8554 *J. Phys. Chem. A* **2004,** *108,* 8554−8558

Scientific Contributions of Gert Due Billing

Michael Baer, Cecilia Coletti, George C. Schatz,* Søren Toxvaerd, Lichang Wang

Gert D. Billing's most significant contribution to the field of molecular dynamics simulations was the development of the classical path method and its extensive application in studies of energy transfer and reactive scattering. A full quantum description of dynamics can only be applied to small systems, at present consisting of up to four atoms. For larger systems, two alternative ways are widely used for describing collision dynamics. One way is to simplify the system by reducing dimensionality or using simplified potentials such as a harmonic oscillator potential to represent the interactions among atoms. The second way is to develop approximate yet accurate dynamics methods for the system in full dimensionality and without simplifying the potential. The classical path method belongs to the second category and has been successfully applied to various large systems.

Classical Path Method

The classical path method, which is also denoted as the semiclassical or quantum-classical method, was developed by Billing to study energy transfer processes in large systems where quantum effects cannot be neglected.[1−5] The classical path method was perhaps first used by Niels Bohr.[6] It was then used in the studies of energy transfer between an atom and a collinear diatomic molecule.[7]

In the classical path treatment, a large system is divided into two subsystems with one treated quantum mechanically and the other classically. Consider a collinear collision of an atom, A, and a diatomic molecule, BC, within the classical path treatment. The distance between the atoms B and C of the diatomic molecule, r, is treated quantum mechanically and the distance between the atom A and the center of mass of the diatomic molecule BC, R, classically. The working equations are

$$i\hbar\frac{\partial\varphi(\boldsymbol{r},\boldsymbol{t})}{\partial\boldsymbol{t}} = [\hat{\boldsymbol{H}}_0 + \boldsymbol{V}(\boldsymbol{r},\boldsymbol{R}(\boldsymbol{t}))]\varphi(\boldsymbol{r},\boldsymbol{t}) \tag{1}$$

$$\dot{\boldsymbol{R}} = \boldsymbol{P}_R(\boldsymbol{t})/\mu \tag{2}$$

$$\dot{\boldsymbol{P}}_R(\boldsymbol{t}) = -\frac{\partial}{\partial\boldsymbol{R}}\langle\varphi|\boldsymbol{V}(\boldsymbol{r},\boldsymbol{R})|\varphi\rangle|_{R=R(t)} \tag{3}$$

where $\hat{\boldsymbol{H}}_0$ includes the kinetic energy operator of the diatomic molecule BC and the potential describing the interaction between the B and C atoms, $V(r,R)$ is the potential describing the interaction between the atom A and the diatomic molecule BC, $\varphi(r,t)$ is the time-dependent wave function of the quantum subsystem, μ is the reduced mass of the diatomic molecule BC, and P_R is the momentum.

One problem associated with eqs 1−3 is that they do not fulfill the principle of detailed balance.[8,9] Indeed, comparisons between the classical path results and the exact solutions were quite disappointing in early applications.[7,10] To overcome this problem, Billing suggested using an intermediate relative velocity, also denoted as the symmetrized Ehrenfest approach.[10,11] Tests showed that the classical path method with the symmetrized Ehrenfest method is very accurate for describing inelastic rotational and vibrational transitions.[12]

From 1979 to 1988, Billing developed the classical path method to study energy transfer processes for polyatomic systems, ranging from simple three-body systems to organic molecules. These were published in a series of 14 papers in *Chemical Physics*.[13−26] In addition, Billing made great effort to implement his theory so that it could be applied to various problems. Prior to 1990, many of his applications were focused on studies of energy transfer processes.[27−36] Billing and co-workers investigated energy transfer processes in a wide range of systems, such as $He-H_2$,[5,37] H_2-H_2,[38−47] Li^+-H_2,[48] $H_2/D_2-{}^4He$,[49] HF−HF/DF,[50−57] $CO-N_2$,[58−61] N_2-N_2,[62−66] CO−CO,[67−73] Li^+-CO_2/N_2O,[17] N_2-CO_2,[19,74] CO−He,[75−79] $CO-D_2$,[80,81] CO_2-Ne/He,[15,21,82,83] $CO-p\text{-}H_2/o\text{-}H_2$,[84,85] $Ar-SF_6$,[22] $Ar-CO_2$,[21,23] NH_3-He/H_2,[86−94] NH_3-NH_3,[95,96] SF_6-SF_6,[97] Li^+-N_2,[98] H^+-CF_4,[99] $N_2^+-{}^4He$,[100] O_2^+-Kr,[101,102] O_2-O_2,[103−106] He/Ar−glyoxal,[26] N_2-O_2,[107] CO−Ar cluster,[108] O_3-Ar,[109] NO^+-CH_4,[110] NO^+-He,[111] HCl−HCl,[112] Ar_2-Ar_2.[113]

In the late 1980s, Billing shifted the focus of his research to reactive scattering. In a series of five papers,[114−118] he extended the classical path method to treat reactive scattering.[119] Billing, together with Muckerman,[120−122] used hyperspherical coordinates in their treatment of reactive scattering in the classical path theory. This is one of the earliest uses of hyperspherical coordinates in the studies of reactive scattering.[117,123−126] The approximations involved in the classical path treatment of reactive scattering are in general more severe than for inelastic collisions and cannot be corrected by simply resorting to a symmetrized Ehrenfest approach. To remedy the problem, Billing introduced quantum corrections as multitrajectory or Hermite corrections,[12,127−131] used wave packets,[132−137] or explored slightly different avenues, as in the formulation of the reaction volume approach,[138−141] a development on his earlier reaction path theory.[142,143]

Billing and co-workers applied the classical path method and its variations to study reactive scattering for a wide variety of systems, including: $H_2+OH\rightarrow H_2O+H$,[120,144−146] $Cl^-+CH_3Cl\rightarrow ClCH_3+Cl^-$,[121,147,148] $O+O_3\rightarrow 2O_2$,[149] $2O_2\rightarrow O+O_3$,[150,151] $O_2+O+Ar\rightarrow O_3+Ar^*$,[152] $CO+OH\rightarrow CO_2+H$,[153,154] $Ar^++H_2\rightarrow ArH^++H$,[155,156] $H_2+CN\rightarrow H+HCN$,[157] the B^++H_2 reaction including charge transfer,[158] $F+H_2/D_2$,[159−161] and the photodissociation[162] of HF,[163] O_3,[164] and CH_2[165,166] and many laser-induced processes.[167−172] His research in energy transfer and reactive scattering using the classical path treatment was reviewed in 1994 by him.[173]

In addition to the studies of gas-phase processes, Billing devoted a great deal of effort to studies of molecule−surface dynamics. In 1982, Billing extended the classical path method to study energy transfer in collisions of gas-phase atoms or molecules with solid surfaces.[174−176] After that, he was actively involved in studies of atom/molecule-surface scattering. Much of the methodology of using the classical path method to describe molecule−surface dynamics was summarized in his review article in 1990[177] and in his book in 1999.[178] Billing and co-workers were among the first to include the effects of

10.1021/jp040488g CCC: \$27.50

surface temperature and electron−hole pair excitation in the studies of molecule−surface scattering problems.[179−186] The classical path method was applied to many molecule−surface dynamics problems, including Ar/W(110),[187] CO/Pt(111),[188,189] C+O→CO on Pt(111),[190] CO+O→CO_2 on Pt(111),[191] H_2/Cu surfaces,[192−196] N_2→2N on a Rh surface,[197,198] CO/Cu(111),[199] D_2/Cu(111),[200,196] N_2→2N on a Ru surface,[201] N_2→2N on Fe(111),[202] He/Cu(001),[203] O_2 on silica,[204] dissociation of O_2 into different electronic atomic O states on a Ag(111) surface,[205] H+H→H_2 on graphite,[206] and the catalytic synthesis of ammonia.[207−209]

An important contribution from Billing was the effective use of second quantization in scattering problems,[210,16,211−213] in particular for molecule−surface scattering problems.[178] This allows one to treat more complex systems. In nearly all the molecule−surface scattering problems studied by Billing, second quantization was used for describing surface phonons.

It is also worth mentioning the long collaboration with Jolicard on many methodological aspects of quantum and semiclassical treatments of collision dynamics,[214−218] including multiphoton dissociation,[219,220] quantum chaos,[221] the use of Bloch's operators[222−224] and Floquet formalism.[225]

A short description of the classical path method, quantum corrections, and variations can be found in the chapter of the *Encyclopedia of Computational Chemistry*[129] devoted to quantum-classical methods.

Fundamental Principles

After the early derivation of the classical path theory, Billing's research focused on two closely related lines. The first was concerned with the introduction, as shown above, of corrections (multitrajectory approach, Hermite corrections, etc.) and variations (i.e., the second quantization) to the original classical path method in order to be able to treat approximately, yet effectively, a wide variety of quantum phenomena, even in complex systems. The second was the derivation of the seemingly ad hoc classical path method from the first principles, i.e., Schrödinger equation. This latter point eventually became a dominant goal for Gert as it meant the availability of a completely general rigorous theory that could both justify and explain the approximations involved in the primitive quantum classical approaches. An early derivation was provided in 1978 based on the Feynmann path integral.[226] Subsequently, Gert developed a formulation based on expressing the wave function in a Gauss−Hermite basis set in the collision coordinate R multiplied by an internal state basis function for the other coordinates.[126] In this approach for the simple case of collinear A + BC scattering, the wave function of the system is expressed as

$$\Psi(R,r,t) = \sum_{nk} a_{nk}(t)\varphi_n(r)\psi_k(R,t) \tag{4}$$

where ψ_k is a Gauss−Hermite function and the internal basis set is chosen to be eigenfunctions of $\hat{H}_0$, i.e.,

$$\hat{H}_0\varphi_n(r) = E_n\varphi_n(r) \tag{5}$$

We can approximate the wave function (eq 4) by using only one basis function in the Gauss−Hermite basis set. The wave function has now a product-type form and becomes

$$\Psi(R,r,t) = \psi_0(R,t)\sum_{nk} a_{nk}(t)\varphi_n(r) \tag{6}$$

where $\psi_0(R,t)$, the "ground state" of the Gauss-Hermite basis set, is an ordinary Gaussian wave packet:

$$\psi_0(R,t) = \exp\left[\frac{i}{\hbar}(\gamma(t) + P_R(R - R(t)) + A(t)(R - R(t))^2)\right] \tag{7}$$

Inserting eq 6 into the time-dependent Schrödinger equation, one can derive the following equations:

$$i\hbar\dot{a}_n(t) = E_n a_n(t) + \sum_m V_{nm}(R(t))a_m(t) \tag{8}$$

$$\dot{P}_R(\mathrm{t}) = -\sum_{nm} a_n^*(t)a_m(t)\frac{\partial V_{nm}(R)}{\partial R}\bigg|_{R=R(t)} \tag{9}$$

$$\dot{R} = P_R(t)/\mu \tag{10}$$

$$\dot{A}(t) = -\frac{2}{\mu}A(t)^2 - \frac{1}{2}\sum_{nm} a_n^*(t)a_m(t)\,\frac{\partial^2 V_{nm}(R)}{\partial R^2}\bigg|_{R=R(t)} \tag{11}$$

$$\dot{\gamma}(t) = \frac{P_R(t)^2}{2\mu} + \frac{i\hbar A(t)}{\mu} - V(R(t)) \tag{12}$$

where the matrix element V_{nm} is defined by

$$V_{nm}(t) = \langle\varphi_n|V(R, r)|\varphi_m\rangle \tag{13}$$

Equations 8−10 are identical to eqs 1−3. The classical path equations have been obtained together with two equations, 11 and 12, for the width $A(t)$ and the phase $\gamma(t)$ of the Gaussian wave packet. Thus, the classical path method has been recovered from a general quantum approach by making a separability approximation between r and R throughout the whole propagation and assuming a particular "shape" (a Gaussian wave packet) for the wave function of the collision coordinate (eq 6). This explains why the classical path method is especially successful when used, for example, in the investigation of energy transfer processes, where the coupling between classical (translation and rotations) and quantum (vibrations) degrees of freedom is often very weak and their separability can be assumed.

Quantum Dressed Classical Mechanics

Billing's adventure in searching for accurate classical path theories ultimately led him to the quantum dressed classical mechanics, which is a rigorous quantum theory that has the appealing feature of having classical mechanics as one limit. The method could be called the "time dependent Gauss−Hermite discrete variable representation"; however, the less formal "quantum dressed classical mechanics", pointing out that quantum corrections to classical mechanics can be obtained by adding grid points around the classical trajectory, immediately suggests the flavor of the theory.

Although many of the ingredients of this method, i.e., the use of the Gauss−Hermite basis set, of a discrete variable representation, and of time-dependent grids, had already been used in Billing's other works (as described above), the complete formulation differs in many important details and leads to a very compact, general, and flexible approach to dynamics. An accurate and complete description of the method, including its mathematical derivation, the underlying philosophy, and differences with other approaches can be found in refs 227 and 228.

The scheme is in effect a development of the earlier Gauss−Hermite derivation,[229−232] partly illustrated in the above section. As suggested by its name, it can be obtained by expanding the wave function in a Gauss−Hermite basis set and then switching to a discrete variable representation in the solution of the equations for the amplitudes of the Gauss−Hermite basis functions.[233,234] As shown above, the use of a Gauss−Hermite basis set leads to classical-like equations (eqs 9−12) for the evolution in time of the parameters of the Gaussian wave packet, where not only the initial parameters of the Gaussian wave packet but also the expression for the effective forces, V' and V'', driving the packet are undefined. However, it turns out that if the basis set is complete (i.e., an infinite number of wave functions is used), those forces vanish. If the basis set is truncated, we can instead expect some advantage by letting it follow the dynamical evolution of the system, by using, for instance, classical forces, or more accurate generalized forces obtained through quantum corrections to the classical ones. The solution of the problem is in any case independent of the effective potential. Only the convergence pattern is affected, in terms of number of basis functions. So in the simplest and most useful scheme classical forces can be used to propagate the trajectory.

The introduction of a DVR procedure into this scheme allows one to operate with quantum amplitudes at grid points rather than with amplitudes for the basis functions. Classical mechanics arises then in this representation naturally as the limit where only one grid point is used for a given degree of freedom. The grid points follow the classical dynamics in time and explore the space around the trajectories, and since each grid point has a quantum amplitude associated with it, the method allows for quantum delocalization, tunneling, and other quantum phenomena.

The method is accurate and it does converge to the exact quantum result if enough grid points are used, while, in the other limit, classical mechanics is recovered. In this way it is possible to gather grid points only where needed, i.e., only in those degrees of freedom showing quantum behavior. Intrinsic in the formulation is the possibility to monitor the behavior (more or less quantum-like) of each degree of freedom during the propagation and to add accordingly more grid points. Since the parameters required for the classical propagation are the same as those needed by conventional classical dynamics, the method is extremely well suited for use with ab initio electronic structure calculations, providing a systematic way to correct for nuclear quantum effects even in large systems. Furthermore, the scheme is very compact and easy to program.[235]

As is often the case for Gert Billing's methods, quantum dressed classical mechanics has found a very wide range of applications, from simple, prototype cases[236] involving inelastic[234] and reactive collisions,[237−239] to complex systems,[240] with the inclusion of the explicit treatment of nonadiabatic effects[240,241] and molecule−surface scattering.[242]

In his last book *The Quantum Classical Theory*,[243] Gert presented a remarkable collection of the quantum classical methods he developed, together with their derivations and detailed descriptions. Starting from the formulation of rigorous theories (as quantum dressed classical mechanics) from first principles, this text shows how they can be used to derive some very useful approximate schemes and when they can be successfully employed. In this sense this book constitutes the "summit" of his research and a legacy for scientists in this field.

Nonadiabatic Processes and Geometric Phase Effects

Some time in the middle of the nineties (∼1995) Gert extended the application of his time-dependent quantum classical approach to treat electronic nonadiabatic processes coupled with reactive scattering. He started by considering the effect of spin−orbit coupling on the reaction[160] $F(^2P_{3/2},^2P_{1/2})+D_2 \rightarrow DF+D$ and then moved one step further to study the $(Ar+H_2)^+$ system.[158] This study, which was carried out in collaboration with the theoretical group in the Laboratoire des Collision Atomique-Moleculaire (LCAM) at the Paris-sud University in Orsay, considers the three competing processes that take place during the collision of Ar^+ and H_2 or Ar and H_2^+; namely, the exchange (reactive) process, the charge-transfer process, and the (3/2↔1/2) spin transitions. The treatment of these three competing processes is a challenge for any approach, and indeed Gert's quantum classical approach yields state-to-state cross-sections that fit both the experimental results as well as the quantum mechanical cross sections.

At a later stage Gert, as the curious scientist that he was, started looking for new challenges and got interested in the Berry phase and its relevance for molecular systems. He is one of the (very) few scientists who tried to address this phenomenon classically[244−246] and semiclassically[247] and later became also deeply involved in the quantum mechanical aspects of this issue.[248−256] This activity culminated in the editing (together with M. Baer) of a book *The Role of Degenerate States in Chemistry*, which was published in *Advances of Chemical Physics* (Vol. 124, 2002).

References and Notes

(1) Billing Sørensen, G. *Chem. Phys. Lett.* **1972**, *14*, 274.
(2) Billing, Sørensen, G. *J. Chem. Phys.* **1972**, *57*, 5241.
(3) Billing, Sørensen, G. *J. Chem. Phys.* **1974**, *61*, 3340.
(4) Billing Sørensen, G. *Chem. Phys.* **1974**, *5*, 244.
(5) Billing, G. D. *J. Chem. Phys.* **1975**, *62*, 1480.
(6) Bohr, N. *Mater. Phys. Medd. Dan. Vidensk, Selsk.* **1948**, *18*, 8.
(7) Rapp, D.; Kassal, T. *Chem. Rev.* **1969**, *69*, 61.
(8) Billing, G. D. *Chem. Phys. Lett.* **1975**, *30*, 391.
(9) Billing, G. D. *J. Chem. Phys.* **1976**, *64*, 908.
(10) Billing Sørensen, G. D. *J. Chem. Phys.* **1973**, *59*, 6147.
(11) Billing, G. D. *Chem. Phys. Lett.* **1983**, *100*, 535.
(12) Billing, G. D. *J. Chem. Phys.* **1993**, *99*, 5849.
(13) Billing, G. D. *Chem. Phys.* **1979**, *41*, 11.
(14) Billing, G. D. *Chem. Phys.* **1980**, *46*, 123.
(15) Billing, G. D. *Chem. Phys.* **1980**, *49*, 255.
(16) Billing, G. D. *Chem. Phys.* **1980**, *51*, 417.
(17) Billing, G. D. *Chem. Phys.* **1981**, *60*, 199.
(18) Billing, G. D. *Chem. Phys.* **1981**, *61*, 415.
(19) Billing, G. D. *Chem. Phys.* **1982**, *64*, 35.
(20) Billing, G. D. *Chem. Phys.* **1983**, *76*, 315.
(21) Billing, G. D.; Clary, D. C. *Chem. Phys.* **1983**, *80*, 213.
(22) Billing, G. D. *Chem. Phys.* **1983**, *79*, 179.
(23) Billing, G. D. *Chem. Phys.* **1984**, *91*, 327.
(24) Billing, G. D. *Chem. Phys.* **1986**, *104*, 19.
(25) Billing, G. D. *Chem. Phys.* **1988**, *127*, 107.
(26) Billing, G. D. *Chem. Phys.* **1993**, *173*, 167.
(27) Nyland, C.; Billing, G. D. *Chem. Phys.* **1978**, *30*, 401.
(28) Nyland, C.; Billing, G. D. *Chem. Phys.* **1979**, *40*, 103.
(29) Nyland, C.; Billing, G. D. *Chem. Phys.* **1981**, *60*, 359.
(30) Billing, G. D. *Chem. Phys. Lett.* **1983**, *97*, 188.
(31) Billing, G. D. *Comput. Phys. Comm.* **1984**, *32*, 45.
(32) Billing, G. D. *Comput. Phys. Rep.* **1984**, *1*, 237.
(33) Billing, G. D. *Comput. Phys. Comm.* **1987**, *44*, 121.
(34) Nyland, C.; Billing, G. D. *J. Phys. Chem.* **1988**, *92*, 1752.
(35) Billing, G. D.; Jolicard, G. *Chem. Phys.* **1982**, *65*, 323.
(36) Henriksen, N. H.; Hansen, F. Y.; Billing, G. D. *Chem. Phys. Lett.* **1992**, *199*, 186.
(37) Billing, G. D. *Chem. Phys.* **1975**, *9*, 359.
(38) Billing, G. D. *Chem. Phys.* **1977,** *20*, 35.
(39) Billing, G. D. *Chem. Phys. Lett.* **1977**, *50*, 320.
(40) Billing, G. D.; Fisher, E. R. *Chem. Phys.* **1976**, *18*, 225.
(41) Cacciatore, M.; Capitelli, M.; Billing, G. D. *Chem. Phys. Lett.* **1989**, *157*, 305.

(42) Cacciatore, M.; Caporusso, R.; Billing, G. D. *Chem. Phys. Lett.* **1992**, *197*, 92.

(43) Kolesnick, R. E.; Billing, G. D. *Chem. Phys.* **1993**, *170*, 201.

(44) Billing, G. D.; Kolesnick, R. E. *Chem. Phys. Lett.* **1993**, *215*, 571.

(45) Zenevitch, V. A.; Billing, G. D. *J. Chem. Phys.* **1999**, *111*, 2401.

(46) Zenevitch, V. A.; Billing, G. D.; Jolicard, G. *Chem. Phys. Lett.* **1999**, *312*, 530. Zenevitch, V. A.; Billing, G. D.; Jolicard, G. *Mol. Phys.* **2000**, *98*, 1691.

(47) Billing, G. D. *Chem. Phys. Lett.* **1980**, *75*, 254.

(48) Billing, G. D. *Chem. Phys.* **1976**, *14*, 267.

(49) Billing, G. D. *Chem. Phys.* **1978**, *30*, 387.

(50) Poulsen, L. L.; Billing, G. D.; Steinfield, J. I. *J. Chem. Phys.* **1978**, *68*, 5121.

(51) Billing, G. D.; Poulsen, L. L. *J. Chem. Phys.* **1978**, *68*, 5128.

(52) Poulsen, L. L.; Billing, G. D. *Chem. Phys.* **1979**, *36*, 271.

(53) Poulsen, L. L.; Billing, G. D. *Chem. Phys.* **1980**, *45*, 287.

(54) Poulsen, L. L.; Billing, G. D. *Chem. Phys.* **1980**, *53*, 389.

(55) Billing, G. D. *Chem. Phys.* **1987**, *112*, 95.

(56) Billing, G. D.; Zenevich, V.; Lindinger, W. *J. Chem. Phys.* **1992**, *97*, 3274.

(57) Billing, G. D. *J. Chem. Phys.* **1986**, *84*, 2593.

(58) Billing, G. D. *Chem. Phys.* **1980**, *50*, 165.

(59) Gorse, C.; Billing, G. D.; Cacciatore, M.; Capitelli, M.; de Benedictis, S. *Chem. Phys.* **1987**, *111*, 351.

(60) Cacciatore, M.; Capitelli, M.; Billing, G. D. *Chem. Phys.* **1984**, *89*, 17.

(61) Kurnosov, A.; Cacciatore, M.; Billing, G. D. *J. Phys. Chem.* **2003**, *107*, 2403.

(62) Billing, G. D.; Fisher, E. R. *Chem. Phys.* **1980**, *45*, 359.

(63) Capitelli, M.; Gorse, C.; Billing, G. D. *Chem. Phys.* **1980**, *52*, 299.

(64) Nyeland, C.; Poulsen, L. L.; Billing, G. D. *J. Phys. Chem.* **1984**, *88*, 1216.

(65) Billing, G. D. *Chem. Phys. Lett.* **1980**, *76*, 178.

(66) Billing, G. D.; Wang, L. *J. Phys. Chem.* **1992**, *96*, 2572.

(67) Cacciatore, M.; Billing, G. D. *Chem. Phys.* **1981**, *58*, 395.

(68) Billing, G. D.; Cacciatore, M. *Chem. Phys. Lett.* **1983**, *94*, 218.

(69) Cacciatore, M.; Billing, G. D. *Chem. Phys. Lett.* **1985**, *121*, 94.

(70) Balakrishnan, N.; Billing, G. D. *Chem. Phys.* **1996**, *204*, 77.

(71) Coletti, C.; Billing, G. D. *J. Chem. Phys.* **1999**, *111*, 3891.

(72) Coletti, C.; Billing, G. D. *J. Chem. Phys.* **2000**, *113*, 4869.

(73) Billing, G. D.; Coletti, C.; Kurnosov, A. K.; Napartovich, A. P. *J. Phys. B* **2003**, *36*, 1175.

(74) Billing, G. D. *Chem. Phys. Lett.* **1982**, *89*, 337.

(75) Billing, G. D.; Cacciatore, M. *Chem. Phys. Lett.* **1982**, *86*, 20.

(76) Cacciatore, M.; Capitelli, M.; Billing, G. D. *Chem. Phys.* **1983**, *82*, 1.

(77) Price, R. J.; Clary, D. C.; Billing, G. D. *Chem. Phys. Lett.* **1983**, *101*, 269.

(78) Jolicard, G.; Billing, G. D. *Chem. Phys.* **1984**, *85*, 253.

(79) Billing, G. D. *Chem. Phys.* **1986**, *107*, 39.

(80) Billing, G. D.; Poulsen, L. L. *Chem. Phys.* **1982**, *70*, 119.

(81) Billing, G. D.; Poulsen, L. L. *Chem. Phys. Lett.* **1983**, *99*, 368.

(82) Billing, G. D.; Clary, D. C. *Chem. Phys. Lett.* **1982**, *90*, 27.

(83) Billing, G. D. *Chem. Phys. Lett.* **1985**, 117, 145.

(84) Poulsen, L. L.; Billing, G. D. *Chem. Phys.* **1982**, *73*, 313.

(85) Poulsen, L. L.; Billing, G. D. *Chem. Phys.* **1984**, *89*, 219.

(86) Billing, G. D.; Poulsen, L. L.; Dieckersen, G. H. F. *Chem. Phys.* **1985**, *98*, 397.

(87) Billing, G. D.; Dieckersen, G. H. F. *Chem. Phys.* **1987**, *118*, 161.

(88) Billing, G. D.; Poulsen, L. L. *J. Chem. Phys.* **1984**, *81*, 3866.

(89) Billing, G. D.; Diercksen, G. H. F. *Chem. Phys. Lett.* **1985**, *121*, 94.

(90) Billing, G. D.; Diercksen, G. H. F. *Chem. Phys.* **1986**, *105*, 145.

(91) Billing, G. D.; Diercksen, G. H. F. *Chem. Phys.* **1987**, *118*, 161.

(92) Broquier, M.; Picard-Bersellini, A.; Aroui, H.; Billing, G. D. *J. Chem. Phys.* **1988**, *88*, 1551.

(93) Billing, G. D.; Diercksen, G. H. F. *Chem. Phys.* **1988**, *124*, 77.

(94) Aroui, H.; Broquier, M.; Chevalier, M.; Billing, G. D. *Mol. Phys.* **1991**, *74*, 897.

(95) Aroui, H.; Picard-Bersellini, A.; Broquier, M.; Blanchard, S.; Billing, G. D. *J. Chem. Phys.* **1988**, *89*, 5373.

(96) Broquier, M.; Lefevre, G.; Pittman, A.; Picard-Bersellini, A.; Chevalier, M.; Billing, G. D. *Mol. Phys.* **1993**, *79*, 1103.

(97) Billing, G. D.; Clary, D. C. *Chem. Phys.* **1989**, *135*, 91.

(98) Billing, G. D. *Chem. Phys.* **1979**, *36*, 127.

(99) Billing, G. D. *J. Chem. Soc., Faraday Trans.* **1990**, *86*, 1663.

(100) Zenevich, V.; Lindinger, W.; Billing, G. D. *J. Chem. Phys.* **1992**, *97*, 7257.

(101) Zenevich, V.; Lindinger, W.; Billing, G. D. *Chem. Phys. Lett.* **1992**, *197*, 99.

(102) Gianturco, F. A.; Serna, S.; Palma, A.; Billing, G. D.; Zenevich, V. *J. Phys. B* **1993**, *26*, 1839.

(103) Billing, G. D.; Kolesnick, R. E. *Chem. Phys. Lett.* **1992**, *200*, 382.

(104) Wang, L.; Billing, G. D. *J. Phys. Chem.* **1993**, *97*, 2523.

(105) Balakrishnan, N.; Dalgarno, A.; Billing, G. D. *Chem. Phys. Lett.* **1998**, *288*, 657.

(106) Coletti, C.; Billing, G. D. *Chem. Phys. Lett.* **2002**, *356*, 14.

(107) Billing, G. D. *Chem. Phys.* **1994**, *179*, 463.

(108) Sevryuk, M. B.; Billing, G. D. *Chem. Phys.* **1994**, *185*, 199.

(109) Gross, A.; Billing, G. D. *Chem. Phys.* **1996**, *202*, 321.

(110) Hansel, A.; Oberhofer, N.; Lindinger, W.; Zenevich, V. A.; Billing, G. D. *Int. J. Mass Spectrosc. And Ion Processes* **1999**, *185*, 559.

(111) Zenevich, V. A.; Pogrebnya, S. K.; Lindinger, W.; Cacciatore, M.; Billing, G. D. *J. Chem. Phys.* **1995**, *102*, 6669.

(112) Billing, G. D.; Wang, L. *Chem. Phys. Lett.* **1992**, *168*, 315.

(113) Goubert, L.; Billing, G. D.; Desoppere, E.; Wieme, W. *Chem. Phys. Lett.* **1994**, *219*, 360.

(114) Billing, G. D. *Chem. Phys.* **1984**, *89*, 199.

(115) Billing, G. D. *Chem. Phys.* **1989**, *135*, 423.

(116) Billing, G. D. *Chem. Phys.* **1990**, *146*, 63.

(117) Billing, G. D. *Chem. Phys.* **1992**, *159*, 109.

(118) Billing, G. D. *Chem. Phys.* **1992**, *161*, 245.

(119) Billing, G. D. *Trends Chem. Phys.* **1991**, *1*, 133.

(120) Muckerman, J. T.; Gilbert, R. D.; Billing, G. D. *J. Chem. Phys.* **1988**, *88*, 4779.

(121) Billing, G. D.; Muckerman, J. T. *J. Chem. Phys.* **1989**, *91*, 6830.

(122) Billing, G. D.; Muckerman J. T. *Int. J. Quantum Chem.* **1989**, *135*, 423.

(123) Markovic, N.; Billing, G. D.; Muckerman, J. T. *Chem. Phys. Lett.* **1990**, *172*, 509.

(124) Markovic, N.; Billing, G. D. *Chem. Phys. Lett.* **1992**, *159*, 53.

(125) Markovic, N.; Billing, G. D. *Chem. Phys.* **1993**, *173*, 385.

(126) Billing, G. D.; Markovic, N. *J. Chem. Phys.* **1993**, *99*, 2674.

(127) Billing, G. D. *J. Chem. Phys.* **1997**, *107*, 4286.

(128) Billing, G. D. *Chem. Phys.* **1994**, *189*, 523.

(129) Billing, G. D. In *Encyclopedia of Computational Chemistry*; Schleyer, P. v. R., Ed.; Wiley: New York 1988; p 1587.

(130) Adhikari, S.; Billing, G. D. *Chem. Phys.* **1998**, *238*, 69.

(131) Adhikari, S.; Billing, G. D. *Chem. Phys. Lett.* **1999**, *305*, 109.

(132) Markovic, N.; Billing, G. D. *J. Chem. Phys.* **1994**, *100*, 1085.

(133) Markovic, N.; Billing, G. D. *Chem. Phys.* **1995**, *191*, 247.

(134) Markovic, N.; Billing, G. D. *Chem. Phys. Lett.* **1996**, *248*, 420.

(135) Billing, G. D.; Markovic, N. *Chem. Phys.* **1996**, *209*, 377.

(136) Markovic, N.; Billing, G. D. *Chem. Phys.* **1997**, *224*, 53.

(137) Hansen, F. Y.; Henriksen, N. H.; Billing, G. D. *J. Chem. Phys.* **1989**, *90*, 3060.

(138) Billing, G. D. *Mol. Phys.* **1996**, *89*, 355.

(139) Billing, G. D. *J. Chem. Soc., Faraday Trans.* **1997**, *93*, 833.

(140) Koch, A.; Billing, G. D. *J. Chem. Phys.* **1997**, *107*, 7242.

(141) Coletti, C.; Billing, G. D. *Phys. Chem. Chem. Phys.* **1999**, *1*, 4141.

(142) Balakrishnan, N.; Billing, G. D. *J. Chem. Phys.* **1996**, *104*, 9481.

(143) Billing, G. D. *Chem. Phys.* **2002**, *277*, 325.

(144) Balakrishnan, N.; Billing, G. D. *Chem. Phys.* **1994**, *189*, 499.

(145) Balakrishnan, N.; Billing, G. D. *Chem. Phys. Lett.* **1995**, *233*, 145.

(146) Balakrishnan N.; Billing, G. D. *J. Chem. Phys.* **1994**, *101*, 2785.

(147) Billing, G. D.; Mikkelsen, K. V. *Chem. Phys.* **1994**, *182*, 249.

(148) Nymand, T.; Mikkelsen, K. V.; Astrand, P.-O.; Billing, G. D. *Acta Chem. Scand.* **1999**, *53*, 1043.

(149) Balakrishnan, N.; Billing, G. D. *Chem. Phys. Lett.* **1995**, *244*, 68.

(150) Gross A.; Billing, G. D. *Chem. Phys.* **1993**, *173*, 393.

(151) Gross A.; Billing, G. D. *Chem. Phys.* **1994**, *187*, 329.

(152) Aguillon, F.; Sizun, M.; Sidis, V.; Billing, G. D.; Markovic, N. *J. Chem. Phys.* **1996**, *104*, 4530.

(153) Balakrishnan, N.; Billing, G. D. *J. Chem. Phys.* **1996**, *104*, 4005.

(154) Billing, G. D.; Muckerman, J. T.; Yu, H. G. *J. Chem. Phys.* **2002**, *117*, 4755.

(155) Sizun, M.; Aguillon, F.; Sidis, V.; Zenevich, V.; Billing, G. D.; Markovic, N. *Chem. Phys.* **1996**, *209*, 327.

(156) Gross A.; Billing, G. D. *Chem. Phys.* **1997**, *217*, 1.

(157) Coletti, C.; Billing, G. D. *J. Chem. Phys.* **2000**, *113*, 11101.

(158) Vijay, A.; Billing, G. D. *J. Chem. Phys.* **1997**, *107*, 2974.

(159) Azriel, V. M.; Billing, G. D.; Rusin, L. Yu.; Sevryuk, M. B. *Chem. Phys.* **1995**, *195*, 243.

(160) Billing, G. D.; Rusin, L. Yu.; Sevryuk, M. B. *J. Chem. Phys.* **1995**, *103*, 2482.

(161) Markovic, N.; Billing, G. D. *Mol. Phys.* **2000**, *98*, 1771.

(162) Billing, G. D.; Jolicard G. *J. Phys. Chem.* **1984**, *88*, 1820.

(163) Guldberg, A.; Billing, G. D. *Chem. Phys. Lett.* **1991**, *186*, 229.

(164) Balakrishnan, N.; Billing, G. D. *J. Chem. Phys.* **1994**, *101*, 2968.

(165) Kroes, G.-J.; van Hemert, M. C.; Marc, C.; Billing, G. D.; Neuhauser, D. *J. Chem. Phys.* **1997**, *107*, 5757.

(166) Kroes, G.-J.; van Hemert, M. C.; Billing, G. D.; Neuhauser, D. *Chem. Phys. Lett.* **1997**, *271*, 311.

(167) Billing, G. D. *Chem. Phys. Lett.* **1986**, *127*, 269.

(168) Billing, G. D. *Chem. Phys.* **1987**, *115*, 229.

(169) Billing, G. D.; Henriksen, N. E. *Chem. Phys.* **1988**, *119*, 205.

(170) Billing, G. D.; Henriksen, N. E.; Leforestier, C. *Phys. Rev. A* **1992**, *45*, R4229.

(171) Guldberg, A.; Billing, G. D. *Chem. Phys. Lett.* **1992**, *191*, 455.

(172) Johnson, M. S.; Billing, G. D.; Groudis, A.; Janssen, M. H. M. *J. Phys. Chem.* **2001**, *105*, 8672.

(173) Billing, G. D. *Int. Rev. Phys. Chem.* **1994**, *13*, 309.

(174) Billing, G. D. *Chem. Phys.* **1982**, *70*, 223.

(175) Billing, G. D. *Surf. Sci.* **1988**, *195*, 187.

(176) Billing, G. D. *J. Phys. Chem.* **1995**, *99*, 15378.

(177) Billing, G. D. *Comput. Phys. Rep.* **1990**, *12*, 383.

(178) Billing, G. D. *Dynamics of Molecule-Surface Interactions*; John Wiley & Sons: New York, 1999.

(179) Billing, G. D. *Chem. Phys.* **1987**, *116*, 269.

(180) Billing, G. D. *Surf. Sci.* **1988**, *203*, 257.

(181) Rakovshik, M.; Billing, G. D. *Chem. Phys. Lett.* **1991**, *185*, 1.

(182) Billing, G. D. *J. Chem. Phys.* **2000**, *112*, 335.

(183) Mengel, S. K.; Billing, G. D. *J. Phys. Chem.* **1997**, *101*, 10781.

(184) Laursen, C. M.; Christoffersen, E.; Billing, G. D. *Chem. Phys. Lett.* **1998**, *290*, 275.

(185) Billing, G. D. *Chem. Phys. Lett.* **1998**, *290*, 150.

(186) Billing, G. D. *Plasma Phys. Rep.* **1999**, *25*, 32.

(187) Billing, G. D. *Chem. Phys.* **1983**, *74*, 143.

(188) Billing, G. D. *Chem. Phys.* **1984**, *86*, 349.

(189) Wang, L.; Ge, Q.; Billing, G. D. *Surf. Sci.* **1994**, *304*, L413.

(190) Billing, G. D.; Cacciatore, M. *Chem. Phys. Lett.* **1985**, *113*, 23.

(191) Billing, G. D.; Cacciatore, M. *Chem. Phys.* **1986**, *103*, 137.

(192) Cacciatore, M.; Capitelli, M.; Billing, G. D. *Surf. Sci.* **1989**, *217*, L391.

(193) Cacciatore, M.; Billing, G. D. *Surf. Sci.* **1990**, *232*, 35. Erratum **1990**, *238*, 350.

(194) Wang, L.; Ge, Q.; Billing, G. D. *Surf. Sci.* **1994**, *301*, 353.

(195) Cacciatore, M.; Billing, G. D. *Pure Appl. Chem.* **1996**, *68*, 1075.

(196) Freiesleben Hansen, B.; Billing, G. D. *Surf. Sci. Lett.* **1997**, *373*, 333.

(197) Henriksen, N. E.; Billing, G. D.; Hansen, F. Y. *Surf. Sci.* **1990**, *227*, 224.

(198) Billing, G. D.; Guldberg, A.; Henriksen, N. E.; Hansen, F. Y. *Chem. Phys.* **1990**, *147*, 1.

(199) Ge, Q.; Wang, L.; Billing, G. D. *Surf. Sci.* **1992**, *277*, 237.

(200) Billing, G. D.; Cacciatore, M. *Faraday Discuss.* **1993**, *96*, 33.

(201) Tully, C.; Billing, G. D. *Chem. Phys. Lett.* **2002**, *365*, 530.

(202) Henriksen, N. E.; Hansen, F. Y.; Billing, G. D. *Chem. Phys. Lett.* **1995**, *244*, 350.

(203) Billing, G. D. *Appl. Surf. Sci.* **1999**, *142*, 7.

(204) Cacciatore, M.; Rutigliano, M.; Billing, G. D. *J. Thermophys. Heat Transfer* **1999**, *13*, 195.

(205) Wang, L.; Billing, G. D. *Chem. Phys.* **1997**, *224*, 65.

(206) Rutigliano, M.; Cacciatore, M.; Billing, G. D. *Chem. Phys. Lett.* **2001**, *340*, 13.

(207) Hansen, F. Y.; Henriksen, N. H.; Billing, G. D.; Guldberg, A. *Surf. Sci.* **1992**, *264*, 225.

(208) Henriksen, N. H.; Billing, G. D.; Hansen, F. Y. *J. Phys. Chem.* **1992**, *96*, 223.

(209) Hansen, F. Y.; Henriksen, N. H.; Billing, G. D. *Surf. Sci.* **1995**, *324*, 55.

(210) Billing, G. D. *Chem. Phys.* **1978**, *33*, 227.

(211) Billing, G. D. *Chem. Phys.* **1999**, *242*, 341.

(212) Billing, G. D. *Phys. Chem. Chem. Phys.* **1999**, *1*, 4687.

(213) Adhikari, S.; Billing, G. D. *J. Chem. Phys.* **2000**, *112*, 3884.

(214) Jolicard, G.; Billing, G. D. *Chem. Phys.* **1982**, *64*, 123.

(215) Billing, G. D.; Jolicard, G. *Chem. Phys. Lett.* **1983**, *102*, 491.

(216) Jolicard, G.; Billing, G. D. *Chem. Phys.* **1986**, *104*, 357.

(217) Jolicard, G.; Billing, G. D. *J. Chem. Phys.* **1992**, *97*, 997.

(218) Billing, G. D.; Jolicard, G. *Chem. Phys. Lett.* **1994**, *221*, 75.

(219) Jolicard, G.; Billing, G. D. *Chem. Phys. Lett.* **1985**, *119*, 162.

(220) Jolicard, G.; Billing, G. D. *J. Chem. Phys.* **1989**, *90*, 346.

(221) Billing, G. D.; Jolicard, G. *Chem. Phys. Lett.* **1989**, *155*, 521.

(222) Jolicard, G.; Billing, G. D. *J. Phys. B* **1990**, *23*, 3457.

(223) Jolicard, G.; Billing, G. D. *Chem. Phys.* **1991**, *149*, 261.

(224) Jolicard, G.; Tuckey, P. A.; Billing, G. D. *J. Chem. Phys.* **1997**, *107*, 6290.

(225) Jolicard, G.; Billing, G. D. *J. Chem. Phys.* **1994**, *101*, 9429.

(226) Billing, G. D. *Introduction to the theory of inelastic collisions in chemical kinetics*, Doctoral Thesis, University of Copenhagen, 1978.

(227) Billing, G. D. *J. Chem. Phys.* **2001**, *114*, 6641.

(228) Billing, G. D. *Phys. Chem. Chem. Phys.* **2002**, *4*, 2865.

(229) Billing, G. D. *J. Chem. Phys.* **1999**, *110*, 5526.

(230) Adhikari, S.; Billing, G. D. *Chem. Phys. Lett.* **1999**, *309*, 249.

(231) Adhikari, S.; Billing, G. D. *J. Chem. Phys.* **1999**, *111*, 48.

(232) Billing, G. D. *J. Mol. Struct. (THEOCHEM)* **2000**, *501−502*, 519.

(233) Adhikari, S.; Billing, G. D. *J. Chem. Phys.* **2000**, *113*, 1409.

(234) Billing, G. D. *Chem. Phys.* **2001**, *264*, 71.

(235) Billing, G. D. *Chem. Phys. Lett.* **2001**, *339*, 237.

(236) Adhikari, S.; Billing, G. D. *J. Chem. Phys.* **2000**, *113*, 1409.

(237) Billing, G. D. *Int. J. Quantum Chem.* **2001**, *84*, 467.

(238) Coletti, C.; Billing, G. D. *Chem. Phys. Lett.* **2001**, *342*, 65.

(239) Feilberg, K. L.; Billing, G. D.; Johnson, M. S. *J. Phys. Chem. A* **2001**, *105*, 11171.

(240) Coletti, C.; Billing, G. D. *Chem. Phys. Lett.* **2003**, *368*, 289.

(241) Billing, G. D. *Chem. Phys. Lett.* **2001**, *343*, 130.

(242) Billing, G. D. *J. Phys. Chem. A* **2001**, *105*, 2340.

(243) Billing, G. D. *The Quantum Classical Theory*; Oxford University Press: New York, 2003.

(244) Adhikari, S.; Billing, G. D. *Chem. Phys. Lett.* **1998**, *284*, 31.

(245) Adhikari, S.; Billing, G. D. *Chem. Phys. Lett.* **1998**, *289*, 219.

(246) Adhikari, S.; Billing, G. D. *J. Chem. Phys.* **1997**, *107*, 6213.

(247) Adhikari, S.; Billing, G. D. *J. Chem. Phys.* **1999**, *111*, 40.

(248) Billing, G. D.; Kupperman, A. *Chem. Phys. Lett.* **1998**, *294*, 26.

(249) Baer, M.; Lin, H. S.; Alijah, A.; Adhikari, S.; Billing, G. D. *Phys. Rev. A* **2000**, *62*, 32506-1.

(250) Adhikari, S.; Billing, G. D.; Alijah, A.; Lin, H. S.; Baer, M. *Phys. Rev. A* **2000**, *62*, 32507-1.

(251) Adhikari, S.; Billing, G. D. *Chem. Phys.* **2000**, *259*, 149.

(252) Baer, M.; Billing, G. D. *J. Phys. Chem. A* **2001**, *105*, 2509.

(253) Baer, M.; Mebel, M.; Billing, G. D. *J. Phys. Chem. A* **2002**, *106*, 6499.

(254) Avery, J.; Baer, M.; Billing, G. D. *Mol. Phys.* **2002**, *100*, 1011.

(255) Baer, M.; Mebel, M.; Billing, G. D. *Int. J. Quantum Chem.* **2002**, *90*, 1577.

(256) Adhikari, S.; Billing, G. D. In *Advances in Chemical Physics*; Billing, G. D., Baer, M., Eds.; Wiley: New York, 2000.