail can be found in previous annual progress reports, abstracts of the annual workshops, and in the publications, only a very brief highlights of the research are given below:

Spherical low-Rydberg atom collisions

Mechanisms for excitation and deexcitation of low-Rydberg Na(ns, $6 < n < 10$) atoms in thermal energy collisions with He have been described and cross sections calculated for a range of collision velocities relevant to experimental measurements carried out at 425K. (A semi-classical MO approach, including electron-translation effects, was used.) A propensity rule $ns \rightarrow n'f$ is indicated by the calculations. This work is an extension of earlier research carried out in this group; improvements in methods and computational techniques led to corrections to earlier published results (see Kumar et.al. 1989, below) and new findings. Similar studies of Na(8s) collisions, carried out at higher velocity resolution, show "intracollisional interference" effects in the form of Stueckelberg oscillations arising from avoided (adiabatic) curve crossings. In a two-state fully quantum mechanical calculation, we observed quasi-vibrational resonances, associated with broad minima in some of the adiabatic potential energy curves that define the system wave function.

Non-spherical low-Rydberg atom collisions

Our studies of spherical atoms have been carried over to non-spherical Na(np) atoms in thermal energy collisions with He. The cross sections have been calculated as a function of alignment of the initial p orbital relative to the initial relative velocity. It has been shown that the Stueckelberg oscillations which occur in the cross sections are sensitive functions of the orbital alignment. The dependence can be explained in terms of the relative contributions of σ and π adiabatic collision states.

Alignment effects in collisions of Na(3p) with He⁺ ions

Cross sections for electron capture in collisions of excited Na(3p) atoms with He⁺ ions have been calculated at relatively low collision velocities 0.1 to 7 keV/u. (A semi-classical MO approach, including electron-translation effects, was used.) The cross sections are found to be similar in magnitude to those for the ground-state atom Na(3s), in agreement with experiment. Charge-transfer cross sections as well as final orientation and alignment parameters for He(2¹,3P) have been calculated as functions of the initial alignment angle for the Na(3p) orbital. The sensitivity can be understood in terms of the relative contribution of σ and π adiabatic collision states, as was found with the low-Rydberg studies.

Near-resonant electron capture at very low energies

Quantum mechanical calculations have been carried out of cross sections for the slightly non-resonant (1.3 meV energy defect) electron-capture process $^3\text{He} + ^4\text{He} \leftrightarrow ^3\text{He} + ^4\text{He}^+$ in the relative velocity range $0.0001 < v < 0.002$ au. Both cross sections exhibit sharp quasi-vibrational resonances. Rate coefficients for both forward (endothermic) and backward (exothermic) processes were calculated and compared with recent measurements. The agreement with the measurements is satisfactory, but the experimental error is fairly large for these extraordinarily difficult measurements.

Ion-atom and ion-molecule collisions, including electron capture

In collaboration with Professor Ronald Stebbings and his experimental group at Rice, we have calculated differential direct, as well as single- and double-charge transfer cross sections for several systems, including most recently collisions of He⁺⁺ with He, and H⁺ with H₂, at keV energies. These have been found to compare favorably with experiment. Independent of these studies, we have calculated cross sections for He⁺(2p) excitation through electron capture in He⁺⁺ collisions with H₂ molecules at low energies. (Much of this work was completed under the previous grant.)

Low-energy processes involving collisions of H⁺ with He, Na, K atoms

Cross sections and rate coefficients have been calculated for total scattering, momentum-transfer, and electron capture (radiative and non radiative) in collisions of H⁺ with He and for electron capture (radiative and non-radiative) in collisions of H⁺ with Na and K atoms. The rate coefficients are useful in astrophysical models. In the case of Na and K, the radiative cross

section, which falls off rapidly with energy, and the non-radiative cross section, which rises steeply with increasing energy, cross one another in the vicinity of 10eV and 3eV, for Na and K, respectively. The non-radiative cross sections exhibit Stueckelberg oscillations.

Other research

Preliminary calculations have been carried out on a number of other collision systems. Although it has not been possible to complete all these projects during the time of the grant, it is hoped that in time they will be completed and additional papers will result. Any publications arising from work supported in full or in part by the grant, will, of course acknowledge the DOE.

PUBLICATIONS SINCE 1991

(Most of the work reported in 1991 publications was completed under the earlier grant)

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