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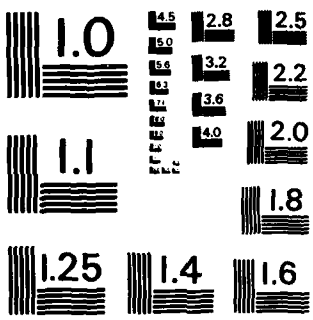

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Theoretical studies were performed on a variety of topics related to collisions involving Rydberg atoms. Progress was made towards the understanding of ion-Rydberg atom, Rydberg atom-Rydberg atom, and ground state atom-Rydberg atom collisions. A strong dc electric field was incorporated in calculations of electron capture and ionization cross sections for ion-Rydberg atom collisions. Electron capture collisions in the presence of a strong laser field were also investigated.

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FINAL REPORT
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November 28, 1984

THEORETICAL STUDIES OF RYDBERG ATOM COLLISIONS

by

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CONTENTS

ABSTRACT

During the five years of this grant, theoretical studies were performed on a variety of topics related to collisions involving Rydberg atoms. Progress was made towards the understanding of ion-Rydberg atom, Rydberg atom-Rydberg atom, and ground state atom-Rydberg atom collisions. Cross sections were calculated for electron capture, ionization, and excitation processes and parametrized in terms of atomic parameters. A strong dc electric field was also incorporated in calculations of electron capture and ionization cross sections for ion-Rydberg atom collisions and studies were made on collisions of ions with state aligned Rydberg atoms. Progress was also realized in the incorporation of strong laser fields in ion-atom collisions.

INTRODUCTION

At present, it is possible to detect single photons having wavelengths $\leq 1\ \mu m$. Conventional methods fail at longer wavelengths because the reduced quantum efficiency leads to signal-to-noise problems. However, because Rydberg atoms have large absorption cross sections for long-wavelength radiation ($> 10\ \mu m$), these atoms can be used to convert infrared and microwave photons into either visible photons or ions that can be detected with conventional techniques. The basic idea of a Rydberg atom infrared or microwave detector is to make a target of these atoms that is optically thick to the radiation to be detected at wavelength λ_o. Thus, the photons with wavelength λ_o will be absorbed in the target of Rydberg atoms in a single n, ℓ quantum state. The atoms that absorb the photons undergo a transition to a state of different parity and energy. This state can then easily be detected by taking advantage of either the different wavelength of the optical radiation, which is subsequently emitted by the atom, or the difference in the field ionization of the two states.

The first question that must be asked is: Is it possible to make an optically thick target of Rydberg atoms for long-wavelength radiation? Experimental work has demonstrated that a Rydberg atom detector for infrared and microwave radiation is, in principle, quite feasible. Moreover, it can be orders of magnitude more sensitive than other available detectors, a condition that can lead to a variety of possible applications. It is only necessary to produce a $10^8\ cm^{-3}$ density of Rydberg atoms in a parent gas density of $\sim 10^{11}\ cm^{-3}$ in order to obtain a 1-cm-long optically thick target of Rydberg atoms. This density of Rydberg atoms is now being produced with dye laser technology in many laboratories.

Because it is apparent that the detector will work in principle, it is necessary to determine its limitations. Obvious problems are collisional processes that either destroy the population of the Rydberg atom in a specific (n,ℓ) electronic level, thereby reducing the steady state population density, or induce the same transition that is used to observe the long-wavelength radiation, thereby producing spurious signals. For either case, the collision mean free time must be comparable to the radiative lifetime of the Rydberg atom, which has been observed to be empirically given by

$$\tau = \frac{n^3}{2.4 \times 10^{+8}} \text{ sec.} \qquad (1)$$

As an example of the magnitudes of the collisional deactivation cross sections that are necessary to compete with the radiative lifetime, one can use a system in which the Rydberg atom is in the $n = 20$ level. From Eq. (1), one finds that the Rydberg atom has a radiative lifetime of 3.3×10^{-5} sec. If a Rydberg atom density of 10^8 cm^{-3} in a ground state density of 10^{11} cm^{-3} is realistically assumed, it is easy to show that collisional deactivation would be comparable to the radiative process if the Rydberg atom-ground state atom deactivation cross section were on the order of 10^{-11} cm^2. Because the geometric cross section for a Rydberg atom is $\pi n^4 a_o^2$, for a Rydberg atom that is in the $n = 20$ level the deactivation cross section conceivably could be comparable to the geometric cross section, or $\sim 1 \times 10^{-11}$ cm^2. Such a cross section is extremely plausible, especially for alkali atoms, which have large electron scattering lengths and high dipole polarizabilities and can thus easily induce the Rydberg electron to other close-lying electronic levels. It appears, therefore, that collisional processes must be seriously considered in the design of a Rydberg atom long-wavelength photon detector.

During the last five years, we completed a series of theoretical studies directed towards Rydberg atom collisional processes. This work includes ion-Rydberg atom, Rydberg atom-Rydberg atom, and ground state atom-Rydberg atom scattering along with collisions in strong electric fields. The work has been very productive and has resulted in the publication of two Physical Review Letters, six major Physical Review A papers, and a review chapter in the book Rydberg States of Atoms and Molecules. We thank the Office of Naval Research for their support in these studies. The results of the research are briefly described in the next section.

RESEARCH PROGRESS REPORT

Early in the grant period, we have focused our attention on several Rydberg atom collision problems. Originally, our theoretical techniques were directed toward ionization collisions involving two Rydberg atoms in the same principal quantum numbers n:

$$A^{***}(n) + A^{**}(n) \rightarrow A^{+} + \ldots \quad . \qquad (2)$$

A four-body classical-trajectory Monte Carlo (CTMC) code was written and applied to Reaction (2) for collision velocities, v/v_e, from 10^{-2} to 10^{1} (note: v_e = 1/n a.u. = 2.2 x 10^{8}/n cm/s). It is of particular interest that the ionization cross sections for (2) were approximately an order of magnitude larger than the geometric value $\pi\, n^4 a_o^{\,2}$ at thermal energies. Also, the CTMC code inherently predicted the importance of the dipole-induced dipole forces at low velocities and showed that the cross section increases as the velocity decreased, as $v^{-2/3}$. The work was published under the title "Ionization Cross Sections by Rydberg-Atom-Rydberg-Atom Collisions" in Phys. Rev. Lett., 43, 126 (1979). Just recently, Wing's group confirmed the validity of the calculations in a report of experimental studies at the 1984 DEAP meeting.

The CMTC code was also applied to collisions of ions with Rydberg atoms in the v/v_e range of 1 to 10. Both electron capture,

$$A^{+q} + B(n) \rightarrow A^{+q}(n') + B^{+} \quad , \qquad (3)$$

and ionization,

$$A^{+q} + B(n) \rightarrow A^{+q} + B^{+} + e^{-} \quad . \qquad (4)$$

cross sections were calculated. The cross sections were conveniently presented in terms of simple analytical expressions containing the collision velocity, incident ion charge state, and electronic level of the Rydberg atom. The most interesting aspect of the calculations was the determination of the $A^{+q-1}(n')$ product ion distributions after the electron capture reaction (3). The net result was that the Rydberg atoms's electron after capture tries to preserve its original dimensions and orbital energy. Consequently, the most probable final state can be expressed by

$$n' = nq^{3/4} \quad . \tag{5}$$

The work was published under the title "Ion-Rydberg Atom Collision Cross Sections" in J. Phys. B, 13, 483 (1980).

In the area of ion-Rydberg atom excitation transfer collisions,

$$A^{+q} + B(n) \rightarrow A^{+q} + B(n') \quad , \tag{6}$$

we recently completed benchmark calculations on the $N^{+3} + H^{**}(n)$ system in the range of n = 9 to 24. These calculations were motivated by the work of Kim and Meyer [Phys. Rev. Lett. 44, 1047 (1980)] who ignored the excitation transfer process in the analysis of their data and incorrectly thought that their observed cross sections were due to the ionization process. A paper that is concerned with the excitation transfer process, "Excitation Transfer in Ion-Rydberg Atom Collisions," was published in Phys. Rev. A 23, 3338 (1981). It was shown the collisionally formed high-lying Rydberg levels lead to fake signals occasioned by Stark ionization of the beam by deflector plates. Thus, the cross section dependences predicted in theoretical papers by several different authors are not in doubt.

A major portion of our time during 1980 and 1981 was spent in writing a review on theoretical methods and results as applied to Rydberg atom collisions. The review covers "ℓ-changing" collisions between Rydberg atoms and ground state neutral atoms, associative ionization processes between Rydberg and ground state atoms, ion-Rydberg atom collisions, and Rydberg atom-Rydberg atom processes. It has been included as a chapter in the book entitled Rydberg States of Atoms and Molecules, edited by R. Stebbings and F. Dunning and published by Cambridge University Press in 1983. Our chapter is entitled: "Theoretical Approaches to Low Energy Collisions of Rydberg Atoms with Atoms and Ions" and is co-authored with A. P. Hickman and J. Pascale.

An interesting research topic for which we have completed calculations is the effect of strong dc electric fields on ion-Rydberg atom ionization and electron capture cross sections. This is a problem that was extremely pertinent to a far infrared photon detector that is based on Rydberg atoms, with the use of field ionization to determine the product state. In fact, because the cross sections are so highly dependent on the magnitude of the electric field, these cross sections probably determine the efficiency of the field ionization in the detector.

The electric field calculations were accomplished by the CTMC method. Model problems on Rydberg atoms in the n = 10 and n = 20 states were solved, and the calculated cross sections were parameterized in terms of the quantum levels of the Rydberg atom, the electric field strength, and the collision velocity. Interestingly, the electric field caused the cross sections for electron capture to decrease by up to fourfold, while the ionization values increased by up to two orders of magnitude. A paper entitled "Ion Collisions with Rydberg Atoms in Strong Electric Fields," which is co-authored with A. D. Mackellar, was published in Phys. Rev. Lett. 46, 1451 (1981).

The effects of core interactions in "ℓ-changing" collisions of Rydberg atoms with rare gases was investigated theoretically using coupled-channel and Born approximation calculations. For low Rydberg levels, $n \leq 10$, the core interactions were found to be negligible. The work was performed by A. Hickman and published in J. Phys. B 14, L419 (1981) under the title "The Effect of Core Interactions in ℓ-Mixing Collisions of Rydberg Atoms with Rare Gases".

We continued our investigations of ion-Rydberg atom collisions to include state-selected Rydberg atoms. Our calculations are on collisions of protons with Rydberg atoms in the $n = 10$ $\ell = 9$ level with $m_\ell = 0$ and 9. The ionization cross sections were found to be relatively insensitive to changes in the m_ℓ levels; however, the electron capture cross section showed considerable enhancement if the Rydberg electron was oriented in a plane parallel to the direction of the incident projectile. The result was unexpected, but following the time-evolution of individual trajectories revealed the reason. It was simply a case of velocity matching between the incident proton and the target electron which lead to the enhanced cross section. The paper describing this research was published in Phys. Rev. A 28, 2526 (1983) with the title "Ion Scattering from State-Selected Rydberg Atoms" by G. Kohring, A. Wetmore and R. Olson.

As a general piece of work to improve our theoretical tools, we have incorporated the calculation of differential cross sections into the classical-trajectory Monte Carlo method. The procedure was benchmarked against $H^+ + H$ scattering cross sections in the intermediate energy region. The tests were completely successful and have led to a major paper prepared by R. E. Olson entitled "Ion-Atom Differential Cross Sections at Intermediate Energies". The paper was published in Phys. Rev. A 27, 1871 (1983). Possible extensions of this work rest primarily in the propagation of directed particle beams.

During this last year, we directed our efforts towards the understanding of electron capture collisions in the presence of a strong laser field. We used pseudo-potential molecular structure calculations and coupled-channel scattering calculations to investigate a system which can realistically be studied theoretically: $K^+ + Na$. Of importance is we are the first to include both the normal dynamical coupling and the laser-assisted coupling terms in a single calculation. This study displayed the regions of importance of both terms and indicated quantal interference effects at intermediate velocities. A major paper has been accepted by Physical Review A, entitled "Laser-Assisted Charge-Transfer Collisions: $K^+ + Na$" and authored by Y. Hsu, M. Kimura and

R. Olson.

The above research greatly increased our capabilities and lead to a nice series of theoretical papers on Rydberg atom collisions. I would like to thank the Physics Division of the Office of Naval Research for stimulating and supporting this research.

LIST OF PUBLICATIONS

The titles and authors of the papers attributed to this contract over the last five years are listed below. The covering pages to the papers follow.

a. "Ionization Cross Sections for Rydberg Atom-Rydberg Atom Collisions," R. E. Olson, Phys. Rev. Lett. 43, 126 (1979).

b. "Ion-Rydberg Atom Collision Cross Sections," R. E. Olson, J. Phys. B 13, 483 (1980).

c. "Excitation Transfer in Ion-Rydberg Atom Collisions," R. E. Olson, Phys. Rev. A 23, 3338 (1981).

d. "Ion Collisions with Rydberg Atoms in Strong Electric Fields," R. E. Olson and A. L. Mackellar, Phys. Rev. Lett. 46, 1451 (1981).

e. "The Effect of Core Interactions in ℓ-Mixing Collisions of Rydberg Atoms with Rare Gases," A. P. Hickman, J. Phys. B 14, L419 (1981).

f. "Ion-Atom Differential Cross Sections at Intermediate Energies," R. E. Olson, Phys. Rev. A 27, 1871 (1983).

g. "Theoretical Approaches to Low Energy Collisions of Rydberg Atoms with Atoms and Ions," A. P. Hickman, R. E. Olson, and J. Pascale, chapter for *Rydberg States of Atoms and Molecules*, ed. by R. F. Stebbings and F. B. Dunning (Cambridge University Press, 1983) pp. 187-227.

h. "Ion Scattering for State-Selected Rydberg Atoms," G. A. Kohring, A. E. Wetmore, and R. E. Olson, Phys. Rev. A 28, 2526 (1983).

i. "Laser-Assisted Charge-Transfer Collisions: K^+ + Na," Y. P. Hsu, M. Kimura, and R. E. Olson, Phys. Rev. A (in press).

ABSTRACTS FROM PUBLICATIONS

a.

VOLUME 43, NUMBER 2 PHYSICAL REVIEW LETTERS 9 JULY 1979

Ionization Cross Sections for Rydberg-Atom–Rydberg-Atom Collisions

R. E. Olson

Molecular Physics Laboratory, SRI International, Menlo Park, California 94025

(Received 23 April 1979)

A classical-trajectory Monte Carlo method has been applied to collisions of two Rydberg atoms. Numerical calculations were made for velocities $v = 0.01v_e$ to $10v_e$, where the Rydberg electron's velocity v_e(a.u.) = $1/n$ and n is the principal quantum number of the Rydberg atom. The total ionization cross sections scale as n^4 and show a $v^{-0.66}$ dependence at low v, a slight maximum around v_e, and a rapid decrease at high v. The cross sections are almost an order of magnitude larger than $\pi n^4 a_0^2$ at thermal energies.

b.

J. Phys. B: Atom. Molec. Phys. 13 (1980) 483–492. Printed in Great Britain

Ion–Rydberg atom collision cross sections

R E Olson

Molecular Physics Laboratory, SRI International, Menlo Park, CA 94025, USA

Received 19 June 1979, in final form 29 August 1979

Abstract. Classical-trajectory Monte Carlo calculations have been performed for collisions of ions in charge states $q = +1, +2, +5$ and $+10$ with hydrogenic atoms in principal quantum levels $n = 1, 2, 5, 10$ and 20. The collision velocity range investigated was $1 \leqslant v/v_e \leqslant 10$ where v_e is the orbital velocity of the Rydberg electron ($1/n$ in atomic units). Both charge-exchange and impact ionisation cross sections were calculated with impact ionisation found to be the dominant channel for $v/v_e \geqslant 2$. For $v/v_e \geqslant 5$, the sum of the charge-exchange (CEX) and impact ionisation (ION) cross sections may be represented by $\sigma_{CEX+ION}(a_0^2) = 6\pi n^2 q^2/v^2$, where v is in atomic units. Analysis of the electronic levels produced after charge exchange by the ion indicates the capture proceeds into excited levels which tend to preserve the energy and orbital size of the initial Rydberg atom.

c.

PHYSICAL REVIEW A VOLUME 23, NUMBER 6 JUNE 1981

Excitation transfer in ion–Rydberg-atom collisions

R. E. Olson*
Molecular Physics Laboratory, SRI International, Menlo Park, California 94025
(Received 11 September 1980)

Recently, electron-loss cross sections were presented by Kim and Meyer [Phys. Rev. Lett. 44, 1047 (1980)] for 40 keV/amu $N^{3+} + H^{**}(n)$ collisions which scaled as $n^{2.12}$, where n is the principal quantum number of the excited H^{*}. Such results are in contrast to an n^{3} scaling predicted by classical and first Born theoretical methods. Our calculations indicate that a major component of the experimentally observed ion signal was due to Stark ionization by deflector grids of highly excited H^{*} produced in excitation-transfer collisions. Inclusion of the excitation process in a theoretical interpretation reveals qualitative agreement between theory and experiment and stresses the importance of excitation transfer in ion–Rydberg-atom collisions.

d.

VOLUME 46, NUMBER 22 PHYSICAL REVIEW LETTERS 1 JUNE 1981

Ion Collisions with Rydberg Atoms in Strong Electric Fields

R. E. Olson[a]
Molecular Physics Laboratory, SRI International, Menlo Park, California 94025

and

A. D. MacKellar
Department of Physics and Astronomy, University of Kentucky, Lexington, Kentucky 40506
(Received 2 February 1981)

The classical-trajectory Monte Carlo method has been used to investigate collisions of ions and Rydberg atoms in strong dc electric fields. Cross sections are presented for $n = 10$ and $n = 20$ Rydberg atoms at velocities $1 \leq v/v_e \leq 10$ where $v_e = n^{-1}$ a.u. Electric fields which ionize product Rydberg atoms in states $n' = n + \Delta n$ with $\Delta n = 1$, 2, and 4 were used. The electric field caused the cross sections for electron capture to decrease by up to fourfold while the ionization values increased by up to two orders of magnitude.

PACS numbers: 34.50.Hc, 34.70.+e

e. J. Phys. B: At. Mol. Phys. 14 (1981) L419–L424. Printed in Great Britain

LETTER TO THE EDITOR

The effect of core interactions in l-mixing collisions of Rydberg atoms with rare gases

A P Hickman

Service de Physique Atomique Centre d'Etudes Nucléaires de Saclay, 91191 Gif-sur-Yvette Cedex, France

Received 6 April 1981

Abstract. Coupled-channel calculations for collisions of Na Rydberg atoms with He and Ar have been performed to investigate the effect of the interaction between the Na^+ core and the rare gas. For $n = 10$, channels corresponding to the levels nd and nf are included, and calculations are reported both with and without terms arising from the core. It is found that the inelastic cross sections (10d → 10f) are insensitive to the core interactions, whereas the elastic cross sections (10d → 10d) may change significantly. This result is consistent with the prediction of the first-order Born approximation.

f.

PHYSICAL REVIEW A VOLUME 27, NUMBER 4 APRIL 1983

Ion-atom differential cross sections at intermediate energies

R. E. Olson

Physics Department, University of Missouri–Rolla, Rolla, Missouri 65401

(Received 25 October 1982)

The classical-trajectory Monte Carlo method has been used to calculate $H^+ + H(1s)$ electron-capture and ionization differential cross sections in the range 25–200 keV. The results indicate the importance of including excited product states to describe the small-angle electron-capture scattering. Angular scattering of the electron removed by the ionization process has been studied as a function of ejected-electron velocity v_e. The classical calculations are in reasonable agreement with coupled-channel results of Shakeshaft [Phys. Rev. A **18**, 1930 (1978)] as to the "electron capture to the continuum" (ECC) component of the ionization process where this term is defined as the ejected electron being more closely centered to the projectile than the target nucleus after the collision. The ECC cross section σ_{ECC} was studied as a function of collision energy (50–500 keV/amu) and projectile charge state ($q = 1$–10). At high energies, σ_{ECC} scales as $q^{2.3}/E^{2.5}$. The maximum value for σ_{ECC} was determined to be an energy $E_{max} \cong (56\ \text{keV/amu})q^{0.4}$. Restricting the ECC component to small electron-scattering angles, $\theta_{lab} \leq 5°$, and electron-ejection velocities $v_e = v_p(1.0 \pm 0.1)$, where v_p is the projectile velocity, indicates this process is a minor component of the total ionization cross section at intermediate energies.

g.

Rydberg states of atoms and molecules

Editors
R. F. Stebbings and F. B. Dunning
Department of Space Physics and Astronomy
Rice University

6

Theoretical approaches to low-energy collisions of Rydberg atoms with atoms and ions

A. P. HICKMAN, R. E. OLSON, AND J. PASCALE

Cambridge University Press 1983
Cambridge
London New York New Rochelle
Melbourne Sydney

h.

PHYSICAL REVIEW A VOLUME 28, NUMBER 4 OCTOBER 1983

Ion scattering from state-selected Rydberg atoms

G. A. Kohring, A. E. Wetmore, and R. E. Olson
Physics Department, University of Missouri–Rolla, Rolla, Missouri 65401
(Received 7 April 1983)

Classical-trajectory Monte Carlo calculations have been performed for collisions of protons with state-selected hydrogenic Rydberg atoms. The examples investigated were Rydberg atoms in the $n=10$, $l=9$ level with $m_l=0$ and 9. The collision velocity range was 0.1 to 1.0 a.u. (2.2×10^7 to 2.2×10^8 cm/s). The ionization cross sections were found to be relatively insensitive to changes in the m_l levels. However, the charge-transfer cross sections showed considerable enhancement if the Rydberg electron is orientated in a plane which is parallel to the direction of the incident projectile.

i.

Laser-assisted charge-transfer collisions: $K^+ + Na$

Y. P. Hsu, M. Kimura and R. E. Olson
Dept. of Physics
University of Missouri-Rolla
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A theory has been formulated to characterize charge transfer collisions in the presence of an external laser field. The molecular state expansion method is used to describe the scattering process within the impact parameter formalism. Electron translation factors are included in the molecular state expansion so that the scattering wavefunction satisfies the correct boundary conditions. The theory is applied to the process: $K^+ + Na \rightarrow K + Na^+$. In addition, we have made a detailed analysis of laser-assisted charge transfer for low-energy collisions. In this case, a Landau-Zener formula can be derived which shows that the cross section increases with decreasing incident energy. In general the laser coupling is dominant in the low-energy region, while the dynamical coupling becomes important as the collision energy increases.

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