Project Title: Calculation of Electron Impact Cross Sections from Metastable States in Atomic and Molecular Gases

Project No: G-41-644

Principal Investigator: Dr. M. R. Flannery

Sponsor: Dept. of the Air Force: Eqs. Aeronautical Systems Division (AFSC); USAFA, OH

Agreement Period: From 10/1/75 Until 1/2/76 (Contr. Expiration)

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- $88,340 - Total

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*Partially funded at $30,000 est. to fund project through about 6-30-76.

TECHNICAL MATTERS

Capt., John F. Prince, AFAPL/PPOF (AFSC) Project Engineer

USAFA Aero Propulsion Laboratory (AFSC)
Wright-Patterson AFB, Ohio 45433


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SPONSORED PROJECT TERMINATION

Date: November 13, 1978

Project Title: Calculation of Electron Impact Cross Sections from Metastable States in Atomic and Molecular Gases

Project No: G-41-644 (C/5 Act. = G-41-336)

Project Director: Dr. M. R. Flannery

Sponsor: Department of Air Force; Headquarters Aeronautical Systems Division (AFSC) WPAFB; OH 45433

Effective Termination Date: 1/2/78 (Contract Expiration)

Clearance of Accounting Charges: by 1/31/78

Grant/Contract Closeout Actions Remaining:

- Final Invoice and Closing Documents
- Final Fiscal Report
- Final Report of Inventions
- Govt. Property Inventory & Related Certificate
- Classified Material Certificate
- Other

NOTE: Follow-on Project is G-41-669.

Assigned to: School of Physics (School/Laboratory)

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Contract No. F33615-76-2033, Item No. 001.

Air Force Aero Propulsion Laboratory
Attn: AFAPL/FOP(450)/Capt. Prince
Wright Patterson AFB,
Ohio 45433

Subject: Quarterly R and D Status Report No. 1.
Contract No. F33615-76-C-2033.

Gentlemen:

Transmitted herewith are four (4) copies of the subject reports.

Sincerely,

M. K. Flannery
Principal Investigator and Professor

Encl.

MRF/ja
Calculation of Electron Impact Cross Sections from Metastable States in Atomic and Molecular Gases

Quarterly Progress Report No. 1

Period Covered: July 8, 1975 to December 31, 1975
Contract No. F33615-76-C-2033

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Appendix A: New Semiclassical Treatments of rotational and vibrational transitions in heavy-particle collisions.

Appendix B: Abstract of invited talk to DEAP.

Appendix C: Highly Excited States excite interest in many labs.
1. Introduction

The work involved under the present contract is a natural extension of the research work carried out under a previous contract with number F33615-74-C-4003 which officially terminated on August 8, 1975. The present contract with number F33615-76-C-2033 officially started on October 1, 1975.

However, between August 8, 1975, the termination of the old contract and October 1, 1975, the starting date of the new contract, work on the subject matter did not end but strongly continued as normal. Therefore, in an effort to preserve continuity, the progress achieved during the period 8 July 1975 - 31 December 1975 will be presented here.

Since I only received notification of the present contract only on Jan 6, 1976 giving various details as the contract number, schedule report dates and addresses, today is the earliest possible time for submittal of this present report.

(a). In an effort to explore the importance of vibrational and rotational excitation in electron-excited molecule collisions, theory was developed. Semiclassical approaches were adopted and in order to assess their accuracy, they were initially applied to rotational excitation in heavy particle collisions. The ground state H-H$_2$, and He-H$_2$ systems were studied since detailed quantal calculations were available for these systems. The present theory yields cross sections in very close accord with the corresponding quantal results. The semiclassical approaches are particularly valuable for targets initially in excited rotational and vibrational states, cases prohibitively difficult to a full quantal treatment.

This work was written up and accepted for publication in J. Chem. Phys. It appears as Appendix A of the present report.

(b). The possibility of including the effect of electron exchange in electron-excited atom collisions was investigated. This effect may be important for the proposed e-N and e-O collisions. A suitable modification to the multichannel eikonal treatment incorporating electron-exchange was found and formulae were developed.

(c). Work on the collisions

\[ e + N(2D) \rightarrow e + N^*(2P) \]

and

\[ e + O(1D) \rightarrow e + O^*(1S) \]

has now begun. Highly accurate multiconfiguration wavefunctions are being used and the interaction matrix elements $V_{ij}(R)$, needed as input to the multichannel eikonal treatment, are being calculated. The effect
of exchange on the previous collisions is under investigation.

(d) Work on the collision

\[ e + H_2(v) \rightarrow e + H_2^+(v^1) + e \]

has also been initiated. The continuum wavefunction for the ejected electron moving in the two-centered field of \( H_2^+ \) has been calculated and correctly normalized. The next stage is the calculation of the electronic matrix element for the ionization process, followed by suitable averaging over the initial and final rotational and vibrational states.

3. Publications

(a) August (1975): "A ten-channel eikonal treatment of differential and integral cross sections and of the \((\lambda, \chi)\) parameters for the \( n = 2 \) and \( 3 \) excitations of helium by electron impact."


(b) September (1975): "Ten-channel treatment of electron-metastable-helium collisions: Differential and integral cross sections for \( ^2P \) and \( ^3P \) and \( n = 3 \) excitations from He(\( ^2S \)) and the \((\lambda, \chi, \pi)\) parameters."


(c) November (1975): "A theoretical treatment of atomic collisions at intermediate energies and highly excited states"


Sixteen copies of (a) and (b) were sent to AFAPI on Oct. 1, 1975. Sixteen copies of (c) and (d) are now being sent to AFAPI/POP.

4. **Presentations**

(a) Two papers were presented at the IX. International Conference on the Physics of Electron and Atomic Collisions (ICPEAC) Seattle, Washington, July 24-30, 1975.

(b) A long paper on semiclassical treatments of rotational and vibrational transitions was presented at the 28th Gaseous Electronics Conference (G.E.C.) Rolla, Missouri October 21-24, 1975.

(c) An invited paper was presented at the 7th Annual Meeting of the Division of Electron and Atomic Physics (DEAP), Tuscon, Arizona, Dec. 4-6, 1975. The abstract is given as Appendix B.

5. **Other items**

In the "Search and Discovery" section of the November 1975 issue of Physics Today, the editor devoted considerable attention to the topic "Highly excited atoms excite interest in many labs". In particular he drew attention to the theoretical work of Dr. Flannery on excited atoms and specifically mentioned the theoretical paper of ionization by electron impact that Dr. Flannery presented at IX ICPEAC in Seattle. The Physics Today article is reproduced as Appendix C.
New semiclassical treatments of rotational and vibrational transitions in heavy-particle collisions. I. H-H\textsubscript{2} and He-H\textsubscript{2} collisions\textsuperscript{*}

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(Received 23 July 1975)

Two new semiclassical methods—the multistate orbital treatment and the multichannel eikonal treatment—are proposed for the description of rotational and vibrational excitation in heavy-particle collisions. The first method includes appropriate trajectories determined from a certain optical potential designed to couple the response of the internal structure, which is described by a quantal multistate expansion, to the orbit for the relative motion and vice versa. While this approach is, in general, valid when the quantum imprecision in the classical trajectories is small (as for heavy particles) the second method based on the use of a straight-line eikonal for the relative motion, of different local momenta in the various channels and of a multistate expansion for the internal motions, is valid for scattering mainly about the forward direction. These procedures are applied to representative rotational transitions in H-H\textsubscript{2} and He-H\textsubscript{2} collisions at 0.25–1.5 eV and yield angular distributions and integral cross sections in very close accord with corresponding quantal results. The methods are particularly valuable at higher impact energies when the inclusion of the resulting many rotational and vibrational channels by full quantal treatments is prohibitively difficult. Various approximate schemes—the perturbed-rotating-atom approximation and the effective potential method—are also investigated.

I. INTRODUCTION

Recently, there has been considerable activity\textsuperscript{1–8} in theoretical descriptions of rotational excitation by approximate schemes all based on the full quantal close-coupling treatment.\textsuperscript{7} Classical studies have also been presented.\textsuperscript{8} The following difficulties have, however, prevented corresponding progress in finding adequate semiclassical descriptions of rotational excitation.

(i) The extraction of differential cross sections from the usual impact-parameter treatment (IPT) is not immediately obvious since the resulting first-order coupled equations\textsuperscript{8} naturally follow from the Dirac method of variation of constants,\textsuperscript{6} a time-dependent formulation.

(ii) The consequent use of a straight-line trajectory for the relative motion, adopted for the needed space-time transformation, is clearly quite valid for rotational excitation which mainly occurs at large scattering angles \( \theta \) due to the required repulsive close-encounters, particularly for neutral–neutral collisions.

(iii) While elastic scattering is mainly about the forward direction (\( \theta \leq 40^\circ \)), and while reasonable integral elastic cross sections can be obtained from the usual classical potential–scattering formulas,\textsuperscript{10} the elastic distributions, particularly at the larger scattering angles \( \theta \), can be affected appreciably by inelastic effects.

By appeal to a stationary-state representation of an A–B collision in which the relative motion appropriate to each channel is described by an eikonal approximation and the internal structure by an expansion of eigenstates of the isolated species, Flannery and McCann have recently developed\textsuperscript{11,12} a multichannel eikonal treatment (MET) which reduced to the Born-wave approximation in the weak-coupling limit and to the customary impact-parameter (IP) equations\textsuperscript{9} in the heavy-particle/high-energy limit. When compared with measurements and other theoretical results, this treatment achieved notable success\textsuperscript{11} for \( e-\text{H} \) and \( e-\text{He} \) inelastic collisions at intermediate energies for which the dominant contribution to the integral cross sections arose from scattering through small angles \( \theta \leq 40^\circ \), where electron exchange was negligible. Electronic excitation in heavy-particle collisions at keV energies, which involved only small scattering angles \( \theta \leq 5^\circ \), was also described,\textsuperscript{12} although lack of experiment and of full quantal calculations prevented complete assessment of the resulting angular distributions.

Rotational excitation is, however, quite different in that detailed quantal results are available\textsuperscript{5,6} for H-H\textsubscript{2} collisions and that inelastic scattering by angles \( \theta \geq 90^\circ \) is significant. Rather than modifying MET so as to properly include the different physical trajectories for appropriate interactions, we propose, in this paper, a corresponding generalization to the heavy-particle limit of MET, i.e., to the impact-parameter prescription. The accuracy of the resulting time-dependent multistate orbital treatment, while applicable to heavy-particle collisions in general, will be assessed by comparison with the detailed quantal treatments of the collision processes\textsuperscript{5,6}.

\[
\begin{align*}
H(1s) \\ H(1s')
\end{align*}
\]

Integral and differential cross sections for these collisions as a function of scattering angle \( \theta \) will be investigated. It is worth noting that the present status of rotational excitation has been studied from various aspects...
of molecular collisions in several recent reviews, an acknowledgment of the increased theoretical interest in this subject. In this paper, we present new developments and very recent progress in theoretical, semi-classical descriptions of rotational (and vibrational) excitation.

These treatments are termed semi-classical in that the relative motion of the collision partners is initially treated separately or decoupled from the internal structure. The classical Hamilton–Jacobi equation (i.e., the Schrödinger equation in the \( h = 0 \) limit) or Hamilton's equations, eikonal, JWKB, or even Born approximations describe the decoupled relative motion in the static field. This motion is then coupled to the motion of the internal degrees of freedom which are described by full quantal procedures.

II. THEORY

We propose here two new descriptions of atomic and molecular collisions—the multistate orbital treatment, valid when classical trajectories can be accurately defined (with small quantal imprecision), and the multichannel eikonal treatment, valid for scattering mainly in the forward direction and which includes different local momenta in the various excitation channels. The overlap of the two methods is difficult to assess without resort to detailed calculation.

A. The multistate orbital treatment

The time-dependent response of the internal structure of collision partners \( A \) and \( XY \) with unperturbed Hamiltonian \( \mathcal{H}_0 \) and associated eigenstates and eigenenergies \( \phi_n \) and \( \epsilon_n \), respectively, to the mutual \( A-XY \) electrostatic interaction \( V(R(t), r) \), with time dependence generated by variation of the channel coordinate \( R(X, Y, Z) \) with time \( t \), can be described in atomic units by

\[
\Psi(r, t) = S a_\ell(t) \phi_n(r) \exp(-i\epsilon_n t),
\]

(2)

where \( r \) denotes the collective coordinates associated with the internal degrees of freedom relative to each parent center. Substitution of (2) into the time-dependent Schrödinger equation,

\[
\mathcal{H}_0 \Psi = \mathcal{H}_0 \Psi(t) = [\mathcal{H}_0 + V(R(t), r)] \Psi(t) = i \hbar \partial \Psi(t)/\partial t,
\]

(3)

results in the following set of first-order coupled differential equations for the transition amplitudes:

\[
\frac{\partial \alpha_j(t)}{\partial t} = \sum_n \alpha_j(t) V_{nj}(R(t)) \exp(i \epsilon_n t),
\]

(4a)

where \( \epsilon_n = \epsilon_n - \epsilon_a \) are the internal states, \( n = 1, 2, \ldots \), and in which the interaction matrix elements are

\[
V_{nj}(R(t)) = \langle \phi_n(r) | V(R(t), r) | \phi_j(r) \rangle. \tag{4b}
\]

The customary procedure in this description then adopts a straight-line (SL) trajectory

\[
R(t) = \rho + v_\rho t,
\]

(5)

in terms of the impact parameter \( \rho(X, Y) \) and incident velocity \( v_\rho \), taken constant, and directed along the \( Z \) axis (say), so as to effect the transformation \( Z = v_\rho t \). This produces the transition amplitudes \( \alpha_j(t) \), which can then be solved for the (asymptotic) transition probabilities \( |\alpha_j^{\text{SL}}(\rho, Z = \infty)|^2 \) by standard numerical procedures to give

\[
|\alpha_j^{\text{SL}}(\rho, Z = \infty)|^2 = \int_0^\infty |\alpha_j^{\text{SL}}(\rho, \infty)|^2 \, dp
\]

(6)

for the integral cross section for excitation of state \( j \). By appeal to a stationary-state (rather than to a time-dependent) description of the collision, it can be shown that the heavy-particle limit to the multichannel eikonal treatment (to be discussed in Sec. II.B) yields, in terms of these \( |\alpha_j^{\text{SL}}| \),

\[
\frac{d\sigma_j^{\text{sc}}}{d\Omega} = \frac{h_j}{k_i} \int_0^\infty J_j(k_i \rho \sin \theta) |\alpha_j^{\text{SL}}(\rho, \infty) - \delta_{ij}| \, d\rho \tag{7}
\]

for the differential cross section for scattering through small angles \( \theta \), where \( h_j \) and \( k_i \) are the initial and final wavenumbers of relative motion, and \( J_j \) are Bessel functions of integral order \( M_j - M_h \), the change in azimuthal quantum number of the internal structures. In the high-energy limit \( k_i = k_f \gg 1 \) and for small angle collisions (7), when integrated over all solid angles \( \Omega \) reproduces the excitation cross section (6). For larger-angle scattering, a major improvement, proposed here, is based on the use of Hamilton's equations

\[
\frac{dq_j}{dt} = \oint \frac{\partial \mathcal{U}}{\partial q_j}, \quad j = 1, 2, 3
\]

(8)

and

\[
\frac{dp_j}{dt} = \oint \frac{\partial \mathcal{U}}{\partial q_j}, \quad j = 1, 2, 3
\]

(9)

to determine the actual variation with time \( t \) of the generalized coordinates \( q_j \) and associated conjugate momenta \( p_j \), of a particle of mass \( \mu \) with the "averaged" Hamiltonian

\[
\mathcal{U} = \sum_j \frac{p_j^2}{2 \mu} + \langle \psi(r, t) | \mathcal{H}_0(r) + V(R(t), r) | \psi(r, t) \rangle, \tag{10}
\]

where the first term in the rhs is the kinetic energy of the \( A-XY \) relative motion, \( p_i^2/2\mu_i \), and the second term, the expectation energy of internal motions under the interaction \( V \) at time \( t \), is, with (2) inserted,

\[
\mathcal{U}(R(t), \Theta) = \frac{1}{\pi} \sum_r \left| \langle a^\ast_n(t) a_n(t) V_m(R) \rangle \right|^2 \tag{11}
\]

an averaged Hamiltonian or optical potential \( \mathcal{U} \) designed to couple the response (2) of the internal \( A-XY \) structure to the perturbation \( V \) back to the relative motion and vice versa. Thus, the relative motion of the collision partners \( A, XY \) is coupled at all times \( t \) to the \( A-XY \) internal motions and is given by the classical solution of a particle moving under the "optical" potential \( \mathcal{U} \) instantaneously generated at time \( t \) by the internal structure averaged over its degrees of freedom. The uncoupled limit neglects this \( \mathcal{U} \), and from (8) \( p_j \) is therefore constant, such that (5) is recovered. With this effective Hamiltonian (9), Eq. (8) therefore yields the set of additional coupled equations

\[
\frac{dq_j}{dt} = \frac{\partial \mathcal{U}(R(t))}{\partial q_j}, \quad j = 1, 2, 3
\]

(11a)

and

\[
\frac{dp_j}{dt} = \oint \frac{\partial \mathcal{U}(R(t))}{\partial q_j}, \quad j = 1, 2, 3
\]

(11b)
whose solutions are coupled at all times $t$ to the solution of (4a). For central potentials, scattering is in a plane (X$\Sigma$, say), and when $N$ eigenstates are used in (2), this procedure effectively increases the number of coupled equations from $N$ to $N+4$. Hence, (4) and (11) are solved for the column matrix $a(t) = \{a_i\}$ subject to the initial ($t=0$) boundary conditions that $X(0) = \rho$, $Z(0) = -\infty$, $(dX/dt)(0) = 0$, $(dZ/dt)(0) = u_i$, and $a_n(0) = \delta_{ni}$, where $i$ denotes the initial state.

An essential feature of the method is that the "effective" Hamiltonian $\mathcal{H}$ is time independent. Thus, total energy is conserved and is continually being redistributed between the relative motion and the internal degrees of freedom, i.e., the time derivative

$$\frac{d\mathcal{H}}{dt} = \sum_{i,j=1}^{\infty} p_i p_j + \sum_{n=1}^{N} [\delta_{mn} q_n + \delta_{mn} + i\epsilon q_n a_n] \frac{H_n}{\epsilon},$$

with the aid of (8) and (11a) reduces to $\delta\mathcal{H}/\epsilon t$, which in turn vanishes by subsequent use of (4a) for $a_i$ and $\delta_{ii}$.

For an incident flux of $N$ particles/cm$^2$/sec, a number $|q_i|^2 N d\Omega$ are scattered in channel $f$ within solid angles $d\Omega$, related to $dp$ by

$$dp = (dp/d\Omega) d\Omega,$$

where $(dp/d\Omega)_{f \sigma}$ is the Jacobian of the $(\rho, \sigma)$ transformation, is the classical differential cross section for elastic scattering by the optical potential $U$ determined previously from the solutions $a(t)$ in (10).

Hence

$$\frac{d\mathcal{H}}{dt} = |q_i(X', Y', Z')|^2 \left(\frac{dp(t)}{d\Omega}\right)_{f \sigma},$$

(14)

is the differential cross section for $i-f$ transitions in this time-dependent orbital method. In a one-channel case ($N=1$), the classical formulas$^{10}$ for potential scattering by $V_{\mu}(R(t))$ are recovered. Hence this treatment is the natural semiclassical generalization of classical potential scattering to include inelastic processes.

Thus, (7) and (14) are complementary, in the sense that (7) pertains only to small angle scattering while (14) is strictly valid for well-defined trajectories $R$, i.e., when the angular uncertainty $\delta \theta = (k_{\rho} R)^{-1}$ is much less than either the scattering angle $\theta = 2\mu \nu_{\mu}(R)/k_{\rho}^2$, when small, or unity, otherwise. Thus (14) will eventually fail in a range of very small $\theta$, a range which is covered by the multichannel eikonal treatment (7). The angular range of overlap between the above two treatments, (7) and (14), is difficult to assess without resort to explicit calculation.

B. The multichannel eikonal treatment

In an effort to clarify more fully the basis of this method, and in the interest of providing full semiclassical descriptions of collisions involving rotational transitions, we develop in this section the present multichannel eikonal treatment by a method alternative to that previously described. The wavefunction for the mutual scattering of an atom (or ion) $A$ and a molecular species $XY$, in general, by their mutual interaction $V(r, R)$ at nuclear separation $R(X, Y, Z)$, is, in the cm frame,

$$\psi^* (r, R) = \psi^* (r) e^{iS - \gamma} + \int d\tau' dR' G(r, R; r', R') \times V(r', R') \psi^* (r', R'),$$

(15)

where the two-particle Green's function $G_{\mu}$ appropriate to $\mathcal{H}$, the Hamiltonian of the unperturbed system of energy $E_i$ at infinite $R$, satisfies

$$(E_i - \omega_{\mu} + i\epsilon) G_{\mu}(r, R; r', R') = \delta(r-r') \delta(R-R'),$$

(16)

In which the composite internal coordinates are denoted by $r$ relative to each parent nucleus. The free particle Green's function, which propagates the effect of the interaction $V$ at $(r', R')$ to $(r, R)$, can be expanded in terms of the complete set of eigenfunctions of $\mathcal{H}$ as

$$G_{\mu}(r, R; r', R') = \lim_{\epsilon \to 0} \frac{1}{(2\pi)^3 \sqrt{2\mu}} \int \phi_\mu^+(r') \phi_\mu^-(r') e^{i\Sigma_{\mu}(r'-r)} / \rho_{\mu}(r') + \int d\tau' dR' G_{\mu}(r, R; r', R') \times V(r', R') \phi_\mu^+(r'),$$

(17a)

where $\phi_\mu(r)$ describes the internal structure at infinite nuclear separation $R$, where the relative motion is planar with propagation vector $k_{\rho} = k_{\rho} X + k_{\rho} Y$. For heavy-particle collisions and for electron-atom inelastic collisions at intermediate and high impact energy, scattering about the forward direction contributes most to the total cross section, and it is therefore a good approximation to assume that the major contributions to the propagator (17a) arises only those waves at $Z' < Z$ with $k^2 = k_{\rho}^2 + k_{\rho}^2$ such that

$$G_{\mu}(r, R; r', R') = \lim_{\epsilon \to 0} \frac{1}{(2\pi)^3 \sqrt{2\mu}} \int \phi_\mu^+(r') \phi_\mu^-(r') \int e^{i\Sigma_{\mu}(r'-r)} / \rho_{\mu}(r') + \int d\tau' dR' G_{\mu}(r, R; r', R') \times V(r', R') \phi_\mu^+(r'),$$

(17b)

The reduction of (17a) to (17c) can also be obtained by the method of stationary phase (cf. Schiff$^{17}$). The multichannel eikonal approximation follows by setting

$$\psi^* (r, R) = \sum_{\mu} A_{\mu}(r, Z) \phi_\mu^+(r) e^{iS_{\mu}(r)}$$

(18)

where the eikonal $S_{\mu}$ for the relative motion in excitation channel $\mu$ under the static interaction

$$V_{\mu}(R) = \phi_\mu^+(r) \mid V(r, R) \mid \phi_\mu^-(r),$$

(19)
the diagonal elements of $4(b)$, satisfies

$$\rho S_\omega^2 - i h (\nabla^2 S_\omega) - \frac{k_\omega^2}{4} V_m = \frac{2 \mu}{h^2} V_m = 3 \alpha_n (R) \tag{20}$$

which, on differentiation, yields

$$\frac{d}{dz} \left[ A_n (e^{i \phi_s}) \right] = - i \frac{\mu}{\hbar^2} \frac{S_\omega}{\rho} A_n (p, Z) V_m (R) e^{i \phi_s} \tag{23}$$

The projection of (21) onto the orthonormal set $\phi_s (r)$ is

$$\langle \phi_s (r) e^{i \phi_s (\phi)} \rangle | V (r, R) \rangle = \frac{\rho}{\hbar^2} \frac{S_\omega}{\rho} A_n (p, Z) V_m (R) e^{i \phi_s} \tag{24}$$

The above derivation therefore shows that the multichannel eikonal treatment. The transition matrix for rearrangement collisions between the projectile at $R$ and a target particle at $r_i$ is obtained from (25a) by the $R \rightarrow r_i$ interchange in the wavefunction for the final state $f$.

The above derivation therefore shows that the multichannel eikonal treatment is based on the following three assumptions: (a) the Green's function (17c); (b) $| V S_n | = \alpha_n$; and (c) a straight-line trajectory, all included within a restricted basis set of $N$ target states. This treatment can be immediately generalized, so that the eikonal $S_n$ in (20) is the integral of $\alpha_n$ along the actual classical trajectory $s_n$ appropriate to $V_m$, with the result that the coupled set (24) are then obtained with $Z$ replaced by $s_n$.

The basic formula (25a) for the scattering amplitude can be further reduced for two-particle interactions for which $V_f (R) = V_f (p, Z) e^{i \phi_s}$, where $\Delta$ is $(M_f - M_l)$, the change in azimuthal quantum number. This property permits the following definition of a phase $\Phi$-independent transition amplitude $B_f$:

$$B_f (p, Z) = A_f (p, Z) \exp (-i \Delta \Phi) \exp i \int (x_f - k_f) dZ . \tag{26}$$

After some algebraic manipulation, the scattering amplitude in (25a) can be expressed as

$$f_s (\theta, \phi) = -i e^{i \phi_s} \int J_n (K') \left[ I_n (0, \theta) - H_n (0, \theta) \right] dK' \tag{27}$$

where $J_n$ and $H_n$ are Bessel functions of integral order $(M_i - M_l)$ and $K'$ is the $Y^2$ component of the momentum change $K = k_f - k_i$. The collision functions

$$I_1 (\theta, \phi) = \int_0^\infty \left[ \frac{3B_f (p, Z)}{8Z} \right] \exp (i \alpha Z) dZ \tag{28}$$

and

$$I_2 (\theta, \phi) = \int_0^\infty \left[ \frac{X_f (x_f - k_f)}{8Z} \right] B_f (p, Z) \exp (i \alpha Z) dZ \tag{29}$$

contain a dependence on the scattering angle $\theta$ via

$$\alpha = k_f (1 - \cos \theta) = 2 k_i \sin \frac{1}{2} \theta . \tag{30}$$

The difference between the $Z$ component of the momentum change $K$ and the minimum change $k_i - k_f$ in the collision. The coupling (phase $\Phi$-independent) amplitudes $B_f$ are solutions of the following set of $N$-coupled differential equations

$$\frac{i \hbar^2}{\mu} \frac{S_\omega}{\rho} (p, Z) \frac{d B_f (p, Z)}{dZ} = \left[ \frac{R^2}{\mu} \frac{S_\omega}{\rho} \frac{S_\omega}{\rho} (x_f - k_f) + V_f (p, Z) \right] B_f (p, Z) \tag{31}$$

solved subject to the asymptotic boundary conditions $B_f (p, - \infty) = 0$.

We note that in the absence of all couplings, except that directly connecting the initial and final channels, i.e., $B_2 = 0$, either (25b) or (27) yields

$$f_s (\theta, \phi) = - \frac{1}{4 t_0} \int V_f (R) \exp (i K \cdot R) dR \tag{32}$$

which is the Born-wave scattering amplitude. Obviously, a variety of useful approximations readily follow, and these have already been outlined in Ref. 11. In particular, it can also be shown in the heavy-particle limit, i.e., when the wavenumbers can be expanded as

$$k_f = k_i - \frac{\epsilon_f}{\hbar v_i} \left( 1 + \frac{\epsilon_f}{2 \mu v_i} + \cdots \right) \epsilon_f = - \epsilon_i, \ h k_i = \mu v_i . \tag{33}$$
\[ f_i(j, \theta, \phi) = -i^{i+1} h \int_{\Delta_0} J_i(K) \left[ C_0(\rho, \alpha) - \delta_{ij} \right] \rho \, d\rho, \]  

(34a)

where \( K^2 = -K^2 + \frac{e^2}{\alpha} / \rho \) and the amplitudes \( C_0 \) satisfy

\[ ih \left( \frac{\partial}{\partial \rho} \right) C_0(\rho, Z) \exp \left( \frac{i \Delta E Z}{\hbar} \right). \]  

(34b)

We note on the adoption of the space–time transformation \( Z = \sqrt{2} t \) that the set of coupled equations (34b) is identical with (4a), which followed from a time-dependent formulation. Also, when the impact energy is sufficiently high \( (E_i = E_f) \) and when the scattering is into small angles \( \theta \) such that \( K^2 = 2k(1 - \cos \theta) \) can be used in (34a) together with

\[ \int_{\Delta_0} n_{\Delta_0 = \pi} J_i(\Delta_0) J_{\Delta_0}(\Delta_0) K \, dK = \frac{1}{\rho} (\rho - \rho') \]  

(35)

such that (6) for the excitation cross section can be recovered from (34a) when squared and integrated over all solid angles.

### C. Interaction potentials

#### H–H₂ system

In order to directly compare the above semiclassical treatments with the detailed quantum results of Chu and Dalgarno, we adopt the same interaction as proposed by Wolken et al. for the H–H system in the ground electronic state, i.e., the Porter–Karplus (PK) semiempirical short-range potential surface\(^{16}\) joined to the correct long-range van der Waals \( R^{-8} \) interaction. The PK surface was expanded according to

\[ V_{\text{PK}}(R, \theta) = \sum_{\lambda = 1}^\infty \sum_{\mu = -\lambda}^{\lambda} v_{\lambda, \mu}(R) P_\lambda(\cos \theta), \quad \cos \theta = \cos \lambda, \]  

(36)

where \( R \) and \( \theta \) are, respectively, the nuclear separation vector of H₂ and the relative channel vector. For the pure rotation transitions in (1), the interaction averaged over the ground vibrational level to give

\[ V_{\text{PK}}(R, \theta) = \sum_{\lambda = 1}^\infty \sum_{\mu = -\lambda}^{\lambda} v_{\lambda, \mu}(R) P_\lambda(\cos \theta), \]  

(37)

where the coefficients \( v_{\lambda, \mu}(R) \) have been tabulated\(^{15}\) for \( \lambda = 0, 2, \ldots, 12 \). Chu and Dalgarno\(^{6}\) have shown that the first two terms of (37) dominate the scattering and are sufficient for an accurate determination of the rotational transitions in (1).

The short-range PK contribution (37) is joined to the following correct long-range interactions\(^{17}\) (in eV):

\[ v_{\text{PK}}^{\alpha}(R) = 511.088 \exp(-1.9R) - 251.546/R^6 \] (eV), \n
(38a)

\[ v_{\text{PK}}^{\beta}(R) = 346.664 \exp(-2R) - 27.8767/R^6 \] (eV), \n
(38b)

with \( R \) in \( a_0 \). Thus the H–H₂ interaction, adopted here and in the full quantum treatment, has the following form:

\[ V(R, \theta) = \sum_{\lambda = 0}^\infty \sum_{\mu = -\lambda}^{\lambda} v_{\lambda, \mu}(R) P_\lambda(\cos \theta), \]  

(39)

where 4.6 \( a_0 \) and 7 \( a_0 \) are the respective intersections of \( v_\alpha^\beta \) with \( v_\beta^\beta \) and of \( v_\alpha^\beta \) with \( v_\beta^\alpha \). We note, however, that the slopes are discontinuous at the intersection points, with the result that a nonphysical rainbow effect will be evident in the semiclassical cross sections (14).

In order to compare also with the detailed quantum results of Hayes et al.,\(^{5}\) we also use the H–H₂ interaction of Tang,\(^{16}\) i.e., (37) with

\[ v_0(R) = (90.2/R^4) \exp(-0.617R + 1.234) \] \n
(40a)

\[ -251.546/R^6 \] (eV) \n
(40b)

and

\[ v_0(R) = 92.04 \exp(-1.87R) \] \n
(40b)

Although this potential is weaker and less anisotropic than (39) and hence will yield smaller elastic and inelastic cross sections, it is nevertheless continuous, such that no nonphysical rainbow will be evident. So as to ensure that the calculations are internally consistent, the energy levels\(^{13}\) used with (39) and (40) are those associated with the Morse curve.\(^{15}\)

#### Matrix elements

Since H₂ is a linear molecule in the \( \Sigma_g^+ \) ground electronic state, the eigenfunctions for the rotational state \((J, M)\) are therefore given by the pure spherical harmonics \( Y_{J\lambda}(\hat{\Omega}) \). With the aid of (36), the interaction matrix elements (4b) appropriate for rotational transitions reduce to

\[ \langle JM | V | J_0 M_0 \rangle = \sum_{\lambda = 0}^J \frac{4\pi}{2J+1} v_{\lambda, M}(R) \sum_{\mu = -\mu}^{\mu} Y_{\lambda\mu}(\hat{\Omega}) \] \n
\[ \int Y_{\lambda\mu}^*(\hat{\Omega}) Y_{\lambda\nu}(\hat{\Omega}) Y_{J_0 \nu}(\hat{\Omega}) d\Omega, \]  

(41)

where

\[ \int Y_{\lambda\mu}^*(\hat{\Omega}) Y_{\lambda\nu}(\hat{\Omega}) Y_{J_0 \nu}(\hat{\Omega}) d\Omega = \left\{ \begin{array}{c} \frac{(2J+1)(2\lambda+1)}{4(2J+1)} \end{array} \right\}^{1/2} \langle J_\lambda | J_\lambda \rangle \langle J_\lambda | J_\lambda \rangle \langle J_\lambda | J_\lambda \rangle \]  

(42)

in terms of the Clebsch–Gordan coefficients \( \langle i,j_1,i_2 | m_1,m_2 | J,M \rangle \) and where \( Y_{\lambda\mu} = (-1)^M Y_{\lambda\mu} \) defines the phase convention. The inclusion of an extra rotational level \( J \) in the basis expansion (2) therefore implies an additional \((J+1)\) degenerate substates such that the number of coupled equations to be solved grows rapidly with each addition of \( J \). Rabitz\(^{18}\) has, however, introduced an effective potential method which eliminates the \( M \) dependence in (41), thereby permitting considerable reduction in the number of equations that require solution. Thus, the orientation dependent interaction (41) is replaced by

\[ \langle J | V_{\text{eff}} | J_0 \rangle = (-1)^J (2J+1)(2J_0+1)^{1/2} \] \n
\[ \times \sum_{\lambda = 0}^J \frac{v_{\lambda, M}(R)}{(2\lambda+1)^{1/2}} \left( \begin{array}{c} J \lambda \nu \end{array} \right)_{\lambda \nu}, \]  

(43)

where

\[ \eta = \frac{1}{2} \left( |J_0 - J| + J_0 + J \right) = \max(J, J_0), \]  

(44)

and the 3\( j \)-symbol is either
for integral $g = \frac{1}{2} (J + \lambda + J_0)$, or zero otherwise. Thus the use of the effective potential (43) causes a reduction, in a case where $J = 0, 2, 4, 6$ rotational levels (with 16 substates) are included, from 20 coupled equations [(4a), (11a), (11b)] to 8. We also note that this procedure provides a spherical symmetric optical potential for (10) such that the solution of (11) becomes more simple than that involved with orientation-dependent interactions.

The above suggestion of Rabitz will, however, be tested in this paper by performing both explicit 16-state and four-level semiclassical calculations with the use of (41) and (43), respectively. In addition, the accuracy of the present semiclassical procedures will be assessed by direct comparison of the resulting integral and differential cross sections for various rotational transitions in the H–H$_2$ collision system at several impact energies $E(eV)$, the interactions and of Chu and Dalgarno [who used (43)] within a quanlal treatment which closely coupled the $J = 0, 2, 4, 6$ levels.

He–H$_2$ system. For direct comparison with the full quantal results of McGuire and Kouri, the hypersurface determined by Krauss and Mies is adopted for the He–H$_2$ interaction. Hence, for He remaining in the ground vibrational state, the He–H$_2$ interaction in eV is given by (39) with $v_{0x}^2$ and $v_{2x}^2$ replaced by

$$v_{0x}^2(R) = 0.806 \exp[-3.49(R-1.6)] \text{ (eV)}$$

$$v_{2x}^3(R) = 0.157 \exp[-3.47(R-1.6)] \text{ (eV)}$$

for all $R$ measured in Å.

Direct comparison with the quantal results also necessitates the use of a corresponding semiclassical analogue to the "$j_+$-conserving method." During a slow collision, there is a natural tendency, especially for intermediate encounters with impact parameter $\rho$ in the range $\rho_1 < \rho < \rho_2$, for the quantization axis of each collision partner is reluctant to follow the rapid rotation of $R$. These effects are, of course, theoretically acknowledged by including couplings between the $M$ substates but are ignored in the perturbed rotating atom approximation (PRA). Therefore, the PRA approximation (or its quantal equivalent, the $j_+$-conserving method) is expected to be invalid for distant encounters in small-angle scattering, and (b) for very close encounters in large-angle scattering, and (c) for cases involving sufficiently high relative speeds so that the quantization axis cannot adjust itself to the rapid rotation of $R$. The PRA approximation will be investigated further by direct calculation for He–H$_2$ collisions.

III. RESULTS AND DISCUSSION

A computer program was written for the solution of the coupled equations (4), (11), and (34) by the Burlish–Stoer rational extrapolation technique, which provided great accuracy and efficiency. A cubic spline interpolation routine was used for the interpolation of both the Porter–Karpus surface and of $(dp/d\theta)$. Multistate orbital treatments (MOT) and multichannel eikonal treatments (MET) developed previously in Secs. II.A and II.B, respectively, are applied to the following collision processes:

$$H(1s) \rightarrow H_2(X^1\Sigma^+_g, v=0, J) \rightarrow H(1s)$$

at several impact energies $E$(eV), the interactions and approximations being chosen so as to provide direct comparison with corresponding full-quantal treatments carried out previously for these processes.

H–H$_2$ collisions: Figure 1 displays differential cross sections for 0–0 elastic collisions at 1 eV determined from (a) a four-level $(J=0, 2, 4, 6)$ optical-potential orbital treatment MOT, (b) a one-state pure classical orbital description $(i a_i=1)$, and (c) a four-channel eikonal treatment MET, together with (d) the corresponding multi-channel quantal results $Q$ of Chu and Dalgarno (who used the technique of Rabitz). The influence of inelastic effects on elastic scattering is manifested quite clearly by the difference between (a) and (b). Thus, while essential agreement exists at small angles between (a) and (b), inelastic effects become increasingly important with larger scattering angles $\theta$, as expected, since rotational excitation primarily results from close encounters. Comparison between (a) and (d) demon-
strates that inclusion of inelasticity, as in (a), is required so as to preserve accord with the full quantal results (d) for the "background" scattering (i.e., with quantal oscillations, due to phase-shift interference, removed) for \( \theta > 30^\circ \). Also, in spite of the validity in describing large-angle scattering, a one-state pure classical orbit is therefore inadequate, e.g., calcula-

TABLE I. Integral cross sections (A) for H+H₂ (J) → H+H₂ (J') at impact energies E(eV) = 0.5, 1, and 1.5.

<table>
<thead>
<tr>
<th>J → J'</th>
<th>0.5</th>
<th>1.0</th>
<th>1.5</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(A)</td>
<td>(B)</td>
<td>(C)</td>
</tr>
<tr>
<td>0--0</td>
<td>37.2</td>
<td>39.0</td>
<td>41.2</td>
</tr>
<tr>
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<td>40.8</td>
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</tr>
<tr>
<td>4--4</td>
<td>41.9</td>
<td>43.7</td>
<td>43.3</td>
</tr>
<tr>
<td>6--6</td>
<td>41.6</td>
<td>43.1</td>
<td>45.4</td>
</tr>
<tr>
<td>0--2</td>
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<td>4.27</td>
<td>4.02</td>
</tr>
<tr>
<td>2--0</td>
<td>0.81</td>
<td>0.85</td>
<td>0.88</td>
</tr>
<tr>
<td>2--4</td>
<td>1.07</td>
<td>0.96</td>
<td>0.63</td>
</tr>
<tr>
<td>4--2</td>
<td>0.60</td>
<td>0.66</td>
<td>0.45</td>
</tr>
<tr>
<td>4--6</td>
<td>0.25</td>
<td>0.22</td>
<td>0.14</td>
</tr>
<tr>
<td>6--4</td>
<td>0.18</td>
<td>0.21</td>
<td>0.18</td>
</tr>
<tr>
<td>0--4</td>
<td>0.228</td>
<td>0.192</td>
<td>0.104</td>
</tr>
<tr>
<td>4--0</td>
<td>0.020</td>
<td>0.023</td>
<td>0.017</td>
</tr>
<tr>
<td>4--2</td>
<td>0.014</td>
<td>0.010</td>
<td>0.0004</td>
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<tr>
<td>6--2</td>
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<td>0.008</td>
<td>0.0006</td>
</tr>
<tr>
<td>0--6</td>
<td>2.14e0</td>
<td>1.32e0</td>
<td>4.24</td>
</tr>
<tr>
<td>6--0</td>
<td>1.0e0</td>
<td>1.3e0</td>
<td>8.5e0</td>
</tr>
</tbody>
</table>

Column (A): Present semiclassical four-level orbital treatment $\text{MOT}(J=0, 2, 4, 6)$.
Column (B): Elastic scattering. Four-level eikonal approximation $\text{MET}(0 \leq \theta \leq 20^\circ)$ combined with MOT for $\theta = 20^\circ$.
Column (C): Inelastic scattering. Present semiclassical MOT cross sections $\sigma(\theta)$.

Exponent gives power of 10 by which entry is to be multiplied.

Effective potential method used in all treatments.

Column (C): Corresponding four-level quantal treatment $\sigma(\theta)$.

TABLE I shows that the probability for H₂ remaining rotationless at $\theta=80^\circ$ is $0.5$, rather than unity, as in (b).

Figure 2, a display of the 2--2 and 4--4 transitions at two impact-energies, shows again that inclusion of inelasticity is needed to introduce harmony with the quantal distributions. The inelastic effects are controlled by the variation with $\theta$ and $E$ of the cross sections for rotational excitation. The semiclassical and integral cross sections given in Fig. 2 show good agreement.

However, the one oscillation observed in the semiclassical distributions, in both Figs. 1 and 2, at $-10^\circ$ arises directly from a nonphysical rainbow effect introduced by the artificial graft of the short-range potential (37), to yield (39) as the full interaction $P$. Figure 3 isolates this rainbow, by exhibiting an infinite $|dp/d\theta|$ in the variation of $p$ vs $\theta$ for H-H₂ collisions at 1 eV. The rainbow angle (at which $dp/d\theta=0$) decreases with increase of $E$. Figure 3 also shows that this nonphysical rainbow vanishes on using the Tang interaction $T$, Eq. (40), which is continuous. The full quantal values based on $p$ and $\theta$ given at $5^\circ$ intervals in Figs. 1 and 2 also contain this effect, although in contrast to the semiclassical results, all angular momenta (or $p$) contribute here to a given $\theta$, thereby causing a suppressed effect, evident in the inset of Fig. 1. Because of this peculiarity, strict comparison between (a) MOT, (b) MET, and (d) the quantal results are precluded in the angular range $5^\circ - 20^\circ$ about the rainbow angle. The inset in Fig. 1 illustrates the very slow divergence of the semiclassical MOT results for scattering in the forward direction. With the aid of Fig. 3, the quantal imprecision to scattering by small angles $\sim (h\theta R)^{-1}$ at 1 eV.

In order to obtain the actual inelastic cross sections $\sigma$ from those $\sigma$ determined directly from the effective po-
FIG. 5. Differential cross sections ($\lambda^2$/sr) for $J = 0 - 0$ elastic H-H$_2$ ($J$) collisions at 0.25 eV. SC: Present four-state ($J, M = 0, 2, 0, 2, 1, 2, 2$) orbital treatment. ME: Present four-state eikonal treatment. Q: Corresponding quantal results. SC: integral SC cross section. Q(ESC): integral cross section determined from combination of ME for $0 \leq \theta \leq 50^\circ$ and of SC for $\theta > 30^\circ$.

TABLE II. Integral cross sections $\sigma_{J' J}$ given by a 16-state, four-state, and four-level semiclassical orbital treatments of the collision H + H$_2$($J, M$) - H + H$_2$(J, M) at 1 eV with interaction $T$.

<table>
<thead>
<tr>
<th>$J_J' M_J$</th>
<th>$J_J' M_J$</th>
<th>16-State</th>
<th>Four-state</th>
<th>Four-level</th>
</tr>
</thead>
<tbody>
<tr>
<td>00 - 0</td>
<td>0.185</td>
<td>0.197</td>
<td>0.197</td>
<td>0.197</td>
</tr>
<tr>
<td>00 - 2</td>
<td>0.451</td>
<td>0.471</td>
<td>0.471</td>
<td>0.471</td>
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<tr>
<td>00 - 0</td>
<td>0.673</td>
<td>0.909</td>
<td>0.913</td>
<td>0.913</td>
</tr>
<tr>
<td>00 - 4</td>
<td>4.16$^a$</td>
<td>5.93$^a$</td>
<td>5.93$^a$</td>
<td>5.93$^a$</td>
</tr>
<tr>
<td>00 - 1</td>
<td>8.17$^a$</td>
<td>8.17$^a$</td>
<td>8.17$^a$</td>
<td>8.17$^a$</td>
</tr>
<tr>
<td>00 - 2</td>
<td>8.43$^a$</td>
<td>8.43$^a$</td>
<td>8.43$^a$</td>
<td>8.43$^a$</td>
</tr>
<tr>
<td>00 - 3</td>
<td>5.59$^a$</td>
<td>5.59$^a$</td>
<td>5.59$^a$</td>
<td>5.59$^a$</td>
</tr>
<tr>
<td>00 - 4</td>
<td>1.44$^a$</td>
<td>1.44$^a$</td>
<td>1.44$^a$</td>
<td>1.44$^a$</td>
</tr>
<tr>
<td>00 - 6</td>
<td>2.78$^a$</td>
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<tr>
<td>00 - 4</td>
<td>2.03$^b$</td>
<td>2.03$^b$</td>
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<tr>
<td>00 - 6</td>
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<tr>
<td>00 - 1</td>
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</tr>
<tr>
<td>00 - 2</td>
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<td>1.02$^b$</td>
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<td>7.55$^b$</td>
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</tr>
<tr>
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<td>00 - 6</td>
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</tr>
<tr>
<td>00 - 4</td>
<td>4.71$^b$</td>
<td>4.71$^b$</td>
<td>4.71$^b$</td>
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</tr>
</tbody>
</table>

$^a$ Combined transition to substates $+ M_J$ and $- M_J$. See note added in proof.
$^b$ Inclusion of all substates $+ M_J$ of $J = 0, 2, 4$, and 6 rotational levels.
$^c$ Inclusion of all substates of $J = 0$ and 2 rotational levels.
$^d$ Inclusion of $J = 0, 2, 4$, and 6 levels within effective potential method of Rabitz.
$^e$ Transition between rotational levels specified.
$^f$ Exponent gives power of 10 by which entry is to be multiplied.

tential method, the following transformation was adopted:

$$\frac{d\sigma}{d\Omega}(J_1 \rightarrow J_2) = \left[ \frac{2J_1 + 1}{2J_2 + 1} \right]^{1/2} \frac{d\sigma}{d\Omega}(J_1 \rightarrow J_2), \quad J_2 > J_1.$$  (49)

In Table I we present values of the integral cross sections

$$\sigma_{J' J} = \int |a_J|^2 \frac{d\sigma(a)}{d\Omega} d\Omega = 2\pi \int_0^\pi |a_J|^2 \rho d\theta$$  (50)

and of $(h_k / \hbar) \sigma_{J' J}$ for all the elastic and inelastic transitions in (48). Although the ratio $(h_k / \hbar) \sigma_{J' J}$ does not follow exactly from the present semiclassical treatment, its inclusion nonetheless introduces even better accord of the semiclassical cross sections with the full quantal results. The agreement is very good for the elastic and the stronger inelastic transitions ($\Delta J = 2$) and is improving for the weaker $\Delta J = 4$ transitions as $E$ increases $\sim 1.5$ eV.

Figure 4, a display of the angular distribution in H-H$_2$ collisions at 1.5 eV, confirms that the nonphysical rainbow present with interaction $P$ vanishes on using the continuous interaction $T$ (see also Fig. 1). It also shows that the uncertainty in the H-H$_2$ interaction, as evidenced by the differences entailed in using $P$ and $T$, introduces error greater than the neglect of inelastic effects. Potential $T$ is (a) less anisotropic than $P$, such that inelastic effects are reduced and elastic scattering is therefore enhanced at large $\theta_j$ and is also (b) weaker...
at large distances with, consequently, less elastic scattering by $\theta \leq 20^\circ$ which provides most of the integral cross section which is therefore reduced.

Figure 5 exhibits the impressive agreement of the quantal differential cross sections of Hayes et al. (who used potential $T$) at 0.25 eV with both the corresponding four-state eikonal and four-state orbital treatments, in which the substates $(J, \pm M) = (0, 0), (2, 0), (2, 1)$, and $(2, 2)$ are closely coupled. The eikonal treatment successfully describes scattering by $\theta < 30^\circ$, while the orbital description is excellent for $\theta > 5^\circ$.

Figure 6 shows that the differential cross sections for the $J=0-2$ rotational transition in $H_2$ by $H$ impact at 0.25 eV are much smaller than those for the elastic case, in accord with the small inelastic effects evident in Fig. 4 when interaction $T$ is used. The present semiclassical results are finite for scattering in the forward direction (since $\alpha_\gamma$ tends to zero for these distant encounters) and are in general harmony with the quantal distribution $Q$. While the procedure of Rabitz introduces a deeper minimum in the angular distribution, the integral cross sections are essentially unaffected. The semiclassical integral cross sections $\sigma_{SC}$ displayed in Fig. 6 are calculated from (12) alone; multiplication of (12) by the ratio $(k_f/k_i)$ yields $\sigma_{SC} = 0.34 \text{ Å}^2$, which is closer...
He–H₂
E = 0.9 eV
0–0

σ(SC) = 4.34 Å²
σ(RA) = 3.33 Å²
σ(R) = 2.93 Å²
σ(O) = 1.13 Å²

He–H₂ (b)

Since T is less anisotropic than P, the corresponding integral cross sections are therefore much smaller (cf. also Table I). The figures also show that rotational excitation in neutral–neutral collisions mainly arises from repulsive encounters causing scattering through large angles. The spikes in Fig. 7(b) arise from the nonphysical rainbow in potential P.

Finally, in order to test both the convergence of the wavefunction expansion in rotational states and the effective potential method R of Rabitz, a 16-state treatment which closely coupled all the ±M substates of the J = 0, 2, 4, and 6 rotational levels, a four-state treatment which ignored all levels with J ≥ 4, and a four-level treatment within method R were all performed. In Table II we present the integral cross sections for all the included transitions at 1 eV. The four-state treatment agrees well with the more elaborate 16-state calculations, since the transitions probabilities to the higher J = 4 and 6 states are extremely small. Curiously enough, while the four-level treatment within R provides reasonable cross sections for the ΔJ = 2 and 4 transitions, it considerably underestimates both the elastic and the much weaker ΔJ = 6 transitions.

It is important to note that the inclusion of the J substates necessitates the determination of the semiclassical trajectory under an orientation-dependent optical potential (10). It is also worth noting that all the present computations on all the various transitions in the H–H₂ collision entailed ~1 h Cyber 74 (CDC 6600 processor).

He–H₂ collisions: Part of this investigation will in-

accord with the quantal result.

Figures 7(a)–7(c) illustrate differential cross sections for ΔJ = 2 transitions with interactions T and P, re-

FIG. 9. Differential cross sections (Å²/sr) for J = 0–0, 0–2, and 0–4 transitions in He–H₂ (J = 0) collisions at 1.2 eV. 
- : corresponding quantal results.
- - : corresponding quantal results.

spectively, and for ΔJ = 4 transitions with interaction T. Since T is less anisotropic than P, the corresponding integral cross sections are therefore much smaller (cf. also Table I). The figures also show that rotational excitation in neutral–neutral collisions mainly arises from repulsive encounters causing scattering through large angles. The spikes in Fig. 7(b) arise from the nonphysical rainbow in potential P.

Finally, in order to test both the convergence of the wavefunction expansion in rotational states and the effective potential method R of Rabitz, a 16-state treatment which closely coupled all the ±M substates of the J = 0, 2, 4, and 6 rotational levels, a four-state treatment which ignored all levels with J ≥ 4, and a four-level treatment within method R were all performed. In Table II we present the integral cross sections for all the included transitions at 1 eV. The four-state treatment agrees well with the more elaborate 16-state calculations, since the transitions probabilities to the higher J = 4 and 6 states are extremely small. Curiously enough, while the four-level treatment within R provides reasonable cross sections for the ΔJ = 2 and 4 transitions, it considerably underestimates both the elastic and the much weaker ΔJ = 6 transitions.

It is important to note that the inclusion of the J substates necessitates the determination of the semiclassical trajectory under an orientation-dependent optical potential (10). It is also worth noting that all the present computations on all the various transitions in the H–H₂ collision entailed ~1 h Cyber 74 (CDC 6600 processor).

He–H₂ collisions: Part of this investigation will in-

FIG. 8. Differential cross sections (Å²/sr) (a) for elastic scattering and (b) J = 0–2 transitions in He–H₂ (J = 0) collisions at 0.9 eV. SC: Present nine-state orbital treatment alone, with rotating axis approximation RA, or with effective potential method R. σ: Corresponding integral cross sections obtained from p integration in (50). Q: quantal J₂-conserving treatment.
volve direct comparison with the full quantal results of McGuire and Kouri, who closely coupled the \( J = 0, 2 \), and 4 rotational levels within the \( J \)-conserving method and who adopted the interaction \( \gamma \) (46) determined from that given by Krauss and Mies. The accuracy of the rotating axis approximation PRA will also be assessed.

In Table III we present differential cross sections for elastic scattering in \( \text{He-H}_2 \) (\( J = 0 \)) collisions at 0.9 eV determined from the following three semiclassical schemes: (a) the present multistate orbital treatment MOT with all the substates \( J, M = 0, 0; 0, 2; 2, 2; 4, 0; 4, 1; 4, 2; 4, 3; 4, 4 \) closely coupled, (b) MOT with the \((0, 0)\) and \((4, 0)\) substates closely coupled within the PRA approximation, and (c) MOT with the \((J = 0, 2, 4)\) and 4 levels closely coupled within the effective potential method. Columns 2–4 in Table III illustrate the effect of the above approximations (a)–(c), respectively, the accuracy of which can be assessed by comparing with the full quantal results (d) and (e) in columns 5 and 6, which are the respective quantal analogues of approximations (a) and (b). The agreement between (a) and (d) and between (b) and (e) can be considered as excellent. We note that the PRA approximation (b) is somewhat inadequate for small-angle scattering, i.e., distant encounters, as expected. Its closeness with (a) for large-angle scattering simply indicates that the required repulsive encounters are not sufficiently close to cause breakdown of this approximation. The PRA approximation which causes some decrease in the scattering through small angles is therefore apparently successful in describing elastic collisions at 1 eV, since here both the distant and close encounters, at which it fails, are not important. On the other hand, the effective potential method consistently overestimates the cross section.

Table IV presents similar findings for the 0–2 rotational transitions of \( \text{He-H}_2 \) collisions at 1.2 eV and shows excellent agreement with the detailed quantal angular distributions. These methods would prove particularly valuable for high-energy collisions when the inclusion of the many rotational and vibrational channels, which is prohibitively difficult and time consuming in full quantal treatments, is relatively straightforward. Moreover, as mentioned in the introduction and in Sec. II.B, the multichannel eikonal treatment can be generalized so as to include appropriate trajectories with the result that electron–molecule collisions can be described. Also, preliminary semiclassical treatments of other collisions, e.g., the Li⁺–\( \text{H}_2 \), Li⁺–N\(_2\), and H–HF systems for which the interactions are stronger, have longer range and are more anisotropic than those evident in the present neutral–neutral collisions, reveal that the present procedures are extremely successful when compared with full quantal treatments.

![Table III](image-url)

<table>
<thead>
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<th>( \theta ) (degree)</th>
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<th>Quantal treatment</th>
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<td>( \text{PRA} )</td>
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<td>165</td>
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</tbody>
</table>

\(^{a}\) Present nine-state \((0, 0; 2, 0; 4, 0)\) rotating-atom orbital treatment.

\(^{b}\) Three-state \((0, 0; 2, 0; 4, 0)\) rotating-atom orbital treatment.

\(^{c}\) Three-level \((J = 0, 2, 4)\) orbital treatment within effective potential method.

\(^{d}\) Nine-state close-coupling quantal method.

\(^{e}\) \( j \)-Conserving quantal method.

\(^{f}\) Exponent gives power of 10 by which entry is to be multiplied. Parentheses denote values at 0.6° which are closest to the quantal values at 0° scattering.

\(^{g}\) Obtained from \( \theta \) integration in (50).

Note added in proof: For a given rotational level \( J \), the associated \((2J + 1)\) substates \( M \) are explicitly acknowledged in the basis set expansions (2) and (18). The
\( \frac{1}{4}(J^*+1)(J^*+2) \) coupled equations (4a) and (31) resulting from such an expansion which included \( J=0, 2, 4, \ldots J^* \) levels were, in the present study, reduced by parity arguments, to \( \frac{1}{4}(J^*+2)^2 \) coupled equations with explicit account taken of nonzero couplings between all the \( M \) sublevels.

*This research was sponsored by the Air Force Research Laboratories, Air Force Systems Command, United States Air Force, Contract F 33615-74-C-4003.

24L. A. Collins and N. F. Lane, Phys. Rev. (to be published).
Two new semiclassical methods\textsuperscript{1,2} - the multistate orbital treatment and the multichannel eikonal treatment - are presented for the description of elastic and inelastic scattering in heavy-particle collisions. The first method includes appropriate trajectories determined from a certain optical potential designed so as to couple the response of the internal structure, which is described by a quantal multistate expansion, to the orbit for the relative motion and vice-versa. While this approach is, in general, valid when the quantal imprecision in the classical trajectories is small, (as for heavy-particle systems), the second method, based on the use of a straight-line eikonal for the relative motion, of different local momenta in the various channels, and of a multistate expansion for the internal motions, is valid for scattering mainly about the forward direction. Results of these procedures will be illustrated by displays of angular distributions of atom-atom and ion-atom collisions in the keV energy-range. A quantal theoretical method\textsuperscript{3}, designed, in particular, for collisions involving atoms initially in (highly) excited states, will also be presented. The method appears particularly promising for improved theoretical descriptions of collisional ionization.

\textsuperscript{*Supported by AFOSR, Contract No. F33651-74-4003.}
Early type stars blow bubbles in the interstellar medium," assert John Casor, Richard McCray and Robert Weaver of the Joint Institute for Laboratory Astrophysics and the University of Colorado in a recent paper in *Astrophysical Journal Letters*. These bubbles are seen as arising when the strong wind emanating from a star sweeps up the surrounding interstellar gas and compresses it into a shell. After a million years, such a bubble would have a radius of 30 parsecs with a shell, about 4 pc thick, that expands at about 20 km/sec. While this theory is supported by a variety of observational data, it is being compared with an earlier theory postulating hot interstellar tunnels, which explains some of the same data—some of it apparently with greater accuracy.

Bubbles provide a possible explanation for the UV spectroscopic data sought back by the *Copernicus* astronomical satellite. In particular, they explain the presence of OVI ions (OII+), the absorption lines of which were studied by Edward Jenkins and Debrahrounc. The radial dependence of density (black line) and temperature (color) in a typical structure surrounded by Edward Jenkins and Debrahrounc is shown in the figure.

Highly excited atoms excite interest in many labs

"Since I'm an atom as big as bacteria, I've got to take microwaves serious. No matter what I do, I'll be cryin'... After they hit me, I'll be an ion." This beginning a poem written last year by Peter Koch during the stresses of preparing his PhD thesis. With James Bayfield, Koch had been studying highly excited atoms (often called "atoms in high Rydberg states," or even simply "Rydberg atoms"); the interesting behavior of such atoms has become the subject of current experimental and theoretical work at many centers around the world.

The unusual properties of these Rydberg atoms (some of which have been prepared with principal quantum number \( n \) as high as 105) arise because of their large size and small ionization potential. With electron orbital radii approaching \( 10^{-5} \) cm, such atoms are, as Koch's poem suggests, the size of simple bacteria, and they therefore have very large geometric collision cross sections. Because these high states lie so close to the continuum, ionization potentials become very low; when \( n \) exceeds about 23, the ionization potential falls to less than 25 millielectron volts. When left to itself, a Rydberg atom has a relatively long lifetime, increasing roughly as \( n^2 \), for a fixed orbital angular-momentum quantum number \( L \). Yet even thermal collisions can transfer enough energy to ionize the atom. On the other hand, it also appears possible that a neutral could pass through a Rydberg atom without causing serious perturbation.

As well as the intrinsic interest generated by such systems, there are applications of these studies in radioastronomy (for example, hydrogen transitions involving atoms with \( n \) up to more than 250 have been observed in interstellar regions), possibly for laser-induced isotope separation, and for basic atomic physics such as the determination of fundamental constants to improved accuracy.

A session on collisions of Rydberg atoms was organized as part of the Ninth International Conference on the Physics of Electronic and Atomic Collisions, held last July at the University of Washington in Seattle, and papers have been appearing in *Physical Review Letters* and elsewhere this past August and September. The first modern work on highly excited atoms was reported by A. C. Riviere and D. R. Sweetman (Culham, UK) in 1963, when they extended their technique developed earlier for molecular hydrogen ions to investigate the Rydberg states of hydrogen atoms excited with \( n \) up to 22.

When an atom sees an externally applied electric field, the potential-energy surface of the electron-nucleus system is distorted so that there may be a region outside the atom with potential equal to or lower than the potential of one or more of the bound electrons. Then tunnelling of an electron through the potential barrier to the continuum will occur if the tunnelling time is short compared to the lifetime of the bound state. This condition is met for the long-lived high Rydberg states in moderate fields, and Riviere and Sweetman (and many other experimenters since) have used this "field-ionization" method to detect Rydberg atoms. They...
studied highly excited hydrogen atoms formed by electron capture on protons accelerated to 25-100 keV; the ionizing field was of the order 10^5 volts/cm, strong enough to ionize atoms with n greater than 9 in 10^{-10} sec.

In 1967 H. Hotop and A. Niehaus (University of Freiburg) reported work on collisions between highly excited atoms and ground-state molecules in which the atom is ionized by energy transferred in the (thermal) collision. And at Argonne in 1969, W. A. Chupka and Joseph Berkowitz became involved with high Rydberg states when studying photoionization in the ion source of a mass spectrometer.

With the availability of tunable dye lasers came the ability to pump high Rydberg states one at a time with great precision; hence the recent growth of interest exhibited at the Seattle meeting. Rydberg states are produced at both Stanford Research Institute and the Massachusetts Institute of Technology by two-step laser excitation, whereas at Rice University one stage of excitation is followed by a single stage of photoionization.

SRI. The work on high Rydberg states at SRI was reported at the Seattle meeting by Thomas Gallagher, Stephen Edelstein and Robert Hill. They use two dye lasers pumped by a single nitrogen laser to excite sodium atoms; first from the 3s ground state to the 3p state, then, after a 4-ns delay of the second laser pulse, from the 3p level to high-lying s and d states in any level from n = 5 on up. Detection of the Rydberg atoms in this series of experiments was accomplished by monitoring the fluorescence from the upper s or d state back to the 3p state. The lifetimes of the higher s and d states were found to follow $n^{2\alpha}$ and $n^{2\alpha}$ power laws respectively.

More interesting, however, was the observed variation of the lifetime as other gases were added to the sodium in the experimental region. Addition of helium, argon and neon was found to lengthen the lifetimes of the high Rydberg states, not shorten them as might be expected from experience with quenching. The proposed explanation is that collisions between the sodium Rydberg atoms and the added gas atoms result in collisional mixing of the sodium atom in angular momentum states with $l = 2$ or more. The observed lifetime is then the statistically averaged lifetime of all states for which $l$ is not less than 2 and increases as $n^{2\alpha}$. In the range of $n$ from 5 to 10, the cross section for this process increases as $n^4$, as does the geometrical cross section of the excited sodium atom.

An exciting possibility, being examined at SRI, is that Rydberg atoms could be used for laser-induced isotope separation. In a two-step photoexcitation sequence, the first stage would be isotope-specific and the second stage would produce a Rydberg atom easily ionized by a relatively low electric field. Yield is expected to be high because the two laser beams are pumping bound, not ionizing, states; the final field-ionization stage is practically 100% efficient.

At MIT, Richard Freeman, Theodore Ducas, Michael Littman and Daniel Kleppner are also working with sodium atoms excited to high Rydberg states ($n = 20-50$) with two-step laser excitation. They differ from the SRI procedure, however, in using an atomic beam and in detecting by field ionization. Each sublevel of a given term ionizes at its own characteristic field value. The scale for the over-all pattern varies as $1/n^4$ for the $n = 30$, S-state of sodium, the field is 386 volts/cm. With this technique the MIT group examined the Stark shift at the onset of ionization, an interesting phenomenon because it represents the extreme case of distortion of a free atom by an electric field.

Kleppner pointed out in PHYSICS TODAY that it should be possible to devise a detector based on transitions in high Rydberg states that would allow photon counting in the infrared and millimeter range. One application could be as a diagnostic tool to examine radiation from tokamaks and other plasma-fusion devices. He also outlined a scheme for using these high n transitions for a determination of the Rydberg in frequency units with eventual accuracy that could reach 1 in $10^{11}$. By providing a frequency calibr-
tion for the hydrogen spectrum, the measurement would in effect serve to create an optical frequency standard. An improved value of the Rydberg would also yield a better value of the electron–proton mass ratio.

The work at Rice University is motivated, appropriately enough for a department of space physics and astronomy, by an interest in transitions such as the anomalous recombination lines seen in the radiofrequency spectra of HII regions. These lines arise from high Rydberg states of hydrogen with values of \( n \) up to one or two hundred. Colin Latimer, Phil West, Thomas Cook, Barry Dunning and Ronald Stebbings reported to the Seattle meeting their work with xenon atoms in high Rydberg states (\( n \) now up to about 40) in collisions with SF6 molecules, which Stebbings says is a relatively easy system suitable for tuning up their experimental techniques in preparation for an eventual attack on the astronomically more important collisions of hydrogen Rydberg atoms.

The Rice group excite Rydberg states in a two-step process, but unlike SRI and MIT, their method is to excite first to the metastable \( ^3P \) level of Xe by electron collision, and follow this up with laser photoexcitation to selected pure Rydberg states. Detection is by field ionization, for the lifetime studies: absolute cross sections for ionization of Rydberg atoms in collisions with various targets are now being determined.

At Yale University several years ago, James Bayfield’s group had extended the earlier work at Culham while studying electron-transfer collisions of protons with hydrogen atoms. The fast neutral beam, prepared by passing a unique fast-beam method. Koch, Larry Gardner and Bayfield reported Flannery a \( \frac{1}{2} \) neutralization as a source of atoms for a merged-beam approach assumes that ionization of SF6, containing hot gas of very low density, is seen as being permeated by a grid of interconnected tunnels containing hot gas of very low density. McCray suggests, be visible aspects of such wind-driven circumstellar shells have been reported by Peter Brand and William Zealey of the University of Edinburgh in a paper provocatively titled “Cloud structure in the galactic plane: a cosmic bubble bath?”

An alternative theory was published last year by Donald Cox and Barham Smith of the University of Wisconsin (Madison), in which the interstellar medium is seen as being permeated by a mesh of interconnected tunnels containing hot gas of very low density. These tunnels are said to have formed by the interaction of bubble-like supernova remnants, which have lifetimes of about four million years and remain hot during this entire time. When a younger supernova remnant encounters an older one it reheat it, saving it from destruction. This process continues so that roughly half of interstellar space is estimated to be now filled with the resulting tunnel network. This theory yields an approximately correct value for the soft x-ray background and explains the approximate intensities of collisions with highly excited atoms because of applications to recombination in astrophysics and laboratory plasmas. They started in 1964 with classical-trajectory methods like those of molecular dynamics, to obtain ionization and charge-transfer cross sections, the latter being in reasonable agreement with the experiments of Bayfield and Koch. More recently they used a variety of classical, semiclassical and quantal methods to obtain excitation cross sections for incident energies between \( 50/n^2 \) eV and \( 10^5 \) eV. The key to their methods lies with Heisenberg’s 1924 form of the old Bohr correspondence principle, which equates quantal matrix elements and Fourier components of classical motion, with errors of no more than about (100/n) per cent.

Comparison of this and other theories suggests that resultant errors in cross sections of no more than 5 per cent need be made, with a variety of approximations within well defined ranges of validity. However, outside its range of validity, the binary-encounter approximation, for example, can be out by about two orders of magnitude.
Contract No. F33615-76-2033, Item No. 001.

Air Force Aero Propulsion Laboratory
Attn: AFAPL/POP(450)/Capt. Prince
Wright Patterson AFB, Ohio 45433

Subject: Quarterly R and D Status Report No. 2
Contract No. F33615-76-C-2033.

Gentlemen:

Transmitted herewith are four (4) copies of the subject reports.

Sincerely,

M. R. Flannery
Principal Investigator and Professor

MRF:rh

Enclosures
Calculation of Electron Impact Cross Sections from Metastable States in Atomic and Molecular Cases

Quarterly Progress Report No. 2

Period Covered: Jan. 1, 1976 to March 31, 1976

Contract No. F33615-76-C-2033

Principal Investigator

Professor M. R. Flannery
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Technical Monitor

Capt. John F. Prince, AFAPL/POP
Project Engineer
USAF Aero Propulsion Laboratory (AFSC)
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Program Address

Air Force Aero Propulsion Laboratory
Attn: AFAPL/POP(450)/Capt. Prince
Contract No. F33615-76-2033. Item No. O0X
Wright-Patterson AFB, Ohio 45433
Theoretical investigations of the collisional excitation processes,

\[ e + N(2^D) \rightarrow e + N^*(2^P) \]

and

\[ e + O(1^D) \rightarrow e + O^*(1^S) \]

are proceeding. Also theoretical studies of the ionizations

\[ e + X^* \rightarrow 2e + X^+, \quad X \equiv \text{He, Ne, Ar}, \]

are still continuing. In response to the letter of March 4 from Capt. J. F. Prince, work on the ionization processes

\[ e + \begin{cases} 
  \text{Kr}^* \\  \text{Xe}^* 
\end{cases} \rightarrow 2e + \begin{cases} 
  \text{Kr}^+ \\  \text{Xe}^+ 
\end{cases} \]

has begun.

In contrast to collisions involving ground-state atoms, the Born approximation, when applied to excitation and ionization processes involving excited-state species, is now considered to yield satisfactory cross sections, particularly for ionization. Figure 1 displays the current status of the ionization of H(2s) by electron-impact and is taken from a recent paper (Proc. Roy. Soc. London A 343 (1975) 333) by Dixon, von Engel and Harrison. It displays the comparison of several theoretical models (classical, close-coupling, Bethe, Born) with the recent experiment. The Born approximation yields results in much better agreement with experiment than the close-coupling method.
Figure 2 displays the cross sections for
\[ e + \text{He}^* \rightarrow 2e + \text{He}^+. \]

The triangles and circles are measurements of Dixon, Harrison and Smith (in preparation), LG are data of Long and Geballe (Phys. Rev., Al (1970) 260), and FB are measurements of Fite and Brackman (Proc. 6th Intern. Conf. on Ionization Phenomena in Gases, 1963, p. 21-26).

The curves (a), (b), (c), (d) are the present theoretical data of Flannery, That and Manson.

(a) Binary-encounter method for He(2 \(^1\)S) ionization
(b) Born approximation for He(2 \(^1\)S) ionization
(c) Binary-encounter method for He(2 \(^3\)S) ionization
(d) Born approximation for He(2 \(^3\)S) ionization

Since the experimental results refer to a predominantly 2 \(^3\)S-target, the extent of the agreement is rather encouraging. For this reason, it is extremely important to present highly accurate and reliable Born theoretical cross sections. These cross sections are sensitive functions of the initial and final wavefunctions, which must therefore be chosen with care.

As the enclosed two figures indicate, the Born approximation for ionization of metastable atoms is apparently rather good.
Figure 6. A comparison between the measured cross section for electron impact ionization of H(2S) and various recent theoretical values. The measurements listed in brackets in table 1 are plotted as O (thick error bars) whereas the remainder are shown as I (thin error bars). The error bars represent the standard error of the mean Q(2S). Where errors are small they are shown by the size of the solid symbols. The theoretical curves are: c.c., the close-coupling approximation of Burke & Taylor (1965); B.B., the Born B and B.-ex., the Born-exchange approximation evaluated by Pressal (1966); c. is the classical Monte Carlo calculation of Abrines, Percival & Valentine (1966) and Be. is the Bates approximation of Vriens & Bonson (1963). Levels of the n = 2 state of H taken from Series (1957) are shown inset.
Figure 2
Contract No. F33615-76-2033, Item No. 003.

Air Force Aero Propulsion Laboratory
Attn: AFAPL/FOP(450)/Capt. Prince
Wright Patterson AFB, Ohio 45433

Subject: Quarterly R and D Status Report No. 3
Contract No. F33615-76-C-2033.

Gentlemen:

Transmitted herewith are four (4) copies of the subject reports.

Sincerely,

M. R. Flannery
Principal Investigator and Professor

MRF/jia

Enclosures
Calculation of Electron Impact Cross Sections from Metastable States in Atomic and Molecular Gases

Quarterly Progress Report No. 3

Period Covered: April 1, 1976 to June 30, 1976

Contract No. F33615-76-C-2033

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Project Engineer
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Program Address

Air Force Aero Propulsion Laboratory
Attn: AFAPL/POP(450)/Capt. Prince
Contract No. F33615-76-2033. Item No. 003
Wright-Patterson AFB, Ohio 45433
1. Introduction

Apart from the Born and binary-encounter treatments (see Appendix) for the ionization of metastable helium by electron-impact, little is known about the cross sections for ionization of metastable rare-gas atoms $X(\text{Ne}^*, \text{Ar}^*, \text{Kr}^*, \text{Xe}^*)$ by electron impact. This information is important in the modelling and feasibility studies of certain excimer lasers. In particular, very recent observations\textsuperscript{1-5} of high-power laser emission from a new class of molecules - the noble-gas monohalides $(\text{ArF}^*, \text{KrF}^*, \text{and XeF}^*)$ - have demonstrated the potential of a new class of high-power, high-efficiency and partially tunable lasers operating around 3000 Å. The lasing transition originates on an excited molecular state $XF^*$ formed directly by $X^*-F_2$ rearrangement collisions or indirectly by three-body ion-ion recombination between the positive ions $X^+$ produced by electron-impact ionization of $X^{(*)}$ and the negative ions $F^-$ formed by dissociative attachment in $(e-F_2)$ collisions. In either event, the ionization of metastable rare gas atoms plays a key role.

In this report, cross sections for the ionization of $\text{Ne}^*$, $\text{Ar}^*$, $\text{Kr}^*$ and $\text{Xe}^*$ by electron impact are provided as a function of the collision energy $E$ and are compared with experiment when available.
2. Results

In response to the letter of March 4 from Capt. J F. Prince, theoretical treatments of the ionization processes

\[ e + X^* \rightarrow 2e + X^+, \quad X = \text{He, Ne, Ar, Kr, Xe} \]

have been initiated and the associated cross sections have now been calculated in the Born and the binary encounter approximations as a function of electron-impact energy \( E \). A paper describing excitation and ionization in \( e^- \, \text{He}^* \) collisions has been written up and appears as an Appendix to this report.

The full description as provided in this Appendix was then applied to ionization in \( e^- \, (\text{Ne}^*, \text{Ar}^*, \text{Kr}^* \text{and Xe}^*) \) collisions. The numerical cross sections are tabulated as functions of \( E \) in tables 1, 2 and 3 of this report. Here, BF and BH are Born-approximations computed by integrating respectively over the full-range \( F \) and half-range \( H \) of energy of the ejected electron (see Appendix pages 4, 12), the binary encounter approximation for outer-shell ionization is denoted by \( \text{BE} \) and additional contributions from inner-shell ionization are included in \( \text{BEI} \).

Computer-drawn graphs of the ionization cross sections are displayed in figures 1-4. For \( e^- \, \text{He}^* \) (in Appendix), \( \text{Ne}^*, \text{Ar}^* \) collisions, the measurements of Dixon, Harrison and Smith (their \( \text{He}^* \) results are to be published in J. Phys. B. (1976), the \( \text{Ne}^* \) and \( \text{Ar}^* \) results are unpublished) are also shown, for comparison. We note that agreement with experiment is fairly good particularly for intermediate \( E \leq 100 \text{ ev} \), when inner shell ionization has not assumed importance. A paper describing the \( e^- \, (\text{Ne}^*, \text{Ar}^*, \text{Kr}^*, \text{Xe}^*) \) work in fuller detail is at present being written up.
3. Reprints and Preprints

Enclosed with this report are six (6) reprints of the paper
"The Multichannel Eikonal Treatment of electron-atom collisions",
published as Chapter 24 of the book "Electron and Photon Interactions
with atoms" (1976) edited by H. Kleinpoppen and M. R. C. McDowell.

Enclosed as an appendix to this report is a preprint entitled
"Cross Sections for Excitation and Ionization in e - He (2\textsuperscript{1,3}s)
collisions", which is to be submitted for publication to J. Phys. B:
Atom. Molec. Phys.

4. References

4. E. R. Ault, R. S. Bradford, Jr. and M. L. Bhaumik, Appl. Phys. Letts. 27,
   413 (1975).
   28, 326 (1976).
Table 1: Born (full-range BF and half-range BH) and Binary encounter (BE) cross sections ($10^{-16}$ cm$^2$) for the ionization of the outer-shell of metastable Ne$^*$ ($2p^53s$) and Ar$^*$ ($3p^54s$) by electrons with energy E(a.u.). The cross sections BEI include additional inner-shell contributions as determined from the binary-encounter approximation.

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Table 2: Born (full-range BE and half-range BH) and Binary encounter (BE) cross sections ($10^{-16}$ cm$^2$) the ionization of the outer-shell of metastable Kr$^*$ ($4p^5 5s$) and Xe$^*$ ($5p^5 6s$) by electrons with energy $E$(a.u.). The cross sections BEI include additional inner-shell contributions as determined from the binary-encounter approximation.

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<tr>
<th>$E$(a.u.)</th>
<th>BF</th>
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<th>BEI</th>
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Table 3: Born (full-range BF and half-range BH) and Binary Encounter (BE) Cross Sections ($10^{-16}$ cm$^2$) for the Ionization of He(2 $^1S$, $^3S$) by Electrons with Energy E(a.u.).

<table>
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<tr>
<th>E(a.u.)</th>
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<th>$^3S$ BE</th>
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<td>-</td>
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FIGURE 1

\[ e + Ne^* (2p)^5 (3s) \rightarrow 2e + Ne^+ (2p)^5 (2p)^4 (3s) \]

Total cross section for electron-impact ionization versus incident electron energy

\( Q \times 10^{-16} \text{ cm}^2 \)

Collision energy (eV)

THEORY: Ne\(^+\) (2p)\(^5\)  ⨁: Born approximation (with full range integration)
□: Born approximation (with half-range integration)
△: Binary encounter method
Ne\(^+\) (2p)\(^4\)(3s)  γ: Binary encounter method, (2p) electron alone
x: Binary encounter method, (2p)\(^5\) shell
Ne\(^+\) (2p)\(^5\) and (2p)\(^4\)(3s)  +: Binary encounter method, sum
(all atomic wavefunctions were obtained with the Herman-Skillman program)

EXPERIMENT:
o: Dixon et al.
$$e + \text{Ar}^* (3p)^5(4s) \rightarrow 2e + \text{Ar}^+ (3p)^5(3p)^4(4s)$$

Total cross section for electron-impact ionization versus incident electron energy

**THEORY:**
- $\text{Ar}^+(3p)^5$ : Born approximation (with full range integration)
- $\Diamond$: Born approximation (with half-range integration)
- $\Delta$: Binary encounter method

$\text{Ar}^+(3p)^4(4s)$
- $\Upsilon$: Binary encounter method, $(3p)$ electron alone
- $\times$: Binary encounter method, $(3p)^5$ shell

$\text{Ar}^+(3p)^5$ and $(3p)^4(4s)$ $+$: Binary encounter method, sum

(all atomic wavefunctions were obtained with the Herman-Skillman program)

**EXPERIMENT:**
- $\circ$: Dixon et al.
\[ e + \text{Kr}^* (4p)^5 (5s) \rightarrow 2e + \text{Kr}^+ (4p)^5 (4p)^4 (5s) \]

Total cross section for electron-impact ionization versus incident electron energy

THEORY: \( \text{Kr}^+ (4p)^5 \)
- \( \diamond \): Born approximation (with full range integration)
- \( \square \): Born approximation (with half-range integration)
- \( \triangle \): Binary encounter method

\( \text{Kr}^+ (4p)^4 (5s) \)
- \( \gamma \): Binary encounter method, (4p) electron alone
- \( \times \): Binary encounter method, (4p) shell

\( \text{Kr}^+ (4p)^5 \) and (4p)^4 (5s) +: Binary encounter method, sum

(all atomic wavefunctions were obtained with the Herman-Skillman program)
FIGURE 4

\[ e + \text{Xe}^* (5p)^5(6s) \rightarrow 2e + \text{Xe}^+ (5p)^5(6s) \]

Total cross section for electron-impact ionization versus incident electron energy

**THEORY:**
- \( \text{Xe}^+ (5p)^5 \) : Born approximation (with full range integration)
- \( \square : \) Born approximation (with half-range integration)
- \( \Delta : \) Binary encounter method
- \( \text{Xe}^+ (5p)^4 (6s) \) : Binary encounter method, (5p) electron alone
- \( x : \) Binary encounter method, (5p) \( ^5 \) shell
- \( \text{Xe}^+ (5p)^5 \) and (5p) \( ^4 (6s) \) : Binary encounter method, sum

(all atomic wavefunctions were obtained with the Herman-Skillman program)
Appendix

Cross Sections for excitation and Ionization in e - He(2\(^{13}\)S) collisions
Cross Sections for Excitation and Ionization
in e − He\(^{1,3}\text{S}\) Collisions

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Georgia State University
Atlanta, Ga. 30303, U.S.A.

Abstract: Cross sections for the \(2\^{1,3}\text{S} \rightarrow n\^{1,3}\text{L}(n = 2-5, L = \text{S-}\text{F})\) excitations and for single ionization in e − He\(^{1,3}\text{S}\) collisions are determined by using the Born approximation. For a given \(n\), the \(2\^{1,3}\text{S} \rightarrow n\^{1,3}\text{D}\) transitions dominate at intermediate energies and this trend is continued for transitions to the near continuum. For larger energies \(\varepsilon\) of the ejected electron, higher angular momentum \(l\)-waves progressively dominate. While up to 9 partial waves are in general required for convergence of the bound-free form factor, as many as 30 are needed when momentum changes are in the vicinity of the Bethe ridge for large \(\varepsilon\). Binary encounter results for ionization are also obtained and agree with the Born values at intermediate energies. The present calculations and the recent measurements of Dixon et al. are in satisfactory accord at intermediate energies. The contributions to ionization arising from multiple processes are also discussed.
1. Introduction.

Relatively little is known about electron-metastable rare gas atom collisions which are of direct importance to gaseous discharges, astrophysical and atmospheric plasmas, the development of excimer and monohalide rare gas lasers (cf. Flannery et al. 1975) and of ion-engines (cf. Martin 1974). Nor is much known concerning the validity of the Born approximation for electron collisions with excited atoms containing a loosely bound electron. The recent ten-channel eikonal results of Flannery and McCann (1975) for the $2^{1,3}S - n^{1,3}L$ ($n = 2, 3; L = S, P, D$) collisional excitations of helium approach the corresponding Born values (Flannery et al. 1975) at relatively modest impact energies $E \sim 100$ eV which are, however, $>> \Delta E$, the transition-energy. Also, contrary to expectation, both the Born and ten-channel eikonal treatments predict that transitions to the optically-forbidden $3^{1,3}D$ and $3^{1,3}S$ states are much stronger, by up to an order of magnitude, than the dipole allowed $2^{1,3}S - 3^{1,3}P$ collisional excitations, except of course at high energies where the dipole transitions predominate.

In an effort to determine whether this behaviour is characteristic of transitions to higher discrete and continuum states, and to obtain information on the corresponding cross sections (as functions both of impact energy $E$ and of the angular momentum of the final target-state) the following collisional excitation and ionization processes

$$e + He(2^{1,3}S) \rightarrow e + He(n^{1,3}L); n = 4, 5; L = 0 - (n - 1),$$

$$\rightarrow 2e + He^+$$

will be studied in the Born approximation. The cross sections for ionization (1b) can be compared with the recent measurements of Dixon et al. (1976a) and with
results to be obtained from the binary-encounter approximation, considered applicable since the averaged electronic separation in He is $\approx 5a_0$ (Pekeris 1962). It is worth noting that the Born (Prasad 1966) and classical (Abrines et al. 1966) predictions for ionization of metastable hydrogen are in close accord with the experimental data (Dixon et al. 1975) over a wide range of electron-impact energies.

2. Theory.

The cross section for ionization of a singly excited atom $B$ with mass $M_B$ and ionization potential $I$ by an incident particle $A$ with mass $M_A$, speed $v$ and relative energy $E$ is, in the (elastic) binary encounter approximation given by (cf. Vriens 1969, Flannery 1971)

$$
\sigma_{n\ell}^I (E) = \frac{\pi}{M_{Ae} v^2} \int \frac{d\epsilon_T}{\epsilon_T} \int f_{n\ell}(u) \frac{du}{u} \int \frac{4}{P^4} \Gamma(P) \, dP \tag{2}
$$

where the distribution in speed $u$ of the valence electron described by a spatial wavefunction $\phi_{n\ell m}(r)$ is, with all quantities in atomic units,

$$
f_{n\ell}(u) = \frac{1}{(2\ell+1)} \sum_{m=-\ell}^{\ell} \left| \int \frac{1}{(2\pi)^{3/2}} \int \phi_{n\ell m}(r) e^{-i\mathbf{p}\cdot\mathbf{r}} \, dr \right|^2 u^2 \frac{du}{u} \tag{3}
$$

and $M_{Ae}$ is the reduced mass of the $(A-e)$ binary system.

For a specified energy-transfer $\epsilon_T$ to the valence electron, the momentum-change $P$ can vary between the lower limit,

$$
P^- = \max \left[ M|u' - u|, M_{AB}|v' - v| \right]; \quad M = m \left(1 + \frac{m}{M_i}\right), \quad M_{AB} = \frac{M A_B}{M_A + M_B}, \tag{4}
$$

where $m$ and $M_i$ are the electronic and ionic masses respectively, and the upper limit,

$$
P^+ = \min \left[ M(u' + u), M_{AB}(v' + v) \right] \tag{5}
$$
where the post binary-collision speeds of the projectile and target particles are respectively,

\[ v^- = (v^2 - 2e_T /M)^{1/2} \]  \hspace{1cm} (6)

and

\[ u^- = (u^2 + 2e_T /M)^{1/2} \]  \hspace{1cm} (7)

In the general expression (2), the function \( \Gamma(P) \), which represents the departure of the differential cross section for \((A - e)\) elastic scattering from the Rutherford value, is set to unity for direct \((e - e)\) collisions for which \( M \approx M_{AB} \approx m \).

The Born approximation to the cross section for single ionization of a 2-electron atom by an incident electron is written, with all quantities in atomic units, as

\[ \sigma_{nl} (E) = \int \frac{1}{(2\ell+1)} \sum_{m=-\ell}^{\ell} \sigma_{n\ell m} (E, \hat{k}_{ie}) \, dk_{ie} \]  \hspace{1cm} (8)

in terms of

\[ \sigma_{n\ell m} (E, \hat{k}_{ie}) = \frac{8\pi}{k_1} \left( \frac{K^+}{K} \right) \left| \langle \psi_f (E, \hat{k}_{ie}; \lambda_{ie}) \rangle \right|^2 \sum_{i=1}^{2} \left| \frac{2}{\psi_{1} (n_1, L_1, m, \lambda_{1})} \, e^{i\hat{k}_{i} \cdot \hat{k}_{ie}} \right|^2 \, \frac{dK}{K^3}, \]  \hspace{1cm} (9)

the differential cross section for ejection of the electron with momentum \( k_{ie} \) into unit solid angle and unit energy interval. The limits to the momentum-change \( (\hat{k}_{\lambda} - \hat{k}_{\lambda}) \) of the scattered electron are,

\[ \frac{1}{K^+} = (2E)^{1/2} \frac{1}{4} [2(E - I - \epsilon)]^{1/2} \]  \hspace{1cm} (10)
where the energy $e$ of ejection is $\frac{1}{2}k_e^2$ a.u. and where the kinetic energy transferred to the ion is neglected. While the parameter $a$ in the $e$-integration limit in (8) is unity for ionization involving distinguishable particles, Rudge and Seaton (1965) have shown that, for ionization of atomic hydrogen by electrons with random spin orientations, $a = 0.5$ when electron-exchange effects are fully neglected. This choice ensures that the faster of the scattered and ejected electrons is always described by a plane wave. For electron-impact ionization of helium (when neither choice is rigorously based), the spatial wavefunctions for the initial discrete and final continuum states are,

$$\psi_1(n, 1, 3; \kappa_e) = \frac{1}{\sqrt{2}} \left[ \phi_{1s}^{(n)}(\kappa_1) \phi_{n\ell m}(\kappa_2) + \phi_{1s}^{(n)}(\kappa_2) \phi_{n\ell m}(\kappa_1) \right]$$

and

$$\psi_f(\epsilon, \kappa_e, 1, 3; \kappa_e) = \frac{1}{\sqrt{2}} \left[ \phi_{1s}^{(n)}(\kappa_1) \phi_{n\ell m}(\kappa_2) + \phi_{1s}^{(n)}(\kappa_2) \phi_{n\ell m}(\kappa_1) \right]$$

in which the He$^+$($1s$) core is considered frozen. The one-electron orbitals $\phi_{n\ell m}$ are chosen to form an orthonormal set such that the form factor in (9) is simply

$$\langle \psi_f | e^{i\mathbf{k} \cdot \mathbf{r}_1} + e^{i\mathbf{k} \cdot \mathbf{r}_2} | \psi_1 \rangle = \langle \phi_\epsilon(\kappa_e; \kappa_e) | e^{i\mathbf{k} \cdot \mathbf{r}} | \phi_{n\ell m}(\kappa_e) \rangle$$

The continuum orbital for the ejected electron is now expanded as

$$\phi_\epsilon(\kappa_e; \kappa_e) = \sum_{\ell=0}^{\infty} \sum_{m=-\ell}^{\ell} \frac{1}{\ell} e^{-i\ell \kappa_e} F_{\ell m}^\mathbf{r} Y_{\ell m}^2(\kappa_e) Y_{\ell m}^1(\kappa_e)$$
in which the radial part has asymptotic behavior,

\[ F_{\epsilon \ell'}(r) \sim \left( \frac{2}{\pi k}\right)^{1/2} \sin \left[ k_e r - \frac{1}{k_e} \ln(2k_e r) - \frac{1}{2} \ell' \pi + \eta_{\ell'} \right], \]

where the additional phase shift \( \delta_{\ell} \) is a measure of the departure of the electron-ion interaction from pure Coulomb. The amplitude \( 2^{1/2} \frac{1}{n} \frac{1}{k} \) of (15) is chosen so as to fulfill the normalization condition,

\[ \langle \phi_{\epsilon}(k_e; \ell) | \phi_{\epsilon'}(k'_e; \ell') \rangle = \delta(\epsilon - \epsilon') \delta(k_e - k'_e) \]

where \( \epsilon = \frac{1}{2} k_e^2 \) is in atomic units. With \( \epsilon = \frac{2}{k_e^2} \) in Rydberg units, then the amplitude of (16) would be \( n^{-1/2} \epsilon^{-k} \) in order to satisfy (16).

Alternatively, the differential cross section per unit momentum interval \((d\sigma/dk_e)\) is given by (9) with the asymptotic amplitude of \( F_{\epsilon \ell'} \) chosen as \( (2^{1/2} n^{-1/2} k) \) thereby ensuring that

\[ \langle \phi^{\dagger}(k_e; \ell) | \phi(\hat{k'}_e; \hat{\ell}) \rangle = \delta(k_e - \hat{k'}_e) \]

rather than (16).

By writing the orbital for the bound \((n \ell m)\) state as

\[ \phi_{n\ell m}(r) = \frac{1}{r} P_{n\ell}(r) Y_{\ell m}(\hat{\ell}) \]

and with the aid of

\[ e^{i\hat{k} \cdot \hat{\ell}} = 4\pi \sum_{\ell''=0}^{\infty} \sum_{m''=-\ell''}^{\ell''} 1^{\ell''} J_{\ell''}(Kr) Y_{\ell'' m''}(\hat{k}) Y_{\ell m}(\hat{\ell}) \]

rather than (16).
where \( j_{\ell} \) denotes the spherical Bessel function then, in terms of the Wigner 3j-symbols (cf Messiah 1966), the form factor (13) is

\[
F_{\ell e, n \ell m}^{\hat{\kappa}_e, n \ell m}(K) = (4\pi)^{\frac{1}{2}} \sum_{\ell' m' \ell'' m''} i^{l''} M_{\ell e, n \ell m}^{\ell' m'}(K) Y_{\ell m}(\hat{\kappa}_e) Y_{\ell'' m''}^{*}(K)
\]

(20)

\[
(-1)^{m'} [(2\ell'+1)(2\ell''+1)]^\frac{1}{2} \begin{pmatrix} \ell & \ell' & \ell'' \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} m & m' & m'' \end{pmatrix}
\]

where the radial matrix element

\[
M_{\ell e, n \ell m}^{\ell' m} = \int_0^\infty e^{-|\hat{r}|} \bar{F}_{\ell e, n \ell m}(r) j_{\ell'}(Kr) P_{\ell} \, dr
\]

(21)

The partial cross section (9) therefore entails, in general, eight summations and a double integration. However, when the modulus of (20) is squared, integrated over \( |\hat{\kappa}_e| \) (assumed to be uncorrelated with \( |\hat{K}| \)), and summed over \( m \) and \( m' \), then it follows that

\[
\frac{1}{(2\ell+1)} \sum_{m=-\ell}^{\ell} \left| F_{\ell e, n \ell m}(K) \right|^2 \frac{d\hat{\kappa}_e}{\hat{\kappa}_e} = \sum_{\ell' m'} (2\ell'+1)(2\ell''+1) \begin{pmatrix} \ell & \ell' & \ell'' \\ 0 & 0 & 0 \end{pmatrix}^2 \left| M_{\ell e, n \ell m}^{\ell' m'}(K) \right|^2
\]

(22)

for a given momentum change \( K \). For initial s-states, then, from (8) and (9),

\[
\sigma_{ns}(e) = \frac{8\pi}{k_1^2} \int_1^{K^+} \sum_{\ell' = 0}^{\infty} (2\ell'+1) \left| M_{\ell e, n s}^{\ell'}(K) \right|^2 \frac{dK}{K^3}
\]

(23)
is the differential cross section per unit ejected-energy interval. The integral Born cross section for excitation of discrete level \((n',\ell')\) is also given by each \(\ell'-\)term of (23) but with \(F_{\ell'}\) in (21) replaced by the bound-state function \(P_{n',\ell'}\). For excitation, additional allowance will be made for core relaxation.

3. Method

In the present study, the electronic discrete and continuum wavefunctions \(\psi_{n_1, n_2} \) for helium are antisymmetrized products of one-electron orbitals \(\phi_{n'm'}(r)\) forming an orthonormal set obtained from a central field approximation. The corresponding radial functions \(P_{n\ell}\) and \(F_{\ell\ell}\) are therefore the appropriate solutions of the radial Schrödinger equation,

\[
\frac{d^2 P_{n\ell}}{dr^2} + 2 \left[ E - V(r) - \frac{l(l+1)}{2r^2} \right] P_{n\ell} = 0, \quad E = \begin{cases} \epsilon_{n\ell} < 0 \\ \epsilon > 0 \end{cases}
\]

which are normalized to unity for \(E < 0\) and per unit energy interval for \(E > 0\) as implied by (16). The spherical interaction \(V(r)\) which tends to \((-2/r)\) and \((-1/r)\) as \(r \to 0\) and \(\infty\), respectively, is determined from the Hartree-Slater approximation i.e. the Hartree method modified by Slater (1951) to take some account of electron-exchange. This approximation for \(V\) as determined by Herman and Skillman (1963) for ground state atoms alone has been extensively used with encouraging results (cf. Fano and Cooper 1968, Dehmer et al. 1975, Manson and Purcell 1976, and references therein). In this paper, some variations to the standard treatment are explored by adopting the following choices for \(V\):

(A) A self-consistent field (SCF) iteration for the ground-state configuration \((1s)^2\) provides \(V_0\) which is used to generate \(P_{n\ell}\) and \(P_{\ell\ell}\) which are automatically orthogonal. This \(V_0\) is the standard selection tabulated by Herman and Skillman (1963).
(B) A SCF iteration for the initial-state \((1s)(2s)\) configuration yields \(\tilde{V}_i\) which, when inserted in (24), provides orthonormal \(P_{n\ell}\) and \(F_{\epsilon\ell}\) for \(n = 1-5\), \(\ell = 0 - (n - 1)\), \(\ell' = 0-30\) and for a wide range (0-110 eV) of energies \(\epsilon\) for the ejected electron.

(C) SCF iterations for the three configurations \((1s)(2s), (1s)(2p), (1s)(3d)\) yield three potentials \(V_i^{(L)}\) \((L = 0, 1, 2)\) which are used to generate \(P_{n, \ell=0,3,4}^{(0)}, P_{n, \ell=1}^{(1)}\) and \(P_{n, \ell=2}^{(2)}\) respectively for the \((n\ell)\) states of the \(n = 1-5\) levels. The form factor in (9) for bound-bound transitions is, for the \(L = 1\) and 2 cases, given by

\[
F_{fi}^{(K)} = \langle \psi_f(n,1,3,L) | \sum_{i=1}^{2} e^{iK \cdot r_i} | \phi_{1s}^{(L)} \rangle \langle \phi_{1s}^{(0)} \phi_{2s}^{(0)} \rangle \pm \langle \phi_{1s}^{(L)} \phi_{1s}^{(0)} \rangle \phi_{1s}^{(0)} \rangle
\]

where the \(\phi_{1s}^{(L)}\) which respectively result from \(V_i^{(L)}\), \(L = 1\) and 2, are in general different from \(\phi_{1s}^{(0)}\) and are not orthogonal to both \(\phi_{1s}^{(0)}\) and \(\phi_{2s}^{(0)}\) because some core relaxation has been provided for those less-penetrating orbitals with non-zero angular momentum. The \(f\) and \(g\) orbitals are generated in \(V^{(0)}\) for simplicity since it is found that little improvement is gained by further modification.

(D) SCF iterations for the \((1s)(nk)\) configuration of the final bound state \(\psi_f(n,1,3,L)\) provide \(V_f^{(L)}\) which are used to generate \(P_{n\ell}\) for the lower bound states.

(E) SCF independent iterations for the initial and final configurations, \((1s)(2s)\) and \((1s)(nk)\) respectively, yield independent SCF solutions \(\psi_i(2,1,3S)\) and \(\psi_f(n,1,3,L)\) with orthogonality satisfied by restricting the final bound states to \(L \neq S\).

In the above five procedures, explicit dependence on electron spin appears only in the symmetry of the total spatial wavefunctions (11) and (12) and not in the interaction potential (which includes equal allowance for spin in the
singlet and triplet cases). Spin effects are also acknowledged by the use of the corresponding experimental values of the excitation and ionization energies in the momentum-change limits (10) for the cross section (9).

All of the above possibilities were fully investigated via appropriate modifications to the standard Herman-Skillman program. By comparison of the resulting excitation energies (in cases A, B, D), form-factors, Born electron-impact cross sections for the \(2\,^1S - 2\,^1P, \, 3\,^1S, \, 3\,^1P, \, 3\,^1D\) and \(4\,^1P\) transitions with those previously determined (Kim and Inokuti 1969, Flannery et al. 1975) from highly accurate correlated functions (Weiss, 1967) we have concluded that for the triplet transitions, method B achieves very good agreement (with cross sections to within 2.5% over a wide energy-range) which is improved somewhat by modification C. For singlet transitions, method A yields the closest, but yet rather poor, agreement (with cross sections to only within 40%). The reasons for this dissimilarity in agreement are twofold and are rather instructive.

The (B, C) combination provides an adequate account of core relaxation while ensuring that all of the excited \(^3S, \, ^3P\) and \(^3D\) states are orthogonal to all lower states of the same symmetry. The same degree of orthogonality between the singlet states, especially that between the \(2\,^1S\) and the \(1\,^1S\) singlet ground state, is obtained via method A which however fails to account satisfactorily for core relaxation of the excited states and which fails to satisfy the Sharma-Coulson (1962) requirement that variational calculations of the (1s) and (ns) orbitals of helium should not be orthogonal for excited \(^1S\) states. Moreover, while the exchange interaction is attractive for the triplet states and repulsive for the excited singlet states (as predicted from perturbation theory which gives zero exchange effect for \(^1S\)), the Slater prescription assigns an exchange correction which is always negative (even for \(^1S\) state) and which is therefore very biased towards the triplets.
The above considerations help explain the difference in quality of the present agreement obtained for the singlet and triplet cases. Also, the above \((B, C)\) combination is an extremely useful (and fast) method for obtaining accurate orthogonal wavefunctions for all the triplet (excited) states of helium.

4. Results and Discussion.

4.1 Excitation:

In table I are displayed the excitation energies (from method B) and cross sections for the processes

\[
e + \text{He}(2^3S) + e + \text{He}(n^3L); \quad n = 2-5, \quad L = S-G
\]

at impact energies \(E\) in the \(5 \text{ eV} \leq E \leq 1000 \text{ eV}\) range. These cross sections agree with the available highly accurate cross sections (Kim and Inokuti 1969, Flannery et al. 1975) to within 2.5\% over the entire energy-range, except for the \(2^3S - 3^3P\) transition for which the largest departure is 8.5\%. The more accurate results, where available, are presented in the Table for reference. The cross sections for the remaining transitions from the metastable level to the \(n = 4\) and 5 levels are presumably even more accurate since the central field approximation improves as the valence electron becomes more loosely bound and much less correlated with the core electron, as exhibited in the Table by the closeness between the experimental and derived excitation energies. The Table also shows that the optically-forbidden transitions to the D-states of the \((3-5)\) levels dominate all other transitions, except at the highest energies when the \(S-P\) dipole excitations gain in relative importance, in accord with Bethe's approximation. This behaviour is even true for transitions to the ionization threshold and is preserved in more elaborate treatments (Flannery et al. 1975, Flannery and McCann 1975) of excitation.
The data in the Table are useful and directly relevant to current modelling of excimer and monohalide rare gas lasers.

4.2 Ionization.

In figure 1 are displayed present cross sections determined from the binary-encounter formulae (1) - (7) for the ionization process,

\[ e + He(2^1,3S) \rightarrow 2e + He^+(1s) \]  

(28)

in which the ion is left in the ground state. The frozen core \( 2^1,3S \) Hartree-Fock functions of Cohen and McEachran (1967a,b) and the \( 2^1,3S \) Hartree-Slater functions obtained from both methods (A) and (B) above are used in (3) to generate the velocity distributions \( f_{nl}(u) \) for the valence electron. The cross sections associated with the rather accurate Hartree-Fock functions are in excellent agreement, especially at low energies, with the cross sections derived from method (B) for \( 2^3S \)-ionization, and in good agreement with those derived from method (A) for \( 2^1S \)-ionization, respectively. This closeness provides an additional check on the quality of our present \( 2^3S \) wavefunction, and indicates that the present \( 2^1S \) Hartree-Slater function is quite adequate for bound-free transitions, the form factor (13) for which tends to be controlled by interelectronic distances \( r_{12} \) larger than those important for bound-bound transitions. Also shown for completeness are previous binary-encounter results of Vriens (1964) based on an exponential velocity distribution (Gryzinski 1965) which is, however, completely at variance (cf. Burgess and Percival 1968) with any quantal distribution. The recent experimental data of Dixon et al. (1976a) for ionization of a predominantly \( 2^3S \)-target are also displayed; the earlier measurements of Fite and Brackman (1963), Long and Geballe (1970) and modifications thereof have already been discussed extensively by Dixon et al. (1976a).
In order to achieve convergence of the differential and integral cross sections (23) and (8) for ionization over the impact-energy range \( E \), it is necessary to compute the Born matrix element \( M_{e l', 2s}^{l'}(K) \) for a wide range of energy \( \epsilon \) and angular momentum \( l' \) of the ejected electron and also of the momentum change \( K \) of the scattered electron. This necessitates the calculation of 9 to 30 partial \( l' \)-waves for each set of \((\epsilon, K)\) values appropriate to impact energies \( E \) up to 100 eV. This determination together with the triple \((r, K, \epsilon)\) integration in (21), (23) and (8) and the \( l' \)-summation requires extremely lengthy computation which can be reduced by the use, where possible, of various semi-analytical methods particularly for low values of \( l' \). The resulting cross sections obtained from two different computer programs are in close accord, thereby providing a test of the numerical accuracy.

In addition, we have verified that the relationship

\[
\lim_{n \to \infty} (n - \delta)^3 F_{i,n} (K) = \frac{dF_{ic}}{d\epsilon} (K) \tag{29}
\]

involving the bound-bound and bound-free form factors \( F_{in} \) and \( F_{ic} \) respectively, and the asymptotic quantum defects \( \delta \) is satisfied on crossing the ionization threshold.

In figure 2 are presented the Born (full-range) and the 'modified' Born (half-range) cross sections, (8) with \( \alpha = 1 \) and \( \frac{1}{2} \) respectively, for the ionization processes (28) at collision energies \( E \) up to 100 eV and 200 eV respectively. Although the former choice is correct for ionization involving distinguishable particles and the latter is used for electron-impact ionization of \( H \) with exchange effects fully excluded, neither choice is rigorously based for
helium and the correct $\alpha$ would demand a detailed account of exchange effects.

In the present case, the full-range results $F$ are in closer accord than the half-range results $H$ with the experimental data for ionization of the predominantly $2^3S$-target at low and intermediate impact energies $E$. Calculations (Ton-That and Flannery 1976a) for bigger targets ($Ne^*$, $Ar^*$) show an evolution from this trend, favouring the half-range over the full range results. While convergence between $F$ and $H$ is obtained at the higher $E$, as expected, the measurements are up to 30% higher. This discrepancy which may be due to the theoretical neglect of additional processes will be studied in further detail below by means of a Bethe plot. The half-range cross sections for ionization of the triplet states agree to within a few percent with the corresponding Born-calculations (not shown) of Peach (1976) who used a Clementi-type fit for the initial state and, for continuum waves, hydrogenic functions, except the $s$, $p$ and $d$ partial waves.

Figure 3 provides a comparison of the measured cross sections with the present Born full-range values to 100 eV and with the binary encounter results to 1000 eV. Also shown is an additional binary encounter contribution which arises from direct ejection of the inner-shell electron such that the product is left as $He^+(2s)$. This mechanism does not fully account for the discrepancy with the observations at collision energies $E \gtrsim 200$ eV.

This difference is further amplified in figure 4 where Bethe plots - i.e. $\sigma_1(E) \times E$ versus $\ln E$ - of the various results are displayed. (The inclusion of dipole allowed transitions yields an $E^{-1} \ln E$ dependence for the ionization cross section $\sigma_1(E)$ at large impact energies $E$, thereby resulting in a straight line Bethe plot at high $E$). The lower set of experimental data includes a
correction made to the higher original set for charge exchange reactions between metastable He and residual molecular ions in the experimental collision region (see Dixon et al. 1976a). The remaining difference between the theoretical Born curve corresponding to a ground-state He\(^+\) (1s) product ion and the measurements cannot be attributed to the 2 \(^1\)S/2 \(^3\)S admixture in the target gas since the difference between the cross sections for singlet and triplet ionization decreases as E increases (see figure 3).

Other processes, neglected theoretically, which enter as E increases are (a) inner-shell ionization, (b) autoionization over certain small discrete regions of E, (c) double ionization (which is also not included in the measurements) and (d) ionization of one electron with simultaneous excitation of the other electron. Of all the above processes, (a) is expected to be the largest contributor over a wide energy-range since it can be regarded as a single scattering process, while the remaining wide energy-range processes are multiple processes. The contribution of (a) here determined by the Born approximation is shown in figure 4, is in good agreement with the corresponding binary-encounter results of figure 3, and is smaller than outer-shell ionization by one order of magnitude. A simple treatment of the multiple processes adopts the sudden approximation wherein the additional excitation or ionization involved is described by a transition probability factor \( P(i, f) \) which is the overlap squared of selected initial and relaxed final (bound or continuum) orbitals. Thus, for example, when exchange is ignored in the target wavefunction, the cross sections for the multiple processes (c) and (d) are \( P(1s, c) Q(2s, c) \) and either \( P(1s, n\ell) \times Q(2s, c) \) or \( P(2s, n\ell) Q(1s, c) \), respectively, in terms of the cross section \( Q(n\ell, c) \) for ionization of the \((n\ell)\) orbital by a single process. Byron and Joachain (1966) used correlated atomic wavefunctions in a Born treatment of the
ionization of ground-state helium and found that the contribution (DI) arising from double ionization is less than that for single ionization (SI) by about two orders of magnitude, in keeping with a sudden collision model (Mittleman 1966) and with experiment (Schram et al. 1965, Briglia and Rapp 1966). Calculations (Gillespie 1972 and references therein) based on the sudden approximation for He(1S) show that simultaneous excitation and ionization (SICE) are lower than SI by about three orders of magnitude (or at best by two orders, cf. Dixon et al. 1976b). Although excited states of helium have lower thresholds for multiple processes than the ground state, the correlation between the core and valence electrons, which is important in multiple processes if the energy transfer is not too large, is much less (the atomic electrons have separation \( \sim 5a_0 \) and binding energies \( \sim 46 \text{ eV} \) and \( 5 \text{ eV} \) respectively). While some doubt has been cast on the usefulness of the sudden approximation for multiple processes (Kaminsky and Popova 1976, see also Dixon et al. 1976b), recent measurements (Schmidt et al. 1976), however, do not suggest that a revision of the experimental results for double ionization of helium in its ground state is necessary.

The above arguments are however difficult to reconcile with the findings of Briggs and Kim (1971) who obtained various Born asymptotic limits for single and double ionization of the triplet states by subtracting the sum of the Born asymptotes for the discrete excitations from the asymptotes of the total inelastic cross section obtained from closure-sum rules. The resulting Bethe \((A \ln E)\) term and the asymptotic Born (half-range) term, \( A \ln E + \gamma + \ldots \), with and without the Mott correction for exchange, are reproduced in figure 4. The difference between the curves H3 and BA should arise from the effects of inner shell ionization (ISI) and, to a lesser extent, of double ionization (DI), and single ionization with core excitation (SICE). Results from our present Born treatment of ISI are shown explicitly; and, when added to the SI-results H3 and F3, yield the dotted lines in the figure. The remaining difference, if all is well, must arise from (unacceptably large) contributions from DI and SICE. The asymptotic procedure has been estimated by Briggs and Kim (1971) to be reliable only for collision energies
E > 400 eV such that the correct plots for smaller energies may deviate from those shown. We note however that the difference between BA and H3 can mainly be resolved by assigning some value, to the second parameter \( \gamma \) smaller than given by Briggs and Kim (1971). This parameter is difficult to evaluate correctly and involves a sum of integrals of the generalized and optical oscillator strengths (Inokuti et al. 1967).

In summary, figure 4 depicts (a) full Born calculations of both SI and ISI, the SI results agreeing rather closely with similar calculations of Peach (1976), (b) Born asymptotes of Briggs and Kim (1971) including all contributions (SI, ISI, DI, SICE) to ionization, and (c) measurements of the combined SI, ISI and SICE contributions. Accord between (a) and (c) is introduced at high energies \( E \) only if the yield from the (neglected) multiple SICE process is roughly three times that from the single ISI process. Also the apparent agreement between (b) and (c) at large \( E \) suggests that DI is negligible in comparison to SICE, an unlikely event. The resolution of the discrepancies between (a), (b) and (c) can only be obtained if the various contributions to ionization are arranged in order of importance as SI, SICE, ISI and DI, an order at variance with that naturally assumed.

The theoretical cross sections obtained from the Born and the binary encounter approximations for ionization of the \( 2^1\,^3S \) states are presented in table 2. The full-range and the half-range Born results mutually converge with increasing impact energy \( E \), as expected. In the intermediate energy range \( 1 < E(\text{a.u.}) < 8 \), the binary encounter values, when compared with the full-range Born results, display fair agreement which originates from the overall agreement in magnitude and shape between the quantum mechanical and the binary encounter results for FFS, the form factor (20) squared, or for the associated generalized oscillator strength \'(GOS) away from the optical
K + 0 limit. The increasing discrepancy in the GOS for small momentum changes $K \approx \varepsilon_1/v$ associated with larger impact energies $E$ yields an $E^{-1}$ asymptotic limit for the binary encounter cross sections rather than the correct $E^{-1} \ln E$ limit obtained when proper account is taken of the dipole transitions for such low momentum changes.

4.3 The rate of convergence of the partial wave expansion.

For most values of $\varepsilon$ and $K$, the number of the $l$ partial waves required for convergence of FFS w.r.t. $l$ is relatively small, 9 or less in the present case. However, for large $\varepsilon$, the required number increases sharply near the Bethe ridge defined by

$$K_{BR}^2 = 2\varepsilon_T = 2(1 + \varepsilon), \quad (30)$$

where up to 30 partial waves are required. This is interpreted as follows. The region where the initial bound state orbital $P_\ell(r)$ is substantial defines for our purposes the maximal region of overlap. The continuum orbital $F_{\ell}(r)$ and the spherical Bessel function $j_{\ell''}(Kr)$ (cf. (21)) initially increase in amplitude from the origin as $(\sqrt{2\varepsilon})^{\ell'+1}$ and $(Kr)^{\ell''}$ respectively. These sharp dependencies on $\ell'$ and $\ell''$ can be regarded as centrifugal-barrier effects. $F_{\ell}(r)$ and $j_{\ell''}(Kr)$ then evolve into oscillating functions. An increase in $\ell$ (i.e. $\ell'$ and $\ell''$) results in a shift of the pattern of these functions out of the overlap region together with a stretching of their first lobe.

When $\varepsilon$ is large enough, the continuum orbital $F_{\ell'}$ oscillates many times within the overlap region.

If either $K^2 << 2\varepsilon_T$ or $K^2 >> 2\varepsilon_T$, then, due to the varying relative phase of $j_{\ell''}$ and $F_{\ell'}$, the contribution from their oscillatory regions is small and

---

Hereafter $l$ is used to denote either $\ell'$ or $\ell''$ interchangeably.
the effective region of overlap is therefore reduced to the inner lobes of \( j_{\ell -} \) or \( F_{\ell -} \). As \( \ell \) increases, the slower-oscillating of the two functions shifts out of this reduced region of overlap more quickly than this region can widen such that fast convergence with \( \ell \) is obtained.

When \( K^2 \sim K^2_{BR} = 2 \epsilon_T \), the resonance which can occur between \( F_{\ell -} \) and \( j_{\ell -} \) in the oscillatory region implies that this region can contribute substantially to (21) and yields a peak w.r.t. \( K \) and \( \epsilon_T \), the so-called Bethe ridge (Inokuti 1971). The effective region of overlap is now the maximal overlap region. Moreover, the greater \( \epsilon \) is, the larger is the number of initial low-\( \ell \) oscillations of \( F_{\ell -} \) in the overlap region and the larger is the value of \( \ell \) attained before the oscillatory pattern of \( j_{\ell -} F_{\ell -} \) is pulled out of the overlap region. This results in a slow convergence w.r.t. \( \ell \) in the vicinity of the Bethe ridge — the \((2\ell' + 1)\) factor even favours an initial rise w.r.t. \( \ell \) eventually subdued by the stretching of the first lobe of \( j_{\ell -} F_{\ell -} \) and therefore a broad peak w.r.t. \( \ell \) is exhibited.

On the other hand, as \( \epsilon \to 0 \), \( F_{\ell -} \) oscillates only a few times in the maximal region of overlap. Increasing \( \ell \) quickly pulls \( F_{\ell -} \) out of this region thereby ensuring that, even if there is an initial rise with \( \ell \), fast convergence w.r.t. \( \ell \) soon sets in, whatever \( K \). This convergence can be accelerated for small or large \( K \) by the centrifugal barrier effect in \( j_{\ell -} \) or by a reduction of the effective region of overlap.

Overall, the rate of convergence w.r.t. \( \ell \) is relatively slower where the FFS is relatively large (as a function of \( K \)). Whatever \( \epsilon \), the optical transitions dominate the sum (23) over \( \ell' \) in the limit of small momentum transfer \((K \to 0)\).
Just above the threshold of ionization, the s-d transition still dominates as it does for bound-bound excitation, and therefore indicates that for a given \( L \), the number of oscillations within the range of the initial state does not vary much for final orbitals whether bound or near-threshold continuum. These arguments can indeed be extended to bound-bound transitions (Ton-That and Flannery 1976b). Even at, or below the threshold of ionization, the dominant transitions (here, s-d) still tend to peak w.r.t. \( K \) near the Bethe ridge \( K^2 \sim 2\xi_T \) and so does the FFS summed over \( L \). This fact seems to be a peculiarity of initial s-states.

In accordance with our previous discussion, the dominant transition shifts from s-d to s-f, and on to s-g, etc. as \( \xi \) increases while \( K \) remains in the vicinity of the Bethe ridge.

5. Conclusion

In summary, we have calculated electron-impact Born excitation, Born and binary encounter ionization cross sections of \( \text{He}(2,3 \, \text{s}) \). The cross sections for excitation from \( \text{He}(2,3 \, \text{s}) \) given here are presumed to be accurate Born values. The results for ionization of \( \text{He}(2,3 \, \text{s}) \) are in fair agreement with the measurements (Dixon et al. 1976b) at low and intermediate impact energy \( E \) when the helium ion is left in its ground state. The present results in conjunction with those of Briggs and Kim (1971) raise some questions about the magnitude of multiple processes involving ionization of \( \text{He}(2,3 \, \text{s}) \). We have also discussed the relation between the orbital angular momentum of the final state and the relative importance of its contribution to a partial-wave expansion of the form factor squared, emphasizing the limitation imposed by the range of the initial state orbital.
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### Table 1: Transition Energies $\Delta E$(eV) and Born Cross Sections $(\pi a_0^2)$ for the $(2^3S - n^3L)$ Collisional Excitations of Helium by Electrons of Energy $E$(eV).

<table>
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<th>$n^3L$</th>
<th>$2^3P$</th>
<th>$3^3S$</th>
<th>$3^3P$</th>
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<td>8.25(\dagger)</td>
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<td>3.37-1 (\dagger)</td>
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(a) From Moore (1949)
(b) From method B in text.
(c) Extrapolated from $5^3F$.
\(\dagger\) From Kim and Inokuti (1969), Flannery et al. (1975).

Exponent gives power of ten by which entry is to be multiplied.
Table 2: Born (full-range BF and half-range BH) and Binary Encounter (BE) Cross Sections \(10^{-16} \text{ cm}^2\) for the Ionization of He\((2^3S, 2^1S)\) by Electrons with Energy \(E\) (a.u.).

<table>
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<th>(E) (a.u.)</th>
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<th>(2^1S)</th>
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<tbody>
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<tr>
<td>8</td>
<td>-</td>
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Figure Captions

Figure 1: Comparison of present binary-encounter treatments of the cross sections for outer-shell ionization of He(2 1,3 S) with different orbital-velocity distributions as determined: from methods A and B in text, and from Hartree-Fock functions C of Cohen and McEachran, (1967a,b). V are results of Vriens (1964) using the exponential distribution of Gryzinski (1965). The associated numerals 1 and 3 refer to 2 1 S and 2 3 S targets respectively. The circles (open and full) are measurements of Dixon et al. (1976a) for the sum of all electron-impact processes leading to single (but not double) ionization of a predominantly He(2 3 S) target.

Figure 2: Comparison of present Born cross sections (F and H) for outer-shell ionization of He(2 1,3 S) by electron-impact, with F and H denoting respective integrations over the full and the lower-half ranges of energy of the ejected electron. The associated numerals 1 and 3 refer to 2 1 S and 2 3 S targets respectively. The circles are measurements, as in figure 1.

Figure 3: Cross Sections for electron-impact ionization of He(2 1,3 S). F and C are the full-range Born and binary encounter treatments respectively. Numerals 1 and 3 refer to 2 1 S and 2 3 S targets respectively. CI3 denotes the present binary encounter treatment of inner shell ionization of He(2 3 S) and the C3 full curve includes this contribution. The circles are measurements, as in figure 1.
Figure 4: Bethe plot (cross section times collision energy $E$ versus $\log E$) for electron-impact ionization of He($2^3S$). F3 and H3 full curves are full-range and half-range Born results for outer-shell ionization alone, FI and HI are the corresponding results for inner-shell ionization, included by the F3 and H3 broken curves. BE, BA and BAE represent cross sections of Briggs and Kim (1971) determined from closure in the Bethe approximation BE, the Born asymptotic with and without exchange BAE and BA respectively. The triangles and circles denote measurements of Dixon et al. (1976a) for the sum of all electron-impact processes leading to single (but not double) ionization of a predominantly He($2^3S$) target.
Collision energy (eV)

FIGURE 1

$2^1S$ - Ionization cross sections ($10^{-16}$ cm$^2$)

Collision energy (eV)
FIGURE 2

\[2^1S-\text{ionization cross sections (}\times 10^{-16} \text{ cm}^2)\]

Collision energy (eV)
FIGURE 3

$2^1,3^1 S$-ionization cross sections ($10^{-16}$ cm$^2$)

Collision energy (eV)
Collision energy (eV)

**FIGURE 4**

![Graph showing the relationship between collision energy and QxE (10^-14 cm^2.eV)](image)

- **BE**
- **BA**
- **BAE**
- **He^+(1s)**
- **F3**
- **H3**
- **Fl**
- **He^+(2s)**
Contract No. F33615-76-2033, Item No. 001

Air Force Aero Propulsion Laboratory
Attn: AFALP/POP(450)/Capt. Prince
Wright Patterson AFB, Ohio 45433

Subject: Quarterly R and D Status Report No. 4
        Contract No. F33615-76-C-2033.

Gentlemen:

Transmitted herewith are four (4) copies of the subject reports.

Sincerely,

M. K. Flannery
Principal Investigator and Professor

Enclosures
Calculation of Electron Impact Cross Sections from Metastable States in Atomic and Molecular Cases

Quarterly Progress Report No. 4

Period Covered: July 1, 1976 to September 30, 1976

Contract No. F33615-76-C-2033

Principal Investigator

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School of Physics
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Technical Monitor

Capt. John F. Prince, AFAPL/POP
Project Engineer
USAF Aero Propulsion Laboratory (AFSC)
Wright-Patterson AFB, Ohio 45433

Program Address

Air Force Aero Propulsion Laboratory
Attn: AFAPL/POP(450)/Capt. Prince
Contract No. F33615-76-2033. Item No. 001
Wright-Patterson AFB, Ohio 45433
1. Results

During this period, the binary encounter treatment of ionization was applied to the processes,

\[ e + N_2^* (\Delta \Sigma_u^+) \rightarrow 2e + N_2^+ (\Delta \Pi_u) \]  
\[(1a)\]

\[ e + N_2^* (\epsilon \Sigma_u^-) \rightarrow 2e + N_2^+ (\Delta \Pi_u) \]  
\[(1b)\]

and to

\[ e + CO^* (\alpha 3\pi) \rightarrow 2e + CO^+ (X \Sigma^+). \]  
\[(1c)\]

The ionization cross sections were calculated as a function of electron impact-energy \(E(eV)\). The results are displayed in figure 1. In general, these ionization cross sections are smaller than those reported in the previous progress report no. 3 for \(He^*, Ne^*, Ar^*, Kr^*\) and \(Xe^*\) and are of the order \((2 - 3) \times 10^{-16} \text{ cm}^2\) in comparison to \((8 - 15) \times 10^{-16} \text{ cm}^2\) for the atomic metastables as in figures 2a - 2(d).

A paper describing this work on the atoms and molecules has been written up and is presented as an Appendix.

In the Table are displayed the numerical values of the cross sections for processes (1a - 1c). These are also graphically displayed in figure 1.

In figures (2a - 2d) and (3a - 3d) are shown the cross sections \(\sigma\) versus impact energy \(E\) and Bethe plots \((\sigma \times E)\) versus \(\log_{10} E\) for the ionization of metastable (a) \(Ne^*\) (b) \(A^*\) (c) \(Kr^*\) and (d) \(Xe^*\). The circles shown in figures 2 and 3 refer to the experimental measurements of Dixon, Harrison and Smith (private communication) for the ionization of \(Ne^*\) and \(Ar^*\). See the Appendix for the description of other curves.
Table: Electron-impact ionization cross sections ($10^{-16} \text{ cm}^2$) (Binary encounter method) for the ionization processes 1(a) - 1(c).

<table>
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<tr>
<td>6.</td>
<td>1.54</td>
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</table>

(a) $e + N_2^* (A \ ^3 \Sigma^+ \text{u}) \rightarrow 2e + N_2^+ (A \ ^3 \Pi \text{u})$ (1a)

(b) $e + N_2^* (a' \ ^1 \Sigma^- \text{u}) \rightarrow 2e + N_2^+ (A \ ^2 \Pi \text{u})$ (1b)

(c) $e + CO^* (a \ ^3 \Pi) \rightarrow 2e + CO^+ (X \ ^2 \Sigma^+) \ (1c)$
Figure 1

Ionization cross section in $10^{-16}$ cm$^2$

(a) $N_2^* \left( \chi^3 \Sigma_u^+ \right) \rightarrow N_2^* \left( \chi^2 \Pi_u \right)$

(b) $N_2^* \left( \alpha' \Sigma_u^- \right) \rightarrow N_2^* \left( \chi^2 \Pi_u \right)$

(c) $CO^* \left( \alpha^3 \Pi \right) \rightarrow CO^* \left( \chi^2 \Sigma^+ \right)$

Collision energy $E$ [eV]
Figure (2a)
Figure (2b)
Figure (2d)
Figure (3a)
Figure (3b)
Figure (3c)
Figure (3d)
Cross Sections for Ionization of Metastable Rare Gas Atoms
(Ne*, A*, Kr*, Xe*) and of Metastable N2*, CO* Molecules by Electron Impact

D. Ton-That and M. R. Flannery
School of Physics
Georgia Institute of Technology
Atlanta, Ga. 30332

Cross sections for the collisional ionization of the metastable atoms Ne*, A*, Kr* and Xe* by electrons with impact energy E in the range 6 eV < E < 250 eV are determined in the Born and the binary encounter approximations. For low energies \( \epsilon \) of ejection, the s-d and s-f bound-free transitions dominate the associated form factor while transitions to continuum states with progressively higher angular momentum gain importance with increasing \( \epsilon \). While up to 9 partial waves are normally sufficient for convergence of the bound-free form factor at a given energy \( \epsilon \) and momentum-change \( K \) of the ejected electron, as many as 30 are required for those \( K \) in the vicinity of the Bethe ridge at large \( \epsilon \). These properties are the origin of the overall closeness obtained between the Born and binary-encounter cross sections. Also inner-shell ionization, as described by the binary encounter treatment, becomes increasingly important as the target atom becomes more complex. Cross sections for ionization of metastable N2* and CO* are also determined. Good agreement with available measurements (for Ne* and Ar*) is obtained.
1. Introduction

Apart from the Born and binary-encounter treatments for the ionization of metastable helium by electron-impact, little is known about the cross sections for ionization of metastable rare-gas atoms \(X (\text{Ne}^*, \text{Ar}^*, \text{Kr}^*, \text{Xe}^*)\) by electron impact. This information is important in the modelling and feasibility studies of certain excimer lasers. In particular, very recent observations of high-power laser emission from a new class of molecules - the noble-gas monohalides (\(\text{ArF}^*, \text{KrF}^*, \text{and XeF}^*\), for example) - have demonstrated the potential of a new class of high-power, high-efficiency and partially tunable lasers operating around 3000 Å. The lasing transition originates on an excited molecular state \(XF^*\) formed directly by \(X^*-\text{F}_2\) rearrangement collisions or indirectly by three-body ion-ion recombination between the positive ions \(X^+\) produced by electron-impact ionization of \(X(^*)\) and the negative ions \(F^-\) formed by dissociative attachment in (e-\(\text{F}_2\)) collisions. The ionization of metastable rare gas atoms therefore plays a key role, as a mechanism for depletion of atomic metastables in the former case and as a source of production of metastable excimers via ionic recombination in the latter case.

In this paper, cross sections for the ionization of metastable \(\text{Ne}^*, \text{Ar}^*, \text{Kr}^*, \text{and Xe}^*\) by electron impact are determined as a function of the collision energy \(E\) by means of the Born approximation and the binary encounter method which is also applied to the ionization of metastable \(\text{N}_2^*\) and \(\text{CO}^*\). It is worth noting that the corresponding theoretical treatments of electron-impact ionization of \(\text{H}(2s)\) and \(\text{He}(2^1,3^1S)\) yield cross sections in good agreement with recent measurements.
2. Basic Formulae

The cross section for ionization of a singly excited atom $B$ with mass $M_B$ and ionization potential $I$ by an incident particle $A$ with mass $M_A$, speed $v$ and relative energy $E$ is, in the (elastic) binary encounter approximation given by\textsuperscript{11,12}

$$
\sigma_{n\ell}^I(E) = \frac{\pi}{M_{AE} v^2} \int_0^\infty \int_0^{p^+} \int_0^{p^-} \frac{4}{p^4} I(P) \, dP \, u \frac{f_{n\ell}(u)}{u^2} \left( \frac{\partial}{\partial u} \right) \left( \frac{E}{I} \right) \frac{dE}{I} \, du
$$

where the distribution in speed $u$ of the valence electron described by a spatial wavefunction $\phi_{n\ell m}(\mathbf{r})$ is with all quantities in atomic units,

$$
f_{n\ell}(u) = \frac{1}{(2\pi)^{3/2} u^{3/2}} \left| \int \phi_{n\ell m}(\mathbf{r}) \frac{-i}{\hbar} \frac{e^{-i\mathbf{K}\cdot\mathbf{r}}}{\mathbf{r}} \, d\mathbf{r} \right|^2 u^2 \, du
$$

and $M_{AE}$ is the reduced mass of the ($A - e$) binary system.

For a specified energy-transfer $\epsilon_T$ to the valence electron, the momentum-change $P$ can vary between the lower limit,

$$
P^- = \max [M |u' - u|, M_{AB} |v' - v|]; \quad M = m \left( 1 + \frac{m}{M_i} \right), \quad M_{AB} = \frac{M_AM_B}{(M_A + M_B)},
$$

where $m$ and $M_i$ are the electronic and ionic masses respectively, and the upper limit

$$
P^+ = \min [M(u' + u), M_{AB}(v' + v)]
$$
where the post binary-collision speeds of the projectile and target particles
are respectively,

\[ v' = (v^2 - 2\varepsilon_T/M)^{1/2} \] (5)

and

\[ u' = (u^2 + 2\varepsilon_T/M)^{1/2} \] (6)

In the general expression (1), the function \( \Gamma(P) \), which represents the departure
of the differential cross section for \((A-e)\) elastic scattering from the
Rutherford value, is set to unity for direct \((e-e)\) collisions for which
\( M \approx M_{AB} \approx m \).

The Born approximation to the cross section for single ionization of an
atom with \( N \) electrons by an incident electron can be written, with all quantities
in atomic units, as

\[ \sigma_{1n}^{I}(E) = \alpha(E-I) \int d\varepsilon \frac{1}{(2\pi)^2} \sum_{m=-\infty}^{\infty} \int \sigma_{1nkm}^{I}(\varepsilon, \hat{k}_e) d\hat{k}_e \] (7)

in terms of the differential cross section for ejection of the electron with
momentum \( \hat{k}_e \) per unit solid angle and unit energy interval,

\[ \sigma_{1nkm}^{I}(\varepsilon, \hat{k}_e) = \frac{8\pi}{k_i^2} \int \left| \langle \psi_{i}(\varepsilon, \hat{k}_e; r) \right| \sum_{i=1}^{N} e^{iK_{f}r_i} \left| \psi_{f}(\varepsilon, \hat{k}_f; r) \rangle \right| dK \] (8)

where \( \psi_{i,f}(r) \) are the initial and final electronic wavefunctions for the rare-
gas atom with electronic coordinates denoted collectively by \( r \). The limits to
the momentum-change \((\hat{k}_{1} - \hat{k}_{f})\) of the scattered electron are,
\[ K^T = (2E)^{1/2} \mp [2(E - I - \varepsilon)]^{1/2} \]  

where the energy \(\varepsilon\) of ejection is \(\frac{1}{2} k^2\) a.u. and where the kinetic energy transferred to the ion is neglected. While the parameter \(a\) in the \(\varepsilon\)-integration limit in (7) is unity for ionization involving distinguishable particles, Rudge and Seaton have shown that, for ionization of atomic hydrogen by electrons with random spin orientations, \(a = 0.5\) when electron-exchange effects are fully neglected. While this choice however ensures that the faster of the scattered and ejected electrons is always described by a plane wave, neither choice is rigorously based for ionization of atoms more complex than hydrogen. The spatial wavefunction \(\psi_\uparrow(x)\) for the initial bound metastable state of the rare gas atom is taken as a simple product of the one-electron orbitals with quantum numbers \((n,l,m)\)

\[
\phi_{nlm}(x_j) = \frac{1}{r_j} P_{nl} (r_j) Y_{lm}(\hat{x}_j), \quad j = 1, 2, \ldots, N
\]

where the \(Y_{lm}\) are spherical harmonic functions. The corresponding wavefunction \(\psi_\uparrow(x)\) for the final ionized state includes the orbital

\[
\phi_\varepsilon(x; \hat{k}_e) = \sum_{l'} \sum_{m=-l'}^{l'} \frac{\eta_{l'}}{r} e^{-i\eta_{l'}} F_{\varepsilon l'}(r) Y_{l'm'}(\hat{x}) Y_{l'm'}^*(\hat{k}_e) \]

for the electron ejected in direction \(\hat{k}_e\) with energy \(\varepsilon\). In this study, these orbitals are obtained from a central field approximation such that the above radial functions \(P_{nl}\) and \(F_{\varepsilon l}\) are appropriate solutions of the radial Schrödinger equation,
where the spherically symmetric interaction $V(r)$, which tends to $(-N/r)$ as $r \to 0$ and to $(-1/r)$ as $r \to \infty$ and which is frozen for all the orbitals, is determined in a manner described in the following section. All the bound and continuum orbitals therefore form an orthonormal set such that the form factor in (9) reduces to a one-electron form factor involving only the ejected electron.

The bound solutions $P_{n\ell}$ to (12) are normalized to unity (for $E < 0$) and the radial continuum function behaves asymptotically as

$$F_{n\ell}^\infty(r) \sim \left(\frac{2}{\pi k_e}\right)^{1/2} \sin \left[ k_e r - \frac{1}{k_e} \ln(2k_e r) - \frac{1}{2} \ell \pi + \eta_{n\ell} \right]$$

where the additional phase shift $\delta_{n\ell}$ is a measure of the departure of the electron-ion interaction from pure Coulomb. The amplitude $\frac{1}{4\pi} \frac{1}{2} \frac{1}{4}$ is chosen so as to fulfill the normalization condition,

$$\langle \phi_e(k_e; \ell) | \phi_e(k_e'; \ell') \rangle = \delta (\varepsilon - \varepsilon') \delta (k_e - k_e')$$

where $\varepsilon = \frac{1}{2} k_e^2$ is in atomic units such that the ionization cross section (7) is obtained by integrating (8) over $k_e$ and $\varepsilon$. By first performing the $k_e$-integration, then it can be shown that the differential cross section per unit $\varepsilon$ is, for initial s-states,
where the radial matrix element to be evaluated is

\[
M_{\ell',\ell,ns}(\varepsilon,k) = \int_0^\infty e^{i\varepsilon r} F_{\ell',\ell}(r) j_{\ell'}(kr) p_{ns}(r) dr
\]  

in which \( j_{\ell'} \) is the spherical Bessel function.

Antisymmetry of the target wavefunction is omitted although some account of exchange effects in the target is introduced by the use of the Hartree-Slater approximation for the interaction potential \( V(r) \).
3. Wavefunctions for the Rare Gas Atoms

The singly-excited states of rare gas atoms, in general, follow neither the pure LS nor the jj angular momentum coupling schemes. These excited states are obtained by single excitation of one of the np outer-shell electrons of the ground-state configuration to a n' ℓ' state. The binding energy of (or effective charge seen by) this excited valence electron is substantially less than that for the np electrons, and the excited electron is, on the average, relatively distant from all the core electrons, including the np-shell electrons. The spin-orbit interaction of the electrons of the atomic core can therefore be greater than the electrostatic interaction of these electrons with the excited electron and this effect manifests itself in the distinctive "core splitting" structure of the spectrum. The intermediate coupling scheme which describes the angular momentum structure is therefore closest to the pure jℓ (or jK) coupling wherein the orbital angular momentum ℓ of the valence electron is coupled to the total angular momentum ℓ of the atomic core and their resultant K is in turn coupled to the valence electron spin to give the total angular momentum J of the atom.

Whatever the coupling, pure or intermediate, the total angular momentum J of the atom is a good quantum number and remains invariant under the various recoupling transformations. For the lowest excited configuration np^5(n + 1)s, J can take the values 0, 1 (twice) and 2. Since dipole transitions within the same configuration are parity-forbidden and since the ground state is 1S0, the (np)^5(n+1)s states with J = 0 and J = 2 are therefore metastable. Expansion of these states from an intermediate coupling to a jK basis projects onto the \( ^2P_{1/2} \) \( [K = \frac{1}{2}, J=0] \) state, for J = 0, and onto the \( ^2P_{3/2} \) \( [K = 3/2, J=2] \) state, for J = 2.
Alternatively an expansion to an LS basis projects onto $^3P_0$ for $J = 0$ and $^3P_2$ for $J = 2$. Since no mixing is involved in each case, either set of pure-state notations provides convenient labeling of the metastable states. Of the remaining LS states, $^1P_1$ and $^3P_1$, the possible metastability arising from $^3P_1$ is lost by its mixing with $^1P_1$ which is dipole-connected with the $^1S_0$ ground state.

In this study the effective field (potential) $V(r)$ is the same for all target electrons. Moreover the atomic core is regarded as frozen and the continuum electron moves in the same field as the initially bound excited valence electron. This potential is determined in the Hartree-Slater approximation via the following modification to the standard program of Herman and Skillman.\textsuperscript{15} A self-consistent field (SCF) iteration for the initial excited-state configuration $[1s^2 \, 2s^2 \, \ldots \, np^5 \, (n+1)s]$ yields $V_i(r)$ which when inserted in (12) provides orthonormal $P_{(n+1)s}$ for a wide range of angular momentum $l$ (from 0 to about 30) and of energies $\varepsilon$ of the ejected electron. This procedure which differs from the standard one, based on an interaction $V_o(r)$ obtained from a SCF iteration of the ground-state configuration, has been found\textsuperscript{1} to yield considerable improvement for the helium excited states with symmetry different to that of lower-lying states. This improvement is attributed to the allowance of core relaxation whilst the symmetry condition avoids the necessity of explicit orthogonalization of the wavefunctions to those of lower-lying states, the ground state, in particular. In the present case the excited and ground state configurations $np^5 \, (n+1)s$ and $np^6$ are orthogonal because of the orbital angular momentum of the valence electron. As the number of atomic electrons increases, the Hartree-Slater approximation for exchange is expected to improve. On the other hand, core relaxation in
the excited states diminishes whilst relativistic effects such as spin-orbit coupling, which are not included in the present treatment, become important. The same orbitals are used for the \( ^3P_2 \) and \( ^3P_0 \) (or \([3/2]_2\) and \([1/2]_0\)) states since the major contribution to their splitting comes from spin-orbit coupling in the core, an effect not included in the present treatment.

Knowledge of the bound and continuum orbitals permits evaluation of the Born radial matrix element (16) and of the speed distribution (2) for use in the binary-encounter formula (1).

Whilst preserving or introducing the following approximations of (a) omission of antisymmetry, (b) magnetic quantum-number independence of the radial orbitals \( P_{n\ell}(r) \) and \( F_{\epsilon\ell'}(r) \), (c) explicit \( k_e, \xi \) dependence of the continuum orbitals (cf. 11) i.e. uncoupling of the orbital angular momentum \( \ell' \) of the ejected continuum electron from all other angular momenta and (d) lack of selection of the angular momentum configuration of the residual ion, one finds that the introduction of explicit coupling of the individual angular momenta (spin and orbital) of the target electrons in an intermediate, pure \( jK \) or other coupling scheme, leaves the expression (15), initially derived without coupling, unchanged.
4. Results and Discussion

4.1 Ionization of Ne*, Ar*, Kr*, Xe*.

The ionization cross sections of metastable Ne, Ar, Kr and Xe versus the impact energy E are displayed in Fig. 1a-d. The same cross section is assigned to the \( ^3P_2 \) and \( ^3P_0 \) metastable states which are treated as degenerate. The ionization potentials I used in the expressions (1), (7) and (9) are the weighted average of the potentials of the \( ^3P_{2,0} \) states for ionization without core transitions. The values so derived from the Tables of Moore\(^16\) are given in Table 1, together with the \( p^5 \) inner-shell ionization potentials.

As the target varies from Ne* to Xe*, its physical size increases, its ionization potential decreases and the ionization cross sections exhibit the systematic increase in Fig. 1a-d. Moreover, the binary encounter curves BE drop in relation to BF, the Born cross sections obtained from integration over the full-range of energy \( \xi \) of the ejected electron i.e. \( \alpha = 1 \) in (7). For example, the separation between BE and BF (which is remarkably small for He*\(^1\) and Ne*) widens with variation of the target from Ne* to Xe* until BE eventually comes into close agreement with BH, the "half-range" Born cross sections i.e. (7) with \( \alpha = \frac{1}{2} \).

The origin of the general agreement in shape and magnitude between the binary encounter and the Born curves has already been fully investigated\(^1\) (for the ionization of He*) and is rather instructive. For most values of the energy \( \xi \) and the momentum-change \( K \) of the ejected electron, the number of \( \ell' \) partial waves required for convergence of the matrix element (16) or form-factor with respect to \( \ell'' \), the angular momentum of the ejected electron, is relatively small, 9 or less. However, for large \( \xi \), the required number increases sharply near the Bethe ridge defined by,
where a "resonance" can occur between \( F_{E_{\lambda}} \) and \( j_{\lambda'} \) in (16) and where up to 30 partial waves are required for convergence. Moreover, for low \( \lambda' \), the \( s-d,f \) (i.e. \( \lambda' = 2,3 \)) transitions dominate (16). As \( \lambda' \) is increased, the dominant transition shifts from \( s-d,f \) to \( s-g, s-k, \) etc., for those \( K \) near the Bethe ridge where a broad distribution of (16) over \( \lambda' \) is exhibited. For large \( \lambda' \), these contributions maximize w.r.t. \( K \) in the vicinity of the Bethe ridge. These properties can be interpreted mainly in terms of the spatial range of the wavefunction of the initial excited state, and of the drift of both the final continuum orbital \( F_{E_{\lambda'}}(r) \) and Bessel function \( j_{\lambda'}(Kr) \) out of this range. The overall agreement between the Born and the binary encounter curves originates from the resulting closeness between the Born and the binary encounter determinations of the form factor squared in (8) away from the optical \( K \to 0 \) limit. The impact energies for which the ionization cross section is relatively large involves momentum transfers \( K \) for which the form factor requires many partial waves and is substantial. This situation, where many partial waves and therefore multipoles are required, is generally well described by the binary encounter method.

Also shown in Fig. 1 is the binary-encounter contribution arising from the \( p^5 \) inner-shell, obtained by multiplying the ionization cross section for ejection of a single \( p \) electron by the occupation number of the \( p^5 \) shell. The outer valence electron remains in its initial excited state. Again, as the target varies from Ne* to Xe*, the inner-shell ionization potentials drops, the inner-shell ionization cross section increases in magnitude, absolute as well as relative to the outer-shell ionization cross section, and its effect on the shape of the excitation (ionization) function becomes more prominent by the display of a secondary maximum.
The results for ionization of Ne* and A* are compared with the experimental data of Dixon et al.\textsuperscript{17} Whereas the measurements are in better agreement with the Born full-range calculations\textsuperscript{1} for ionization of He(2 3S) the experimental data for ionization of Ne* and A* are in closer agreement with the (lower) Born half-range calculations than with the Born full-range results. The data for A* support the increasing importance of p\textsuperscript{5} inner-shell ionization. Dixon et al.\textsuperscript{10} have subjected their measurements of ionization of He (2 3S) to a correction for systematic effects arising from charge exchange reactions occurring in the experimental collision region. This correction, if applicable to the data for Ne* and A*, is not included in the data shown here in Fig. 1a,b; however the correction is appreciable only at impact energies greater than 100 eV, a range which is beyond that of the experimental data shown here.

Also shown for completeness are previous binary encounter calculations of Vriens\textsuperscript{18}, based however on an exponential velocity distribution which is at variance\textsuperscript{19} with any proper quantal distribution, such as the one used here.

4.2 Bethe Plots

The calculations and experimental data for ionization of Ne*, A*, Kr*, and Xe* are also presented in Fig. 2a-d in the form of Bethe plots (σ\textsuperscript{1} x E) versus Log\textsubscript{10} E) which emphasize the behavior in the intermediate and near-asymptotic energy range.

These plots show that the slopes of the Born curves for outer-shell ionization of metastable Ne, A, Kr, and Xe at impact energies beyond 100 eV are quite small in comparison with, say, that\textsuperscript{1} for outer-shell ionization of He(2 3S) (cf. Fig. 4 of Ref. 1) thereby indicating that the outer-shell photo-ionization cross sections of metastable Ne, A, Kr, and Xe are substantially smaller than the corresponding cross section of He 2 3S over an appreciable
range of ejected electron energy beginning from threshold. Thus, a reversal, between target He(2\(^3\)S) on one hand, and metastable Ne, A, Kr, and Xe targets on the other, occurs in the magnitudes of the cross sections depending on whether the outer-shell ionization is caused by electron impact or by photon impact. However, the effective photoionization cross section also involves the contribution from the contiguous inner shell; for Ne\(^*\) - Xe\(^*\) the inner shell (p\(^5\)) has five electrons instead of one for He(2\(^3\)S) and it begins to contribute at lower impact-energies.

For the heavy rare gases the E\(^{-1}\) asymptotic dependence (instead of the correct E\(^{-1}\) Log\(_{10}\) E) of the ionization cross section normally exhibited in the binary encounter approximation does not appear until E \(\approx\) 1 KeV. This late onset, combined with the weakness of the dipole transition oscillator strength discussed earlier, corresponds to the fact that the binary encounter curves for outer-shell ionization remains above the Born curve BF as the impact energy becomes large, instead of crossing below as implied by the asymptotic dependence (cf. figs. 1 and 2).

While there is reasonable accord between the Bethe plots associated with the theoretical and experimental results at low energies when only ionization of the outer-shell occurs, the fig. 2, in contrast to fig. 1, clearly shows that any measure of agreement between the asymptotic slopes is obtained only by inclusion of the p\(^5\) inner shell contributions, especially for A\(^*\). These inner-shell contributions are adequately described by the binary encounter method and for Kr\(^*\) and Xe\(^*\), become larger and increasingly important at the lower impact-energies.

4.3 Tabulation

The numerical values of the electron-impact ionization cross-section of Ne\(^*\), A\(^*\), Kr\(^*\), and Xe\(^*\) calculated in the full-range and half-range Born approxi-
mations (for ionization from the outer-shell alone) and in the binary-encounter approximation (with and without the inner-shell contributions) are given in Tables 2 and 3. Measurements for electron-impact ionization of Kr* and Xe* are not available; the binary encounter results inclusive of p5 shell contributions or, for E ≲ 20 eV, the Born half-range results, should provide reasonable estimates of the cross sections of Kr* and Xe*.

We have not considered multiple processes such as double ionization or ionization with simultaneous excitation. In relation to single processes, multiple processes are small for a He target (≈ 1%) but are less so for heavier rare gases with larger occupation numbers and lower excitation energies (see for instance Schmidt et al. 20). For a precise comparison between theory and experiment, one should include these multiple processes fully in the calculations or else ensure that they do not contribute to the measurements.

4.4 Ionization of Metastable N2 and CO

Ionization of diatomic molecules like N2 and CO in metastable states is also important for gas laser dynamics. We have therefore applied the binary encounter approximation of Section 2 to the electron-impact ionization of N2 in the A 3Σu+ and a 1Σg+ metastable states and of CO in the a 3Π metastable state.

The electronic configurations of the metastable states of N2 and CO as well as those of the low-lying states of N2+ and CO+ are shown in Table 3. It shall be assumed that single processes, involving only one target electron, are more important than multiple processes requiring multi-electron target transitions. This assumption also makes the ionization problem tractable within the framework of the binary-encounter approximation of Section 2; otherwise, inclusion of multiple processes would require consideration of energy exchange with the residual ion. We shall therefore restrict ourselves to consideration of the following single-process transitions,
\[ e + N_2 (A^3\Sigma_u^+) \rightarrow 2e + N_2^+ (A^2\Pi_u) \quad [I = 10.79 \text{ eV}] \quad (17) \]
\[ e + N_2 (a'^1\Sigma_u^-) \rightarrow 2e + N_2^+ (A^2\Pi_u) \quad [I = 8.56 \text{ eV}] \quad (18) \]
\[ e + CO (a^3\Pi) \rightarrow 2e + CO^+ (X^2\Sigma^+) \quad [I = 8.2743 \text{ eV}] \quad (19) \]

The molecular orbitals used are the orbitals previously derived by Richardson\textsuperscript{21} for \( N_2^* \) and Huo\textsuperscript{22} for \( CO^* \), who used Slater-type-orbital (STO) bases. (Similar orbitals have also been calculated by Rose and McKoy\textsuperscript{23} using Gaussian-type-orbital (GTO) bases.) Hence, the appropriate Fourier transforms are obtained and averaged over angles as in (2) to give the initial speed distribution \( f(u) \). Although a spherical distribution may be a poor approximation because of the molecular axis, the effects of rotation and random orientation of the molecular target partially offset this inadequacy. The ionization potentials used in (1) for each molecular orbital are the vertical ionization potentials derived from the data of Gilmore\textsuperscript{24} for \( N_2^* \) and Krupenie\textsuperscript{25} for \( CO \), at the equilibrium internuclear distance of the initial state, even though the molecular orbital may have been derived at the nuclear separation of the ground state. The cross sections for the ionization processes (17)-(19) at impact energies below 200 eV are shown in Fig. 3.

Since Richardson\textsuperscript{21} gives the same \( 1\pi_g \) orbital for \( N_2 (A^3\Sigma_u^+) \) and \( N_2 (a'^1\Sigma_u^-) \), expression (1) adopts the same speed distribution for these two states. The only difference arises from the ionization potentials and the main effect is a reduction in the magnitude of the ionization cross section.

On the other hand, \( N_2 (a'^1\Sigma_u^-) \) and \( CO (a^3\Pi) \) have very similar potentials for single-process ionization. Any slight differences between the curves for ionization of \( N_2 (a'^1\Sigma_u^-) \) and \( CO (a^3\Pi) \) are therefore attributed to differences.
between the $1\pi_g$ orbital of $N_2^*$ and the $2\pi$ orbital of $CO^*$. Any similarity between the properties of $N_2$ and CO usually originates from the isoelectronic character of these molecules, while any difference arises from the heteronuclear aspect.

While the ground states of $N_2$ and CO have similar electronic configurations, $N_2^* (a^* 1\Sigma_u^\text{−})$ and $CO^* (a^3 \Pi)$, and for that matter $N_2^+ (A^2 \Pi_u)$ and $CO^+ (X^2 \Sigma^+)$, have slightly different electronic configurations. However, since the binary encounter method of Section 2 relies mainly on the properties of the valence electron ($1\pi_g$ or $2\pi$), the difference between the configurations of the remaining electrons is reflected only indirectly through its effect on the valence electron ($1\pi_g$ or $2\pi$).

Metastable $N_2$ and CO have greater ionization potentials than metastable rare gases and have correspondingly smaller electron-impact ionization cross sections as seen by comparing fig. 1 with fig. 3.

5. Conclusion

Electron-impact ionization cross sections of [np$^5$ (n+1)s] metastable Ne, Ar, Kr, and Xe have been calculated in the Born and the binary encounter approximations. The further approximations used here make explicit coupling of the target angular momenta — to reflect the peculiarities of the heavy rare gas spectra — unnecessary.

As the atomic number of the target increases, the cross section for ionization from the outer shell increases. Ionization of the inner p$^5$ shell also becomes increasingly important with increasing target complexity. Moreover, the binary encounter results drop in relation to the Born results.

The maximum of the experimental data of Dixon et al.$^{17}$ is in fair agreement with that of the Born half-range calculations. Beyond the maximum the inner-shell contribution becomes evident. The Bethe plots suggest that the outer-
shell photoionization cross sections of metastable Ne, A, Kr, and Xe are overall smaller than that of He$^*(2^3S)$.

The binary encounter approximation has also been used for the single-process ionization of the π valence electron of metastable N$_2$ and CO. Their greater ionization potentials lead to cross sections smaller than for He$^*$, Ne$^*$, A$^*$, Kr$^*$, and Xe$^*$. 
Table 1: Potentials $I_0$ and $I_\perp$ for Ionization of the ns Outer-shell and the (n-1) $p^5$ Inner-shell of Metastable Rare Gas Atoms

<table>
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<tr>
<th>$X^*$ (ls$^2$, 2s$^2$ ... (n-1) $p^5$, ns)</th>
<th>$I_0$ (eV)</th>
<th>$I_\perp$ (eV)</th>
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<tr>
<td>Ne$^*$ (3s)</td>
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</tr>
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<td>Ar$^*$ (4s)</td>
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<td>20.88</td>
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<td>Kr$^*$ (5s)</td>
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<td>18.16</td>
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<tr>
<td>Xe$^*$ (6s)</td>
<td>3.44</td>
<td>15.41</td>
</tr>
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</table>
Table 2: Born (full-range BF and half-range BH) and Binary encounter (BE) cross sections (10^{-16} \text{ cm}^2) for the ionization of the outer-shell of metastable Ne* (2p^5 3s) and Ar* (3p^5 4s) by electrons with energy E(a.u.). The cross sections BEI include additional inner-shell contributions as determined from the binary-encounter approximation.

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<th>BH</th>
<th>BE</th>
<th>BEI</th>
<th>Ar* BF</th>
<th>BH</th>
<th>BE</th>
<th>BEI</th>
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Table 3: Born (full-range BE and half-range BH) and Binary encounter (BE) cross sections \(10^{-16} \text{ cm}^2\) for the ionization of the outer-shell of metastable Kr* \((4p^5 5s)\) and Xe* \((5p^5 6s)\) by electrons with energy \(E(\text{a.u.})\). The cross sections BEI include additional inner-shell contributions as determined from the binary-encounter approximation.

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<th>(E(\text{a.u.}))</th>
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**Table 4: Electronic Configurations for N$_2^*$, N$_2^+$, CO$^*$, CO$^+$**

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<th>$^1\Sigma_u^-$</th>
<th>$^2\Sigma_g^+$</th>
<th>$^2\Pi_u$</th>
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</table>

$^\dagger$ cf. Gilmore, Ref. 24

$^\ddagger$ cf. Krupenie, Ref. 25

References


Figure Captions

**Fig. 1.** Cross Sections (10^{-16} \text{ cm}^2) for collisional ionization of metastable (a) Ne*, (b) A*, (c) Kr* and (d) Xe* by electrons with impact energy E(eV).

BF and BH are the present Born results for outer-shell ionization obtained from integrations over the full and lower-half ranges of energy $\varepsilon$ of the ejected electron i.e. $\alpha = 1$ and $\frac{1}{2}$ respectively in (7) of text. The binary encounter (quantal distribution) cross sections are denoted by BEO for outer-shell ionization, by BE1 for ionization of one of the electrons in the np$^5$ shell, by BE5 for the total ionization of the np$^5$ shell and by BE for the sum of BEO and BE5. Previous binary encounter (exponential distribution) results of Vriens are represented by V. Q: measurements (Dixon, Harrison and Smith$^{17}$).

**Fig. 2.** Bethe plots (cross section times collision energy $F$ versus $\log_{10} E$) for electron-impact ionization of metastable (a) Ne* (b) A* (c) Kr* and (d) Xe*. Labelling of curves as in fig. 1.

**Fig. 3.** Binary-encounter cross sections (10^{-16} \text{ cm}^2), (a), (b) and (c), for the ionization of metastable N$_2^*$ ($A^\text{3}\Xi_u^+$), N$_2^*$ ($a^\text{1}\Xi_u^-$) and CO* ($a^\text{3}\pi$) respectively by electrons with energy E(eV). The final state of the residual ion is indicated.
Figure 1.
Figure 2.
Figure 3.
Calculation of Electron Impact Cross Sections from Metastable States in Atomic and Molecular Gases

Quarterly Progress Report No. 5

Period Covered: October 1, 1976 to December 31, 1976

Contract No. F33615-76-C-2033

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1. Results

During this contract period, we considered the collision,

\[ e + X^*(n_{i}l_{i}) \rightarrow e + X^{**}(n_{f}l_{f}) \quad (1) \]

at intermediate energies in an effort to determine whether one could, given an initial state \((n_{i}l_{i})\) and a final principal quantum number \(n_{f}\), select which final angular momentum state \(l_{f}\) was populated favorably in such a collision. Such information is critical in determining, for example, what states should be closely coupled in the multichannel eikonal treatment of atomic collisions. Previous theoretical investigation on the excitation

\[ e + He(2^1,3S) \rightarrow e + He(3^1,3L) \quad (2) \]

\[ + e + He(n_{f}^1,3L) \quad (3) \]

showed us that the \(3^1,3D\) state was preferentially populated at low and intermediate energies, rather than the \(3^1,3P\) state which is dipole connected to \(2^1,3S\). (At much higher energies \(\sim\) hundreds of eV, the \(2^1,3S - 3^1,3P\) cross section dominates, as expected). The preferential population of the \(3^1,3D\) state at low and intermediate energies was predicted by both Born and Multichannel Eikonal Treatments of the collision \((2)\). Also this fact was experimentally confirmed by the recent measurements of A. Garscadden and M. L. Lake. Also, we have performed Born calculations of \((3)\) and these showed us that the \(2^1,3S - 4^1,3D\), and the \(2^1,3S - 5^1,3D\) transitions were the most favored for transitions into the \(n_{f} = 4\) and \(n_{f} = 5\) shells respectively. In fact, for electron-induced transitions out of \(He(2^1,3S)\), those final discrete \(1,3D\) states are preferred for a given \(n_{f}\), up to the ionization threshold.

We consider these observations to be extremely important in any analysis or interpretation of processes involving excited states in general; since,
according to rules of thumb developed for ground-state atoms, the dipole transitions i.e. S-P, would be preferred. Accordingly, we considered whether the above behavior is unique to \( \text{He}^* (2 \, ^1\!S) \). We have found that the behavior is quite characteristic of excited states in general and have discovered several rules whereby one could ascribe the most probable \( l_f \) in (1). Applying these rules to \( \text{H}(n\ell) \), we have found that

<table>
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<th>final states ((n_f,\ell_f))</th>
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</tr>
<tr>
<td>2p</td>
<td>3d, 4d, 5d, ... ( \infty )d</td>
</tr>
<tr>
<td>3(s,p,d)</td>
<td>4f, 5f, 6f, ... ( \infty )f</td>
</tr>
<tr>
<td>4(s,p,d,f)</td>
<td>5g, 6g, 7g, ... ( \infty )g</td>
</tr>
</tbody>
</table>

are the dominant transitions out of a given initial state \((n\ell)\) to a final state with principal quantum number \( n_f \). In general, we found that the most probable \( \ell_f \) depends not on the initial value \( \ell_i \) but on \( n_i \), which characterizes the physical size of the initial excited atom. Whether a transition is dipole or not is immaterial. It may occur however that the 3d-4f and 4f-5g transitions in (4) are dipole, but this is not the essential reason for their dominance. We have found that the preferred \( \ell_f \) is given by the simple formula

\[
\ell_f^{(\text{max})} = n_i^{\frac{1}{2}} - \frac{1}{2} \quad \text{as} \quad n_i \to \infty
\]

In an effort to ascertain whether the predictions as given by (4) were evident from the previous literature, but had not been observed as such, we conducted a literature search and found on examining the Born calculations on \( e - \text{H}(3\ell) \) scattering by McCoyd, Milford and Wahl (Physical Review 119 (1960) 149) that the predictions (4) are correct i.e. that the 4f and 5f states are populated the most for transitions to \( n_f = 4 \) and \( n_f = 5 \) respectively.
A preliminary survey of the above work was presented at the Division of Electron and Atomic Physics Meeting of the American Physical Society held at Lincoln Nebraska 6-8 December 1976. The abstract appears on page 4.

2. Publications

(a) The paper "Cross sections for ionization of metastable rare gas atoms (Ne*, Ar*, Kr*, Xe*) and of metastable N2*, CO* molecules by electron impact by D. Ton-That and M. R. Flannery has been accepted for publication in Physical Review A and is due to appear in the Feb. 1977 issue.

(b) The paper "Cross Sections for excitation and ionization in e—He(2 1,3S) collisions" by D. Ton-That, S. T. Manson and M. R. Flannery has been accepted for publication in J. Phys. B: Atom. Molec. Phys. (London).
On Populating $\ell'$-states in Intermediate-impact-energy, Direct-scattering $n\ell + n'\ell'$ Excitation. D. TON-THAT and M.R. FLANNERY, Ga. Inst. of Tech.†—As the value of the orbital angular momentum $\ell'$ of the final state increases, the magnitude of the inelastic form factor, overall, rises until for $\ell' > \ell'_{\text{max}}$ it drops sharply. One finds the value $\ell'_{\text{max}}$ to be insensitive to those of $n'(>n)$ and $\ell$, but, for $n, \approx n$ or somewhat greater, for large $n$. These features can be regarded as resulting from the drift of the pattern of the final radial orbital out of the effective range of the initial radial orbital. For $H$, the effective range is $nr_0$ where $r_0$ is some appropriate length unit, whilst the drift rate is $\Delta r/\Delta \ell = (n'r_0-n'0)/(1+(n'-1))$, $(n'-1)-0 \approx r_0$; so, $\ell'_{\text{max}} = nr_0/r_0 = n$. For heavier atoms, $\ell'_{\text{max}}$ may be smaller. These considerations are relevant to the subject matter.

†Supported by AF Aero Prop. Lab., AF Syst. Command, USAF, Contract F33615-76-C-2033

Submitted by

M. R. Flannery

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Calculation of Electron Impact Cross Sections from Metastable States in Atomic and Molecular Gases

Quarterly Progress Report No. 6

Period Covered: January 1, 1977 to March 31, 1977

Contract No. F33615-76-C2033

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ATTN: AFAPL/POP(450)/Capt. Prince
Contract No. F33615-76-2033. Item No. 001
Wright-Patterson AFB, Ohio 45433
1. Research

In contrast to the excitation of atoms, to discrete levels, the only present-day theoretical treatments of electron-impact ionization are provided by the Bonn and the binary encounter (first-order) approximations. Also there are apparently no reliable methods for the description of ionization accompanied by core excitation.

Accordingly, Dr. McCann and myself are in the process of extending and modifying the multichannel eikonal treatment (designed originally for discrete excitation) so as to formulate a treatment of ionization, more accurate than Bonn's approximation. We are also including specifically the effects of core excitation accompanied by ionization as in,

\[ e + \text{He}(2^13_s) \rightarrow \text{He}^+(nl) + e \]  

(1)

where the ion is left, not why in the ground state, but also in excited levels nl which are accessible at that given impact energy.

2. Publications


(b) "Cross Sections for Excitation and Ionization in e-He(2^13_s) collision" by Ton-That, Manson and Flannery has been published in J. Phys. B: Atom. Molec. Phys. 10 (1977) 621-635. Reprints will be sent when available.
Calculation of Electron Impact Cross Sections from Metastable States in Atomic and Molecular Cases

Quarterly Progress Report No 7

Period Covered: April 1, 1977 to June 30, 1977

Contract No. F33615-76-C2033

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(A) During this period, Dr. Ton-That and myself wrote a paper entitled "On Populating \( \ell' \)-States in Intermediate Impact-Energy, Direct-Scattering Atomic \( n\ell \rightarrow n\ell' \) Excitation". This research explored the reason why, for example, was the \( 2^1,3_s - 3^1,3_d \) transition in helium induced by electron-impact more favorable at intermediate energies than the dipole \( 2^1,3_s - 3^1,3_p \). We concluded that this "peculiarity" is quite general, that it appears in e - H(n\ell) collisions and in collisions involving singly excited Rydberg states. A copy of the paper is attached.

(B) Also, Dr. McCann modified the "Multichannel Eikonal Program" by incorporating the highly accurate Burlisch-Stoer technique for the solution of the coupled equations, rather than the Adams-Moulton integrator previously used. This modification permitted a greater improvement in the efficiency and accuracy with which calculations of the excitation of metastable helium by low-energy electrons could be performed. Accordingly, in Tables I and II are presented ten-channel \( \left[ (1^1s) 2^1,3_s, 2^1,3_p, 3^1,3_s, 3^1,3_p, 3^1,3_d \right] \) eikonal cross sections for

\[
e + \text{He}(2^1,3_s) \rightarrow e + \text{He}(n^1,3_L), n = 1,2,3.
\]

at energies of 15 eV, 20 eV, 21.5 eV and 25 eV. These energies are additional to those previously presented. Each energy point requires 1 hr Cyber 74 (CDC 6600) computer time.

Dr. R. Flannery is presenting a paper entitled "The Ionization of Metastable Rare Gas Atoms (He*, Ne*, Ar*, Kr* and Xe*) by Electron and by Photon Impact" by Flannery, McCann and Ton-That to X International Conference on the Physics of Electronic and Atomic Collisions, Paris, 20-27 July. An abstract of the paper is enclosed.
Table 1: Cross Sections (10^{-16} \text{ cm}^2) for the 2'S-n'L Transitions in Helium by Collisions with Electrons of Energy $E(\text{eV})$

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The number in parenthesis indicates the power of 10 by which the entry is to be multiplied.
Table 2: Cross Sections ($10^{-16} \text{cm}^2$) for the $2^3S$-$n^3L$ Transitions in Helium by Collisions with Electrons of Energy $E$(eV)

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The number in parenthesis indicates the power of 10 by which the entry is to be multiplied.
THE IONIZATION OF METASTABLE RARE GAS ATOMS (He*, Ne*, Ar*, Kr* AND Xe*)
BY ELECTRON AND BY PHOTON IMPACT

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Little is known about the cross sections for ionization of metastable rare-gas atoms (He*, Ar*, Kr*, Xe*) by electron and by photon impact. This information is important in the modelling and feasibility studies of certain excimer lasers. In particular, very recent observations of high-power laser emission from electron-beam excitation of the noble gas/halide mixtures at high pressures have demonstrated the potential of a new class of high-power, high efficiency and partially tunable lasers operating in the 2000 Å-3000 Å range of wavelength. The search for possible optical windows in the photoionization of metastables in this wavelength range is therefore very important.

The cross sections for the ionization of the metastables by electron and photon impact will be presented and a number of important features will be discussed.

On populating $l'$-states in intermediate-impact-energy, direct-scattering atomic $n\ell \rightarrow n'l'$ excitation

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and

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It is observed that the magnitude of the largest peak with respect to momentum transfer of the inelastic hydrogenic form factor for an $n\ell \rightarrow n'\ell'$ transition overall rises as the value of the orbital angular momentum $l'$ of the final state increases, until for $l' > l'_{\text{max}}$ it drops sharply. The value $l'_{\text{max}}$ is insensitive to the values of $n'$ ($> n$) and $\ell$, but depends mainly on $n$. This trend in the form factor also appears in available ($n \leq 3$) calculated and measured cross sections for electron-impact excitation of H and He at intermediate impact energies near the cross section peak. It generally differs from that suggested by the Bethe dipole-favoring high energy limit.


†Present address
On populating \( l' \)-states in intermediate-impact-energy, direct-scattering atomic \( n \rightarrow n' l' \) excitation

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It is observed that the magnitude of the largest peak with respect to momentum transfer of the inelastic hydrogenic form factor for an \( n \rightarrow n' l' \) transition overall rises as the value of the orbital angular momentum \( l' \) of the final state increases, until for \( l' > l'_{\text{max}} \) it drops sharply. The value \( l'_{\text{max}} \) is insensitive to the values of \( n' (> n) \) and \( l' \), but depends mainly on \( n \). This trend in the form factor also appears in available \( n \leq 3 \) calculated and measured cross sections for electron-impact excitation of H and He at intermediate impact energies near the cross section peak. It generally differs from that suggested by the Bethe dipole-favoring high energy limit.


†Present address
I. INTRODUCTION

Recent progress in the study of excited, Rydberg states of atoms allows an investigation of collisional changes of the orbital angular momentum $l$ of the outer electron. Collisional excitation to states with high values of $l$ is an important mechanism because these states have relatively long radiative lifetimes. A useful guideline in high energy inelastic electron-atom collisions is provided by the high collision energy $E$, asymptotic behavior of the Born cross section, the leading term of which is proportional to the modulus squared of the dipole matrix element of the transition. This Bethe asymptotic cross section goes as $E^{-1} \ln E$ and favors larger cross sections for dipole-allowed transitions than for dipole-forbidden transitions, the cross sections for which decrease as $E^{-1}$.

Calculations for electron impact excitation of atoms initially in excited states exhibit a departure from the above trend near the peaks of the cross sections, where the Bethe limit is not yet predominant. We now present some observations on the behavior of the hydrogenic form factor for an $nlm-n'l'$ transition as the final $l'$ varies. These observations suggest a detailed and systematic pattern for this departure.
II. OBSERVATIONS

A. Form factor for excitation

A study$^5$ of the convergence rate of the ejected-electron partial-wave expansion of the form factor for electron-impact ionization suggested that the form factor for excitation might also exhibit interesting regularities near its peaks. Indeed such a regularity exists. The present calculations use the general expression for the form factor $\langle n', \ell', m' | e^{i\mathbf{K}\cdot\mathbf{r}} | n, \ell, m \rangle - \mathbf{r}$ is the electron position— for an $n\ell\rightarrow n'n'\ell'$ hydrogenic transition derived by Flannery$^6$; this expression is somewhat simpler than other available expressions.$^7$

By searching for the largest peak, with respect to the momentum transfer $\mathbf{K}$, of the form factor (modulus squared, summed over $m'$ and averaged over $m$, where $n, m, n'$ are the magnetic quantum numbers) for a given $n\ell\rightarrow n'n'\ell'$ transition, one finds that the magnitude $F$ of this peak goes up and down as $\ell'$ increases, but that on the average it slowly rises before exhibiting a final precipitate decrease after $\ell' = \ell'_{\text{max}}$. This is shown for various values of $n, \ell, n'$ in Fig. 1-3.

The dependence of $\ell'_{\text{max}}$ itself on $n, \ell, n'$ is also interesting. For fixed $n$ and $\ell$, the value $\ell'_{\text{max}}$ is quite insensitive to that of $n'(\neq n)$ within a small margin $\Delta \ell'$ substantially smaller than $\ell'_{\text{max}}$ (see Fig. 1, 2, 4). Moreover, for fixed $n$ and $n'$, the value $\ell'_{\text{max}}$ is also insensitive to that of $\ell$ (see Fig. 2, 3) within a small margin, with a smaller $\ell'_{\text{max}}$ corresponding to a higher $\ell$. For large $n$ a unique average value $\ell'_{\text{max}}$ can be obtained by averaging over $\ell$ before varying $\mathbf{K}$ and $\ell'$ (see Fig. 4, 5); this average removes most of the up and down behavior.
and makes the rise and fall about $l^{'}_{\text{max}}$ most evident (see Fig. 5).

Thus $l^{'}_{\text{max}}$ is approximately independent of $n'$ and $t$ and dependent on $n$ only. A plot of $l^{'}_{\text{max}}$ against $n$ (see Fig. 6) suggests an almost linear dependence. Also shown in Fig. 6 is the asymptotic relation

$$l^{'}_{\text{max}} \sim n^\frac{1}{2}$$  \hspace{1cm} (1)

derived for large $n,n'$ by Ton-That using a model which also provides a simple interpretation of the insensitivity of $l^{'}_{\text{max}}$ to the values of $n'$ and $t$. The overall rise in the magnitude of the form factor before $l^{'}_{\text{max}}$ can be regarded as arising from the $(2l'^{'+1})$ statistical factor of the final state $n'^n$. Although asymptotic, relation (1) unexpectedly maintains its agreement with the computed $l^{'}_{\text{max}}$ even for small $n$. The computed $l^{'}_{\text{max}}$ is not shown for $n>10$ due to algorithm breakdown which makes it unreliable.

B. Excitation cross sections

The observed trend in the form factor acquires further relevance if it is reflected in the cross sections. Moreover it suggests a specific method of comparison, namely varying $l^'$ whilst keeping $n,l,n'$ fixed, then varying $n'$ or $t$ and finally repeating the process for various $n$. Cross section calculations which consider a variety of $l^',l$ and $n'$ are at present available only for low $n$. For $n=1,2,3$ the form-factor $l^{'}_{\text{max}}$ is 1,2 and 4 (p,d and g levels) respectively.

Calculated electron-impact cross sections of H(1s), He(1s)$^2$ on one hand and of H(2s), He(1s)(2s) $^{2,3}s$ on the other are, near their
maxima, largest for excitation to the $n'p$ and $n'd$ levels respectively (see Fig. 1, 7), although the $s-p$ dipole-allowed transitions prevail at higher energies. Figures 1 and 7 show that the drop after a certain $l'$ in the cross sections, at a given impact energy (near their maxima), is similar to that in $F$, at a value of $K$ adjusted to produce a maximum. For $n=3$, $n'=4$, calculations of McCoyd et al. 10 show that e-H excitation cross sections, near their peaks, overall increase with $l'$ up to the $f$ levels whatever $l$; $l'_{\text{max}}=g$ and the sharp drop after $l'_{\text{max}}$ cannot be verified in this case as consideration of higher $l'$ would be required.

Measured 11 cross sections support $l'_{\text{max}}=1$ ($p$ level) for electron-impact $1s+n'$ excitation of $H(1s)$ and $He(1s)^2 1S$—for $He$, in direct (as opposed to exchange) scattering—. Similarly, the data of Lake and Garscadden 12 indicate that at intermediate energies (20eV) the cross section of $He(1s)(2s) 2 3S$ is larger for excitation to the $3 3D$ level than to the $3 3S,P$ levels, in agreement with $l'_{\text{max}}=2$ for $n=2$.

Thus the trend in the form factor can appear in electron-impact excitation cross sections especially near the maxima, at impact energies below the region of applicability of the Bethe limit. At such energies the lower $K$ limit of the Born cross section integral expressed in terms of the form factor remains sufficiently removed from the optical limit $K=0$, such that the magnitude of the form factor does influence that of the Born cross section. Although the Born approximation is often poor near the excitation cross section peaks, a sharp drop is likely to persist in other direct-scattering approximations whilst the initial rise
due to the \((2t'+1)\) statistical weight is essentially approximation-independent. Note that according to (1), the sharp drop itself will not be observed unless \(n' > n/2 + \frac{1}{2}\). For \(n \leq n' \leq n/2 - \frac{1}{2}\), only the overall initial rise with \(t'\) is noticeable.
This trend in the form factor, and presumably in certain collision-energy ranges of the electron-impact cross sections as well, agrees with previous observations\(^3,5,13\) that dipole-forbidden transition cross sections become non-negligible and even important in relation to their dipole-allowed counterparts favored by the Bethe limit. It also provides details about the manner of this departure, emphasizing the importance and yet limitations of the role of statistical factors and expressing the dependence of these limitations on \(n, \ell, n'\). Such information is relevant to modelling studies of highly excited \(\text{(Rydberg)}\) atomic \(\ell\) states in astrophysics, plasma physics, gas dynamics and collision problems in general. It also suggests that the large cross sections, at low collision-energies, for electron impact excitation of dipole-allowed transitions from \(\text{H}(1s)\) and \(\text{He}(1s)\)\(^2\) are not to be ascribed to some optical-like selection mechanism\(^{14}\) but rather to a particular case of a more general\(^{15}\) trend. A model to interpret this trend will be presented in a subsequent report.\(^8\) The ability to select and detect a particular \(n\) \(\text{Rydberg}\) state should permit an experimental test of the present remarks.

**Acknowledgement**

One of us (D. T. T.) is grateful to Professor Bernd Crasemann for his generous support.
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This trend for e - He \((2^1,3S \rightarrow 3^1,3S,P,D)\) was interpreted in Ref. 3(a) as arising from the 2s - 3p line strength weakness in He compared to H, i.e. a peculiarity of He \(2^1,3S\). It is now clear, however, that the trend also applies to H \(2s\) and is not peculiar to He \(2^1,3S\).
Figure Captions

Figure 1: Magnitude $F$ of the largest peak, with respect to momentum transfer $K$, of the $nl \rightarrow n'l'$ hydrogenic form factor (modulus squared, summed over $m'$ and averaged over $'m$) plotted against $t'$, for $n = 1$, $t = 0$ and several values of $n'$. In this figure only, the dashed lines correspond to the Born electron-impact excitation cross section $^4\sigma$ of H for $n' = 2-4$ and collision energies of 51.02, 43.05 and 45.41 eV respectively.

Figure 2: $n = 2$, $t = 0,1$ and $n = 4$, $t = 0-3$ (see Fig. 1).

Figure 3: $n = 8$, $t = 0-7$, $n' = 13$ (see Fig. 1).

Figure 4: As in Fig. 1, but the form factor is averaged over $t$ as well, before varying $K$ and $t'$. $n = 3$, $n' = 4-12$.

Figure 5: $n = 6 \rightarrow n' = 11$, $n = 8 \rightarrow n' = 14$ (see Fig. 4).

Figure 6: $t_{\text{max}}$ (see text) plotted against $n$. The straight line is the theory $^8$ for large $n,n'$ (cf. Eq. (1)). The full circles are from computation (including an average over $t$, cf. Fig. 4, 5). The open circles denote sample values of $t'$ for the peak second in magnitude to $F$.

Figure 7: Electron impact cross section $^5$ of He (1s)(2s) $2^3S$ for excitation to the He (1s)(n$'l'$) $^3L(= l')$ states, plotted against $t'$, for impact energies near the cross section peak (50 eV) and well above (1 keV).
Figure 2
$n = 8 \rightarrow n' = 13$

Figure 3
f.f. averaged over $l$
$n = 3$
Figure 5

f.f. averaged over \( l \)

\( n = 6 \rightarrow n' = 11 \)
\( n = 8 \rightarrow n' = 14 \)
Figure 6

Theory for large $n, n'$

Computation
Figure 7
Calculation of Electron Impact Cross Sections from Metastable States in Atomic and Molecular Gases

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Contract No. F33615-76-2033, Item No. 001
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1. Work in Progress

(a) We are investigating the resonance transition \((n+1)s \rightarrow (n+1)p\) arising in electron impact excitation of rare gas atoms initially in their \(3P_{0,2}\) metastable states. The theoretical methods under consideration are the Born approximation and the multichannel eikonal treatment.

(b) We are also investigating certain peculiarities that might be observed in \(n\mathcal{L} - n'\mathcal{L}'\) collisional transitions in general. Attached is an abstract and some discussion on certain systematic behavior of the form factor

\[
F_{fI}(\mathbf{K}) = \langle \psi_f(\mathbf{r}) | e^{i\mathbf{K} \cdot \mathbf{r}} | \psi_i(\mathbf{r}) \rangle
\]

(1)

a matrix element which is essentially the "driver" in most theoretical treatments of the collision process. We find that behavior exhibited by (1) is also reflected in the cross sections for \(n\mathcal{L} - n'\mathcal{L}'\) collisional transitions. This behavior will assume significance in laser modeling.

In Figs. 1-10, we have plotted the maxima of the form factor squared versus \(\mathcal{L}'\), the final angular momentum state in \(n\mathcal{L} - n'\mathcal{L}'\) transitions for \((n = 4, \mathcal{L} = 0-3)\) and \([n' = 5,6,8,10,20; \mathcal{L}' = 0-(n'-1)]\). The key feature here is that dipole transitions do not in general dominate, and that \(|F|^2\) shows a well defined maximum as a function of \(\mathcal{L}'\) (e.g., see figs. 7-10) and after this maximum, a precipitous decrease is observed.

In Figs. 11-20, we have plotted the corresponding excitation cross section peaks for \(n\mathcal{L} - n'\mathcal{L}'\) transitions induced by electron impact. These results indicate that the behavior pattern in Figs. 1-10 are reflected in the cross sections, e.g., for all transitions out of the \(n = 4\) level the state with final angular momentum \(\mathcal{L}' = 5\) (if accessible) is populated the most.

A paper on this subject is at present being written up.
2. **Papers presented at conferences**

Two invited papers were presented at two workshops on (a) Kinetic Processes in Rare Gas-Halide Lasers and (b) electron-excited state collisions at 30th Annual Gaseous Electronics Conference, Palo Alto, California, 18-21 October 1977.
Figure 1. Maxima in the Form-Factor-Squared as a function of $\ell'$ for $n\ell-n'\ell'$ transitions.
Figure 2. Results of Figure 1 averaged over the initial angular momentum states $l$. 

$n=4 \rightarrow n'=5$
Figure 3. As in Figure 1.
Figure 4. As in Figure 2.

\[ |F_{\text{max}}|^2 \]

\[ 10^{-1} \]

\[ 10^{-2} \]

\[ 10^{-3} \]

\[ n=4 \rightarrow n'=6 \]

$\ell'$
Figure 5. As in Figure 1.
Figure 6. As in Figure 2.
Figure 7. As in Figure 1.
Figure 8. As in Figure 2.
Figure 9. As in Figure 1.
Figure 10. As in Figure 2.
Figure 11. The cross section peaks for ni-n'\ell' electron-impact excitations as a function of final-state angular momentum \ell'.
Figure 12. Results of Figure 11 averaged over the initial angular momentum state $\ell$. 

\[ e + H(4) \rightarrow e + H(5 \ell') \]
Figure 13. As in Figure 11.
Figure 14. As in Figure 12.
Figure 15. As in Figure 11.
Figure 16. As in Figure 12.
Figure 17. As in Figure 11.
Figure 18. As in Figure 12.

\[ e + H(4) \rightarrow e + H(10 \ell') \]
Figure 19. As in Figure 11.
Figure 20. As in Figure 12.
The population of angular momentum states $l'$ in $n^2-n'^2$ direct collisional excitations

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Abstract

The unexpected result that the $2^1S-n^1D, e^1D$ transitions dominate the cross sections both for the excitation of the $n = 3-5$ levels and for ionization to continuum energy $e$ in e-He($2^1S$) collisions at intermediate impact-energies is examined and is found to originate from general characteristic trends exhibited by the inelastic form factor. It is found that the magnitude of the largest peak (with respect to momentum transfer) of the form factor for $n^2-n'^2$ transitions oscillates on a background which rises as the angular momentum $l'$ of the final state is increased until it drops sharply for a unique value $l'_{max}$ which is strongly dependent only on the initial level $n$ and which is fairly insensitive to both $l$ and $n'$. This behavior is directly reflected in the available theoretical and measured cross sections for e-H(n$l$) and
e-He\(^{2,3\text{S}}\) inelastic collisions at intermediate energies. It leads to prediction of the most probable value of \(l'\) that is likely to dominate final-level population in \((n\pi-n'\pi')\) collisional excitation both by electrons and by heavy-particles at low and intermediate impact-energies. The prediction in general differs from that suggested by the Bethe asymptotic limit which favors dipole transitions, and assumes significance in situations where excited states are important as in laser modeling and in astrophysical plasmas.

PACS CLASSIFICATION NUMBERS:
I. INTRODUCTION

It is becoming increasingly apparent in collisions involving an atom initially excited that a certain useful guideline developed with ground-state atoms in mind is not of general validity. Contrary to expectation, both the Born and the more elaborate ten-channel eikonal treatments of \( e-\text{He}(2^{1,3}S) \) collisions, for example, predict that transitions to the optically forbidden \( 3^{1,3}D \) and \( 3^{1,3}S \) states are much stronger, by up to an order of magnitude, than the dipole-allowed \( 2^{1,3}S-3^{1,3}P \) collisional excitations, except, of course, at high impact-energies \( E \) when the cross sections for dipole transitions, with their characteristic \( E^{-1} \) and \( E \) slower-varying asymptotic-dependence, eventually dominate. Also the \( 2^{1,3}S-3^{1,3}D \) transition is the major contributor from \( 2^{1,3}S \) to the population of the \( n=3 \) level of He. These findings were verified by the recent measurements of Lake and Garscadden. The obvious analogy with ground-state species and with optical properties of the atom would suggest (erroneously) that the \( 2^{1,3}S-3^{1,3}P \) transition in \( e-\text{He}(2^{1,3}S) \) collisions would dominate the population of the \( n=3 \) level at all impact-energies. (It is however worth noting that the \( (2^{1,3}S-2^{1,3}P) \) cross section is much larger than those for excitation out of the \( n=2 \) level.)

Also a study of excitation to the \( n=4 \) and \( 5 \) levels in \( e-\text{He}(2^{1,3}S) \) collisions revealed that transitions to the \( 1^{1,3}D \) states are again predominant for a given \( n \), except at the highest energies when the S-P dipole excitations gain in relative importance in accord with Bethe's approximation. Moreover, this trend continues for higher \( n \), even for the bound-free transitions in He where we have found, for low energies \( \epsilon \) of electron-ejection, that the ionization cross section is again dominated by the contribution arising from the ed partial-wave describing the
In an effort to probe the origin of this behavior and to establish certain general patterns observed in variation of the initial excited state, the $n_2-n'_2$ transition in hydrogenic systems will be studied. In so doing, we will attempt to examine, for a fixed value of $n'$, the variation of that value of $l'$ which yields the largest cross section with change of $n$ and of $l$, the quantum numbers for the initial state.

As well as being of fundamental significance in themselves, the findings may have rather important and far-reaching implications in situations where excited states are of particular significance, as in, for example, laser modeling, gaseous discharges, recombination and astrophysical plasmas. In H I and H II regions of hot stars, the interpretation of the observed emission between highly excited Rydberg states $n=110\rightarrow n'=109$, for example, is based on evaluation of the coefficients describing departure from thermodynamic equilibrium. These coefficients are functions of cross sections for collisional transitions between excited states. In both this case and in laser-modeling studies, it has been tacitly assumed that the dipole transitions $l'=l\pm1$ dominate at all energies and, in the absence of any further information, the cross sections are determined by the Bethe limit, or some modification thereof.

In this paper we shall show that this assumption is rather questionable at intermediate impact energies, and that the result observed for $e-\text{He}(2^1\text{S})$ collisions is but a particular case of a more general trend.
II. THE INELASTIC FORM FACTOR IN COLLISIONAL TRANSITIONS

The Born approximation to the transition matrix element $T_{fi}$ for inelastic scattering of a structureless particle by a target atom with composite electronic coordinates denoted by $\xi$ and with electronic wave function $\psi_n(\xi)$ is

$$T^B_{fi}(k_i, k_f) = \langle \psi_f(\xi)e^{ik_i \cdot R} | V(\xi, R) | \psi_i(\xi) e^{ik_f \cdot R} \rangle_{\xi, R},$$

where $k_i$ and $k_f$ are the initial and final wave vectors of relative motion of the collision partners moving under their mutual electrostatic interaction $V(\xi, R)$ at nuclear separation $R$. This matrix element (1) is usually written as,

$$T^B_{fi} = \mathcal{F} \{ V_{fi}(R) e^{iK \cdot R} \} \equiv \mathcal{F} \{ V_{fi}(R) \} , \quad K = k_i - k_f ,$$

the Fourier transform of the interaction

$$V_{fi}(R) = \langle \psi_f(\xi) | V(\xi, R) | \psi_i(\xi) \rangle$$

coupling the initial and final states of the target. However, on changing the integration-variables in (1) from $(\xi, R)$ to $(\xi, \xi_{12} = \xi - R)$, the transition-matrix element simply reduces to,

$$T^B_{fi}(K) = \mathcal{F} \{ \xi_{12} \} \left[ \mathcal{F} \{ V(r_{12}) e^{iK \cdot \xi_{12}} \} \right]$$

$$= \mathcal{F} \{ \xi_{12} \} T^B_{\text{el}}(K),$$

(4)

a product of the $T$-matrix element for elastic scattering of the projectile by a fixed potential $V(r_{12})$, provided by the interaction of the structureless projectile with the "frozen" target-electrons, and

$$F_{fi}(K) = \langle \psi_f(\xi) | e^{iK \cdot \xi} | \psi_i(\xi) \rangle,$$

(5)
the inelastic form-factor for the \( i \rightarrow f \) transition in the target. Expression (4) yields valuable insight to the collision process in that it separates the (target) property responsible for the actual transition, and characterized by the inelastic form factor, from the background potential scattering. This separation of properties is not readily apparent from the usual expression (2).

Moreover, the form of (4) suggests that an improvement\(^6,7\) to Born's approximation is given by,

\[
T_{fi}(k) = F_{fi}(k)T_{el}(k)
\]

(6)

where \( T_{el} \) is now the actual (or most accurate available) \( T \)-matrix (off-the-energy-shell!) for potential scattering of the projectile by the static electrons of the target. This feature has been exploited by Flannery\(^8\), in various treatments of collisions involving highly excited states. The relative control of the former property over the latter in the collision process is determined by the sensitivity of \( T_{el}^{B} \) to those momentum-changes \( K \) important to the collision. For a Coulombic interaction \( e^{2}/r_{12} \) (as in e-atom collisions\(^7\)), the potential scattering term \( T_{el}^{B} \) in (4) is simply \( 4\pi e^{2}/K^{2} \), a quantity which exerts a dramatic influence on (4) only in the limit of low momentum-change \( K \). Therefore, for most intermediate \( K \), any unusual variation of the transition matrix \( T_{fi}^{B} \) with excited state \( (n'k') \) will originate largely from the form-factor \( F_{fi} \). This inelastic form factor not only is the driver-mechanism for collisional transitions in Born's approximation, but is also a very basic factor in more elaborate collisional treatments, such as the quantal close-coupling and multichannel eikonal methods. In fact, Flannery\(^9,10\) has already shown how the whole array of interaction matrix elements \( V_{ij}(k) \) which appear in close-coupling formulations may be derived from detailed knowledge of the inelastic form-factor.
Also, a detailed study\textsuperscript{4,11} of the convergence rate of the $l'$ partial-wave expansion for the ejected electron in the associated bound-free form-factor for electron-impact ionization revealed certain regularities (given in ref. 4) which were fully preserved in the corresponding partial $l'$-ionization cross sections. For example, the peaks of the form-factor as a function of momentum-change $K$ exhibited interesting regularities with $l'$, regularities which were reflected in the corresponding cross sections. This suggests that the bound-bound form factor might also exhibit similar displays, and if so, may well have important implication for $n_l-n'l'$ collisional transitions, in general.
III. SYSTEMATIC TRENDS

In an effort to further explore the findings\textsuperscript{1-4} outlined in the introduction for e-He(\textsuperscript{1,3}S) collisions and to predict certain peculiarities not previously observed for transitions between excited states, we shall examine in detail the form factor for \textit{n\ell-n'\ell'} transitions in hydrogen by using the general expression derived previously by Flannery.\textsuperscript{12} This expression is in closed form and is somewhat simpler to use than other available formulae.\textsuperscript{13,14}

By a computer analysis, we search for the largest peak, with respect to the momentum-transfer \(K\), of the form factor (modulus squared, summed over \(m'\) and averaged over \(m\) where \(m\) and \(m'\) are the azimuthal quantum numbers).

Figure 1 displays the variation of this peak \(F\) for the \(1s-n'(=2-12), 2'(=0-n'-1)\) transitions as a function of \(\ell'\), together with the associated excitation cross sections at the impact-energies indicated. Indeed, the \(1s-n'p\) transition dominates \(F\) for a given \(n'\) and this behavior is also reproduced by the Born cross sections.

The analogous Fig. 2, however, clearly indicates that transitions out of the 2s and the 2p states to the \(n' = 4, 7\) and 10 levels are dominated by population of the d-sublevels (\(\ell' = 2\)), while excitation from the \(n = 4\) sublevels (\(\ell = 0-3\)) to the \(n' = 10\) level are controlled by the \(4\ell-10(\ell' = 5)\) transitions. The dominance of the \((2s-n'd)\) transition for a given \(n'\) is in keeping with that already observed for e-He(\textsuperscript{1,3}S) excitational collisions. The dominance of the 2p-nd transition is also predicted not because of its dipole nature, as we shall see later. An important deduction from Fig. 2 is that the inelastic form factor is much more sensitive to variation in the initial value of the principal quantum
number $n$ (which determines the physical size of the atom) than it is to variation of the initial angular quantum number $\ell'$.  

This inference becomes more apparent in Fig. 3 where the $8\ell-13\ell'$ transitions are shown; the choice of transitions being rather arbitrary. Here, $F$ in general oscillates as $\ell'$ is increased but, on the average, it slowly rises to a relatively-large peak value at $\ell' = \ell'_{\text{max}} \approx 11$ exhibiting a final precipitous decrease. This behavior is identical to that already observed in the form factor for $e$-$He(2^1S)$ ionization, a behavior preserved in partial $\ell'$-wave contributions to the ionization cross section. Figure 3 clearly shows that the value of $\ell'_{\text{max}}$ is mainly determined by initial principal quantum number $n$ rather than by $\ell$. As $\ell$ increases, however, there is a slight tendency for $\ell'_{\text{max}}$ to shift to somewhat lower $\ell'$-values.

By averaging over $\ell$ before varying $K$ and $\ell'$, the oscillations can be suppressed and the key issue is then transparent. For example, Fig. 4 illustrates the results for $n = 3 + n' \ell'$ transitions and the rise and rapid fall about $\ell'_{\text{max}} \approx 4$ becomes most evident. Also, in situations where that value of $\ell'_{\text{max}}$ is excluded, as in the $n' = 4$ case, $F$ steadily increases with variation of $\ell'$ from 0 to $(n'-1)$. Thus, from Fig. 4, we predict that population of the $g$ state ($\ell' = 4$) will dominate collisional excitation out of the $n = 3$ levels to a given $n'$ level, except, of course, in cases when the $g$ state is excluded.

Figure 5 further clarifies the situation by showing the slow rise to $\ell'_{\text{max}}$ followed by the precipitous decline. Here we predict that, in collisional transitions out of the $n = 6$ level at intermediate energies, the population of the $n' = 11$ level is mainly determined by transitions to that state with angular momentum 8. Also excitation between the $n = 8$ and
14 levels are controlled by the $8\ell + 14(\ell' = 11)$ array of transitions.

The origin of the behavior, displayed in Figs. 1-5, lies of course in the variation of the overlap between the initial ($\ell_1 t$) and final ($\ell_2 t'$) wave functions, modulated by the phase $\exp(iK \cdot r)$, as $\ell'$ is increased. The general results we have observed here were obtained from a computer analysis of the form-factor. While a rigorous mathematical analysis of the properties of the form factor for transitions between excited-states in general is at present extremely difficult, some physical insight as to the existence of a value $\ell'_{\text{max}}$ is possible.

The region where the initial bound state orbital is substantial defines, for our purposes, the maximal region of overlap. The radial orbital for the final state ($\ell_2 t'$) evolves in the limit of high $n'$ to the Bessel function $J_{2\ell' + 1} (\sqrt{8r})$ which, in general, oscillates many times within the overlap region. The Bessel function increases from the origin as $(\sqrt{8r})^{\ell'}$. As $\ell'$ increases, the oscillating pattern tends to shift out of the overlap region together with a stretching of the first lobe. When this lobe embraces the maximal region of overlap, the overlap exhibits a maximum for this particular value of $\ell' = \ell'_{\text{max}}$. As $\ell'$ increases, the first lobe shifts out of the overlap region and the overlap decreases thereafter. For $\ell' < \ell'_{\text{max}}$, the oscillations of the Bessel function with $\ell'$ therefore yields the oscillations observed in Fig. 3. The $(2\ell' + 1)$ statistical weight factor of the final state ($\ell_2 t'$) favors an initial rise of $F$ with respect to $\ell'$ eventually subdued by the stretching of the first lobe. One of the effects of the modulating factor $\exp(iK \cdot r)$, which can be expanded in terms of spherical Bessel functions $j_{\ell'}(Kr)$ to effectively reduce the region of overlap (because of the varying relative phase of the Bessel functions) and therefore the magnitude of form factor. The important
effect, however, is to introduce the possibility of a resonance between the Bessel functions at a specific value of K. The effective region of overlap is now the maximal region and a peak in the form factor results. As $t'$ increases the first lobe of the resonance pattern approaches the maximal region of overlap, embraces it and then shifts out, thereby yielding a value $l'_{\text{max}}$ at which the form-factor peaks.

A more difficult problem, however, is to find the general variation of $l'_{\text{max}}$ with the initial principal quantum number $n$. Each of us has attempted to make such a deduction and we conclude, in the special limit of high $n$ and $n'$, the conclusion is that

$$l'_{\text{max}} \sim n\sqrt{2} - \frac{1}{2}. \quad (6)$$

Figure 6 compares this prediction with the computed results. The agreement is extremely close, even for small values of $n$. The computed $l'_{\text{max}}$ is not shown for $n > 10$ due to algorithm breakdown. Thus, the general trends (oscillations, $l'_{\text{max}}$, and rapid fall-off) in Figs. 1-5 can in general be understood. Note, however, according to (6), the sharp drop itself will not be observed unless $n' > n\sqrt{2} + \frac{1}{2}$. For $n \leq n' \leq n\sqrt{2} + \frac{1}{2}$ only the overall initial rise with $l'$ is noticed.
IV. EXCITATION CROSS SECTIONS

The prediction here that the peaks in the inelastic form factors for \( n\rightarrow n'\ell' \) transitions occur at \( \ell' = \ell'_{\text{max}} \), a value which depends rather strongly on the initial principal quantum number \( n \) and which is rather insensitive to the initial angular-momentum quantum number \( \ell \), acquires further significance if it is reflected in the corresponding collisional cross sections. The pattern has already been exhibited\(^1-4\) in e-He(\(2^1,3^S\)) collisions involving both excitation to the \( n=3, 4 \) and 5 levels of He and ionization. Not only does this pattern emerge in Born’s approximation but it has already been established in the more elaborate multichannel eikonal treatment\(^2\) and has also emerged in recent experiments\(^3\).

Figure 7 displays some of our results\(^4\) and shows that at intermediate energies the \( 2^1,3^S - n^1,3^D \) transitions dominate (as verified also by experiment\(^3\)) while the S-P dipole-allowed transitions prevail at higher energies. The reasons for this are rather instructive. At these intermediate impact-energies, the lower limit \( (k_i-k_f) \) of the cross-section integral,

\[
\sigma_{if} = \frac{1}{8\pi} \left( \frac{2m}{\hbar^2 k_f} \right)^2 \int \frac{\left| F_{fi}(K) \right|^2 \left| T_{el}(K) \right|^2 KdK}{(k_i-k_f)} , \quad (7)
\]

obtained from (4), or (6), remains sufficiently removed from the optical limit \( K=0 \) such that the magnitude of the form-factor directly influences the cross section \( \sigma_{if} \). For e-atom collisions, the Born "potential-scattering" term \( T_{el}^B \) in (4) is simply \( 4\pi e^2/K^2 \) which only causes a smooth background reduction to the form factor at those intermediate momentum changes \( K \gtrsim 1 \) a.u., of importance to the collision process.

At much higher impact-speeds \( v \), the lower limit \( k_i-k_f \) tends to
(\epsilon_{fi} \equiv \text{transition-energy})/v, such that the potential scattering term T_{e\ell}^B in (4) will considerably amplify the dipole contribution to F_{fi} which can be expanded (for small K) in terms of the various multipoles. At high impact-energies E, the Born cross section approaches the usual Bethe-expression,

$$
\sigma_{if}(E) = \frac{8\pi}{k_i a_0} \frac{2v}{\epsilon_{fi}/v} \left[ \frac{<\psi_f | z | \psi_i >^2}{K} + \frac{K}{4} |<\psi_f | z^2 | \psi_i >|^2 + \ldots \right] dK. \quad (7)
$$

The first term in the RHS of (7) is the dipole term, is the dominant contributor to the integral for the small K, appropriate to high impact-energies E, and yields the customary $E^{-1} \ln E$ dependence in $\sigma_{if}$ for dipole transitions. The remaining terms in (7) are important only for optically forbidden transitions at high impact energies E and decrease rather rapidly with E.

**e-excited atom collisions.** Since these investigations originated with an effort to explore the origin of results 1-4 for e-He(2^1 \, 3^S) collisions and eventually culminated in prediction, we subsequently searched the literature to seek further substantiation for our findings. Indeed, Tables I-III in the review article of Moiseiwitsch and Smith 19 provides partial evidence, although not actually highlighted. These tables (after Vainshtein 20 show, for e-H(n\ell) collisions, that the 2s-n'd (n' = 3-9) transitions dominate population of a given level n' at low and intermediate impact-energies, analogous to our findings for e-He(2^1 \, 3^S) collisions. The tables also show that the cross sections for the 3s-n'\ell' (\ell' = 0,1,2) transitions increase as \ell' is raised. However, the 3s-n'f transition, the strongest, according to our predictions, is not given.

In the (n = 3-n' = 4) work of McCoyd et al. 21 this prediction is
indeed substantiated. Their Figs. 1-4 exhibit quite clearly the dominance of the 3\ell-4f transition in populating the n'=4 level from n=3. These results are also in keeping with our prediction that the value \ell'_{\text{max}} is sensitive only on n and not on \ell. However, these trends were not apparently noticed, as evidenced by an accompanying paper\textsuperscript{22} which was confined only to optically-allowed transitions between the n'=4 and n'=5 levels. We have predicted from Fig. 6 that the 4\ell-n'g (\ell'=5) array of transitions control the population of levels n'. Dipole transitions (\ell = \ell' \pm 1) are also covered by this prediction and the two figures of Fisher et al.\textsuperscript{22} show that 4f-5g and 5f-6g are indeed dominant. But, unfortunately, Fisher et al.\textsuperscript{22} have not included the 4s-ng, 4p-ng, and the 4d-ng transition which, according to our predictions, are also substantial contributors to excitations out of the n=4 level.

\textbf{Atom-excited atom collisions.} Another attractive feature in writing the Born T-matrix as (4), or as the presumably more accurate T-matrix (6), arises in heavy-particle collisions. We note that the essential factor which causes the cross sections to approach dipole character at high impact-energies is T\textsuperscript{B}_el', which, for e-atom collisions is 4\pi e^2/K^2 and is singular as K\to0, the optical limit. In the case of atom-atom collisions, however, T\textsuperscript{B}_el for scattering of the projectile by the "frozen" electrons of the target does not contain such a singularity (e.g. for the screened Coulombic interaction e^2/r^1 e^{-\lambda r} between the projectile and target electrons, T\textsuperscript{B}_el is 4\pi e^2/[K^2 + \lambda^2]). The absence of this singularity at K=0 is to allow the form factor in (4) or (6) to exert even more control in the collision process, than in the e-atom case. Therefore, the above predictions will be even more appropriate to atom-excited atom collisions. The overall range of impact speeds will, in effect,
be extended. Moreover, higher impact-speeds must be achieved for the onset of the Bethe dipole-limit.

Also, in the limit of collisions of very slow atoms with highly excited atoms, the scattering, as described by \( T_{el}(K) \) in (6), of the "frozen" electrons of the target by the slow projectile is isotropic with the result that details of the collisional transition as given by (6) is governed entirely by the form factor.
V. SUMMARY AND CONCLUSIONS

In this paper we have predicted new and interesting "peculiarities" involved in collisional excitation between excited states. The study originated with an effort to explain "unexpected" results\textsuperscript{1-4} for e-He(2\textsuperscript{1,3}S) collisions, which were found, as the investigation progressed, to be particular cases of a more general and basic trend. Our conclusions take the form of certain predictions which are substantiated somewhat by the rather limited cross-section data in the literature. For collisions involving one atom in a singly excited-state (nl), the population of level n' (>n) arising from n-n transitions will be mainly controlled at low and intermediate impact-speeds by the nl-n' (l'\textsubscript{max} \sim n\sqrt{2} - \frac{1}{2}) array of transitions, the probabilities for which are fairly insensitive to the initial angular-momentum quantum number \( l \). These predictions also include excitation from the ground-state where, for this case alone, the s-p dipole transitions will dominate. These transitions will in general also dominate in the limit of high impact speeds, in harmony with the Bethe limit.

A related purpose of this paper is therefore to express a note of caution in following the arbitrary assumption in nl-n'l' collisional excitation that the \( l' = l \pm 1 \) transitions are the strongest and are the ones that only need calculation. This assumption, which is without foundation except at high impact-speeds, has tacitly been accepted in modeling studies of the role of excited states in laboratory (laser) and astrophysical plasmas. The dipole transitions n (l'\textsubscript{max} \pm 1) \rightarrow n'l'\textsubscript{max} are of course covered by our prediction and are therefore strong, as are also the nl \rightarrow n'l'\textsubscript{max} array of transitions, in general.
Also, these findings are very relevant to various experiments which investigate collisional changes of the orbital angular momentum of highly excited Rydberg states of atoms.

Finally, it is worth noting that the general trends given here are expected to be reflected also in heavy particle collisions to an extent greater than that observed for electron-excited atom collisions.
REFERENCES

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FIGURE CAPTIONS

Fig. 1. Magnitude $F$ of the largest peak, with respect to momentum transfer $K$, of the $n\ell \rightarrow n'\ell'$ hydrogenic form factor (modulus squared, summed over $m'$ and averaged over $m$) plotted against $\ell'$, for $n = 1$, $\ell = 0$ and several values of $n'$. In this figure alone, the dashed lines correspond to Born cross sections for electron-impact excitation of $H(n' = 2-4)$ by electrons with 51.02, 43.05 and 45.41 eV energy respectively.

Fig. 2. $n = 2$, $\ell = 0, 1$ and $n = 4$, $\ell = 0-3$ (see Fig. 1).

Fig. 3. $n = 8$, $\ell = 0-7$, $n' = 13$ (see Fig. 1).

Fig. 4. As in Fig. 1, but in addition the form factor is averaged over $\ell$, before the variation of $K$ and $\ell'$ is performed. $n = 3$, $n' = 4-12$.

Fig. 5. $n = 6 \rightarrow n' = 11$, $n = 8 \rightarrow n' = 14$ (see Fig. 4).

Fig. 6. $l'_\text{max}$ (see text) plotted against $n$. The straight line is expression (6) valid for large $n$, $n'$. The full circles are from computation (including an average over $\ell$, cf. Figs. 4, 5). The open circles denote sample values of $\ell'$ for the peak second in magnitude to $F$.

Fig. 7. Electron-impact cross section of $\text{He}(1s)(2s)2^3S$ for excitation to the $\text{He}(1s)(n'\ell'3^5L(=\ell'))$ states, plotted against $\ell'$, for impact-energies near the cross section peak (50 eV) and well beyond, at 1 keV.
Figure 2
Figure 3

$n = 8 \rightarrow n' = 13$
Figure 5
Figure 6
Figure 7