

CORRELATED CHARGE-CHANGING ION-ATOM COLLISIONS

FINAL TECHNICAL REPORT

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I. FINAL REPORT (narrative)

This document comprises the final technical report for atomic collisions research supported by DOE grant No. DE-FG02-87ER13778 from September 1, 2001 through August 31, 2004. The research involved the experimental investigation of excitation and charge-changing processes occurring in ion-atom and ion-molecule collisions. Major emphases of the study were: (1) interference effects resulting from coherent electron emission in H_2 , (2) production of doubly vacant K-shell (hollow ion) states due to electron correlation, and (3) formation of long-lived metastable states in electron transfer processes. During the period of the grant, this research resulted in 23 publications, 12 invited presentations, and 39 contributed presentations at national and international meetings and other institutions. Brief summaries of the completed research are presented below.

1. Interference in electron emission from H_2 by fast ions

In this work, ionization of H_2 by fast ions is investigated to explore effects associated with the fact that the atomic centers are identical. This indistinguishability gives rise to interferences, analogous to Young's two-slit experiment, when an electron is ejected coherently within the two-center molecular field [1,2]. To a good approximation, the ratio of cross sections for electron emission from H_2 compared to the cross sections for emission from independent H atoms exhibits an oscillatory behavior described by the function $[1+\sin(kcd)/kcd]$, where k is the outgoing electron momentum, c is a parameter representing the frequency variation with electron observation angle, and d is the internuclear separation. Thus, one full oscillation is expected to occur in the range $kcd = 0-2\pi$, corresponding to kc varying from 0-4.4 a.u. for $d = 1.42$ a.u. in H_2 .

The initial experimental work leading to confirmation of interference effects [1], as well as much of the subsequent work [3,4], involved a collaborative effort, including the present PI, conducted at the GANIL facility in Caen, France. Additional work [5,6,7], constituting the Ph.D. dissertation research of Mr. Sabbir Hossain, was done at WMU. The most recent results from GANIL [4], for incident 68 MeV/u Kr^{33+} ions, are shown in Fig. 1, and results from WMU [7], for 1, 3, and 5 MeV H^+ ions, are shown in Fig. 2. In these figures, the measured cross sections have been normalized to theoretical CDW-EIS cross sections [8].

The cross section ratios in the upper panels of Fig. 1 (from Refs. [3] and [4]) exhibit an oscillatory structure that clearly varies with the electron ejection angle. Furthermore, the ratios in Fig. 2 (from Ref. [7]), in addition to a strong dependence on the electron ejection angle, show a dependence on the collision velocity. As seen in the figure, this latter dependence is in qualitative agreement with predictions of the Born approximation, and can be traced to a dependence on the minimum momentum transfer as formulated by Nagy *et al.* [9]. A notable feature of the measured cross section ratios is the existence of higher frequency structures superimposed on the main oscillatory structure. In Fig. 1 the cross section ratios in the upper panels were divided by their corresponding fits to give the results plotted in the lower panels. Here, secondary oscillations are clearly revealed and have nearly equal frequencies for each of the electron emission angles. These latter data were again fit to the same functional form to determine the oscillation frequency for the secondary oscillations, giving values 2-3 times larger than those for the primary structures [4]. Similar results have been obtained for the data of Fig. 2

[7]. A theoretical formulation based on wave optics suggests that the secondary oscillations are due to interferences resulting from additional scattering within the molecule [4], an effect that has no analogy in Young's experiment.

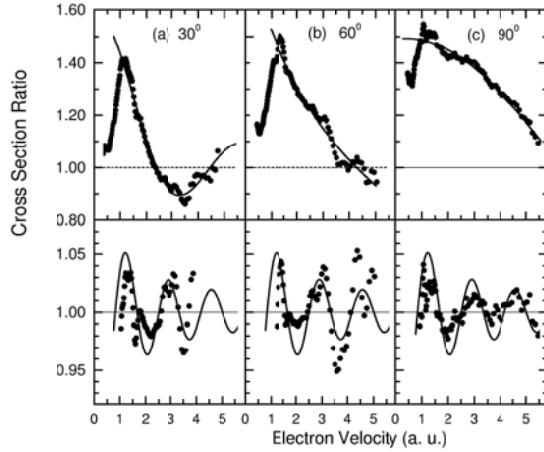


Fig. 1. Upper diagrams: Ratios of experimental to theoretical CDW-EIS [8] cross sections for electron emission by 68 MeV/u Kr^{33+} impact on H_2 for the electron observation angles indicated. Smooth curves are fits to the data (see Ref. [4]). Lower diagrams: cross section ratios divided by the corresponding fit functions and again fit to an oscillating function [4].

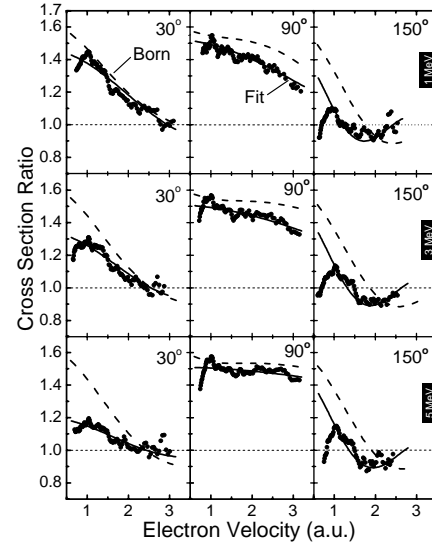


Fig. 2. Ratios of experimental to theoretical CDW-EIS [8] cross sections for electron emission by 1, 3, and 5 MeV H^+ impact on H_2 for the electron observation angles indicated. The dashed curves are Born calculations and the solid curves are fits to the data (see Ref. [7]).

This work, which is still ongoing, formed the Ph.D. dissertation project of Dr. Sabbir Hossain (graduated 08/04). Dr. Hossain participated in some of the measurements at GANIL as well. Dr. Ali Alnaser, whose Ph.D. project involved the electron correlation studies described immediately below, also contributed to some of the early interference work done at WMU. Additionally, two high school students from the Kalamazoo Area Mathematics and Science Center developed a computer code to carry out some of the Born calculations. Eight publications have resulted from this work [1,3,4,5,6,7,10,11].

2. Electron correlation in hollow state formation in three-electron systems

Double-K-shell-vacancy production in atomic Li and ionic Li-like systems by incident ions and electrons was studied to determine the role and manifestations of the electron-electron ($e-e$) interaction in leading to “hollow” states. Associated with the initial promotion of an electron by a charged particle or a photon, the $e-e$ interaction can cause a second electron transition. Promotion of the second electron is often attributed to a *shake* process resulting from the overlap of the initial- and final-state wavefunctions, or to a *dielectronic* process involving the mutual scattering of the two electrons in a binary encounter [12,13]. In this work, electron correlation effects associated with K-shell ionization plus K-shell excitation and double K-shell excitation in the formation of discrete intermediate states was investigated. For incident charged particles, although the nucleus-electron ($n-e$) interaction can produce hollow states, the $e-e$ contribution

becomes dominant as the collision velocity increases and the perturbation strength Z/v decreases (Z is the atomic number and v is the collision velocity in a.u.).

These measurements for Li-like ions colliding with He (Fig. 3) and for atomic Li in collisions with electrons (Fig. 4) show that doubly vacant K-shell states are produced mainly by ionization-excitation events leading to $2l2l'$ and $2l3l'$ configurations, with a small contribution from double-excitation leading to $2l2l'2l''$ configurations. These results can be compared with ion-induced [12,13] and photon-induced [19] spectra for atomic Li. For the Li-like ions hollow-state production is due to a combination of $n-e$ and $e-e$ interactions [14,15,16,17], while for incident electrons at 5 keV hollow-state production can be attributed entirely to the $e-e$ interaction [18]. Analysis of the results indicates that $e-e$ formation of S states is primarily due to shake, while $e-e$ formation of the P states can only be due to dielectronic processes. In the case of 5 keV $e^- + \text{Li}$, for which only the $e-e$ interaction produces hollow states, the experimental identification of the P states thus permits the dielectronic aspect of electron correlation to be separately identified [18]. For Li-like ions the existence of specific hollow states ($2l2l'$, $2l3l'$, or $2l2l'2l''$) and their relative intensities point to significant variations in the electron correlation strength as a function

Fig. 3. High-resolution Auger spectra for Be⁺ and B²⁺ Li-Like ions resulting from collisions with neutral He for energies near 1 MeV/u [14,15,16]

Fig. 4. High-resolution Auger spectrum for 5-keV $e^- + \text{Li}$ collisions [18].

(Caen) (graduated 11/02). Dr. Sabbir Hossain, another Ph.D. student at WMU, also contributed substantially to the former work. Five manuscripts concerning this work have been published [15,16,17,18,20].

3. Excited-state enhancement due to Pauli exclusion in electron transfer

The dynamics of atomic collisions, in the case of charged particles, are governed primarily by the Coulomb interaction of the incident projectile with the target nucleus and by the interactions of these nuclei with the active electrons. On the other hand, apart from the electron-nucleus interaction, atomic structure is determined by electron correlation (i.e., the electron-electron interaction) in conjunction with the Pauli exclusion principle. Because electrons are fermions, Pauli exclusion should play a significant role not only in the structure of atoms, but also in the population of states during collisional interactions between atoms or ions. However, this latter point has been only sparsely investigated.

In the present work, effects of the Pauli principle on collision dynamics were isolated by investigating electron transfer leading to the formation of Auger decaying $1s2s2p$ configurations in highly charged fluorine ions. Single-electron transfer to $F^{7+}(1s2s\ ^3S)$ and double transfer to $F^{8+}(1s)$ to form $1s2l2l'$ configurations have been investigated for 1.1 MeV/u collisions of these ions with He and Ne targets. High-resolution Auger electron emission spectra resulting from the de-excitation of these states exhibit anomalously large intensities for the formation of the metastable $1s(2s2p\ ^3P)\ ^4P$ state compared to the similarly configured $1s(2s2p\ ^3P)\ ^2P$ and $1s(2s2p\ ^1P)\ ^2P_+$ states as shown in Fig. 5. Due to the relatively long lifetime of the 4P state ($\sim 10^{-8}$ s) [21,22], instrumental corrections must be made to the measured intensities of this state to obtain the "true" intensities (all other observed states have sufficiently short lifetimes so that corrections are not needed). The corrected intensities for the 4P state are shown by the dashed curves in Fig. 5.

The large intensity for 4P cannot be explained on the basis of simple spin counting. Instead, the enhancement is attributed to an *electron exchange interaction* between projectile and target electrons having the same spin alignment during the electron transfer process [23]. This exchange results directly from the Pauli exclusion principle that prevents electrons with like spins from simultaneously occupying the $1s$ (or $2s$) orbital. Within the time-dependent Hartree-Fock (TDHF) picture of the collision system, this dynamical Pauli exchange corresponds to a time-dependent exchange potential between similarly aligned electrons. In the case of anti-aligned electrons the exchange potential is zero at all times. Thus, by means of this exchange between electrons having the same spin, the 4P state is enhanced beyond the intensity expected for direct transfer of aligned target electrons to the $2p$ orbital.

A challenge posed by the present observations is the calculation of the effect of the Pauli exchange potential for the collision systems studied here. Such a TDHF-type calculation would determine whether Pauli exchange can quantitatively explain the observed enhancement of the 4P state, or whether dynamical correlation effects must also be considered.

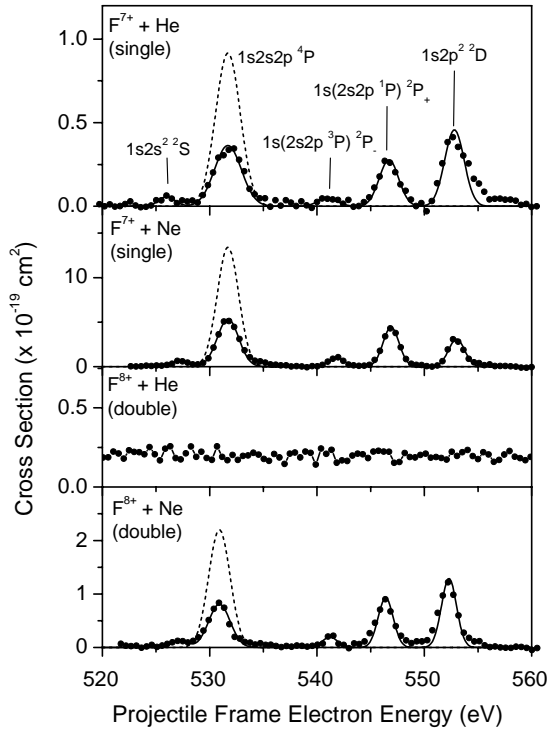


FIGURE 1

Fig. 5. Measured spectra for single electron transfer to F^{7+} and double electron transfer to F^{8+} for 1.1 MeV/u collisions of these ions with He and Ne. The observed $1s2l2l'$ intermediate states are indicated (for $F^{8+} + \text{He}$ no measureable intensity was observed for these states). The dashed curves represent the true intensity of the $1s2s2p^4 P$ state after correction for instrumental effects due to the long lifetime of this state (see text).

The enhancement effect observed here may be useful for controlling the production of certain long-lived ionic excited states. For example, the enhancement of metastable states may be useful in the development of high-energy x-ray lasers [24,25] since the radiative decay rate of such states can be comparable to the nonradiative (Auger) decay rate [21].

Two Ph.D. students (A. Alnaser and S. Hossain) were involved in this work, as well as a WMU undergraduate physics major (D.J. Pole). The work constituted the Honors College thesis project of Mr. Pole.

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II. PERSONNEL

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Dr. John A. Tanis

- Professor, Department of Physics

B. Professional Research Staff

Dr. Allen Landers

- Postdoctoral Research Associate (1999-2002), Assistant Professor (2002-03)

Dr. Stephen M. Ferguson

- Accelerator Physicist

C. Technical Support Staff

Mr. Allan Kern

- Accelerator Engineer

Mr. Richard Welch

- Instrument Maker

Mr. Mark Ely

- Electronics Shop Supervisor (until 9/03)

Mr. Benjamin Gaudio

- Electronics Shop Supervisor (10/03-present)

D. Graduate Students

Mr. Ali Alnaser

- Ph.D. student (graduated 8/02)

Mr. Sabbir Hossain

- Ph.D. student (graduated 8/04)

Mr. Jamie Baran

- Ph.D. student (2004-present)

E. Undergraduate Students

Mr. Richard DeBoer

- WMU physics major (spring 2003)

Ms. Heather Knutson

- Johns Hopkins Univ. physics major (summer 2001)

Mr. Donald J. Pole

- WMU Honors student (graduated 4/02)

Ms. Diane Strohschein

- WMU Honors student (2003-present)

F. High School Students

Mr. Austin Robison

- senior, Kalamazoo Math. and Science Center (2001-02)

Mr. Brian Stamper

- senior, Kalamazoo Math. and Science Center (2001-02)

Mr. Zach Steindler

- senior, Kalamazoo Math. and Science Center (2002-03)

III. PUBLICATIONS

1. "Double-K-Shell Vacancy Production in Li-like C^{3+} Ions Colliding with Helium," A.S. Al-Naser, A.L. Landers, D.J. Pole, H. Knutson, and J.A. Tanis, *Physica Scripta* **T92**, 265 (2001).
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16. "Electron Correlation in the Formation of Hollow Li-like Ions," A.S. Alnaser, A. Landers, D.J. Pole, S. Hossain, E.P. Benis, S.M. Ferguson, and J.A. Tanis, *Physica Scripta T* **110**, 137 (2004).
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19. "Electron Correlation in the Formation of Hollow States Along the Li-like Isoelectronic Sequence," A.S. Alnaser, A.L. Landers, and J.A. Tanis, *Phys. Rev. Lett.* **94**, 023201 (2005).
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22. "Interference Phenomena Associated with Electron Emission from H₂ by 1-5 MeV H⁺ Impact," S. Hossain, A.L. Landers, N. Stolterfoht, and J.A. Tanis, *Phys. Rev. A* **72**, 010701(R) (2005).

23. “Comment on “Interference Effect in Electron Emission in Heavy Ion Collisions with H_2 Detected by Comparison with the Measured Electron Spectrum from Atomic Hydrogen”, J.A. Tanis, S. Hossain, B. Sulik, and N. Stolterfoht, Phys. Rev. Lett. **95**, 079301 (2005).

IV. PRESENTATIONS

A. Invited

1. "Photoelectron Diffraction Mapping: Molecules Illuminated from Within," A.L. Landers, 2001 Annual Meeting of the Division of Atomic, Molecular, and Optical Physics (DAMOP) of the American Physical Society, London, Ontario, Canada, May 2001; Bull. Am. Phys. Soc. **46**, 95 (2001).
2. "Photoelectron Diffraction Mapping: Molecules Illuminated from Within," A.L. Landers, XXII International Conference on Photonic, Electronic, and Atomic Collisions (ICPEAC), Santa Fe, New Mexico, July 2001, Abstracts of Contributed Papers, edited by S. Datz, M.E. Bannister, H.F. Krause, L.H. Saddiq, D. Schultz, and C.R. Vane, (Rinton Press, Princeton, 2001), p. 66.
3. "Double Ionization of Spatially Aligned D₂ by Fast Ions," A.L. Landers, XVII International Seminar on Ion-Atom Collisions (ISIAC), Baja California, Mexico, July 2001.
4. "Interference in Electron Emission from H₂ by Fast Ions," J.A. Tanis, XVII International Seminar on Ion-Atom Collisions (ISIAC), Baja California, Mexico, July 2001.
5. "Superelastic Electron Scattering from Metastable He-like Ions," J.A. Tanis, Seventeenth International Conference on the Application of Accelerators in Research and Industry, Denton, TX, 14 November 2002.
6. "Electron Correlation in the Formation of Hollow Li-like Ions Colliding with Helium," A.S. Alnaser, 2003 Annual Meeting of the Division of Atomic, Molecular, and Optical Physics (DAMOP) of the American Physical Society, Boulder, CO, May 2003; Bull. Am. Phys. Soc. Paper C2.002 (2001).
7. "Electron Correlation Leading to Double-K-Shell Vacancy Production in Li-like Ions Colliding with Helium," A.S. Alnaser, 23rd International Conference on Photonic Electronic and Atomic Collisions, Stockholm, Sweden, July 23-29, 2003.
8. "Electron Correlation in the Formation of Hollow Li-like Ions," A.S. Alnaser, 23rd International Conference on Photonic Electronic and Atomic Collisions, Stockholm, Sweden, July 23-29, 2003.
9. "First- and Second-order Interferences in H₂ Spectra by Heavy Ion Impact," J.A. Tanis, XVIII International Seminar on Ion-Atom Collisions (ISIAC), MS Symphony, Silja Line (Stockholm-Helsinki-Stockholm), 30 July 2003.
10. "Interferences in Electron Emission Spectra for H⁺ Impact on H₂," S. Hossain, XVIII International Seminar on Ion-Atom Collisions (ISIAC), MS Symphony, Silja Line (Stockholm-Helsinki-Stockholm), 30 July 2003.

11. "High Frequency Oscillations in Electron Emission Interferences from H₂," S. Hossain, 8th Workshop on Fast Ion-Atom Collisions, 1-3 September 2004, Debrecen, Hungary.
12. "On Two-center and Interference Effects and Some Related Short Stories," R.D. Rivarola and J.A. Tanis, Keynote Lecture, Atomic Collisions and Electron Spectroscopy: A Seminar to Honor the 65th Birthday of Nico Stolterfoht, Berlin, Germany, 21 March 2005.

B. Contributed

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2. "Double-K-Shell-Vacancy Production in Li by 60 MeV/u Kr³⁴⁺ Ions," J.A. Tanis, A.L. Landers, J. Rangama, J.-Y. Chesnel, F. Frémont, X. Husson, D. Hennecart, A. Cassimi, B. Sulik, B. Skogvall, V. Hoffmann, and N. Stolterfoht, 2002 Meeting of the Division of Atomic, Molecular, and Optical Physics (DAMOP) of the American Physical Society, Williamsburg, Virginia, May 2002; Bull. Am. Phys. Soc. **47**, 62 (2002).
3. "Variation of Electron Correlation in Hollow State Formation in Li-like Ions," A.S. Alnaser, A. Landers, S. Hossain, S. Ferguson, O.A. Haija, J.A. Tanis, and E.P. Benis, 2002 Meeting of the Division of Atomic, Molecular, and Optical Physics (DAMOP) of the American Physical Society, Williamsburg, Virginia, May 2002; Bull. Am. Phys. Soc. **47**, 63 (2002).
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8. "Identification of Dielectronic and Shake Processes for Producing Double K-shell Vacancies in Lithium by Fast Charged Particle Impact," J. Rangama, J.-Y. Chesnel, J.A. Tanis, B. Sulik, B. Skogvall, F. Frémont, X. Husson, D. Hennecart, A. Cassimi, A.L. Landers, V. Hoffmann, and N. Stolterfoht, *19th International Conference on X-ray and Inner-shell Processes*, Rome, Italy, June 2002, Abstracts, p. 115.
9. "Interference Effects in Electron Emission from H₂ by Fast Ion Impact," N. Stolterfoht, J.A. Tanis, B. Sulik, V. Hoffmann, B. Skogvall, J.-Y. Chesnel, J. Rangama, F. Frémont, D. Hennecart, A. Cassimi, X. Husson, A. Landers, M.E. Galassi, and R.D. Rivarola, *19th International Conference on X-ray and Inner-shell Processes*, Rome, Italy, June 2002, Abstracts, p. 115.
10. "Mechanisms Responsible for Hollow State Formation in Li-like Ions Colliding with Helium," A.S. Alnaser, A.L. Landers, S. Hossain, E. Benis, D.J. Pole, S. Ferguson, and J.A. Tanis, *19th International Conference on X-ray and Inner-shell Processes*, Rome, Italy, June 2002, Abstracts, p. 116.
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