DOE/ER/13955--21

DYNAMICS OF COLLISION PROCESSES

Final Report

for the Period August 1, 1988 – July 31, 1996

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March 1997

Prepared for

THE U.S. DEPARTMENT OF ENERGY
AGREEMENT NO. DE-FG02-88ER13955

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I. FINANCIAL STATEMENT OF ESTIMATED COSTS AND UNEXPENDED FUNDS

University of Nebraska-Lincoln

Grant #DE-FG02-88ER13955

For the Budget Period August 1, 1988 through July 31, 1996

1.	Total DOE Funds Obligated	\$522,100
2.	Estimated Total Project Costs through the Current Budget Period chargeable to DOE	<u>\$522,100</u>
3.	Anticipated amount of unexpended DOE Funds	-0-

II. COMPLIANCE

Neither the aims of investigations nor the effort of the principal investigator to this project have deviated from what was proposed. The principal investigator devoted 25% of his time to this project during 1 August 1988 – 31 July 1996.

Anthony F. Starace
Principal Investigator

III. BIBLIOGRAPHY

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IV. SCOPE OF INVESTIGATIONS AND SIGNIFICANT RESULTS OBTAINED

The atomic and molecular processes relevant to the basic energy sciences are governed by the dynamics of interacting charged massive particles. The simplest and most fundamental case of this general correlation problem is the three-body Coulomb problem. This research project is concerned with collision processes which are governed by the dynamics of three interacting charged particles. In what follows we provide the abstract descriptions for each of the works listed in the Bibliography.

A. Low-Energy Features of the $e^- - H(n=2)$ System Exhibited in Fast H^- Detachment Collisions. [Bibliographic Item No. 1]

Quantitative predictions for the doubly differential detachment collisions, $0.5\text{-MeV } H^- + He \rightarrow H(n=2) + He^* + e^-$, were obtained by use of hyperspherical wave functions for the $H-e^-$ system. Individual $H-e^-$ hyperspherical channels were identified as producing distinct "cusp," "shape resonance," and/or "shoulder" features in the laboratory-frame cross sections. Moreover, the first quantitative predictions of the expected Gailitis-Camburg dipole-field-induced oscillations in the cross sections above the $H(n=2)$ threshold were presented and distinguished from $^1P^o$ shape resonance effects.

B. Doubly Differential Detachment Cross Sections for 0.5 MeV H^- on He Including Projectile Excitation to $H(n=2)$. [Bibliographic Item No. 2]

Detailed theoretical results were presented for the electron-detachment cross section, doubly differential in both the electron momentum and angle, for the process $0.5\text{-MeV } H^- + He \rightarrow H(n=2) + e^- + He^*$. As discussed briefly elsewhere [C.R. Liu and A.F. Starace, Phys. Rev. Lett. **62**, 407 (1989)], the laboratory-frame doubly differential cross sections (DDCS's) for electron detachment in the forward direction were shown to depend sensitively on the low-energy states of the $H(n=2)-e^-$ three-body system. In particular, the angular dependence of characteristic cusp and shape resonance features was presented. We found that the projectile frame DDCS for detached electron energies in the vicinity of the shape resonance peak is nearly isotropic. This is due in part to the 1P symmetry of the resonance feature, which limits the angular distribution to constant and $\cos^2\theta$ terms, and in part to cancellation in the integral over momentum transfer on which the coefficient of the $\cos^2\theta$ term depends. We also found that the rapid variation of these cusp and shape resonance features with angle in the laboratory frame requires that experimental angular and energy resolution be accounted for in order to obtain good agreement with the measured results of M.G. Menendez and M.M. Duncan [Phys. Rev. A **36**, 1653 (1987)] on the energy spectrum of detached electrons coincident with the formation of $H(2p)$. The DDCS's for $0.5\text{ MeV } H^- + He \rightarrow H(1s) + e^- + He^*$ were also presented. When these latter results were added to those for producing $H(n=2)$, the sum gave good agreement with the experimental data of M.G. Menendez

and M.M. Duncan [Phys Rev. **20**, 2327 (1979)], which include all final states of the H atom, thereby confirming the important contribution the $H(n=2)$ states make to the total-detachment cross section. We presented evidence on the sensitivity of these results to the choice of the average excitation energy of the helium target. Finally, we pinpointed the origin of our predicted Gailitis-Damburg oscillations in the DDCS near threshold as stemming from a rapid decrease of an analytically known phase appropriate for long-range dipole fields.

C. Quasiatomic Contributions to Molecular-Scattering Form Factors. [Bibliographic Item No. 3]

The scattering form factors for excitation of H_2^+ in fast collisions were calculated in a sequence of approximations revealing their dependence on details of the initial- and final-state wave functions. Each approximate calculation was compared directly to the "exact" results obtained with separable, fixed nuclei wave functions. Even for interproton separations larger than those for which the united-atom limit applies, the quasiatomic character of the final-state orbital dominates transition amplitudes to low-lying dissociative states. Possible application to molecular dissociation by charged-particle impact at high energies were considered.

D. Two-Body Fragmentation Channels of Three-Body Systems. [Bibliographic Item No. 4]

An asymptotic expansion of the wave functions for the fragmentation channels of three-body systems was developed in hyperspherical coordinates. It was shown that, to any finite power in $1/R$, where $mR^2 = \sum_i m_i r_i^2$ is the trace of the inertia tensor, the expanded wave function is an analytic finite sum of Sturmian functions. The expansion was carried out explicitly through order $1/R^4$. These asymptotic states converge (as $R \rightarrow \infty$) to polarized orbitals for two-electron systems, and also provide improved dissociation channels for molecular ions, such as HD^+ . Asymptotic potential curves for the He and $\mu^- - H$ systems were presented as illustrations.

E. Variational Methods for High-Order Multiphoton Processes. [Bibliographic Item No. 5]

Methods for applying the variationally stable procedure for Nth-order perturbative transition matrix elements of Gao and Starace [Phys. Rev. Lett. **61**, 404 (1988); Phys. Rev. A **39**, 4550 (1989)] to multiphoton processes involving systems other than atomic hydrogen were presented. Three specific cases were discussed: one-electron ions or atoms in which the electron-ion interaction is described by a central potential; two-electron ions or atoms in which the electronic states are described by the adiabatic hyperspherical representation; and closed-shell ions or atoms in which the electronic states are described by the multiconfiguration Hartree-Fock representation.

Applications were made to the dynamic polarizability of He and the two-photon ionization cross section of Ar.

F. Doubly Differential Detachment Cross Sections for Fast H^- – Rare Gas Collisions. [Bibliographic Item No. 6]

Detailed theoretical analyses were presented of the projectile energy dependence as well as the target dependence of the electron detachment collision processes, $H^- + T \rightarrow H(n=2) + T^* + e^-$. These analyses were illustrated by calculations of both projectile frame and laboratory frame doubly differential cross sections (DDCS's) for 0.5, 1.0 and 1.5 MeV H^- collisions with He targets and for 0.5 MeV H^- collisions with Ne, Ar, Kr, and Xe targets; in addition, we presented laboratory frame DDCS's for 0.1 MeV H^- collisions with Xe targets. Comparisons with available experimental data were given.

G. One- and Two-Photon Detachment of H^- with Excitation of $H(n=2)$ [Bibliographic Item No. 7]

The cross sections for one- and two-photon detachment of H^- with excitation of the degenerate $H(2s)$ and $H(2p)$ levels have been calculated within an adiabatic hyperspherical representation. Both the partial cross sections, $\sigma(2s)$ and $\sigma(2p)$, and the photoelectron angular distribution asymmetry parameters were obtained. Our one-photon detachment results are dominated by the $^1P^\circ$ shape resonance feature above threshold, which is well known from prior theoretical and experimental work, with which our results were compared in detail. Our two-photon detachment cross sections and angular distribution asymmetry parameters exhibit a half-cycle of dipole-field-induced oscillation in the energy region from the $H(n=2)$ threshold to approximately 34 meV above threshold. The occurrence of a half-cycle of oscillation with significant amplitude over a 34-meV energy region above the $H(n=2)$ threshold is due to two circumstances unique to this two-photon process: first, in contrast to the strongly suppressed oscillations in the $^1S^\circ$ and $^1P^\circ$ attractive dipole-field channels for the $H(n=2) - e^-$ system, the amplitude of oscillation in the channel having $^1D^\circ$ symmetry is not strongly suppressed above threshold; second, electric dipole selection rules for the two-photon process do not permit population of the intense $^1P^\circ$ shape resonance located above the $H(n=2)$ threshold, which would otherwise obscure the half-cycle of dipole-field-induced oscillation. For these reasons, the process of two-photon detachment of H^- is a favorable one for observing this dipole-field-induced oscillation above threshold, which has yet to be observed experimentally in any process.

H. Long-Range Dipole Field Effects on H^- Detachment Processes Above the $H(n=2)$ Threshold [Bibliographic Item No. 8]

In this paper we show how the low-energy states of the fundamental $H(n=2) - e^-$ three-body system influence three very different processes for detaching the H^- ion. We also show that these three different detachment processes are each best suited to make manifest a different one of the properties of the long-range dipole field interactions within the $H(n=2) - e^-$ system. Thus, collisional detachment of H^- with measurement of the detached electron in the forward direction is best suited for demonstrating the finite threshold cross sections of those channels which have an attractive dipole-field interaction at asymptotic distances. This is so because the kinematic transformations from the projectile frame to the laboratory frame lead to a cusp behavior in the laboratory frame DDCS's which is possible only because of the finite threshold cross sections in the projectile frame.

Single photon detachment of H^- with excitation of $H(n=2)$ is ideal for observing the shape resonance feature in one of the $^1P^o$ final state channels above the $H(n=2)$ threshold. Electric dipole selection rules result in only final state channels having $^1P^o$ symmetry. The $^1P+$ channel, in which the shape resonance occurs, has a cross section so much larger than those for other allowed $^1P^o$ channels that the shape resonance feature can be observed very clearly.

Finally, the two-photon detachment of H^- with excitation of $H(n=2)$ is the most likely process in which to observe the long-range dipole-field-induced oscillations above threshold that were predicted by Gailitis and Damburg but which have yet to be observed experimentally in any process. On the one hand, the $^1P^o$ shape resonance feature *is not* populated in this process, and hence cannot obscure these oscillations. On the other hand, the $^1D+$ final-state channel which *is* populated has the largest amplitude for such oscillations of any final state channel. Theoretical calculations predict a half cycle of such oscillation over the energy region from 0.1 meV to about 34 meV above threshold.

I. Variationally Stable Treatment of Two- and Three-Photon Detachment of H^- Including Electron-Correlation Effects [Bibliographic Item No. 9]

We have recently presented a variationally stable, adiabatic hyperspherical treatment of two- and three-photon detachment of H^- . Results were compared with analytic predictions of a zero-range potential model of H^- . Detailed comparisons were made also with other theoretical results which include the effects of electron correlations. We predicted analytically (and demonstrated numerically) an extreme sensitivity of the theoretical predictions to any errors in the value of the electron affinity employed. We also showed that the low-intensity limit of the Keldysh treatment [Sov. Phys. JETP 20, 1307 (1965)] of detachment of an electron bound in a zero-range potential agrees with the results of a perturbative treatment. Our calculated two- and three-photon

detachment cross sections for H^- agree best with those of Mercouris and Nicolaides [Phys. Rev. A **45**, 2116 (1992)]. The results were subsequently found to be in excellent agreement with R-matrix results [J. Purvis et al., Phys. Rev. Lett. **71**, 3943 (1993)].

J. Hyperspherical Coordinate Description of Single- and Multiphoton Processes in Two-Electron Systems [Bibliographic Item No. 10]

Much of our research that has been supported by DOE over many years was recently reviewed critically and placed in context with the theoretical work of others and with experimental measurements.

K. Resonant Two-Color Detachment of H^- with Excitation of $H(n=2)$ [Bibliographic Item No. 11]

We have calculated the cross sections for resonant two-color, two-photon detachment of H^- with excitation of the degenerate $H(2s)$ and $H(2p)$ levels within a semiempirical adiabatic hyperspherical representation. The first photon, with energy $\omega_1 = 0.4017$ a.u., was chosen to be resonant with the well-known Feshbach $^1P^o$ resonance below the $H(n=2)$ threshold. The second photon, with energy $\omega_2 \geq 0.12605$ a.u., was chosen to scan the energy region above the $H(n=2)$ threshold over which long-range dipole-field-induced cross section oscillations are predicted to occur. Such Gailitis-Damburg oscillations have not yet been observed experimentally. Results for various pairs of light polarization for the two-photons were calculated. Our resonant two-color, two-photon detachment cross sections are 8-9 orders of magnitude greater than the corresponding nonresonant, single-color, two-photon detachment cross sections obtained by C.R. Liu, N.Y. Du, and A.F. Starace [Phys. Rev. A **43**, 5891 (1991)]. Unmistakable evidence of long-range dipole field effects was demonstrated over the 5 meV energy range above the $H(n=2)$ threshold. Furthermore, the differential cross sections for right- and left-circularly polarized copropagating photons and especially the circular dichroism differential cross sections were found to have nearly a full cycle of a greatly enhanced dipole-field-induced oscillation extending over the region from threshold to ≈ 34 meV above.

L. Electric-Field Effects on H^- Photodetachment with Excitation of $H(n=2)$ [Bibliographic Item No. 12]

A theory of the photodetachment of hydrogen negative ions in an external static electric field with the production of hydrogen in excited states was developed. We use a combination of the frame-transformation theory and the Green's-function method. The cross section in nonzero fields is expressed in terms of the photodetachment matrix elements for zero field. The latter were calculated using the adiabatic hyperspherical representation. For energies close to the threshold we use a smooth extrapolation of the zero-field photodetachment matrix elements down to threshold

taking care to enforce the proper threshold behavior of cross sections for nonzero fields. The ripples in the *total* cross section due to interference effects are not noticeable for incident photons linearly polarized along the static field direction, but are relatively well pronounced for the case of circularly polarized photons propagating along the static field. On the other hand, all *partial* cross sections corresponding to photodetached electrons having magnetic quantum number $m' = 0$ (with respect to a quantization axis parallel to the static electric field) show significant ripple structure regardless of the light polarization. The partial cross section for detachment with excitation of the $n=2$, $m=1$ Stark substate by circularly polarized photons exhibits a particularly distinct ripple structure. All cross sections in the region of the shape resonance increase with increasing electric field in accordance with experimental observations. This effect is stronger in the case of linear polarization. We also predict a fairly stable value for the width of the $^1P^o$ shape resonance as a function of the electric-field strength over the range from 0.4 to 1 MV/cm. For electric-field strengths below 0.4 MV/cm, the ripple structure in the shape resonance makes it difficult to define a resonance width.

M. Parallels Between High Doubly-Excited State Spectra in H^- and Li^- Photodetachment [Bibliographic Item No. 13].

Eigenchannel R-matrix calculations including effects of long-range multipole interactions outside the R-matrix box have been carried out for photodetachment of H^- and Li^- up to the $n=6$ threshold. Results for H^- give the first complete theoretical confirmation of experimental measurements [P.G. Harris et al, Phys. Rev. Lett. **65**, 309 (1990)] below the $n=5$ and $n=6$ thresholds. Partial cross sections for Li^- are found to be remarkably similar to those for H^- , despite the absence of atomic energy level degeneracy in the case of Li . Density plots for two electron resonances appearing in each spectrum confirm the similarity of the two spectra, indicating that advances in understanding doubly-excited states in two-electron systems have application to doubly-excited state spectra in many-electron systems. A more complete description of the present results, including presentation of the full spectrum for Li^- photodetachment from the $n=2$ to the $n=6$ thresholds, is being prepared for publication.

N. Photodetachment of $^3P^o$ State of H^- [Bibliographic Item No. 14].

Because the $H^- 2p^2(^3P^o)$ state has yet to be observed in the laboratory, it is of interest to make theoretical predictions for processes involving this state. We have completed semiempirical, adiabatic hyperspherical calculations for the photodetachment cross section for the process $H^- 2p^2(^3P^o) + \gamma \rightarrow H(n=2) + e^-$ at photon energies in the range from threshold to 0.125 a.u. above. We found the detachment cross section for this state to have a maximum value of 482 Mb at 25.7 meV above threshold, in good agreement with results of close-coupling calculations by Jacobs, Bhatia, and Temkin [Astrophys. J. **242**, 1278 (1980)]. In contrast to the close-coupling results, however, we find that the detachment cross section rises by a factor of more than 400 from its

threshold value to its maximum. This rather startling prediction has recently been confirmed by J.Z. Tang et al. [Phys. Rev. A **51**, 4694 (1995)].

O. Calculation of the Double Photoionization Cross Section of Helium
[Bibliographic Item No. 15]

In this paper, a MBPT calculation for σ^{++} of helium up to 14 keV photon energy was carried out. This work extends the MBPT calculation of Carter and Kelly [Phys. Rev. A **24**, 170 (1981)], who treated the region from the threshold up to 290 eV, to higher photon energies. Our main aim is to cover photon energies below 4 keV, at which ionization of helium due to Compton scattering is still not a large effect, so that our results can be compared to the measured results including all the contributing processes. To reduce the possible numerical errors involved in treating high energy continuum wave functions, summations were performed over intermediate states implicitly by solving inhomogeneous differential equations whenever this is convenient. By including all important electron correlation effects in the present MBPT calculation, a good overall agreement is obtained with the experimental results for σ^{++}/σ^+ of helium in the photon energy range from 200 eV to 4 keV.

P. Photodetachment of Li^- From the Li 3s Threshold to the Li 6s Threshold
[Bibliographic Item No. 16]

Eigenchannel R-matrix calculations (including effects of long-range multipole interactions beyond the reaction volume) for Li^- photodetachment partial cross sections from the vicinity of the Li 3s threshold to the Li 6s threshold ($3.8 \text{ eV} \leq \hbar\omega \leq 5.65 \text{ eV}$) are presented. Excellent agreement with the relative total cross section measurements of U. Berzinsh et al. [Phys. Rev. Lett. **74**, 4795 (1995)] in the vicinity of the Li 3s and Li 3p thresholds is found. The calculated resonance structures are analyzed in detail. In particular, the energy region between the Li 4s and Li 5p thresholds (for which there are as yet no experimental measurements) is shown to have types of doubly excited resonances which are prominent only because of the nonhydrogenic Li^+ core; such types are weak or absent in higher-energy regions as well as in H^- photodetachment spectra.

Q. Novel Doubly Excited States Produced in Negative Ion Photodetachment
[Bibliographic Item No. 17]

Our DOE-supported work on negative ion photodetachment of H^- and Li^- is critically summarized and compared with available experimental and theoretical work.