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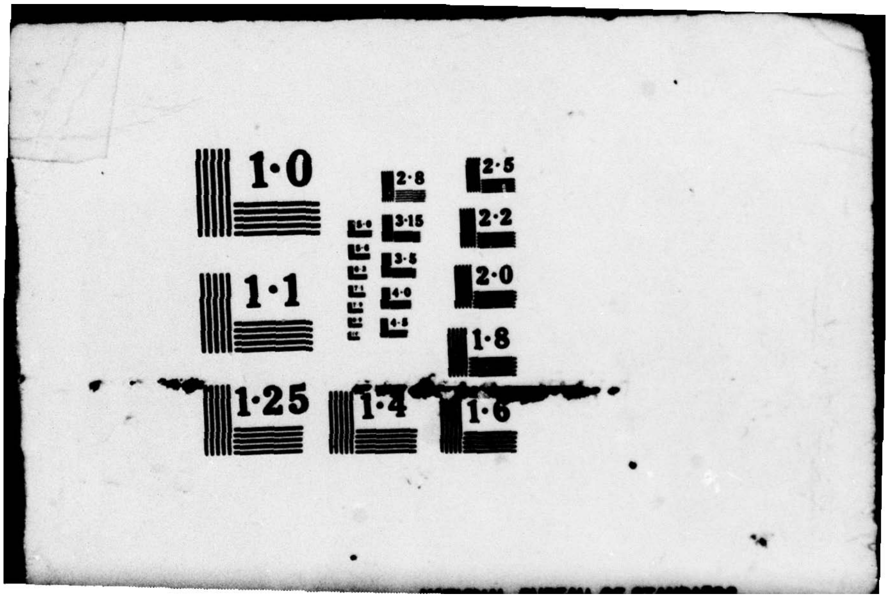
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New methods were used in theoretical studies of the dynamical development of final state populations in atom-molecule reactive collisions. Quantitative calculations of the $H+H_2$ exchange reaction showed that information theory predicts rotational distributions far from equilibrium. A new theory of vibration excitation in electron-molecule scattering was devised. Accurate (1%) cross sections for $e^{(-)}$ He scattering below 19 eV were computed variationally.

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ATOMIC AND MOLECULAR INTERACTION THEORY: FINAL REPORT ON CONTRACT N00014-78-C-0447 (1978-1979)

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I. INTRODUCTION AND BACKGROUND

This is the final report on research supported under Contract N00014-78-C-0447, which terminated May 31, 1979. The general subject matter is atomic and molecular interaction theory. The work reported is continuous with that supported under Contract N00014-72-C-0051, which terminated in March 1978.

A principal emphasis during the contract period was on the theoretical study of atom-molecule reactive collisions. A particular focus of this work was to study the production of entropy in elementary collision processes. This work is intended to elucidate the mechanisms leading to final state population inversions, of importance in chemical laser systems.

Quantum dynamical studies of entropy production in the $H+H_2$ exchange reaction have been carried out in collinear, coplanar and three-dimensional dynamics. These studies have examined implications of quantum dynamics relevant to "surprisal" theory, which applies concepts of information theory to molecular collision processes. A classical dynamical method for the study of collisional entropy production has been formulated, in analogy to the quantum formalism. A new method has been developed for solving the classical collision problem in terms of action-angle variables. This method, which exploits recent developments in the theory of semiclassical quantization of bound states of nonseparable dynamical systems, has been applied to an inelastic collision model (collinear $He+H_2$), and has been formulated for reactive collisions. This method provides a simple definition of semiclassical quantum states, required for studies of entropy production.

Another major area of emphasis is the quantitative theory of electron scattering and attachment by complex atoms and molecules. A new approximate method for the treatment of nuclear motion in electron-molecule scattering has been formulated. This energy-modified adiabatic approximation (EMA) treats the nuclear motion as a perturbation, exploiting the relative simplicity of electron scattering calculations carried out for fixed nuclei. EMA calculations reproduce the complex vibrational excitation structures observed in e^--N_2 scattering. Qualitative implications of the EMA theory have been used to explain recent experimental observations of vibrational threshold excitation peaks and corresponding dissociative attachment structures in electron scattering by polar molecules.

If it were known with sufficient accuracy as a function of energy and scattering angle, the e^--He cross section could serve as a calibration standard for low energy electron scattering experiments. The matrix variational method, developed in the present project, was applied to new calculations of e^--He s- and p-wave phase shifts, designed to give the differential cross section below 19 eV to better than 1% accuracy when higher-order phase shifts are computed from the Born formula. Data from these calculations were published as tables that allow simple interpolation of the e^--He differential cross section to within indicated 1% accuracy for all angles over the energy range up to 19 eV.

II. RESEARCH ACCOMPLISHED: THEORY

Theory of Collisional Entropy Production

Internal state populations of reactive collision products can often be fitted by a simple "surprisal" formula, obtained by applying concepts derived from information theory.[1,2,3] The original proposal by Jaynes,[4] to apply information theory to statistical mechanics, provided a formalism for considering entropy production in an irreversible process. If similar ideas could be applied to an elementary molecular collision, the essential postulate would be that the entropy of the internal state population distribution is maximized not just for separated collision products but throughout the collision process. When a "prior" internal state population distribution,[1,2] defined as an unbiased distribution with no information content, is relevant to the dynamics, the appropriate postulate for entropy production is that the information content, or entropy deficiency relative to the prior distribution, should be minimized.

In order to access the validity of this postulate, it was proposed[5] that quantum dynamical calculations could be reformulated to provide consistent definitions of internal state populations as evolving functions of a reaction coordinate. Results of this approach, applied to calculations of the $H+H_2$ exchange reaction, are described in publications to be discussed in Section III. This work was summarized in a review lecture (Section IV, Publications, No. 4). In these calculations, it was found that surprisal theory, with the postulate of minimum information content, provides a remarkably accurate description of computed internal state populations throughout the exit channel region of a reactive collision.

Surprisal analysis depends crucially on the definition of the prior distribution function $\{P_i^0\}$, where index i refers to an internal state of the colliding system. Levine, Bernstein and collaborators[1,2] have consistently used the microcanonical prior distribution function, for which all points in classical phase space at given energy are equally weighted. Quack and Troe[6] have questioned the use of the microcanonical prior distribution. Pollak[7] has shown that several alternative distribution functions would be compatible with the usual postulates of statistical theory. In collinear classical dynamical calculations, Pollak found that the final vibrational state population distribution was approximately independent of the internal translational energy, in disagreement with the microcanonical distribution.

The present quantum dynamical studies have led to some tentative conclusions about the correct choice of prior distribution function. Since this work indicates that surprisal theory is a direct consequence of collision dynamics, not requiring separate statistical postulates, the prior distribution function must be compatible with the kinematics of scattering. This point was examined in a study of threshold laws relevant to reactive collisions (Section IV, Publications, No. 3). The threshold laws, which are kinematic in nature, determine the dependence of final state populations on product state relative translational energy when the latter is small.

For three-dimensional collision dynamics, the microcanonical prior distribution function is found to be compatible with the threshold law, which excludes alternatives considered by Quack and Troe.[6] For the $H+H_2$ exchange reaction in three dimensions, the rotational population distribution function implied by the simple surprisal formula, using the microcanonical prior distribution function, was found to be in remarkable agreement throughout the product region of the reaction with that computed quantum dynamically (Section IV, Publications, No. 5). In contrast, in collinear dynamics, the microcanonical prior distribution is incompatible with the threshold law. Collinear quantum dynamical calculations of vibrational state populations show clear disagreement with the surprisal formula, if the microcanonical prior distribution is used (Section IV, Publications, No. 6). The threshold law implies an alternative prior distribution that gives consistently better results. In coplanar dynamics, the microcanonical prior distribution is independent of relative translational energy, while the threshold law varies slowly (logarithmic energy dependence). Hence, the coplanar microcanonical function is qualitatively reasonable, but not strictly compatible with the threshold law. These studies have helped to elucidate the problems involved in attempting to use collinear or coplanar calculations to predict three-dimensional results, but indicate a formal difficulty that does not yet have a practical solution.

Although rapid progress is being made toward the implementation of quantum dynamical methods for reactive collisions, full 3-D calculations have as yet been carried out only for the $H+H_2$ exchange reaction.

Quantitative calculations for systems of more than three atoms are unlikely in the foreseeable future. In order to make progress with more complex systems, an analogous classical theory of dynamical entropy production is needed. The conceptual basis of such a theory has been developed (Section IV, Publications, No. 2).

The proposed formalism is intended to provide a classical analog of dynamical entropy as defined through the quantum dynamical S-matrix.[5] The formalism is closely related to semiclassical S-matrix theory.[8,9] An essential concept required for the classical dynamical theory is the definition of a statistical ensemble of trajectories at a common value of a reaction coordinate u rather than at a common time t, as in the classical Liouville equation. Another essential concept is the allocation of these trajectories to quantum states of internal motion defined for fixed u. A striking distinction between this approach and the classical Liouville theory is that dynamical entropy is defined here for a single collision process, treated as a stationary flow problem, in analogy to quantum dynamical solution of the time-independent Schrödinger equation. Although the dynamical equations are time-reversible, a collision process is viewed in this formalism as creating entropy in the form of a nonequilibrium product internal state distribution. Once the collision products separate, so that quantum phase relationships would be lost, the process becomes physically irreversible.

The development of dynamical entropy during a collision process is monitored on each surface of constant collision coordinate u by counting trajectories that lead to each energetically accessible internal state. The probability flux densities define a normalized internal state population distribution function, from which statistical entropy and information content can be computed as in the quantum dynamical theory.

The internal Hamiltonian on surface $u=u_1$ is

$$H_{int} = H - T_u \qquad (1)$$

where H is the full Hamiltonian and T_u is the translational kinetic energy. The definition of approximate eigenstates of H_{int} requires semiclassical quantization of a nonseparable dynamical system. Considerable recent progress has been made on this longstanding problem.[10,11] In the present method, it is necessary to assign internal quantum numbers to a given point in the phase space of internal variables, omitting u and its conjugate momentum p_u. Such a point is defined by the intersection of a given trajectory with the surface $u=u_1$. The Fourier transform method of Percival[10] can be adapted to this problem.

Fourier Transform Method for Atom-Molecule Collisions

It was originally intended to use Percival's method[10] only to define bound states of H_{int}, in conjunction with standard classical trajectory calculations. Subsequently, it was realized that this method, with suitable modifications, could be used for the full collision problem. The resulting new theoretical method has been formulated and applied to a model inelastic collision problem (Section IV, Publication, Nos. 8 and 13). In Percival's method, a Fourier transform procedure is used to compute semiclassically quantized energy levels of a bound nonseparable dynamical system. In the proposed new method, collision dynamics is converted to effective bound state dynamics by introducing an impulse potential barrier or reflecting the scattering potential to create a mirror image potential outside the collision region. Exploratory calculations of collinear He-H_2 inelastic collisions have been carried out successfully.

An iterative Fourier transform method is used to solve partial differential equations for the hypersurface (invariant toroid) in phase space that contains an indefinitely extended classical trajectory. Such a surface is characterized by a complete set of action integrals, which are constants of motion. The invariant toroid is the classical analog of a quantum mechanical wavefunction. On the toroid, all coordinates and momenta are periodic functions of the true angle variables. The extended trajectory appears as a bundle of parallel straight lines, uniformly dense in the primitive hypercube of the angle variables.

These properties reduce the computation of scattering cross sections to simple geometry, the calculation of projected areas, for each invariant toroid with quantized values of the action integrals. When fast Fourier transform and iterative refinement methods are fully exploited, the new method may be able to obtain the ensemble of trajectories that correspond to a semiclassical wave function with less computational effort than present methods.

The bound state method of Percival was used as an iterative procedure to determine parametrized potential energy functions for polyatomic molecules from given vibrational excitation energies (Section IV, Publications, No. 12). Such potential functions were derived for the triatomic molecules NO_2, SO_2 and ClO_2, fitted to the first nine vibrational levels in each case. Twenty-two higher vibrational excitation energies predicted by the model were found to differ from experimental values by at most 3 cm^{-1}. The parametrized potential function determined by vibrational levels alone is not unique. The method must be extended to include rotational perturbations if it is to provide a polyatomic generalization of the well-known RKR method for diatomic molecules.

Variational Methods for Multichannel Scattering.

Most recent quantitative variational calculations of electron scattering by atoms and molecules have used some form of the method of Kohn, introduced in 1948. This method is subject to computational anomalies that cannot easily be removed. Previous versions of the Kohn method for multichannel scattering have either failed to remove all anomalies or have done so at the cost of introducing artificial discontinuities in computed results. Such anomalies or discontinuities make it very difficult to achieve or to verify the convergence of variational calculations when accurate results are required. A new method, RIAF for the "restricted interpolated anomaly-free" method, has been devised (Section IV, Publications, No. 1, 9 and 10). It eliminates both anomalies and discontinuities, and is found otherwise to be as accurate as the best previous methods. A canonical form for transformation matrices occuring in modified versions of the Kohn theory has been established. This canonical form was used in the new variational method.

Energy-Modified Adiabatic Approximation

The large number of internal states (rotation and vibration) of a typical molecule has been a formidable obstacle to the development of quantitative methods in electron-molecule scattering theory. For fixed molecular nuclear geometry, meaningful electron scattering calculations have recently become possible. Additional theoretical analysis is needed to deduce rotational and vibrational transition cross sections from fixed-nuclei calculations. An adiabatic approximation is known to be valid for pure rotational excitation, but not for vibrational excitation, especially when electronic resonances occur. An energy-modified adiabatic approximation (EMA) has been proposed (Section IV, Publications, No. 11). The EMA approximation uses fixed-nuclei electron scattering data, but differs from earlier adiabatic theory by treating the molecular internal kinetic energy as an operator rather than neglecting it. In a trial calculation of e^{-}-N_2 scattering, the EMA method obtained vibrational excitation structure in good agreement with experiment. This structure is due to coupling between an electronic resonance and molecular vibrational motion.

III. RESEARCH ACCOMPLISHED: CALCULATIONS

Quantum Dynamical Studies of Surprisal Theory

In a quantum dynamical S-matrix formalism,[5] population distributions for internal states can be computed as a function of a reaction coordinate for a reactive atom-molecule collision. Quantum dynamical programs of Light and collaborators,[12] using an R-matrix propagator method,[13] were adapted for this purpose and were applied to calculations of the $H+H_2$ exchange reaction (Section IV, Publications, No. 4).

It has been found in many cases,[1,2,3] that final internal state populations of molecular collision products are approximated by

$$P_i = P_i^0 \exp(\alpha-\beta f_i) \tag{2}$$

where α and β are constants and f_i is the fraction of available energy assigned to internal state i. A distribution function of this form is obtained if the information content, defined by

$$I[P|P^0] = \Sigma_i P_i \, \ell n(P_i/P_i^0) \tag{3}$$

is minimized subject to the constraint that the mean value

$$\langle f \rangle = \Sigma_i P_i f_i \tag{4}$$

should have a specified value. The "prior" distribution function $\{P_i^0\}$ is postulated to have no information content. $I[P|P^0]$ is the negative mean value of the "surprisal" $-\ell n(P_i/P_i^0)$, which is a linear function of f_i if Equation (2) holds.

Calculations of the coplanar $H+H_2$ exchange reaction[5] indicated that the quantum dynamical entropy remained close to its maximum value, corresponding to a linear surprisal plot, throughout the product region of the reaction coordinate u. The microcanonical prior distribution function, constant for coplanar dynamics, was used in this work. In collinear dynamics, the microcanonical distribution function contains a factor k^{-1}, which is singular at vibrational thresholds.[7]

Calculations of the collinear $H+H_2$ reaction were carried out, using the program of Walker,[14] adapted for entropy production studies (Section IV, Publications, No. 6). Several different prior distributions were used in surprisal analysis of the internal vibrational state population distribution. If the linearity of the surprisal function is used as a criterion, the quantum dynamical results were incompatible with the microcanonical prior distribution, proportional to k^{-1}, where k is the translational wave number, or momentum in atomic units.

Several choices of the prior function were considered, suggested by quantum dynamical threshold laws.[15] Functions proportional to k or to $k/(k_0^2+k^2)$ gave reasonable results. At collision energies where resonance effects appeared, no choice of prior function was successful. This is compatible with interpretation of the surprisal theory in terms of the motion of wave packets, if mean values of dynamical variables are consistent with surprisal analysis but detailed phase relationships are not.

Calculations of the 3-D $H+H_2$ exchange reaction were carried out, to study rotational excitation (Section IV, Publications, Nos. 5, 7 and 14). In a pure quantum state, with definite total angular momentum $J=0$, for energies below the first vibrational excitation threshold, the internal rotational state population distribution function, computed using the Porter-Karplus potential surface,[16] was found to be in close agreement with that determined by surprisal synthesis. The latter distribution function is governed by a single parameter. This agreement holds throughout the exit channel region, as it had in previous coplanar calculations.[5]

The microcanonical prior distribution function, proportional in 3-D dynamics to $(2j+1)k$, where j is a rotational quantum number, was assumed in this work. Significantly less satisfactory results were obtained if the rotational factor $2j+1$ was omitted, but results were less sensitive to the power of k assumed. In contrast to collinear dynamics, the 3-D microcanonical distribution function is compatible with the quantum dynamical threshold law, and appears to be the correct prior distribution function for use in surprisal theory.

Calculations were also carried out in 3-D dynamics with an improved $H+H_2$ potential derived by Truhlar and Horowitz[17] from the accurate <u>ab initio</u> CI study of Siegbahn and Liu.[18] Although the surprisal formula for the internal rotational state population distribution obtained with this THSL potential surface was accurate for small values of the reaction coordinate u, it did not give a good fit for large values of u. The distinctive difference between the THSL and PK (Porter-Karplus) potentials is that, for the former, the H_3 conformation of minimum energy is nonlinear for large u.

The present dynamical S-matrix method was useful in calling attention to this qualitative distinction between the two potential surfaces. It was found subsequently that the published THSL potential was in error, due to a misprint in Reference 17, and that calculations with the corrected THSL potential gave linear surprisal plots for the final product rotational states (R. B. Walker, private communication). This implies that the uncorrected and corrected THSL potentials provide a model problem that may be extremely valuable for tests of surprisal theory, since surprisal synthesis apparently succeeds or fails in this case depending on the value of one parameter.

The dynamical S-matrix method provided an explanation of an approximate reactive flux rule for $H+H_2$: the sum of unnormalized reaction probabilities into product states with even rotational quantum numbers is approximately equal to the sum of reaction probabilities into odd product states. On the PK potential surface, at the low total energy considered, 0.722 eV, and with total angular momentum J=0, only two internal states are open for small u. These states, described by only even and by only odd basis functions, correlate with the two degenerate bending modes obtained by a normal coordinate expansion near the col. These two bending modes move in identical potential fields and thus contain the same reactive probability flux. Because even and odd states are decoupled for purely inelastic scattering, the separation of probability flux is preserved throughout the exit channel region.

An important general conclusion of these quantum dynamical studies is that the success of surprisal theory must be understood from a detailed examination of collision dynamics, rather than from logically distinct statistical considerations.

Variational Calculations of Accurate e^--He Cross Sections

Calculations were undertaken for the purpose of establishing reliable values of the electron-helium scattering cross section in the elastic scattering range, 0-19 eV (Section IV, Publications, Nos. 15 and 16). To ensure smooth variation with energy, the recently developed RIAF variational method was used (Section IV, Publications, No. 1). Because He can be used universally as a test target, standard cross sections will make it possible to calibrate angular and energy resolution in electron scattering experiments.

The best recent experimental data has a residual error of several percent, with some inconsistency between experiments. The present study was undertaken to verify and refine recent theoretical calculations by other methods, to examine convergence and residual error by systematic use of variational methods developed in the present project, and specifically to provide the needed standard differential elastic cross section to 1% accuracy or better.

It has been shown experimentally, and verified by previous theoretical calculations, that for e^--He scattering below 19 eV the phase shifts η_ℓ, for $\ell \geq 2$, are closely approximated by the partial wave Born formula, which depends only on the electric dipole polarizability of He. To compute the differential cross section to within 1%, this formula can be used for $\ell \geq 2$ if the polarizability itself has the required accuracy. Within this approximation, the scattering amplitude and total elastic cross section can be summed over all ℓ-values and expressed in closed form. The s- and p-wave phase shifts must be obtained from the analysis of experimental data or computed theoretically. These phase shifts are used to correct the Born approximation sums.

Phase shifts η_0 and η_1 were computed by the matrix variational method (RIAF version) for electron wave number k up to 1.1 a_0^{-1} (17 eV). Sequences of variational calculations were carried out in order to examine convergence independently with respect to extension of the orbital basis set, to the level of natural orbital representation of the target atom, and to quadrupole and short-range monopole polarizability effects. The directly computed phase shifts η_0 and η_1 were corrected for systematic errors, evaluated by these convergence studies, which indicate that the scattering amplitude obtained from the corrected phase shifts (estimated values) is accurate to within 1% for $0 \leq E \leq 19$ eV.

The final estimated phase shifts were fitted to smooth functions of energy, making use of analytic effective range formulas to define auxiliary functions that vary slowly over the full energy range considered. These auxiliary functions were fitted by cubic spline functions, with assigned values at the input wave numbers used for variational calculations. Values at k=0 were determined by iterative extrapolation, to make third derivatives of the auxiliary functions continuous at k=0.1 a_0^{-1}. For η_0, this procedure gives the scattering length 1.183511 a_0, in agreement with the experimental value 1.18 a_0, obtained by applying the effective range formula to the experimental momentum transfer cross section for energies 0.008 to 0.1 eV.[19]

The coefficients of these cubic spline fits were published (Section IV, Publications, Nos. 15 and 16) so that η_0 and η_1 could easily be evaluated for any energy value below 19 eV. The partial wave Born formula gives η_2 with sufficient accuracy, and closed formulas can be used for the contribution of all higher partial waves to the scattering amplitude and various cross sections.[20] A simple program has been written that combines these data to give differential, total elastic, and momentum transfer cross sections at any specified energy value up to 19 eV.

Comparison of these results with the best available experimental data indicates general agreement within published error estimates. The indicated level of accuracy of the computed data is compatible with the estimated accuracy (1% or better), which is better than any available experimental data.

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