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Comparison of Numerical Methods for Solving the Second-Order Differential Equations of Molecular Scattering Theory*

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The numerical solution of coupled, second-order differential equations is a fundamental problem in theoretical physics and chemistry. There are presently over 20 commonly used methods. Unbiased comparisons of the methods are difficult to make and few have been attempted. Here we report a comparison of 11 different methods applied to 3 different test problems. The test problems have been constructed to approximte chemical systems of current research interest and to be representative of the state of the art in inelastic molecular collisions. All calculations were done on the same computer and the attempt was made to do all calculations to the same level of accuracy. The results of the initial tests indicated that an improved method might be obtained by using different methods in different integration regions. Such a hybrid program was developed and found to be at least 1.5 to 2.0 times faster than any individual method.

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1. INTRODUCTION

State-to-state chemistry is currently one of the forefront fields of chemical physics. In the past 10 years the ability to study the microscopic behavior of single collision events, both experimentally and theoretically, has developed rapidly. A common feature of nearly all quantum-mechanical methods for studying molecular collisions, whether elastic, inelastic or reactive, is the need to solve coupled sets of linear second-order differential equations.

In the past 15 years there has been a rapid proliferation of methods for solving these sequations. Meaningful comparisons of the methods are very difficult to make because of the number and intangibility of the parameters which should be explored. These include, for example, the computer and compiler used, the accuracy of the solution and the efficiency of the computer codes. The few comparisons which have been made [1–4] usually involved only two or a few methods. Sometimes the calculations were done on different computers,and possibly worst of all, the tests were done on simple model problems. The collinear vibrational problem of Secrest and Johnson [5] and the 9-, and 16-channel rotational problems of Lester and Bernstein [6, 7] are commonly used test cases. These problems, however, reflect the status of reserach in this field 10 years ago. Methods which perform well on those problems may very well not be optimal for problems which are typical of current research in molecular collision theory.

Because of the large number of new methods, the lack of good bases for comparisons and the importance of solving these coupled equations in many branches of molecular collision theory, the National Resource for Computation in Chemistry (NRCC) sponsored a workshop on this topic. The goals of the workshop were to identify the methods and computer codes commonly in use, compare their performance on a fixed set of problems and make tested versions of them available to the scientific community. Volume I of the workshop proceedings [8] contains presentations of 12 different methods and a discussion of the problems used here. Volume II of the proceedings [9] provides a detailed analysis of the test results for 11 different computer codes. All of the codes tested are available from the NRCC[1] [10–22].

The main result of these tests is the development of a new hybrid program which typically runs twice as fast as any individual program and in many cases even faster for a given accuracy. In this paper we briefly describe the methods and test problems, present the main findings of the tests and described the new hybrid program.

Although this has been a large scale study it is by no means complete. The scope of the tests was limited by several factors—the willingness of those who wrote or have codes to invest the necessary time and effort, the cost, especially when overseas scientists are involved and the format of the tests since different methods often require different types of potential matrices and basis set information. We hope that the results of these tests will encourage others to extend them in the future.

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[1] After March 1, 1981 these codes can be obtained from the Quantum Chemistry Program Exchange, Department of Chemistry, Indiana University, Bloomington, Ind. 47405 and the National Energy Software Center, Argonne National Laboratory, 9700 South Cass Avenue, Argonne, Ill. 60439.

2. Coupled Equations

The equations to be solved are

$$\left(\frac{d^2}{dr^2}-\frac{l_i(l_i+1)}{r^2}+k_i^2\right)u_{ij}=\sum_{i'}V_{ii'}(r)\,u_{i'j}(r). \tag{1}$$

The boundary conditions imposed on the solutions are,

$$u_{ij}(0)=0 \tag{2}$$

and

$$\lim_{r\to\infty} u_{ij}(r)=k_i^{-1/2}j_{l_i}(k_ir)\,\delta_{ij}+k_i^{-1/2}\eta_{l_i}(k_ir)\,K_{ij}, \tag{3}$$

where $j_l(kr)$ and $\eta_l(kr)$ are spherical Bessel functions. It should be emphasized that these equations apply to many different types of collisions and many approximate formulations of close-coupling theory. Different systems and approximations differ only in the definitions of the potential matrix elements V_{ij}.

The number of coupled equations which must be solved ranges from one, for elastic scattering from spherically symmetric potentials, to many thousands for large molecules with many closely spaced energy levels. At the present time, up to about 100 coupled equations can be solved routinely and as many as 210 coupled equations have been solved [23]. With improving computer technology and numerical techniques, these numbers will undoubtedly increase significantly in the near future.

3. Methods

Almost as important as the number of equations which must be solved, however, is the number of numerical methods one must chose from for solving them. Table I presents a list of the most commonly used methods for solving differential equations. Those which have an abbreviation or acronym were tested in the present study. Although the development of these methods spans nearly 100 years, most have been developed since 1965. It was about this time that high-speed, digital computers became widely available as research tools, and chemists and physicists began developing specialized techniques for the solution of Eq. (1).

The methods in Table I fall into two broad categories—approximate solution methods and approximate potential methods. With the approximate solution methods, some functional form, usually a polynomial in r, is assigned to the solution, u, and/or its derivatives, over an interval Δr. The parameters of that functional form are then chosen to best satisfy Eq. (1). These methods tend to be numerically simple, but require stepsizes 10 to 20 times smaller than the wavelength of the solution. They are usually advantageous when the wavelength is long compared to the range of the potential. The approximate potential methods assign a functional form to the

TABLE I

Numerical Methods for Integrating Differential Equations[a]

Year	Method name	Library name[b]	Author
1883	Predictor–corrector		Bashforth and Adams [24]
1895	Runge–Kutta		Runge (1895) [24]; Kutta (1901) [24]
1927	Extrapolation methods		Richardson (1927) [25]; Gragg (1965) [25]; Bulirsche and Stoer (1966) [25]
1933	Numerov	MNN	Numerov [26, 27]
1955	DeVogelaere	DEVOG	DeVogelaere [28]
1956	Invariant imbedding		Bellman and Kalaba [29–31]
1966	Amplitude density		Johnson and Secrest [32]
1966	Exponential		Pechukas and Light (1966) [33]; Chang and Light (1969) [34]
1968	Finite difference boundary value method		Diestler and McKoy (1968) [35]; Truhlar and Kuppermann (1970) [36]
1969	Piecewise constant potential		Cheung and Wilson [37]
1969	Integral equations	SAMS	Sams and Kouri [38]
1969	Piecewise analytic reference potentials	GORDON	Gordon [39]
1969	Variable-order, variable-step predictor–corrector	PC	Krogh (1969) [40]; Gear (1971) [25]
1970	Iterative Numerov	MNN	Allison [3]
1973	Log derivative	LOGD	Johnson [41]
1974	Integral equations with piecewise analytic reference potential	INSCAT	Redmon and Micha [42]
1975	Stormer's 2- and 5-step formulas		Choi and Tang [43]
1976	R-matrix propagator	RMAT	Light and Walker [44]; Stechel *et al.* [45]
1977	Renormalized Numerov		Johnson [46]
1978	Exponential fitting		Raptis and Allison [47]
1978	Finite element method		Rabitz, *et al.* [48]
1979	L^2 approach to R-matrix propagator	L2RMAT	Schneider and Walker [49]
1979	Iterative method for a single vector of solutions		Thomas [23]
1979	Variable-interval, variable-step method	VIVS	Parker *et al.* [50]
1980	Hybrid combination of the log derivative and variable-interval, variable-step methods	VIVAS	Parker *et al.* [51, 52]

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[a] This list includes only those methods which have been primarily applied to equations with local potentials. Many other methods have been developed and applied to the integro-differential equations which arise in the theory of electron scattering from atoms and molecules. For these methods the reader is referred to the review articles, Refs. [53–57].

[b] Library name, when given, indicates that the method was tested in this work and the entry is the program name in the NRCC software library.

potential over an interval Δr, usually constant or linear in r, for which exact solutions are known. The solutions are then matched at the interval boundaries to propagate over the entire region. These methods are advantageous when the potential is slowly varying and when the wavelength is small compared to the range of the potential. They also have the advantage that much of the intermediate computational information for a given potential and basis set is energy independent. Therefore, if the potential is energy independent and calculations are done for more than one collision energy, the information from the first calculation can be saved and reused with considerable savings in computation time for other energies. The approximate potential methods in Table I are the methods of piecewise analytic reference potentials, the R-matrix propagator methods and the variable-interval, variable-step method. The others can all be classified as approximate solution methods. For further classification and discussion of the relative advantages and disadvantages of these methods the reader is referred to the articles of Secrest [8, 58].

A very brief description of each of the programs used in the tests follows.

Approximate Solution Methods

PC [10]—*Predictor–corrector* methods are among the oldest in use, but have been significantly improved in recent years [25, 40, 59]. This program uses the variable-order, variable-stepsize predictor–corrector method of Shampine and Gordon [59].

DEVOG [11]—The *DeVogeleare* algorithm was introduced to close-coupling calculations by Lester [60]. It is efficient, simple to program, and stepsize changes of arbitrary value are easy to introduce.

MNN [12]—The *Numerov* method has been widely used in many problems in theoretical physics. Recent modifications to it have been given by Allison [3] and Johnson [46]. The program used in these tests was the Minnesota Numerov (MNN) program of Brandt, Truhlar, Onda, and Thirumalai. This program can do both the regular and iterative [3] Numerov methods and automatically selects the most efficient one.

LOGD [13, 17]—The *log derivative method* [41] uses a special invariant imbedding technique to propagate $u'u^{-1}$ rather than the wavefunction, u, itself. This program also has an option for using a multi-channel WKB approximation [61]. This is useful for very long range problems where the potential is slowly varying over several wavelengths. In this region the WKB approximation is both accurate and efficient.

SAMS [14]—*Sams and Kouri* [38] transformed Eq. (1) into a Volterra integral equation and then developed a numerical technique by introducing an equally spaced quadrature formula. The program used in these tests employs a trapezoidal integration.

INSCAT [15]—This is an integral equation formulation of the piecewise analytic potential method [42]. The differential equations are converted to Volterra integral equations, but a constant potential approximation is made over each integration step. Since the potential is not diagonalized, however, this method is intermediate between the approximate solution and potential methods.

Approximate Potential Methods

GORDON [16]—The piecewise analytic reference potential method of *Gordon* [39] was the first approximate potential method. This is a modified version of the original Gordon code [62].

VIVS [17]—This new *variable-interval, variable-step* method [50] uses piecewise constant potentials and a fixed basis within each interval. Analytic perturbation corrections to the solutions are summed over the steps in an interval and the solutions are carried in R-matrix form.

RMAT [18]—This is a modification [63] of the R-*matrix propagation method* [44, 45] in which the analytic R-matrices are perturbatively corrected in each sector, but propagated as in the original method [44, 45].

L2RMAT [19]—This is a modification of the R-matrix propagation method which uses an L^2 expansion of the wavefunction in the region of rapidly varying potential [49]. The original R-matrix propagation method [44, 45] is then used for the remainder of the integration range.

VIVAS [17]—This is a hybrid program [51, 52] which uses the log derivative method in the inner region where the potential is rapidly varying, and uses the variable-interval, variable-step method for the outer region.

4. TEST PROBLEMS

In constructing the test problems, an attempt was made to pick only realistic, three-dimensional problems of current research interest. With this in mind, three chemical systems—He–H_2, Li^+–CO, and e^-–N_2—were chosen.

Test 1. This is the problem of rotational and vibrational excitation of H_2 by He impact. An accurate configuration-interaction (CI) potential energy surface is available and converged close-coupling calculations of the inelastic transition probabilities have been done [64]. The study of this system is therefore representative of the present state of the art in inelastic collision studies. The case we report here is an 18-channel problem for total angular momentum, $J = 4$, and relative kinetic energy, $E = 0.0224$ hartree. When H_2 is in its ground state, this corresponds to an asymptotic impact parameter of $b = 0.38\, a_0$ and a wavelength of $0.6\, a_0$. Table II gives the complete set of basis quantum numbers.

Test 2. This is the problem of rotational excitation of rigid-rotor CO molecules by Li^+ impact. The problem was chosen because of the extremely long range of the interaction potential. The charge–dipole interaction leads to off-diagonal coupling matrix elements which asymptotically are proportional to r^{-2}. A high quality CI potential energy surface is available [65] and experimental measurements of the differential cross sections in the energy range 4.0–7.0 eV have been done [66, 67]. Because of the close spacing of the CO rotational energy levels, converged close-coupling calculations at these energies are not possible with existing methods.

TABLE II

Basis Set Quantum Numbers for Tests 1, 2, and 3[a]

Test 1					Test 2				Test 3			
No.	η	j	l	Target energy[b]	No.	j	l	Target energy[b]	No.	j	l	Target energy[b]
1	0	0	4	0.0	1	0	25	0.0	1	0	5	0.0
2		2	2	0.001616	2	1	24	$1.76E-5$	2	2	3	$5.46E-5$
3			4		3		26		3		5	
4			6		4	2	23	$5.28E-5$	4		7	
5		4	0	0.005329	5		25		5	4	1	$1.82E-4$
6			2		6		27		6		3	
7			4		7	3	22	$1.06E-4$	7		5	
8			6		8		24		8		7	
9			8		9		26		9		9	
10		6	2	0.011009	10		28		10	6	1	$3.82E-4$
11			4		11	4	21	$1.76E-4$	11		3	
12			6		12		23		12		5	
13			8		13		25		13		7	
14			10		14		27		14		9	
15	1	0	4	0.018971	15		29		15		11	
16		2	2		16	5	20	$2.64E-4$	16	8	3	$6.56E-4$
17			4		17		22		17		5	
18			6		18		24		18		7	
					19		26		19		9	
					20	6	19	$3.70E-4$	20		11	
					21		21		21		13	
					22		23					

[a] Blank entries indicate a repeat of the above entry.

[b] Energies are in atomic units $e = m = \hbar = 1$.

Therefore, the full cross section problem is really beyond the scope of exact present day methods. However, we can still test the speed and performance of the methods on unconverged basis sets. The problem we chose is a 22-channel problem with $J = 25$ and $E = 0.00195$ hartree. For CO in its ground state, this corresponds to an asymptotic impact parameter $b = 4.0\, a_0$ and a wavelength of $1.0\, a_0$. The complete basis set used is shown in Table II.

Test 3. This is the problem of rotational excitation of N_2 by electron impact. We wanted to test the codes on an electron–molecule system, but without the complications of the exchange interaction. For this reason we used the potential energy surface of Onda and Truhlar [68] which employs, a local approximation to the exchange potential, valid only for a collision energy of 1.1025 hartree. The total angular momentum is $J = 5$. The asymptotic impact parameter is $b = 3.37\, a_0$ and the wavelength is $4.23\, a_0$. The complete basis set for this test is contained in Table II.

TABLE III

Most of the Important Physical Attributes of Molecular Collision Problems

Attribute	Range[a]
Chemical systems	H_e–H_2, Li^+–CO, e^-–N_2
No. of channels	2–32
No. of closed channels	0–21
Integration range needed for convergence	16–1000 a_0
Collision energy	0.779*E*–4–1.1025 hartree
Asymptotic wavelength	0.6–5.0 a_0
Transition probabilities	0.91*E*–10–0.47
Reactive scattering	Not tested
Multiple electronic surfaces	Not tested
Very high energy (very short wavelength)	Not tested
Coulomb potentials	Not tested
Large No. of channels (~100)	Not tested

[a] The range, when given, indicates the range covered by the test problems.

Table III summarizes most of the physical attributes which are important in molecular collision calculations. For the tested group, we list the range of the attribute covered by the tests. Tests 1–3 do not cover the full ranges listed in Table III. They are only a representative subset of the 24 test problems studied [9]. However, examination of the full set of results does not change the conclusions reached here based on the above three tests.

5. Results and Discussion

In order to get a meaningful comparison, all calculations were done on the same computer (LBL's CDC 7600), and all programs were compiled on the LBL FTN4 compiler with optimization level 2. The programs were all written in fairly portable FORTRAN. No assembly language routines were used.

Possibly the most difficult quantity to assess in a comparison such as this is the accuracy of the solution. For any given method, the difference between two- and three-significant-figure accuracy is often a factor of 2 or 3 in computer time. Therefore, it is important to compare times only for calculations which result in similar accuracy. Unfortunately, precise accuracy control is very difficult to obtain—especially when the exact answer is not known ahead of time as with these test problems. An attempt was made in each case to achieve two-significant-figure accuracy in one specified transition probability for each test. The actual values computed and the correct values are shown in Table IV. It can be seen that while the

TABLE IV

Selected Transition Probabilities, $|S_{ij}|^2$, for Each Test and Method[a]

Program	Test 1 (1,15)[b]	Test 2 (1,4)[b]	Test 3 (1,2)[b]
PC	0.1184 − 8[c]	0.2036 − 1[c]	0.2864 − 2[c]
DEVOG	0.1183 − 8	0.1722 − 1	0.2874 − 2
MNN	0.1181 − 8	0.1666 − 1	0.2874 − 2
LOGD	0.1185 − 8	0.1715 − 1	0.2868 − 2
SAMS	0.1181 − 8	0.1752 − 1	0.2876 − 2
INSCAT	0.1149 − 8	0.2154 − 1	0.2887 − 2
GORDON	0.1278 − 8	0.1702 − 1	0.2860 − 2
VIVS	0.1213 − 8	0.1732 − 1	0.2867 − 2
RMAT	0.1193 − 8	0.1709 − 1	0.2885 − 2
L2RMAT	0.1170 − 8	0.1718 − 1	0.2865 − 2
VIVAS	0.1164 − 8	0.1710 − 1	0.2861 − 2
Correct	0.1184 − 8	0.1715 − 1	0.2873 − 2
$r_{\min}$	2.5	3.0	0.01
$r_{\max}$	45.0	800.0	120.0

[a] The correct answers for Tests 1 and 3 are accurate to four significant figures. For Test 2, the correct answer is only guaranteed to two significant figures. $r_{\min}$ and $r_{\max}$ indicate the integration range used to calculate the correct answer.

[b] The channel numbers i and j correspond to those in Table II.

[c] − indicates the power of 10.

results are reasonably uniform, there are differences in the accuracies achieved. Therefore, it is quite possible that the times reported for specific methods could be improved on some tests by fine tuning. Nonetheless, we believe the comparisons are meaningful for two reasons. First, as we show below, the spread in computer time over all methods for each test is a factor between 20 and 50. This is a significant difference which could not be eliminated by refining the accuracy. Second, the accuracies and times reported here are typical of those which would be chosen for a new research problem where the accuracy of each partial wave could not be individually fine tuned.

Most workers in this field have realized that the computational effort to integrate Eq. (1) is different in different regions of the integration variable, r. For example, when r is near the origin the potential is often large and rapidly varying. The solutions have exponential behavior requiring small stepsizes. When r is large, the potential is small and slowly varying. The solutions are oscillatory and numerically stable to integrate. Therefore, rather than simply compare the total integration times for all of the methods, it is much more revealing to see which methods are fastest in different regions of r. We do this in the following fashion. By dividing the total time by the total number of integration steps we get the average computer time per step. Then by dividing the entire integration range into smaller intervals and counting the number of steps taken in each interval, we get a histographic approximation to dt/dr, the computer time to integrate a unit of distance, i.e.,

$$\frac{dt}{dr} \cong \frac{T_{\text{total}}}{N_{\text{total}}} \frac{N(r+\Delta r)-N(r)}{\Delta r}, \tag{4}$$

where $N(r)$ is the number of steps required to integrate from $r_{\min}$ to r. Note that the reciprocal of this, dr/dt, is the "velocity" at each point, r, with which the computer is integrating the coupled equations.

The results from all the methods tested are shown in Tables V–VII and selected results in Figs. 1–3. It was not convenient to compute dt/dr at exactly the same points, r, for all methods. Therefore, linear interpolation was used to calculate the numbers in Table V–VII. The time at the bottom of each table is the total time for the region bounded by the first and last values of r listed.

Figure 1 and Table V show the results for Test 1. High absolute accuracy is required on this problem because the transition probability being calculated is very small. For many of the programs this was the most difficult problem to solve. For small r, LOGD is most efficient by a fairly wide margin, and VIVS at the second energy is best for large r. For first energy calculations, however, INSCAT has the smallest total time.

Figure 2 and Table VI show the results for Test 2. This is a long range problem and the total times are dominated by the value of dt/dr at large r. The results are only shown for $r \leqslant 150.00\, a_0$ even though $r_{\max} = 800.0\, a_0$ was necessary for the

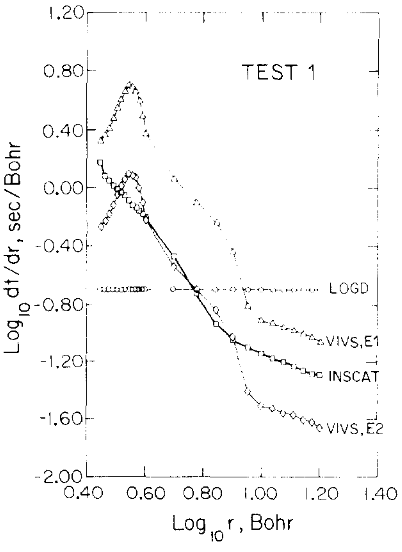

FIG. 1. Test 1. Comparison of dt/dr for the LOGD, VIVS, and INSCAT programs. E1 and E2 indicate first and second energy calculations, respectively.

TABLE V

dt/dr (sec/a_0) for Test 1[a]

r	PC	DEVOG	MNN	LOGD	SAMS	INSCAT	GORDON $E1$	VIVS $E1$	RMAT $E1$	L2RMAT $E1$	GORDON $E2$	VIVS $E2$	RMAT $E2$	L2RMAT $E2$
2.8	4.241	1.020	0.810	0.202	2.208	1.500	10.396	2.130	5.019	7.607	2.043	0.542	1.920	0.520
2.9	3.540	0.912	0.719	0.202	2.211	1.196	12.184	2.354	4.988	6.966	2.394	0.600	1.908	0.580
3.0	2.839	0.805	0.628	0.202	2.213	1.105	13.330	2.579	4.957	6.325	2.620	0.657	1.896	0.640
3.1	2.138	0.698	0.537	0.202	2.213	1.040	14.202	3.000	4.927	5.684	2.792	0.764	1.884	0.700
3.2	1.438	0.591	0.446	0.202	2.213	0.979	15.074	3.532	4.896	5.043	2.964	0.899	1.873	0.760
3.3	1.013	0.560	0.391	0.202	2.213	0.932	14.943	4.064	4.865	4.403	2.938	1.033	1.861	0.820
3.4	1.583	0.635	0.412	0.202	2.213	0.882	13.810	4.515	4.835	3.762	2.714	1.147	1.849	0.880
3.5	2.154	0.710	0.434	0.202	2.213	0.820	12.677	4.945	4.804	3.121	2.489	1.256	1.837	0.940
3.6	2.725	0.786	0.455	0.202	2.211	0.758	11.647	4.845	4.589	2.480	2.286	1.231	1.755	1.000
3.7	3.295	0.861	0.477	0.202	2.208	0.725	11.029	4.595	4.375	2.354	2.166	1.169	1.673	0.950
3.8	3.787	0.919	0.492	0.202	2.205	0.694	10.412	3.900	4.160	2.229	2.046	0.993	1.591	0.900
3.9	3.382	0.940	0.481	0.202	2.203	0.661	9.794	3.133	3.945	2.103	1.926	0.798	1.509	0.850
4.0	2.978	0.961	0.470	0.202	2.200	0.623	9.201	2.366	3.731	1.978	1.811	0.603	1.427	0.800
5.0	1.663	1.076	0.439	0.202	2.213	0.341	5.057	1.151	2.095	1.035	0.999	0.293	0.801	0.420
6.0	1.751	1.076	0.439	0.202	1.776	0.186	2.980	0.795	1.278	0.580	0.590	0.202	0.489	0.233
7.0	1.751	1.076	0.439	0.202	1.325	0.117	1.760	0.582	0.869	0.300	0.350	0.148	0.332	0.120
8.0	1.751	1.076	0.439	0.202	1.325	0.090	1.580	0.368	0.715	0.255	0.314	0.094	0.274	0.105
9.0	1.817	1.076	0.439	0.202	1.325	0.079	1.400	0.155	0.715	0.210	0.278	0.039	0.274	0.090
10.0	2.531	1.076	0.439	0.202	0.994	0.072	1.265	0.123	0.673	0.197	0.251	0.031	0.258	0.083
11.0	1.821	1.076	0.439	0.202	0.663	0.067	1.174	0.117	0.589	0.183	0.233	0.030	0.225	0.077
12.0	1.552	1.076	0.439	0.202	0.663	0.063	1.083	0.111	0.517	0.170	0.215	0.028	0.198	0.070
13.0	1.828	1.076	0.439	0.202	0.663	0.059	1.006	0.105	0.482	0.162	0.195	0.027	0.184	0.066
14.0	1.754	1.076	0.439	0.202	0.663	0.055	0.934	0.099	0.447	0.154	0.175	0.025	0.171	0.062
15.0	1.754	1.076	0.439	0.202	0.398	0.052	0.868	0.094	0.412	0.146	0.159	0.024	0.158	0.058
16.0	1.764	1.076	0.439	0.202	0.133	0.051	0.821	0.088	0.382	0.138	0.155	0.022	0.146	0.054
Time	25.494	13.799	5.894	2.668	15.825	2.602	39.007	9.298	16.421	9.376	7.667	2.366	6.281	2.779

[a] $E1$ and $E2$ refer to first and second energy calculations, respectively. Time (at bottom of table) is the total time for integrating between the first and last values of r in column 1.

TABLE VI

dt/dr (sec/a_0) for Test 2[a]

r	PC	DEVOG	MNN	LOGD	SAMS	INSCAT	GORDON *E*1	VIVS *E*1	RMAT *E*1	L2RMAT *E*1	GORDON *E*2	VIVS *E*3	RMAT *E*2	L2RMAT *E*2
3.5	14.089	2.521	3.710	0.979	2.136	5.592	4.470	3.209	3.396	16.800	0.610	0.840	1.298	0.790
3.6	12.162	2.157	3.143	0.979	2.136	5.209	4.522	3.322	3.396	16.800	0.633	0.870	1.298	0.790
3.7	10.435	1.793	2.576	0.979	2.136	4.844	4.588	3.434	3.396	15.649	0.661	0.900	1.298	1.029
3.8	8.867	1.496	2.160	0.979	2.136	4.494	4.653	6.519	3.396	14.499	0.690	1.709	1.298	1.268
3.9	7.614	1.325	1.940	0.979	2.136	4.160	4.718	9.033	3.396	13.348	0.719	2.368	1.298	1.506
4.0	6.360	1.154	1.719	0.979	2.136	3.840	4.783	8.812	3.396	12.198	0.747	2.309	1.298	1.745
4.1	5.107	0.983	1.499	0.979	2.136	3.532	4.849	8.590	3.396	11.047	0.776	2.251	1.298	1.984
4.2	3.853	0.812	1.279	0.979	2.136	3.235	4.914	4.727	3.396	9.896	0.804	1.237	1.298	2.223
4.3	3.054	0.708	1.147	0.979	2.136	2.949	4.979	4.480	3.260	8.746	0.833	1.173	1.246	2.462
4.4	2.940	0.786	1.209	0.979	2.136	2.669	5.045	4.660	2.989	7.595	0.861	1.221	1.142	2.701
4.5	2.826	0.865	1.271	0.979	2.136	2.393	5.110	4.757	2.717	6.876	0.890	1.248	1.038	2.762
4.6	2.712	0.944	1.333	0.979	2.136	2.113	4.895	3.895	2.445	6.588	0.852	1.006	0.934	2.647
4.7	2.598	1.023	1.394	0.979	2.136	1.829	4.680	2.914	2.174	6.301	0.814	0.763	0.831	2.531
4.8	2.508	1.089	1.444	0.979	2.136	1.507	4.465	2.015	1.970	6.013	0.776	0.527	0.753	2.416
4.9	2.463	1.091	1.444	0.979	2.136	1.179	4.250	1.858	1.834	5.725	0.738	0.486	0.701	2.300
5.0	2.418	1.093	1.444	0.979	2.136	1.069	4.035	1.702	1.698	5.437	0.700	0.445	0.649	2.184
6.0	2.248	0.976	1.444	0.979	2.136	1.565	2.960	0.629	1.189	3.172	0.510	0.165	0.454	1.276
7.0	1.667	0.804	1.082	0.979	1.897	1.314	2.620	0.461	1.456	1.897	0.454	0.121	0.556	0.765

8.0	1.401	0.691	0.722	0.979	1.658	1.046	2.020	0.372	1.844	1.615	0.354	0.098	0.704	0.651
9.0	1.107	0.624	0.722	0.538	1.418	0.830	1.660	0.328	1.916	1.411	0.289	0.086	0.732	0.568
10.0	0.954	0.587	0.722	0.538	1.179	0.665	1.357	0.285	1.673	1.515	0.234	0.075	0.639	0.610
11.0	0.937	0.565	0.722	0.538	0.940	0.537	1.223	0.254	1.430	1.619	0.212	0.067	0.547	0.652
12.0	0.756	0.553	0.722	0.538	0.837	0.435	1.090	0.223	1.187	1.615	0.190	0.058	0.454	0.649
13.0	0.784	0.545	0.722	0.538	0.735	0.351	0.957	0.192	0.944	1.584	0.168	0.050	0.361	0.636
14.0	0.639	0.541	0.722	0.538	0.632	0.280	0.823	0.162	0.701	1.540	0.146	0.042	0.268	0.619
15.0	0..847	0.538	0.722	0.430	0.53-	0.213	0.754	0.142	0.458	1.486	0.133	0.037	0.175	0.598
16.0	0.550	0.537	0.722	0.430	0.427	0.170	0.705	0.134	0.440	1.414	0.124	0.035	0.168	0.570
17.0	0.612	0.535	0.722	0.430	0.427	0.130	0.657	0.125	0.421	1.241	0.115	0.033	0.161	0.501
18.0	0.651	0.535	0.722	0.430	0.427	0.141	0.608	0.117	0.402	1.068	0.105	0.031	0.153	0.432
19.0	0.788	0.535	0.722	0.430	0.427	0.164	0.560	0.109	0.383	0.895	0.096	0.028	0.146	0.364
20.0	0.707	0.535	0.722	0.430	0.427	0.174	0.512	0.100	0.364	0.807	0.087	0.026	0.139	0.328
25.0	0.871	0.535	0.722	0.074	0.427	0.168	0.318	0.065	0.269	0.476	0.050	0.017	0.103	0.194
30.0	0.649	0.535	0.722	0.061	0.427	0.149	0.272	0.056	0.174	0.257	0.043	0.015	0.067	0.105
35.0	0.555	0.535	0.722	0.052	0.427	0.131	0.226	0.048	0.079	0.024	0.036	0.012	0.030	0.091
40.0	0.716	0.535	0.722	0.046	0.248	0.117	0.179	0.039	0.075	0.191	0.029	0.010	0.029	0.078
45.0	0.658	0.537	0.722	0.041	0.126	0.105	0.133	0.030	0.071	0.158	0.021	0.008	0.027	0.064
50.0	0.658	0.537	0.722	0.036	0.107	0.095	0.086	0.025	0.067	0.125	0.014	0.007	0.026	0.050
100.	0.658	0.537	0.722	0.018	0.107	0.048	0.020	0.013	0.040	0.059	0.003	0.003	0.015	0.023
150.	0.658	0.537	0.722	0.012	0.107	0.032	0.013	0.007	0.025	0.046	0.003	0.002	0.010	0.019
Time	110.176	81.001	108.733	15.137	38.449	22.754	37.945	14.529	28.598	56.514	6.407	3.805	10.927	19.634

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[a] $E1$ and $E2$ refer to first and second energy calculations, respectively. Time (at bottom of table) is the total time for integrating between the first and last values of r in column 1.

TABLE VII

dt/dr (sec/a_0) for Test 3[a]

r	PC	DEVOG	MNN	LOGD	SAMS	INSCAT	GORDON $E1$	VIVS $E1$	RMAT $E1$	L2RMAT $E1$	GORDON $E2$	VIVS $E2$	RMAT $E2$	L2RMAT $E2$
0.2	13.599	3.601	7.160	0.216	0.229	4.254	14.943	1.560	2.677	5.910	2.900	0.380	1.025	0.326
0.4	10.695	2.809	5.371	0.216	0.229	4.316	7.043	1.560	2.677	5.910	1.364	0.380	1.025	0.358
0.6	6.313	1.662	2.921	0.216	0.229	2.868	6.088	2.086	2.590	5.910	1.179	0.508	0.991	0.390
0.8	2.222	0.555	0.800	0.216	0.229	2.213	5.132	2.663	2.244	4.952	0.994	0.649	0.859	0.408
1.0	1.641	0.453	0.650	0.216	0.229	1.776	4.443	3.373	1.898	3.994	0.860	0.822	0.726	0.426
1.2	1.060	0.351	0.500	0.215	0.229	1.401	3.931	3.240	1.552	3.036	0.761	0.789	0.594	0.444
1.4	0.852	0.275	0.409	0.213	0.229	1.224	3.419	2.262	1.205	2.078	0.662	0.550	0.461	0.462
1.6	1.174	0.240	0.385	0.212	0.229	1.073	2.907	1.855	0.981	1.120	0.563	0.452	0.375	0.480
1.8	1.497	0.205	0.361	0.210	0.229	0.951	2.395	1.449	0.878	1.013	0.464	0.353	0.336	0.431
2.0	1.281	0.181	0.321	0.208	0.229	0.868	1.883	1.144	0.774	0.907	0.364	0.280	0.296	0.382
3.0	0.199	0.089	0.178	0.102	0.229	0.582	1.267	0.764	0.430	0.552	0.245	0.186	0.165	0.222
4.0	0.103	0.048	0.169	0.102	0.223	0.442	0.942	0.509	0.344	0.397	0.182	0.124	0.132	0.156
5.0	0.103	0.081	0.089	0.102	0.130	0.352	0.739	0.387	0.258	0.334	0.142	0.094	0.099	0.135
6.0	0.875	0.095	0.089	0.102	0.023	0.293	0.657	0.316	0.258	0.279	0.127	0.077	0.099	0.128
7.0	0.330	0.104	0.089	0.102	0.023	0.250	0.574	0.251	0.258	0.243	0.111	0.061	0.099	0.153
8.0	0.120	0.108	0.089	0.102	0.023	0.221	0.492	0.224	0.172	0.206	0.096	0.055	0.066	0.179

9.0	0.273	0.111	0.089	0.102	0.023	0.193	0.410	0.198	0.172	0.182	0.080	0.048	0.066	0.171
10.0	0.244	0.114	0.089	0.102	0.023	0.177	0.390	0.171	0.166	0.167	0.076	0.042	0.064	0.138
11.0	0.163	0.115	0.089	0.102	0.023	0.162	0.370	0.144	0.155	0.153	0.071	0.035	0.059	0.104
12.0	0.123	0.116	0.089	0.102	0.023	0.146	0.350	0.118	0.143	0.138	0.067	0.029	0.055	0.071
13.0	0.123	0.117	0.089	0.102	0.023	0.135	0.330	0.101	0.131	0.127	0.063	0.025	0.050	0.051
14.0	0.104	0.118	0.089	0.102	0.023	0.127	0.310	0.098	0.117	0.120	0.058	0.024	0.045	0.048
15.0	0.078	0.119	0.089	0.102	0.023	0.119	0.290	0.094	0.103	0.113	0.054	0.023	0.040	0.045
16.0	0.087	0.119	0.089	0.102	0.023	0.110	0.270	0.090	0.089	0.106	0.050	0.022	0.034	0.042
17.0	0.089	0.119	0.089	0.102	0.023	0.102	0.250	0.087	0.076	0.099	0.045	0.021	0.029	0.039
18.0	0.097	0.120	0.089	0.102	0.023	0.097	0.230	0.083	0.069	0.092	0.041	0.020	0.026	0.037
19.0	0.212	0.120	0.089	0.102	0.023	0.093	0.217	0.079	0.069	0.087	0.039	0.019	0.026	0.035
20.0	0.096	0.120	0.089	0.102	0.023	0.089	0.211	0.076	0.069	0.084	0.038	0.018	0.026	0.033
25.0	0.112	0.121	0.089	0.102	0.23	0.070	0.182	0.057	0.063	0.068	0.033	0.014	0.024	0.027
30.0	0.151	0.122	0.089	0.102	0.023	0.059	0.153	0.039	0.052	0.057	0.028	0.009	0.020	0.023
35.0	0.120	0.122	0.089	0.102	0.023	0.050	0.124	0.024	0.040	0.050	0.023	0.006	0.015	0.020
40.0	0.134	0.122	0.089	0.102	0.023	0.044	0.110	0.023	0.034	0.043	0.021	0.005	0.013	0.017
45.0	0.134	0.122	0.089	0.102	0.023	0.038	0.110	0.021	0.032	0.041	0.021	0.005	0.012	0.016
50.0	0.134	0.122	0.089	0.102	0.024	0.035	0.110	0.020	0.031	0.041	0.021	0.005	0.012	0.016
Time	14.422	7.298	7.592	5.321	2.128	9.375	22.095	9.352	7.937	11.778	4.222	2.280	3.038	3.345

[a] *E*1 and *E*2 refer to first and second energy calculations, respectively. Time (at bottom of table) is the total time for integrating between the first and last of *r* in column 1.

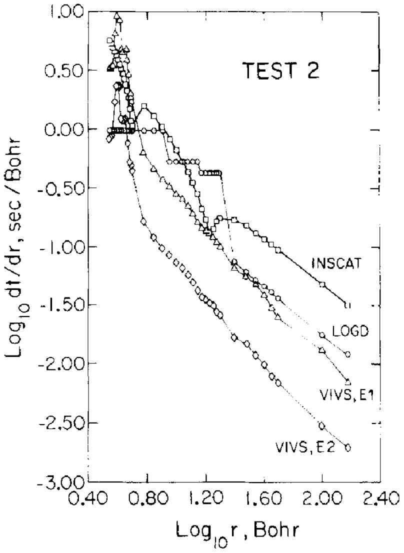

FIG. 2. Test 2. Same as Fig. 1.

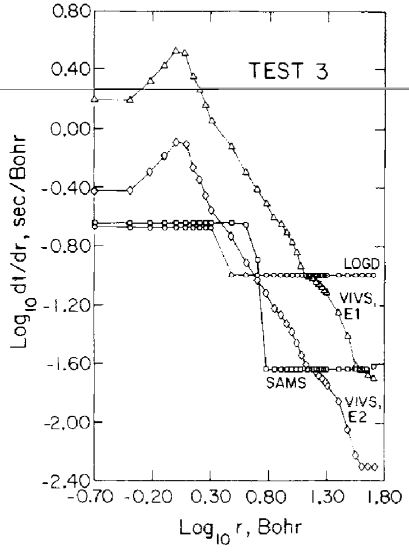

FIG. 3. Test 3. Comparison of *dt/dr* for the LOGD, VIVS, and SAMS programs. E1 and E2 indicate first and second energy calculations, respectively.

correct answer. For first energy calculations, LOGD is still best at small *r*. However, several of the approximate potential methods, especially GORDON, are faster at second energies in this region. For large *r*, VIVS at the second energy is again the fastest. Because of the long range of the potential and the large number of oscillations over this range, the approximate solution methods are impractical for this problem.

Figure 3 and Table VII show the results for Test 3. Although the collision energy is much higher for this problem than for the others, the asymptotic wavelength is still quite long—4.2 a_0—because of the small mass of the electron. Consequently, the approximate solution methods do much better than one might at first expect. For most of the programs, getting the correct answer for this problem presented the fewest difficulties. The potential energy function for the e–N_2 interaction at small r is quite small in contrast to the large barriers which are typical of atom–molecule potentials. In the non-classical region the interaction is dominated by the centrifugal terms. Since these terms are already included without approximation in the SAMS program, SAMS excels on this problem. Its total time is best of all the individual programs, even bettering the approximate potential methods on the second energy. LOGD and SAMS perform equally well at small r and are best in this region. VIVS, at the second energy, is again best for large r. However, we should emphasize that when the potential depends on the collision energy, as is the case for local approximations to the electron exchange potential, the second energy calculations of the

TABLE VIII

Integrated Total Times from Tables V–VII Arranged in Ascending Order on the Time[a]

Test 1			Test 2			Test 3		
Method	Energy	Time (sec)	Method	Energy	Time (sec)	Method	Energy	Time (sec)
VIVAS	2nd	1.0	VIVAS	2nd	2.2	VIVAS	2nd	1.1
HYBRID	2nd	1.3	HYBRID	2nd	3.3	HYBRID	2nd	1.6
VIVAS	1st	1.6	VIVS	2nd	3.8	SAMS		2.1
HYBRID	1st	2.1	VIVAS	1st	5.9	VIVS	2nd	2.3
VIVS	2nd	2.4	GORDON	2nd	6.4	VIVAS	1st	2.9
INSCAT		2.6	HYBRID	1st	9.5	L2RMAT	2nd	3.3
LOGD		2.7	RMAT	2nd	3.0	GORDON	2nd	4.2
L2RMAT	2nd	2.8	RMAT	1st	10.9	HYBRID	1st	4.4
MNN		5.9	VIVS	1st	14.5	LOGD		5.3
RMAT	2nd	6.3	LOGD		15.1	DEVOG		7.3
GORDON	2nd	7.1	L2RMAT	2nd	19.6	MNN		7.6
VIVS	1st	9.3	INSCAT		22.8	RMAT	1st	7.9
L2RMAT	1st	9.4	RMAT	1st	28.6	VIVS	1st	9.3
DEVOG		13.8	GORDON	1st	37.9	INSCAT		9.4
SAMS		15.8	SAMS		38.4	L2RMAT	1st	11.7
RMAT	1st	16.4	L2RMAT	1st	56.5	PC		14.4
PC		25.5	DEVOG		81.0	GORDON	1st	22.1
GORDON	1st	39.0	MNN		108.7			
			PC		110.2			

[a] The hypothetical HYBRID times are the sum of the times for LOGD integrated from $r_{\min}$ to r_0 and for VIVS integrated from r_0 to $r_{\max}$. r_0 is 8.0, 6.0, and 4.0 a_0 for Test 1, Test 2, and Test 3, respectively. The VIVAS times are real computer times from a new hybrid program.

approximate potential methods are not valid for comparison. The advantage these methods normally have for additional collision energies is lost in this case.

Overall, one is struck by the widely varying performances of the individual programs on the different physical problems. For example, INSCAT is the best performer for first energy calculations on Test 1 and a factor of 6 faster than SAMS. On Test 3, nearly the opposite is true. It is also quite clear that the best program would be a hybrid which followed the lowest *dt*/*dr* curve in all regions. Although no single method is best for all tests in any region, the log derivative method is nearly always best for small r and the variable-interval, variable-step method is nearly always best for large r. It therefore was desirable to combine these methods and form a new hybrid program. This is not the ideal combination for all problems, but this hybrid is always better than any individual method. This is evident in Table VIII.

Table VIII summarizes the total times from Tables V–VII, with the methods ordered according to computer time. The HYBRID entry is not from an actual program. It was hand calculated by integrating *dt*/*dr* from r_{min} to r_0 for LOGD and from r_0 to r_{max} for VIVS. It can be seen that the second energy HYBRID time is always smaller than any individual method. For the short range problems the HYBRID result is 1.5 to 2.0 times faster than either of the two methods individually. For the long range problem of Test 2, the improvement is minimal because the total time is dominated by the long range region.

On the basis of these findings a hybrid program, VIVAS, has been developed [17, 51, 52], which combines the variable-interval, variable-step and the log derivative methods. In addition, this program includes significant improvements to the original variable-interval, variable-step program, VIVS. It can be seen in Table VIII that the new program is significantly faster than any of the individual programs.

Finally, regarding Table VIII, we return to the discussion of accuracy. As stated before, improvements in performance of the individual methods can often be made and probably could be made here by refining the input parameters to each program. This means that the ordering of the methods in Table VIII should not be taken literally. However, factors of 20 to 50 are observed between the smallest and largest times. These differences are significant and it is believed doubtful that they could be eliminated by refinements in the numerical parameters of the methods.

REFERENCES

1. B. R. JOHNSON, D. SECREST, W. A. LESTER, JR., AND R. B. BERNSTEIN, *Chem. Phys. Lett.* **1** (1967), 396.
2. R. G. GORDON, *J. Chem. Phys.* **51** (1969), 14.
3. A. C. ALLISON, *J. Comput. Phys.* **6** (1970), 378.
4. W. N. SAMS AND D. J. KOURI, *J. Chem. Phys.* **53** (1970), 496.
5. D. SECREST AND B. R. JOHNSON, *J. Chem. Phys.* **45** (1966), 4556.
6. W. A. LESTER, JR. AND R. B. BERNSTEIN, *Chem. Phys. Lett.* **1** (1967), 207 and 347.
7. W. A. LESTER, JR. AND R. B. BERNSTEIN, *J. Chem. Phys.* **48** (1968), 4896.

8. L. D. THOMAS (Ed.), "Proceedings of the NRCC Workshop on Algorithms and Computer Codes in Atomic and Molecular Quantum Scattering Theory," Vol. I, Lawrence Berkeley Laboratory Report LBL-9501, 1979.
9. L. D. THOMAS (Ed.), "Proceedings of the NRCC Workshop on Algorithms and Computer Codes in Atomic and Molecular Quantum Scattering Theory," Vol. II, Lawrence Berkeley Laboratory Report LBL-9501, 1980.
10. L. D. THOMAS, "National Resource for Computation in Chemistry Software Catalog," Vol. I, Prog. No. KQ10 (PC), 1980.
11. L. D. THOMAS, "National Resource for Computation in Chemistry Software Catalog," Prog. No. KQ11 (DEVOG), 1980.
12. M. A. BRANDT, D. G. TRUHLAR, K. ONDA, AND D. THIRUMALAI, "National Resource for Computation in Chemistry Software Ctalog," Prog. No. KQ12 (MNN), 1980.
13. B. R. JOHNSON, "National Resource for Computation in Chemistry Software Catalog," Prog. No. KQ06 (LOGD), 1980.
14. K. D. MCLENITHAN AND D. SECREST, "National Resource for Computation in Chemistry Software Catalog," Prog. No. KQ07 (SAMS), 1980.
15. M. J. REDMON, "National Resource for Computation in Chemistry Software Catalog," Prog. No. KQ08 (INSCAT), 1980.
16. M. H. ALEXANDER, "National Resource for Computation in Chemistry Software Catalog," Prog. No. KQ05 (GORDON), 1980.
17. G. A. PARKER, J. V. LILL, AND J. C. LIGHT, "National Resource for Computation in Chemistry Software Catalog," Prog. No. KQ04 (VIVAS), 1980. This program contains both the LOGD program of Ref. [13] and an improved version of the VIVS program reported in Tables IV–VIII and Figs. 1–3.
18. T. G. SCHMALZ, "National Resource for Computation in Chemistry Software Catalog," Prog. No. KQ09 (RMAT), 1980.
19. R. B. WALKER AND B. SCHNEIDER, "National Resource for Computation in Chemistry Software Catalog," Prog. No. KQ13 (L2RMAT), 1980.
20. L. D. THOMAS, "National Resource for Computation in Chemistry Software Catalog," Prog. No. KT01 (HEH2), 1980.
21. L. D. THOMAS, "National Resource for Computation in Chemistry Software Catalog," Prog. No. KT02 (LICO), 1980.
22. L. D. THOMAS, "National Resource for Computation in Chemistry Software Catalog," Prog. No. KT03 (EN2), 1980.
23. L. D. THOMAS, *J. Chem. Phys.* **70** (1979), 2979.
24. W. E. MILNE, "Numerical Solution of Differential Equations," Dover, New York, 1970.
25. C. W. GEAR, "Numerical Initial Value Problems in Ordinary Differential Equations," Prentice–Hall, Englewood Cliffs, N.J., 1971.
26. D. R. HARTREE, "Numerical Analysis," Oxford Univ. Press (Clarendon), London/New York, 1952.
27. L. L. BARNES, N. F. LANE, AND C. C. LIN, *Phys. Rev. A* **137** (1965), 388.
28. R. DEVOGELAERE, *J. Res. Nat. Bur. Stand.* **54** (1955), 119.
29. R. E. BELLMAN AND R. E. KALABA, *Proc. Nat. Acad. Sci.* **42** (1956), 629.
30. M. R. SCOTT, "Invariant Imbedding and its Applications to Ordinary Differential Equations. An Introduction." Addison–Wesley, Reading, Mass., 1973.
31. M. E. RILEY AND A. KUPPERMANN, *Chem. Phys.* **51** (1969), 3448.
32. B. R. JOHNSON AND D. SECREST, *J. Math. Phys.* **7** (1966), 2187.
33. P. PECHUKAS AND J. C. LIGHT, *J. Chem. Phys.* **44** (1966), 3897.
34. D. CHANG AND J. C. LIGHT, *J. Chem. Phys.* **50** (1969), 2517.
35. D. J. DIESTLER AND V. MCKOY, *J. Chem. Phys.* **48** (1968), 2941.
36. D. G. TRUHLAR AND A. KUPPERMANN, *J. Chem. Phys.* **52** (1970), 3841.
37. A. CHEUNG AND D. J. WILSON, *J. Chem. Phys.* **51** (1969), 3448.
38. W. N. SAMS AND D. J. KOURI, *J. Chem. Phys.* **51** (1969), 4809.

39. R. G. Gordon, *J. Chem. Phys.* **51** (1969), 14.
40. F. T. Krogh, "VODQ/SVDQ/DVDQ—Variable Order Integrators for the Numerical Solution of Ordinary Differential Equations," TU Doc. No. CP-2308, NPO-11643, Jet Propulsion Laboratory, Pasadena, Calif., May 1969.
41. B. R. Johnson, *J. Comput. Phys.* **13** (1973), 445.
42. M. J. Redmon and D. A. Micha, *Chem. Phys. Lett.* **28** (1974), 341.
43. B. H. Choi and K. T. Tang, *J. Chem. Phys.* **63** (1975), 1775.
44. J. C. Light and R. B. Walker, *J. Chem. Phys.* **65** (1976), 4272.
45. E. B. Stechel, R. B. Walker, and J. C. Light, *J. Chem. Phys.* **69** (1978), 3518.
46. B. R. Johnson, *J. Chem. Phys.* **67** (1977), 4086.
47. A. Raptis and A. C. Allison, *Comput. Phys. Commun.* **14** (1978), 1.
48. H. Rabitz, A. Askar, and A. S. Cakmak, *Chem. Phys.* **29** (1978), 61.
49. B. I. Schneider and R. B. Walker, *J. Chem. Phys.* **70** (1979), 2466.
50. G. A. Parker, T. G. Schmalz, and J. C. Light, Ref. [8], p. 172 and *J. Chem. Phys.* **73**, 1757 (1980).
51. G. A. Parker, B. R. Johnson, and J. C. Light, Ref. [9], P. 101.
52. G. A. Parker, J. C. Light, and R. B. Johnson, *Chem. Phys. Lett.* **73**, 572 (1980).
53. P. G. Burke and M. J. Seaton, *Methods Comput. Phys.* **10** (1970), 1.
54. D. G. Truhlar, J. Abdalla, and R. L. Smith, *Advan. Chem. Phys.* **25** (1974), 211.
55. R. K. Nesbet, *Advan. At. Mol. Phys.* **13** (1977), 315.
56. J. Callaway, *Phys. Rep.* **45** (1978), 89.
57. N. F. Lane, *Rev. Mod. Phys.* **52** (1980), 29.
58. D. Secrest, *in* "Atom-Molecule Collision Theory" (R. B. Bernstein, Ed.), Chap. 8, p. 265 and Chap. 11, p. 377, Plenum, New York, 1979.
59. L. F. Shampine and M. K. Gordon, "Computer Solution of Ordinary Differential Equations," [illegible]
60. W. A. Lester, Jr., *J. Comput. Phys.* **3** (1968), 322.
61. B. R. Johnson, *Chem. Phys.* **2** (1973), 381.
62. R. G. Gordon, QCOL, Program 187, Quantum Chemistry Program Exchange (QCPE), Department of Chemistry, Indiana University.
63. J. C. Light, T. G. Schmalz, and J. V. Lill, Ref. [8], p. 116.
64. A. W. Raczkowski and W. A. Lester, Jr., *Chem. Phys. Lett.* **47** (1977), 45.
65. L. D. Thomas, W. P. Kraemer, and G. H. F. Diercksen, *Chem. Phys.* **30** (1978), 33.
66. R. Böttner, U. Ross, and J. P. Toennies, *J. Chem. Phys.* **65** (1976), 733.
67. W. Eastes, U. Ross, and J. P. Toennies, *Chem. Phys.* **39** (1979), 407.
68. K. Onda and D. G. Truhlar, *Chem. Phys. Lett.* **58** (1978), 512.