CHAPTER 4

The Development of Computational Chemistry in Canada

Russell J. Boyd

Department of Chemistry, Dalhousie University, Halifax, Nova Scotia, Canada B3H 4J3

INTRODUCTION

The development of computational chemistry in Canada is inextricably linked with the emergence of theoretical chemistry as one of Canada's strongest scientific disciplines. For this reason, the present account chronicles the history of both computational and theoretical chemistry in Canada.

The first theoretical chemists were appointed to Canadian universities in the mid- to late 1950s, long after strong traditions for theoretical chemistry had been established in Europe, the United Kingdom and the United States. Furthermore, many of the first Canadian theoreticians often carried out experiments or maintained experimental laboratories in departments of chemistry in which the dominant figures were typically experimental physical chemists or organic chemists with a penchant for natural products and synthesis.

The late development of theoretical chemistry in Canada was a consequence of the relatively weak research base in Canadian universities prior to 1950. In fact, research in Canadian universities during the first half of the twentieth century lagged seriously behind that of the leading universities of the major industrialized nations. Only McGill University and the University of

Toronto had gained international reputations as major research institutions before the Second World War. Although some research was carried out prior to 1950 at about 20 universities across Canada, the majority of the significant research was done in the laboratories of the National Research Council of Canada (NRC) in Ottawa. From its humble beginnings in 1916, the NRC developed into a research organization which by the 1960s operated some of the premier research laboratories in the world.

Long before Canada developed a reputation in theoretical and computational chemistry, the one Canadian scientist who needed no introduction was Gerhard Herzberg. His book on atomic structure[1] was known to many generations of chemistry students, and his book on diatomic molecules[2] and the later one on polyatomic molecules[3] became standard references for theoretical chemists worldwide.

Herzberg was born in Hamburg, Germany, on December 25, 1904 and died in Ottawa on March 3, 1999. After earning his doctorate in engineering physics from the Technical University of Darmstadt in 1928, he did postdoctoral research at the universities of Göttingen and Bristol. In 1930 he returned to Darmstadt and during the next five years his research in atomic and molecular spectroscopy flourished. As his scientific reputation was growing, he received an invitation from Dr. J. W. J. Spinks to join the Department of Physics at the University of Saskatchewan. During his 10 years in Saskatoon he made major applications of spectroscopy to physics and chemistry. In 1945 he was appointed professor of spectroscopy at the Yerkes Observatory of the University of Chicago. After three years in the United States, Herzberg returned to Canada to take up a position at the National Research Council laboratories in Ottawa. When he was awarded the Nobel Prize in chemistry in 1971, the citation stated,

> Dr. Herzberg is generally considered to be the world's foremost molecular spectroscopist, and his large institute in Ottawa is the undisputed centre for such research.

In 1994 on the occasion of Herzberg's ninetieth birthday, the *Canadian Journal of Physics* published a special issue. The festschrift, edited by Donald Betts and John Coxon of Dalhousie University, contains 72 papers by many of the world's leading spectroscopists and theoretical chemists. In his acknowledgment, Nobel laureate Dudley Herschbach summarized Herzberg's inestimable impact with the following tribute:[4]

> With pleasure we dedicate this paper to Gerhard Herzberg. For us, as for a host of others, he has through his books served as a superb, inspiring mentor. As well as showing us the exhilarating scope of spectroscopic methods, he has taught us to orient our work toward basic questions and

to strive for treatments that take account of complications as simply as possible.

An article entitled simply "G. H." by Boris Stoicheff is recommended to those who are interested in learning more about Herzberg's remarkable career and scientific achievements. Stoicheff's article appeared in the April 1972 special issue of *Physics in Canada*, published in honor of Gerhard Herzberg on the occasion of his receipt of the Nobel Prize.

Another landmark in the history of theoretical chemistry in Canada pertained to funding. To stimulate research in Canadian universities, the NRC established a grants program and awarded research grants on a competitive basis to university researchers. During his presidency (1952–1962), E. W. R. Steacie established the principle that NRC's extramural budgets for grants and fellowships should rise to match the intramural budget ($215 million in 1962–1963). Ultimately the NRC was in the difficult position of having to choose between funding its own laboratories and the peer-reviewed applications from external (university) researchers. To alleviate this conflict of interest, the Canadian government transferred in 1978 the research grants and scholarships programs of the NRC to a new federal granting agency, the Natural Sciences and Engineering Research Council of Canada (NSERC).

Research in Canadian universities expanded rapidly in the 1960s for several reasons. The launching of Sputnik in 1957 by the Soviet Union shocked the Western nations and led to a rapid expansion of the existing universities and the creation of many more. Consequently, funding for research grew dramatically, many young people (including the author) were attracted to science, and universities started to put more emphasis on research. The number of universities offering the Ph.D. degree in science increased from 2 in 1940 to more than 25 in 1970.

With this brief introduction, it is useful to reflect on the state of the development of theoretical chemistry in 1970, little more than a decade after its establishment in Canadian universities.

In 1970 on the occasion of its twenty-fifth anniversary, The Chemical Institute of Canada published a 290-page book entitled *Chemical Canada*. The authors intended the book "to present a general account of the impact of things chemical in Canada up to, and including, the present time." The book consists of three parts, the third part being a 50-page summary of research and development in Canada. The R&D section is organized by topics and devotes only two short paragraphs to theoretical chemistry. These two paragraphs are reproduced here for their interesting historical perspective.[5]

The very rapid increase in the number of theoretical chemists in Canada since 1962 does not reflect the true growth rate, or one which can be expected to continue; the sudden increase in numbers was caused by many

universities hiring their first theoretician. Although there has been a marked reluctance in many chemistry departments to consider theoretical chemistry as a discipline in itself, worthy of being pursued for its own sake, about eight departments having one theoretician have plans to hire another in the near future, and two departments have initiated plans to form centers of theoretical research consisting of about six staff members.

The solution to the problems involved in the theoretical study of the electronic structure of large molecules and reaction rate theory demands very large and very fast computers, larger and faster than those now available in Canada. If these studies are to proceed, greatly expanded computing facilities must be made available. The type of computer envisaged and necessary for these calculations would be of such a large size that Canada could afford only one or two such installations. It is a general feeling that by the establishment of such a central computational facility, Canada could, for a nominal amount, be in the forefront of a number of critical areas in scientific research and development.

The data collected for the 1970 chapter support the authors' statements and their prophetic predictions. Moreover, their comments about computing facilities are still relevant, three decades later.

IN THE BEGINNING THERE WAS QUANTUM CHEMISTRY AND SPECTROSCOPY

The early efforts in quantum chemistry in Canada, as in other countries, were closely connected with electronic spectroscopy. Later the scope expanded to include all forms of spectroscopy. One of the Canadian pioneers whose interests epitomize the history of quantum chemistry is Camille Sandorfy. After graduating from the University of Szeged in his native Hungary and completing a D.Sc. under the supervision of Profs. Louis de Broglie and R. Daudel at the Sorbonne in Paris, Sandorfy took up a postdoctoral fellowship with Dr. R. Norman Jones, a spectroscopist at the National Research Council of Canada laboratories in Ottawa. By the end of 1953, Sandorfy had settled in Montreal, where he began a highly distinguished career, initially as a theoretical chemist and ultimately as a full-time spectroscopist, specializing in the far-ultraviolet and infrared regions. He developed the basis for a very strong group in quantum chemistry that continued to flourish under the leadership of Dennis Salahub and his younger colleagues at the University of Montreal.

In the years 1945–1955 theoretical chemists as well as chemical spectroscopists concentrated their efforts on systems containing π electrons. Such

molecules can be easily treated by simple molecular orbital methods. It was considered a normal procedure in those times to treat the π electrons as a separate quantum chemical problem to which the σ electrons supply a supposedly uniform potential field.

Sandorfy was struck by the artificial character of this approximation. His first target was the series of normal chain saturated hydrocarbons. A preliminary note on his results was published in the *Comptes Rendus de l'Académie des Sciences.*[6] After taking up his position in Montreal, he greatly extended this work by introducing three different approximations, which he called the "C" "CH," and "H" approximations.[7] The first was based only on the atomic orbitals of the carbons, the second used the hybrid sp^3 orbitals of the carbons and the 1s orbitals of the hydrogens, and the third included all the valence atomic orbitals of the carbons and the hydrogens.

These methods appear rather simple, yet they were the starting point of a long evolution. Gilles Klopman, whose research interests at Case-Western Reserve University later turned to modeling bioactive molecules, was the first to use Sandorfy's methods. Kenichi Fukui made extensive use of them in his well-known work on the structures and reactions of saturated hydrocarbons and their derivatives. Fukui added his frontier orbital considerations. Around 1959 the milieu of developments in quantum chemistry contributed to inspire William N. Lipscomb to conceive the extended Hückel method, which was subsequently implemented by Lawrence L. Lohr and Roald Hoffmann.[8a] Soon thereafter, John Pople and his coworkers introduced self-consistent field methods based on the zero-differential overlap approximation.[8b]

The arrival of ab initio programs based on Gaussian-type orbitals and more powerful computers reduced all these developments to historical souvenirs. It should be emphasized, however, that Sandorfy's work constituted the first attempt to go beyond the π-electron approximation, and the first molecular orbital treatment of polyatomic molecules that took into account all valence electrons without using group orbitals.

From among Sandorfy's subsequent theoretical projects, two in particular should be mentioned: the first treatment of the excited states and electronic spectra of saturated hydrocarbons[9] by a new π-electron method of the Pariser–Parr type, and the first attempt to introduce Rydberg orbitals into molecular orbital calculations.[10]

In 1964 Sandorfy and his coworkers initiated an extensive experimental project to explore the electronic absorption spectra of saturated molecules in the far-ultraviolet region. Their first spectra of a series of paraffin hydrocarbons were published in 1967.[11] This paper coincided with an article, also giving spectra of hydrocarbons, by J. Raymonda and W. T. Simpson.[12] While there was general agreement between the recorded spectra of the two groups, the investigators' interpretations were different; it was Sandorfy and his coworkers who put the spectroscopy of the paraffins on the right track by adopting an idea of R. S. Mulliken[13] that related to the Rydberg character of the bands of lowest

frequency found in their spectra. In this case too, a long evolution was forth-coming. The field has been summed up by M. B. Robin[14] and, more concisely, by Sandorfy.[15] Today, after many years of work in several laboratories the spectroscopy of σ-electron systems has advanced to a level previously attainable only for simple π-electron systems; it is possible, in a qualitative manner, to predict the bands that a given organic molecule, saturated or unsaturated, either heteroatomic or not, should possess in the near- or far-ultraviolet regions. Ultraviolet photoelectron spectroscopy helped greatly during this work of exploration.

Camille Sandorfy is acknowledged as the first theoretical chemist to be appointed to the faculty of a department of chemistry in Canada. It is therefore fitting that he was the first theoretician to be honored by a special issue of the *Canadian Journal of Chemistry*. The 750-page special issue for July 1985 contained 117 papers spanning a broad range of topics relating to spectroscopy and theoretical chemistry. The Sandorfy issue is by far the largest issue the *Canadian Journal of Chemistry* has published. It was edited by two of his former Ph.D. students and younger colleagues at the University of Montreal, Gilles Durocher and Dennis Salahub, the former a spectroscopist and the latter a theoretical chemist.

EXPO 67 AND FULLERENES

A key event in the emergence of theoretical chemistry and computational chemistry as strong disciplines in Canada took place in 1965 at the University of Alberta in Edmonton. Fraser Birss, a spectroscopist with a keen interest in theory, and Serafin Fraga, a recent arrival from the Chicago group of Mulliken and Roothaan, organized the first chemistry conference in Canada devoted entirely to theoretical chemistry. Participants in the Alberta Symposium on Quantum Chemistry recall that the main theme of the Edmonton meeting was whether the Hartree–Fock method was good enough. One notable lecture at the conference, often cited and reworded later, was the closing talk by Chris Wahl entitled, "Hartree-Fock Is Here. What's Next?"

While it is impossible to determine the full impact of the Edmonton conference, Ernest Davidson recalls that it was while he was driving back to Seattle through the Canadian Rockies that he got the idea for iterative natural orbitals.[16] Also, Fritz Grein remembers a long discussion with Chris Wahl in the Faculty Club which gave him the idea for the work on MCSCF methods that is cited later in this chapter.

Following a very successful conference in Edmonton, it was decided that a second theoretical chemistry conference would be organized in 1967 in Montreal by Camille Sandorfy (University of Montreal) and Tony Whitehead (McGill University). Montreal provided the perfect venue. Canada was cele-

brating its centennial, and the euphoria from coast to coast exceeded anything Canadians had known previously, or indeed would experience in the following three decades. Nowhere was the excitement greater than in Montreal, where 50 million people flocked to the Expo 67 site on islands in the historic St. Lawrence River. More than 100 pavilions were built to celebrate the theme, *Man and His World,* a title that within a generation would be deemed politically incorrect. The exhibition was testimony to the optimism and faith of Canadians and others in the future, and it celebrated progress and technology. There were pavilions with fantastic shapes, one of which made an indelible impression upon a young spectroscopist and is forever linked to one of the most exciting scientific discoveries of the 1980s.

It is likely that virtually every participant in the Montreal conference visited the United States pavilion at Expo 67. But none could have foreseen that less than 30 years later the 1996 Nobel Prize in chemistry would be awarded for the 1985 discovery of fullerenes by Robert Curl, Harry Kroto, and Richard Smalley.[17] Nor could anyone present have predicted that the name of the third ordered form of carbon was destined to have a Montreal connection. Harry Kroto suggested the name buckminsterfullerene for C_{60} because the giant molecule reminded him of Buckminster Fuller's geodesic dome (Figure 1), which he and his young family had visited during those memorable days in the summer of 1967. The U.S. pavilion, later dubbed Biosphere, became the unofficial symbol of the World's Fair. Thirty years later it still attracts tourists and at a height of more than 200 feet is said to be the largest round structure in the world.

1967 was a remarkable year. The Vietnam War was being fought, Elvis married Priscilla, Che Guevara was assassinated, the hippie movement was beginning, the Six-Day War between Egypt and Israel forced the closing of the
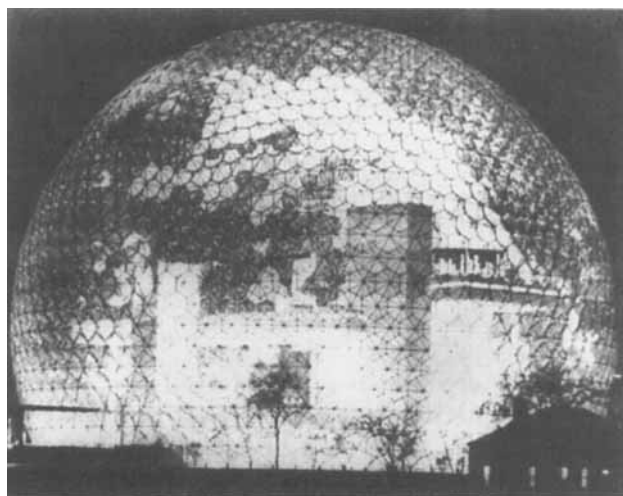
Figure 1 The geodesic dome designed by Buckminster Fuller for the U.S. pavilion at Expo 67 in Montreal.

Suez Canal, Simon and Garfunkel were singing *The Sound of Silence,* the first heart transplant was done in South Africa, and Charles de Gaulle stood on a balcony in Old Montreal and uttered his famous "Vive le Québec libre."

At the Montreal symposium, it was decided to hold the third conference in the series in 1969 at the University of Toronto and to have the conference, like the Montreal meeting, organized by two cochairs from different institutions, a tradition that has been maintained by all subsequent symposia in the series. The Toronto symposium was organized by Richard Bader (McMaster University) and Imre Csizmadia (University of Toronto). Like for the first two symposia, the list of invited speakers included a large number of chemists from abroad. Despite its name, the Canadian Symposium on Theoretical Chemistry is an international conference. Typically about 70% of the invited speakers are from abroad and more than 60% of the participants are from 15 or more countries other than Canada. Attendance is generally in the range of 160–220 registrants.

The fourth symposium, organized by Gulzari Malli (Simon Fraser University) and Bob Snider (University of British Columbia) was held in 1971 on UBC's beautiful campus in Vancouver. Plans for the fifth symposium in the nation's capital, Ottawa, were modified when the First International Congress of Quantum Chemistry was organized in 1973 in Menton, France. David Bishop (University of Ottawa) and Vedene Smith (Queen's University) organized the fifth symposium in Ottawa in 1974. The shift from a biennial to a triennial conference established a natural rotation between the American Conference on Theoretical Chemistry,[18] the Canadian Symposium on Theoretical Chemistry, and the International Congress of Quantum Chemistry. This rotation has continued without interruption for more than two decades.

As indicated in Table 1, the Canadian Symposia on Theoretical Chemistry (CSTC) have continued to flourish more than 30 years after the original conference organized by Fraser Birss and Serafin Fraga. Amid the splendid scenery of the Rockies, it was decided at the Seventh Canadian Symposium on Theoretical Chemistry in 1980 in Banff, Alberta, that the two cochairs should be chosen 6 years in advance. This policy has helped to facilitate future planning and to ensure the continuation of the strong traditions associated with the CSTC. Thus, at the symposium in 1995 in Fredericton, New Brunswick, Fred McCourt (University of Waterloo) and Jim Wright (Carleton University) were chosen to organize the fourteenth symposium in 2001, an auspicious date for computational chemistry.

Although our colleagues in other countries generally have warm memories of what they refer to as the "Canadian Conference," there are very few written records of the CSTC. Indeed, the first published proceedings did not appear until 1984, when André Bandrauk and Russell Boyd arranged to publish 16 of the 36 invited papers presented at the Eighth Canadian Symposium on Theoretical Chemistry in *the Journal of Physical Chemistry.*[19] Similarly, there have been very few accounts written for a more general audience, the one exception being an account[20] of the Halifax meeting. After extolling the plenary lectures

Table 1 Canadian Symposia on Theoretical Chemistry

Number	Location	Year	Cochairs
1	Edmonton, AB	1965	F. W. Birss and S. Fraga
2	Montreal, PQ	1967	C. Sandorfy and M. A. Whitehead
3	Toronto, ON	1969	R. F. W. Bader and I. G. Czismadia
4	Vancouver, BC	1971	G. L. Malli and R. F. Snider
5	Ottawa, ON	1974	D. M. Bishop and V. H. Smith Jr.
6	Fredericton, NB	1977	F. Grein and W. Forst
7	Banff, AB	1980	S. Huzinaga and W. G. Laidlaw
8	Halifax, NS	1983	A. D. Bandrauk and R. J. Boyd
9	Toronto, ON	1986	R. E. Kapral and J. Paldus
10	Banff, AB	1989	B. L. Clarke and A. Rauk
11	Montreal, PQ	1992	D. R. Salahub and B. C. Sanctuary
12	Fredericton, NB	1995	W. J. Meath and A. J. Thakkar
13	Vancouver, BC	1998	G. N. Patey and T. Ziegler
14	Ottawa, ON	2001	F. R. W. McCourt and J. S. Wright

of Gerhard Herzberg and Roald Hoffmann (Figure 2), the 1971 and 1981 Nobel laureates in chemistry, this account noted:

> The technical program for the first day concluded with a late evening poster session. An assortment of liquid refreshments encouraged a high participation rate in the poster session, despite the late hour. Such inaugural benediction on the first day of the conference was, needless to say, the right kind of encouragement for stimulating the atmosphere which prevailed the rest of the week.

Figure 2 André Bandrauk, Roald Hoffmann, Gerhard Herzberg and Russell Boyd at the Eighth Canadian Symposium on Theoretical Chemistry in Halifax.

CANADIAN ASSOCIATION OF THEORETICAL CHEMISTS

At the Seventh Canadian Symposium on Theoretical Chemistry, it was recommended by Bill Laidlaw, and subsequently approved by the approximately 40 persons in attendance at the business meeting, that the chairs of the seventh, eighth, and ninth symposia investigate the possibility of forming a federally incorporated association of theoretical chemists. On the basis of information gathered by Russell Boyd, it was decided at an impromptu meeting on June 1, 1982, in Toronto to apply for incorporation. The meeting was held during the 65th Chemical Conference and Exhibition of the Chemical Institute of Canada and attended by Russell Boyd, Fritz Grein, Ray Kapral, Bill Laidlaw, Joe Paldus, and Vedene Smith, all past or soon-to-be chairs of the Canadian Symposium on Theoretical Chemistry.

On February 10, 1983, the minister of Consumer and Corporate Affairs issued Letters Patent creating a corporation under the name Canadian Association of Theoretical Chemists/Association Canadienne des Chimistes Théoriciens. The objects of the corporation are:

(i) to organize the triennial Canadian Symposia on Theoretical Chemistry;

(ii) to further the development of theoretical chemistry in Canada;

(iii) and to engage in matters relating to theoretical chemistry as deemed appropriate by the Board of Directors.

The application was filed by André Bandrauk, Russell J. Boyd, Sigeru Huzinaga, Raymond E. Kapral, William G. Laidlaw, and Josef Paldus. According to the by-laws of the corporation:

(i) The Board of Directors shall accept applications for membership from any theoretical chemist holding a faculty position in a Canadian university or college who is eligible for grants in aid of research from the Natural Sciences and Engineering Research Council of Canada;

(ii) The Board of Directors shall accept applications for membership from any theoretical chemist actively engaged in research at government, industrial and private institutes and laboratories in Canada;

(iii) Membership shall be for life upon payment of a $5.00 fee.

The $5 lifetime membership fee was levied to cover the roughly $300 cost of incorporation. At any one time, the board consists of the cochairs of the most recent Canadian Symposium on Theoretical Chemistry and the cochairs of the next two symposia.

DEMOGRAPHIC FACTS

One fascinating way to chart the development of computational chemistry in Canada is to consider the chronology of the appointments of theoretical and computational chemists in Canadian universities. Figure 3 plots the number of appointments in two-year intervals, beginning with 1954–1955 up to and including 1996–1997. The most evident feature of Figure 3 is the rapid increase in the early 1960s leading to a peak later in the decade, which was followed by a precipitous decrease in the early 1970s. A second peak, about half as high as the first, occurred about two decades later, in the late 1980s. The second peak appears to have been due to a combination of growth and retirements.

From 1954, when Camille Sandorfy took up his position at the University of Montreal, to 1960, only 6 theoretical chemists were appointed. During the next five-year interval, 16 appointments were made. In the five-year interval 1966 to 1970, inclusive, a total of 35 theoretical and computational chemists were appointed to Canadian universities, a time of unprecedented opportunities. The growth rate was not sustainable. From personal experience, the author knows that appointments for theoretical and computational chemists were extremely rare in Canada for the decade beginning in 1971. Indeed, for the ten-year interval from 1971 to 1980, only 14 appointments were made: 6 in 1971–1975 and 8 in 1976–1980.

While the first peak in Figure 3 is due to the rapid expansion of the Canadian university system in the 1960s, the second peak was at least in part
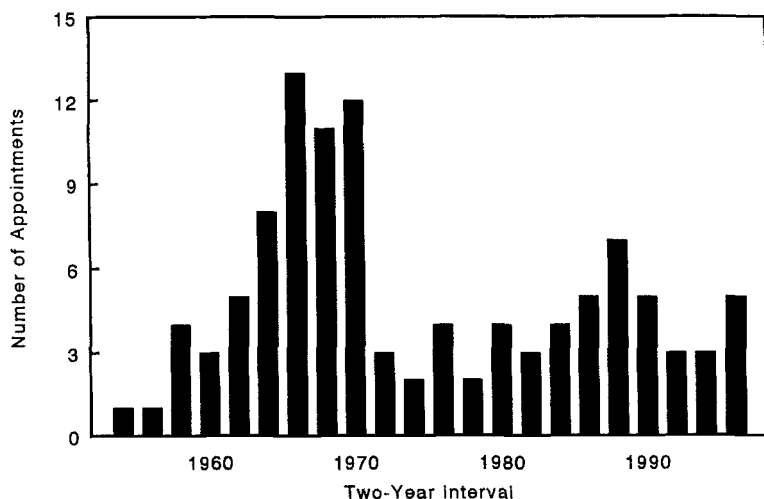
Figure 3 Number of new appointments, in 2-year intervals, of theoretical and computational chemists to the faculties of the departments of chemistry of Canadian universities.

brought about by a Natural Sciences and Engineering Research Council (NSERC) program to maintain a pool of talented young researchers to take up those positions which were expected to be available as the first of the many professors recruited to Canadian universities in the early 1960s began to retire. Thus, at least 10 appointments were made between 1981 and 1989 under the terms of the NSERC University Research Fellowships (URF) program. Without the URF appointments, the situation in the 1980s would have been scarcely better than that of the 1970s. Here it may be remarked that the limited employment prospects noted above for the 1970s were not restricted to theoretical chemistry. In fact, the situation in Canada was much the same in all branches of chemistry and in many of the other sciences.

Figure 3 is based on the information in Table 2, which lists 108 individuals, their year of appointment, and a few biographical details. The criteria for inclusion in Table 2 are:

1. The individual was actively involved in theoretical and computational chemistry at the time of his or her appointment;
2. The appointment must have been in a department of chemistry at the level of assistant professor or higher.
3. The individual held one or more appointments per items 1 and 2 for a minimum of four years.

Not included in Table 2 are the many experimentalists who became active in computational chemistry after their appointments. A few such individuals are discussed later in the chapter.

The brief biographical sketches in Table 2 indicate some interesting facts. Of the 108 appointees, 58 (54%) obtained their first degree in Canada. Nine were undergraduates at the University of Toronto, whereas the Universities of British Columbia (UBC) and Western Ontario each count seven as alumni. McGill and Queen's Universities follow with five each, while the University of Alberta graduated four with B.Sc. degrees. Another 16 Canadian universities graduated the remaining 19. Of the 50 (46%) appointees who obtained their first degrees abroad, 13 graduated from universities in the United States and 12 studied in the United Kingdom. Hungary and the former Czechoslovakia each account for three more. The other 19 received their first degrees from universities in 18 countries.

Table 2 reveals some equally interesting facts on the sources of the Ph.D. degrees of the 108 individuals. Of the 48 (44%) Ph.D. degrees earned in Canada, the University of Toronto leads with 11, followed by McGill University with 6 and McMaster University with 5; UBC and Alberta account for 4 apiece, and Queen's University and the University of Montreal each claim 3. The remaining 12 individuals received their Ph.D. degrees from nine other Canadian universities. Of the 60 (56%) Ph.D. degrees earned abroad, 29 and 19 were received from universities in the United States and the United Kingdom, respectively.

Table 2 Chronology of Appointments[a] of Theoretical and Computational Chemists in Canadian Universities

Name	Year	Biographical Sketch
Camille Sandorfy Univ. of Montreal	1954	B.Sc., 1943, Ph.D., 1946, Univ. of Szeged, Hungary (Prof. A. Kiss); D.Sc., 1949, Sorbonne, Paris (Profs. Louis de Broglie and R. Daudel); PDF, 1951–1953, NRC, Ottawa (Dr. R. N. Jones)
John A. R. Coope Univ. of British Columbia	1957	B.A., 1950, M.Sc., 1953, Univ. of British Columbia; D.Phil., 1956, Oxford Univ. (Prof. C. A. Coulson); PDF, 1956, Univ. of British Columbia (Prof. C. Reid)
Robert F. Snider Univ. of British Columbia	1958	B.Sc., 1953, Univ. of Alberta; Ph.D., 1958, Univ. of Wisconsin (Prof. C. F. Curtiss); PDF, 1958, NRC, Ottawa (Dr. W. G. Henry)
Richard F. W. Bader Univ. of Ottawa, 1959–1963; McMaster Univ., 1963–	1959	B.Sc., 1953, M.Sc., 1955, McMaster Univ.; Ph.D., 1958, Massachusetts Inst. of Technology (Prof. C. G. Swain); NRC PDF, 1958–1959, Cambridge Univ. (Prof. H. C. Longuet-Higgins)
Fraser W. Birss[b] Univ. of Alberta, 1959–1987	1959	B.A., 1953, M.A., 1954, Univ. of Saskatchewan; D.Phil., 1956, Oxford Univ. (Prof. C. N. Hinshelwood); NRC PDF, 1956–1958, Oxford Univ. (Prof. C. A. Coulson); PDF, 1958–1959, Univ. of Rochester (Prof. A. B. F. Duncan)
K. Lenore McEwen Univ. of Saskatchewan, 1959–1977	1959	B.A., 1951, M.Sc., 1953, Univ. of British Columbia; Ph.D., 1958, Cambridge Univ. (Prof. H. C. Longuet-Higgins); NRC PDF, 1957–1959, Univ. of Montreal (Prof. C. Sandorfy)
Wendell Forst Laval Univ., 1961–1986	1961	B.Sc., 1948, Technical Institute of Prague; Ph.D., 1955, McGill Univ. (Prof. C. A. Winkler); PDF, 1958–1961, Univ. of North Carolina (Prof. O. K. Rice)
John B. Moffat Univ. of Waterloo	1961	B.A., 1953, Ph.D., 1956, Univ. of Toronto (Prof. R. L. McIntosh); RS, DuPont, 1956–1961
John P. Valleau Univ. of Toronto	1961	B.A., 1954, M.A., 1955, Univ. of Toronto; Ph.D., 1958, Cambridge Univ. (Prof. H. C. Longuet-Higgins); PDF, 1958–1960, NRC, Ottawa
R. Julian C. Brown Queen's Univ., 1962–1966, 1969–	1962	B.Sc., 1956, M.Sc., 1959, Univ. of Sydney; Ph.D., 1962, Univ. of Illinois (Prof. H. S. Gutowsky)
Friedrich Grein Univ. of New Brunswick	1962	B.Sc., 1956, M.Sc., 1958, Univ. of Göttingen; Ph.D., 1960, Univ. of Frankfurt (Prof. H. Hartmann); NRC PDF, 1960–1962, Univ. of New Brunswick (Prof. R. Kaiser)
Michael Anthony Whitehead McGill Univ.	1962	B.Sc., 1956, Queen Mary College, London; Ph.D., 1960, Univ. of London (Prof. M. J. S. Dewar); PDF, 1960–1962, Univ. of Cincinnati (Prof. H. H. Jaffe)
David M. Bishop Univ. of Ottawa	1963	B.Sc., 1957, Ph.D., 1960, Univ. College, London (Prof. D. P. Craig); PDF, 1960–1962, Carnegie Inst. of Technology (Prof. R. G. Parr)

Table 2 *Continued*

Name	Year	Biographical Sketch
Serafin Fraga Univ. of Alberta	1963	L.Sc., 1954, Ph.D., 1957, Univ. of Madrid (Prof. A. Perez-Masia); PDF, 1958–1961, Univ. of Chicago (Prof. R. S. Mulliken); PDF, 1961–1962, Univ. of Alberta (Prof. F. W. Birss); Asst. Prof., 1962–1963, Royal Military College, St. Jean, Quebec
Imre G. Csizmadia Univ. of Toronto	1964	Dipl.Eng., 1956, Polytechnic Univ., Budapest; M.Sc., 1959, Ph.D., 1962, Univ. of British Columbia (Prof. L. D. Hayward); NATO PDF, 1962–1964, Massachusetts Institute of Technology (Prof. J. C. Slater)
Sandor Fliszar Univ. of Montreal	1964	L.Sc., 1958, D.Sc., 1962, Univ. of Geneva (Prof. Th. Posternak); PDF, 1962–1964, Cyanamid European Research Inst. (Prof. R. F. Hudson)
D. A. Hutchinson Univ. of Victoria, 1964–1967; Queen's Univ., 1967–1986	1964	B.Sc., 1958, M.Sc., 1960, Univ. of British Columbia; D.Phil., 1963, Oxford Univ. (Prof. C. A. Coulson); PDF, 1963–1964, Univ. of Pennsylvania (Prof. H. F. Hameka)
William G. Laidlaw Univ. of Calgary	1964	B.Sc., 1959, Univ. of Western Ontario; M.Sc., 1961, California Inst. of Technology (Prof. R. Mazo); Ph.D., 1963, Univ. of Alberta (Prof F. W. Birss); NRC PDF, Oxford Univ., 1963–1964 (Prof. C. A. Coulson)
Delano P. Chong Univ. of British Columbia	1965	B.S., 1958, Univ. of California, Berkeley; A.M., 1959, Ph.D., 1963, Harvard Univ. (Prof. G. B. Kistiakowsky); NATO PDF, 1963–1964, Oxford Univ. (Prof. J. W. Linnett); RA, 1964–1965, Univ. of Wisconsin (Prof. W. Byers Brown)
Patrick W. M. Jacobs Univ. of Western Ontario	1965	B.Sc., 1941, M.Sc., 1943, Univ. of Natal; Ph.D., 1951, Univ. of London (Prof. F. C. Tompkins); faculty member, 1951–1965, Imperial College, London
William J. Meath Univ. of Western Ontario	1965	B.Sc., 1960, Carleton Univ.; Ph.D., 1965, Univ. of Wisconsin (Prof. J. O. Hirschfelder)
Huw O. Pritchard York Univ.	1965	B.Sc., 1948, M.Sc., 1949, Ph.D., 1951, Univ. of Manchester (Prof. H. A. Skinner); faculty member, 1951–1965, Univ. of Manchester
Margaret L. Benston[b] Simon Fraser Univ., 1966–1981	1966	B.A., 1959, Willamette Univ.; Ph.D., 1964, Univ. of Washington; PDF, 1964–1966, Univ. of Wisconsin
Geoffrey Hunter York Univ.	1966	B.Sc., 1961, Univ. of Salford; M.Sc., 1962, Ph.D., 1964, Univ. of Manchester (Prof. H. O. Pritchard); Welch PDF, 1964–1965, Rice Univ. (Prof. K. S. Pitzer); NRC PDF, 1965–1966, York Univ. (Prof. H. O. Pritchard)
Gulzari L. Malli Simon Fraser Univ.	1966	B.Sc., 1958, Delhi Univ.; M.Sc., 1960, McMaster Univ. (Prof. G. W. King); Ph.D., 1964, Univ. of Chicago (Profs. R. S. Mulliken and C. C. J. Roothaan); PDF, 1964–1965, Yale Univ. (Prof. G. Breit); Asst. Prof., 1965–1966, Univ. of Alberta
Keith A. R. Mitchell Univ. of British Columbia	1966	B.Sc., 1960, Ph.D., 1963, Univ. College London (Prof. D. P. Craig); Turner and Newall Fellowship, 1964–1966, Univ. of Manchester

	Year	
Reginald Paul Univ. of Calgary	1966	B.Sc., 1956, M.Sc., 1957, Univ. of Lucknow, India; M.Sc., 1962, Univ. of Alberta (Prof. F. W. Birss); Ph.D., 1966, Univ. of Durham (Prof. Lord Wynne-Jones and Prof. G. N. Fowler)
Neil S. Snider Queen's Univ. 1966–1995	1966	B.Sc., 1959, Purdue Univ.; M.A., 1961, Ph.D., 1964, Princeton Univ. (Prof. D. Hornig); PDF, 1964–1965, Cornell Univ. (Prof. B. Widom); PDF, 1965–1966, Yale Univ. (Prof. M. Fixman)
John Walkley Simon Fraser Univ.	1966	B.Sc., 1954, Ph.D., 1957, Univ. of Liverpool; PDF, 1957–1960, Univ. of California, Berkeley (Prof. J. H. Hildebrand); faculty member, Imperial College, London, 1960–1966
Robert Wallace Univ. of Manitoba	1966	B.Sc., 1961, Ph.D., 1964, Univ. of Glasgow (Prof. A. L. Porte); NATO PDF, 1964–1965, Uppsala Univ. (Prof. P.-O. Löwdin); NATO PDF, 1965–1966, Univ. of Bristol (Prof. A. D. Buckingham)
Thomas W. Dingle Univ. of Victoria	1967	B.Sc., 1958, Ph.D., 1965, Univ. of Alberta (Prof. H. E. Gunning); PDF, 1965–1966, Oxford Univ. (Prof. C. A. Coulson); PDF, 1966–1967, Univ. of Ottawa (Prof. D. M. Bishop)
Byung Chan Eu McGill Univ.	1967	B.S., 1959, Seoul National Univ.; Ph.D., 1965, RA, 1965–1966, Brown Univ. (Prof. J. Ross); RF, 1966–1967, Harvard Univ. (Prof. M. Karplus)
Che-Shung Lin[b] Univ. of Windsor, 1967–1989	1967	B.Sc., 1956, M.Sc., 1960, National Taiwan Univ.; Ph.D., 1965, Univ. of Saskatchewan (Prof. K. L. McEwen); PDF, 1964–1966, Univ. of Alberta (Profs. F. W. Birss and S. Fraga); PDF, 1966–1967, Indiana Univ. (Prof. H. Shull)
David P. Santry McMaster Univ.	1967	B.Sc., 1960, Ph.D., 1963, Univ. College, London (Prof. D. P. Craig); PDF, 1963–1964, National Physical Lab, England; PDF, 1964–1967, Carnegie-Mellon Univ. (Prof. J. A. Pople)
Vedene H. Smith Jr. Queen's Univ.	1967	B.A., 1955, Emory Univ.; M.Sc., 1957, Ph.D., 1960, Georgia Inst. of Technology (Prof. H. A. Gersch); Fil.Lic, Fil.Dr, Docent, 1967, Uppsala Univ. (Prof. P.-O. Löwdin)
N. Colin Baird Univ. of Western Ontario	1968	B.Sc., 1963, Ph.D., 1967, McGill Univ. (Prof. M. A. Whitehead); Robert A. Welch PDF, 1966–1968, Univ. of Texas (Prof. M. J. S. Dewar)
Jiri Cizek Univ. of Waterloo	1968	M.Sc. (RNDr.), 1961, Charles Univ., Prague, Ph.D. (CSc.), 1965, Czechoslovak Academy of Sciences, Prague (Prof. J. Koutecky)
Sigeru Huzinaga Univ. of Alberta	1968	B.Sc., 1948, Kyushu Univ., Ph.D., 1959, Kyoto Univ. (Prof. G. Araki), RA, 1959–1961, Univ. of Chicago (Prof. R. S. Mulliken)
Josef Paldus Univ. of Waterloo	1968	M.Sc. (RNDr.), 1959, Charles Univ., Prague; Ph.D. (CSc.), 1962, Czechoslovak Academy of Sciences, Prague (Prof. J. Koutecky); PDF, 1962–1964, NRC, Ottawa (Dr. D. A. Ramsay)

Table 2 *Continued*

Name	Year	Biographical Sketch
Walter R. Thorson Univ. of Alberta, 1968–1994	1968	B.S., 1953, Ph.D., 1957, California Inst. of Tech. (Prof. R. M. Badger); NSF PDF, 1956, Harvard Univ. (Prof. W. E. Moffitt); Asst. and Assoc. Prof., 1958–1968, Massachusetts Inst. of Technology
Alan R. Allnatt Univ. of Western Ontario	1969	B.Sc., 1956, Ph.D., 1959, Univ. of London (Prof. P. W. M. Jacobs); NATO PDF, 1959–1961, Univ. of Chicago (Profs. S. A. Rice and M. H. Cohen); faculty member, 1961–1969, Univ. of Manchester
Johannes P. Colpa Queen's Univ., 1969–1991	1969	Candidaat, 1948, Doctoraal, 1953, Ph.D., 1957, Univ. of Amsterdam (Profs. J. A. A. Ketelaar and J. de Boer); PDF, 1961–1962, Cambridge Univ. (Prof. H. C. Longuet-Higgins)
Jack L. Ginsburg[b] Saint Mary's Univ., 1969–1998	1969	B.S., 1962, Temple Univ.; Ph.D., 1968, Rutgers Univ. (Prof. L. Goodman); PDF, 1968–1969, McMaster Univ. (Prof. R. F. W. Bader)
Bryan R. Henry Univ. of Manitoba, 1969–1987; Univ. of Guelph, 1987–	1969	B.Sc., 1963, Univ. of British Columbia; Ph.D., 1967, Florida State Univ. (Prof. M. Kasha), PDF, 1968–1969, NRC, Ottawa (Dr. W. Siebrand)
Alan C. Hopkinson York Univ.	1969	B.Sc., 1963, Ph.D., 1967, Sheffield Univ. (Prof. P. A. H. Wyatt); PDF, 1967–1969, Univ. of Toronto (Profs. K. Yates and I. G. Csizmadia)
Raymond E. Kapral Univ. of Toronto	1969	B.S., 1964, King's College, Wilkes-Barre, PA; Ph.D., 1967, Princeton Univ. (Prof. L. C. Allen); PDF, 1968, Massachusetts Inst. of Technology (Prof. J. Ross)
André D. Bandrauk Univ. of Sherbrooke	1970	B.Sc., 1961, Univ. of Montreal (Loyola College); S.M., 1963, Massachusetts Institute of Technology; Ph.D., 1968, McMaster Univ. (Prof. R. F. W. Bader); NATO PDF, 1968–1969, Oxford Univ. (Prof. C. A. Coulson)
Bruce L. Clarke Univ. of Alberta	1970	B.Sc., 1965, Univ. of Toronto; Ph.D., 1969, Univ. of Chicago (Prof. S. A. Rice); PDF, 1969 Univ. of California, Santa Cruz (Prof. T. L. Hill)
Frederick R. W. McCourt Univ. of Waterloo	1970	B.Sc., 1963, Ph.D., 1966, Univ. of British Columbia (Prof. R. F. Snider); NRC PDF, 1966–1967, Swiss Federal Institute of Technology (ETH), Zürich (Prof. H. Primas); NRC PDF/RA 1967–1969, Kamerlingh Onnes Lab, Leiden (Prof. J. Beenakker)
Arvi Rauk Univ. of Calgary	1970	B.Sc., 1965, Ph.D., 1968, Queen's Univ. (Prof. S. Wolfe); NRC PDF, 1968–1970, Princeton Univ. (Profs. L. C. Allen and K. Mislow)
Stuart M. Rothstein Brock Univ.	1970	B.S., 1960, Univ. of Illinois; M.S., 1966, Ph.D., 1968, Univ. of Michigan (Prof. S. M. Blinder); PDF, 1968–1969, Johns Hopkins Univ. (Prof. H. J. Silverstone); Asst. Prof., 1969–1970, Swarthmore College

Bernard Shizgal Univ. of British Columbia	1970	B.Sc., 1964, McGill Univ.; Ph.D., 1968, Columbia Univ. (Prof. M. Karplus); NRC PDF, 1968–1969, Univ. of Leiden (Prof. P. Mazur); NRC PDF, 1969–1970, Univ. of British Columbia (Prof. R. F. Snider)
David Rodney Truax Univ. of Calgary	1970	B.Sc., 1964, Ph.D., 1969, Univ. of Western Ontario (Prof. R. G. Kidd); PDF, 1969–1970, Univ. of Copenhagen (Prof. C. J. Ballhausen)
Charles H. Warren Dalhousie Univ.	1970	B.Sc., 1964, Univ. of Western Ontario; Ph.D., 1968, McMaster Univ. (Prof. G. W. King); NRC PDF, 1968–1970, Uppsala Univ. (Prof. P.-O. Löwdin)
Stuart G. Whittington Univ. of Toronto	1970	B.A., 1963, M.A., 1967, Ph.D., 1972, Cambridge Univ. (Prof. D. Chapman)
James S. Wright Carleton Univ.	1970	B.S., 1962, Stanford Univ.; Ph.D., 1968, Univ. of California, Berkeley (Prof. A. Streitweiser Jr.); PDF, 1968–1970, Orsay, France (Prof. L. Salem)
Michael C. Zerner[b,c] Univ. of Guelph, 1970–1982	1970	B.Sc., 1961, Carnegie-Mellon Univ.; A.M., 1962, Ph.D., 1966, Harvard Univ. (Prof. M. Gouterman); NIH PDF, 1968–1970, Uppsala Univ. (Prof. P.-O. Löwdin)
Roy E. Kari Laurentian Univ.	1971	B.Sc., 1964, Univ. of Western Ontario; M.Sc., 1966, Ph.D., 1969, Univ. of Toronto (Prof. I. G. Csizmadia); NRC PDF, 1969–1970, Univ. of York (Prof. B. T. Sutcliffe); NRC PDF, 1971, York Univ. (Prof. H. O. Pritchard)
Robert J. Le Roy Univ. of Waterloo	1972	B.Sc., 1965, M.Sc., 1967, Univ. of Toronto (Prof. G. Burns); Ph.D., 1971, Univ. of Wisconsin (Prof. R. B. Bernstein); NRC PDF, 1971–1972, Univ. of Toronto (Prof. J. van Kranendonk)
Saul Goldman Univ. of Guelph	1972	B.Sc., 1964, Ph.D., 1969, McGill Univ. (Prof. G. C. B. Cave); NRC PDF, 1969–1972, Univ. of Florida (Prof. R. G. Bates)
John M. Sichel Univ. of Moncton	1972	B.Sc., 1964, Ph.D., 1968, McGill Univ. (Prof. M. A. Whitehead); PDF, 1967–1969, Univ. of Bristol (Prof. A. D. Buckingham); RA, 1969–1970, Asst. Prof., 1970–1972, Laval Univ.
Russell J. Boyd Dalhousie Univ.	1975	B.Sc., 1967, Univ. of British Columbia; Ph.D., 1971, McGill Univ. (Prof. M. A. Whitehead); NRC PDF, 1971–1973, Oxford Univ. (Prof. C. A. Coulson); Killam PDF, 1973–1975, Univ. of British Columbia
Paul W. Brumer Univ. of Toronto	1975	B.S., 1966, Brooklyn College; Ph.D., 1972, Harvard Univ. (Prof. M. Karplus); Institute Fellow, 1972–1973, Weizmann Inst. (Prof. R. D. Levine); RF, 1973–1974, Harvard College Observatory (Prof. A. Dalgarno)
Dennis R. Salahub Univ. of Montreal, 1976–1999; Steacie Institute of the National Research Council of Canada, 1999–	1976	B.Sc., 1967, Univ. of Alberta; Ph.D., 1970, Univ. of Montreal (Prof. C. Sandorfy); NRC PDF, 1970–1972, Univ. of Sussex (Prof. J. N. Murrell); RA, 1972–1974, Univ. of Waterloo; RS, 1974, Johns Hopkins Univ.; RS, 1975–1976, General Electric, Schenectady

Table 2 *Continued*

Name	Year	Biographical Sketch
Bryan C. Sanctuary McGill Univ.	1976	B.Sc., 1967, Ph.D., 1971, Univ. of British Columbia (Prof. R. F. Snider); PDF, 1972–1974, Univ. of Leiden (Prof. J. Beenakker); Asst. Prof., 1974–1976, Univ. of Wisconsin, Madison
Paul G. Mezey Univ. of Saskatchewan	1977	M.Sc., 1967, Ph.D., 1970 (Prof. F. Torok), M.Sc. (Math) 1972, Univ. of Budapest; PDF, 1973–1976, Univ. of Toronto (Profs. I. G. Csizmadia and K. Yates)
Seamus O'Shea Univ. of Lethbridge	1977	B.Sc., 1967, Univ. College Cork, Ireland; Ph.D, 1973, McMaster Univ. (Prof. D. P. Santry); PDF, 1973–1975, Univ. of Western Ontario (Prof. W. J. Meath); RA, 1975–1977, NRC, Ottawa (Dr. M. L. Klein)
Jean-Pierre Laplante Royal Military College of Canada	1979	B.Sc., 1971, Ph.D., 1976, Univ. of Sherbrooke (Prof. A. D. Bandrauk); PDF, 1976–1977, Free Univ. of Brussels (Prof. G. Nicolis), PDF, 1977–1979, NRC, Ottawa (Dr. W. Siebrand)
Glenn M. Torrie Royal Military College of Canada	1979	B.Sc., 1971, M.Sc., 1972, Ph.D., 1975. Univ. of Toronto (Prof. J. P. Valleau); NSERC PDF, 1975–1976, Univ. of Paris (Prof. D. Levesque)
Grenfell N. Patey Univ. of British Columbia	1980	B.S., 1970, Memorial Univ.; M.Sc., 1972, Ph.D., 1975, Univ. of Toronto (Prof. J. P. Valleau); PDF, 1975–1977, Univ. of Paris (Prof. D. Levesque); RA, 1978–1980, NRC, Ottawa (Dr. M. L. Klein)
Ajit J. Thakkar Univ. of Waterloo, 1980–1984; Univ. of New Brunswick, 1984–	1980	B.Sc., 1973, Ph.D, 1976, Queen's Univ. (Prof. V. H. Smith Jr.); NRC PDF, 1976–1978, Univ. of Waterloo (Profs. J. Paldus and I. Magee); RA, 1978–1980, Queen's Univ. (Prof. V. H. Smith Jr.)
Ross W. Wetmore Univ. of Guelph, 1981–1985	1981	B.Sc., 1972, Univ. of Ottawa; Ph.D, 1977, Univ. of Southern California (Prof. G. A. Segal); PDF, 1977–1979, Univ. of California, Berkeley (Prof. H. F. Schaefer III); RS, 1979–1981, Smithsonian Astrophysical Observatory (Prof. A. Dalgarno)
Tom Ziegler Univ. of Calgary	1981	M.Sc., 1972, Univ. of Copenhagen; Ph.D., 1978, Univ. of Calgary (Prof. A. Rauk); PDF, 1978–1980, Free Univ., Amsterdam (Prof. E. J. Baerends); NSERC PDF, 1980–1981, McMaster Univ. (Prof. D. P. Santry)
Raymond A. Poirier Univ. of Toronto, 1982–1984; Memorial Univ., 1984–	1982	B.Sc., 1975, M.Sc., 1976, Laurentian Univ. (Prof. R. E. Kari); Ph.D., 1980, Univ. of Toronto (Prof. I. G. Csizmadia); NSERC PDF, 1980–1982, CNRS, Paris (Prof. R. Daudel)
John D. Goddard Univ. of Guelph	1983	B.Sc., 1973, Univ. of Western Ontario; M.Sc., 1974, Ph.D., 1978, Univ. of Toronto (Prof. I. G. Csizmadia); NSERC PDF, 1978–1979, Univ. of California, Berkeley, and RA, 1979–1980, Univ. of Texas, Austin (Prof. H. F. Schaefer III); RA, 1980–1982, NRC, Ottawa (Dr. W. Siebrand)

Brian A. Pettitt Univ. of Winnipeg	1983	B.Sc., 1968, M.Sc., 1971, Univ. of Manitoba (Prof. R. Wallace); Ph.D, 1985, Dalhousie Univ. (Profs. R. J. Boyd and R. E. Wasylishen)
Axel D. Becke Queen's Univ.	1984	B.Sc., 1975, Queen's Univ.; Ph.D., 1981, McMaster Univ. (Prof. D. W. L. Sprung); NSERC, Killam and Eastburn PDF, 1981–1984, Dalhousie Univ. (Prof. R. J. Boyd)
T. T. Nguyen-Dang Univ. of Sherbrooke, 1984–1987; Laval Univ., 1987–	1984	B.Sc., 1975, Univ. of Liège; Ph.D., 1980, PDF, 1980–1982, McMaster Univ. (Prof. R. F. W. Bader); PDF, 1982–1984, Univ. of Sherbrooke (Prof. A. D. Bandrauk)
David M. Wardlaw Queen's Univ.	1984	B.Sc., 1975, Ph.D., 1982, Univ. of Toronto (Prof. P. W. Brumer); NSERC PDF, 1982–1984, California Inst. of Technology (Prof. R. A. Marcus)
John M. Cullen Univ. of Manitoba	1985	B.Sc., 1969, M.Sc., 1973, Univ. of Windsor (Prof. G. W. F. Drake); Ph.D., 1981, Univ. of Guelph (Prof. M. C. Zerner); PDF, 1983–1985, Harvard Univ. (Prof. W. N. Lipscomb)
Simon J. Fraser Univ. of Toronto	1986	B.A., 1966, Oxford Univ.; Ph.D., 1970, Cambridge Univ.; Ciba-Geigy fellowship, 1970, Univ. of Paris; SRC fellowship, 1972, Univ. of Sussex
Ian P. Hamilton Univ. of Ottawa, 1986–1992; Wilfred Laurier Univ., 1992–	1986	B.Sc., 1976, Ph.D., 1982, Univ. of Toronto (Prof. P. W. Brumer); PDF and RA, 1982–1986, Univ. of Chicago (Prof. J. C. Light)
Saba M. Mattar Univ. of New Brunswick	1986	B.Sc., 1968, Alexandria Univ.; M.Sc., 1974, American Univ. in Cairo (Prof. G. Habashi); Ph.D., 1982, McGill Univ. (Prof. W. C. Galley); PDF and RA, 1982–1986, Univ. of Toronto (Prof. G. A. Ozin)
Timothy A. Wildman McMaster Univ., 1986–1991	1986	B.Sc., 1977, M.Sc., 1978, Ph.D., 1982, Univ. of Manitoba (Prof. T. Schaefer); RA, 1982–1986, NRC, Ottawa (Dr. W. Siebrand)
Gregory C. Corey Univ. of Montreal, 1987–1993, Saint Mary's Univ., 1995–1999	1987	B.Sc., 1974, Univ. of New Brunswick, M.Sc., 1980, Univ. of Waterloo (Prof. R. J. Le Roy), Ph.D., 1984, Univ. of Waterloo (Profs. F. R. W. McCourt and W.-K. Liu); NATO PDF, 1984–1986, RA, 1986–1987, Univ. of Maryland
Tucker Carrington Jr. Univ. of Montreal	1988	B.Sc., 1981, Univ. of Toronto; Ph.D., 1985, Univ. of California, Berkeley (Prof. W. H. Miller); PDF, 1985–1987, Swiss Federal Institute of Technology (ETH), Zürich (Prof. M. Quack); RA, 1987–1988, NRC, Ottawa (Dr. P. R. Bunker)
Randall S. Dumont McMaster Univ.	1988	B.Sc., 1981, Univ. of Western Ontario; Ph.D., 1987, Univ. of Toronto (Prof. P. W. Brumer); NSERC PDF, 1987–1988, Columbia University (Prof. P. Pechukas)
David M. Ronis McGill Univ.	1988	B.Sc., 1974, McGill Univ.; Ph.D., 1978, Massachusetts Inst. of Technology (Prof. I. Oppenheim); Miller Fellow, 1978–1980, Univ. of California, Berkeley; Asst. and Assoc. Prof., 1980–1988, Harvard Univ.
Katherine V. Darvesh Mount Saint Vincent Univ.	1989	B.Sc., 1980, Ph.D., 1984, Univ. of New Brunswick (Prof. F. Grein); Killam and NSERC PDF, 1984–1989, Dalhousie Univ. (Prof. R. J. Boyd)

Table 2 *Continued*

Name	Year	Biographical Sketch
Mariusz Klobukowski Univ. of Alberta	1989	B.Sc., 1971, Ph.D., 1978, N. Copernicus Univ., (Prof. J. Karwowski); PDF and RA, 1979–1988, Univ. of Alberta (Prof. S. Huzinaga)
Peter G. Kusalik Dalhousie Univ.	1989	B.Sc., 1981, Univ. of Lethbridge; M.Sc., 1984, Ph.D., 1987, Univ. of British Columbia (Prof. G. N. Patey); NSERC PDF, 1987–1989, Australian National Univ. (Prof. D. J. Evans)
Donald F. Weaver Queen's Univ.	1989	M.D., 1981, Ph.D., 1986, Queen's Univ. (Prof. S. Wolfe); neurology residency, 1986–1988, Dalhousie Univ.
Kathleen M. Gough Brock Univ., 1990–1995; Univ. of Manitoba, 1995–	1990	B.Sc., 1971, Loyola of Montreal; M.Sc., 1976, Ph.D., 1984, Univ. of Manitoba (Prof. B. R. Henry); RA, 1984–1986, NRC, Ottawa (Dr. W. Murphy); NSERC PDF, 1987–1989, McMaster Univ. (Prof. R. F. W. Bader)
Baltazar D. Aguda Laurentian Univ.	1991	B.Sc., 1978, Univ. of the Philippines; Ph.D., 1986, Univ. of Alberta (Prof. B. L. Clarke); PDF, 1987, Univ. of Alberta; PDF, 1988–1990, Indiana Univ.–Purdue Univ. Indianapolis (Prof. R. Larter); PDF, 1990–1991, York Univ. (Prof. H. O. Pritchard)
Donald D. Frantz Univ. of Lethbridge, 1991–1995	1991	B.Sc., 1980, Univ. of Alberta; Ph.D., 1989, Harvard Univ. (Prof. D. R. Herschbach); PDF, 1989–1990, Univ. of Rhode Island (Prof. D. L. Freeman)
William J. Pietro York Univ.	1991	B.S., 1978, Polytechnic Inst. of New York; Ph.D., 1982, Univ. of California, Irvine (Prof. W. J. Hehre); PDF, 1982–1985, Northwestern Univ. (Profs. T. J. Marks and M. A. Ratner); Asst. Prof., 1985–1991, Univ. of Wisconsin, Madison
Benoit Roux Univ. of Montreal	1991	B.Sc., 1981, M.Sc., 1984, Univ. of Montreal; Ph.D., 1990, Harvard Univ. (Prof. M. Karplus); PDF, 1990–1991, Atomic Energy Commission, France (Dr. J. Smith)
Gustavo A. Arteca Laurentian Univ.	1992	B.Sc., 1980, M.Sc., 1981, Ph.D., 1985, National University of La Plata, Argentina (Profs. E. A. Castro and F. M. Fernandez); PDF and RA, 1986–1992, Univ. of Saskatchewan (Prof. P. G. Mezey)
David Jack Concordia Univ.	1992	B.Sc., 1979, Univ. of Toronto; M.Sc., 1981, Ph.D., 1986, Univ. of Alberta (Prof. H. J. Kreuzer); NSERC Industrial PDF, 1986–1989, Alcan, Kingston; RA, 1990, Visiting Asst. Prof., 1991, Univ. of Toronto (Prof. J. C. Polanyi)
Alain St-Amant Univ. of Ottawa	1993	B.Sc., 1986, Univ. of Winnipeg, M.Sc., 1988, Ph.D., 1992, Univ. of Montreal (Prof. D. R. Salahub), NSERC PDF, 1992–1993, Univ. of California, San Francisco (Prof. P. A. Kollman)
John R. Gunn Univ. of Montreal	1994	B.Sc., 1987, Univ. of Calgary; Ph.D., 1992, Univ. of California, Berkeley (Prof. K. Dawson); PDF, 1992–1994, Columbia Univ. (Prof. R. A. Friesner)

Name / Institution	Year	Details
Margot E. Mandy Univ. of Northern British Columbia	1994	B.Sc., 1976, Acadia Univ.; M.Sc., 1980, Ph.D, 1991, Univ. of Toronto (Profs. J. E. Dove and P. G. Martin); PDF, 1991–1992, Dalhousie Univ. (Prof. P. G. Kusalik); RA, 1992–1994, Univ. of Toronto (Profs. J. R. Drummond and H.-R. Cho)
Mark R. Roussel Univ. of Lethbridge	1995	B.Sc., 1988, Queen's Univ.; M.Sc., 1990, Ph.D., 1994, Univ. of Toronto (Prof. S. J. Fraser); NSERC PDF, 1994–1995, McGill Univ. (Prof. M. C. Mackey)
René Fournier York Univ.	1996	B.Sc., 1984; Ph.D., 1989, Univ. of Montreal (Prof. D. R. Salahub); PDF, 1989–1991, Iowa State Univ. (Prof. A. E. DePristo); RA, 1991–1994, NRC, Ottawa (Dr. W. Siebrand); PDF, 1995–1996, Univ. of Nevada, Las Vegas (Prof. C. Chen)
Mark Thachuk Univ. of British Columbia	1996	B.Sc., 1986, Univ. of Western Ontario; Ph.D., 1991, Univ. of Waterloo (Prof. F. R. W. McCourt); NSERC PDF, 1991–1993, Northwestern Univ. (Prof. G. C. Schatz); PDF, 1993–1996, Queen's University (Prof. D. M. Wardlaw)
Natalie M. Cann Queen's Univ.	1997	B.Sc., 1989, Univ. of New Brunswick; Ph.D., 1993, Dalhousie Univ. (Profs. R. J. Boyd and A. J. Thakkar); NSERC and Killam PDF, 1993–1997, Univ. of British Columbia (Profs. C. E. Brion and G. N. Patey)
Jeremy M. Schofield Univ. of Toronto	1997	A.B., 1988, Amherst College; Ph.D., 1993, Massachusetts Inst. of Technology, (Prof. I. Oppenheim); PDF, 1993–1995, Univ. of Chicago (Prof. S. A. Rice); PDF, 1995–1997, Northwestern Univ. (Prof. M. A. Ratner)
Igor M. Svishchev Trent Univ.	1997	M.Sc., 1984, Moscow State Univ.; Ph.D., 1988, USSR Academy of Sciences (Prof. Buslaev); PDF and RA, 1991–1997, Dalhousie Univ. (Prof. P. G. Kusalik)

[a] Data through 1997 only. Among the abbreviations used in this table are: PDF, postdoctoral fellow; RF, research fellow; RA, research associate; RS, research scientist.

[b] Deceased

[c] Note added in proofs: Sadly, I note that Mike Zerner died on February 2, 2000.

The remaining 12 Ph.D. degrees were earned in 10 other countries. Of the 29 Ph.D. degrees earned in the United States, Harvard leads with 5, followed by 3 each from Berkeley, MIT, and Wisconsin. The remaining 15 were received from 13 universities, including 2 each from Chicago and Princeton. The University of London tops the U.K. list with 6, followed by Cambridge with 4 and Oxford with 3. The remaining 6 were awarded by five other universities in the U.K.

TOWARD A STEADY-STATE POPULATION

One of the remarkable features of the data in Table 2 is that only five of the 108 individuals are deceased and only 10 others have severed their ties through resignation or retirement. Of the remaining 93, two have taken up full-time administrative appointments and do not play an active part in their respective departments of chemistry. Another 13 have reached retirement age but continue to be associated with their respective departments. This leaves a total of 78 on the permanent teaching faculties of the universities. Since many of the retired professors are active in research and often continue to do some teaching, the number of theoretical and computational chemists active in teaching and/or research is estimated to have been about 85 in 1997. Figure 4 shows the number of active theoretical and computational chemists as a function of time. The nearly exponential growth in the 1960s, followed by slower linear growth in the
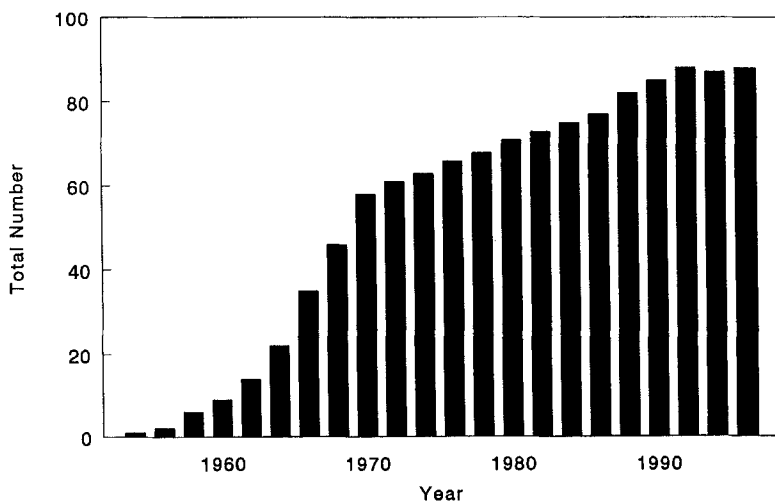
Figure 4 Number of active theoretical and computational chemists on the faculties of the departments of chemistry of Canadian universities as a function of time.

1970s and 1980s, and a leveling off in the 1990s is clearly evident. It must be emphasized that this figure underestimates the true level of activity in computational chemistry because many experimentalists have taken up computational chemistry during the past decade.

FAMILY TREES AND TRENDS

Examination of the brief biographical sketches in Table 2 reveals some interesting trends, notably a shift from the United Kingdom as the preferred location for research experience in the 1950s and 1960s to the United States in the 1980s and 1990s. For example, 11 of the first 20 individuals on the list completed their doctorates or did postdoctoral research in Britian, five at Oxford, three at London, and three at Cambridge. In contrast, none of the last 20 individuals on the list completed any part of their education in the United Kingdom, whereas 9 received doctoral or postdoctoral training in the United States. Seven of the last 20 received all their degrees and postdoctoral in training in Canadian universities, whereas only one of the first 20 on the list received all his degrees in Canada.

There has been a significant increase in the number and duration of postdoctoral positions before taking up permanent positions. The average time spent doing postdoctoral research for the first 20 appointees in Table 2 is just under 2 years, while for the last 20 on the list the corresponding figure is about 4 years.

A number of interesting scientific family trees may be deduced from Table 2. For example, Alain St-Amant and René Fournier can be traced back to Camille Sandorfy through Dennis Salahub. Mark Thachuk can be traced back to Bob Snider through Fred McCourt, and Peter Kusalik can be traced back to John Valleau through Gren Patey.

A significant fraction of the appointees in Table 2 can be traced back to the legendary figures of C. A. Coulson, H. C. Longuet-Higgins, D. P. Craig, A. D. Buckingham, and others in the United Kingdom, M. Karplus, O. K. Rice, R. S. Mulliken, C. C. J. Roothaan, J. C. Slater, J. O. Hirschfelder, W. N. Lipscomb, and others in the United States, and P.-O. Löwdin in Sweden.

In some cases it is difficult to complete the family trees in Table 2 because information is missing. Such omissions do not always indicate that the identity of the supervisor is unknown. John Valleau recounted to this author an amusing tale about his postdoctoral fellowship at the NRC in Ottawa.

I had somehow become interested in ultrasonics and wished to set up some experimental activity in the field. I explained this, as far as I remember, by mail to Bill Schneider (whom I knew quite well, having worked in his lab one summer), and the solution proposed was that I should be

ostensibly attached to Otto Maass. Now Otto, a dear sweet man, had just been retired from McGill and had been taken in by the NRC as a rescue. Of course, Otto had no interest in either theory or ultrasonics, so we had no scientific interaction; also, he never actually cottoned-on to the fact that I was doing experiments. But he wanted to do his social duty to me. So once a week he appeared at the door of my office, stuck his head in, and said, "Well . . . are they giving you enough pencils?" And each week I replied in an equally unvarying way, saying, "Yes; the problem is in finding enough erasers."

DEPARTMENTAL HISTORIES

The following sections outline the history of the development of theoretical and computational chemistry in 21 of the largest departments of chemistry in Canadian universities. The coverage is limited to universities with Ph.D. programs where at least one full-time theoretical or computational chemist was appointed in 1970 or earlier. The sequence is based on the order in which the first appointments were made in each department (see Table 2). Where more than one university is listed in a given year, the order is alphabetical. Space limitations preclude more comprehensive coverage. Nevertheless, it is hoped that this account will give a general sense of the emphasis within each department and an overview of Canada's strength in the various subtopics of computational and theoretical chemistry.

University of Montreal (1954)

As indicated in an earlier section, the first appointment of a theoretical or computational chemist was made in 1954 when Camille Sandorfy became a faculty member at the University of Montreal. Ten years later, Sandor Fliszar joined the Department of Chemistry and developed a research program focused on electron distributions and a variety of charge-dependent properties. His many papers on correlations between atomic charges and NMR chemical shifts, the relationships between atomic charges and bond dissociation energies, and the partitioning of atomic charges into core and valence contributions have been summarized in his monograph.[21]

The University of Montreal showed much foresight with the appointment of Dennis Salahub in 1976. In the next two decades he made several key contributions to the development and applications of density functional (DF) methodology. His early work with the $X\alpha$ method[22] helped to explain diverse complex phenomena in the area of transition metal clusters, their electronic and magnetic properties, and their use as models for chemisorption and catalysis. Explanations emanating from calculations on the reduction of surface mag-

netism following chemisorption and on the lack of thermal expansion in the Fe–Ni Invar alloys are noteworthy from this early period. Salahub was among a handful of workers who recognized early the possibility of bringing DF methods into mainstream quantum chemistry. His group has made seminal contributions to incorporating quantum chemical techniques into the DF methods and software: efficient integral schemes, model core potentials, and analytic gradients have helped to extend the range of applications. He has shown the importance of incorporating nonlocal corrections to the density functionals for treating weaker interactions such as hydrogen bonds.[23] Recently, accurate values for NMR chemical shifts,[24] and even spin-spin coupling constants, have been calculated. Salahub's research has helped to bring the formal and theoretical strengths of density functional theory (DFT) to bear on both qualitative and quantitative aspects of real chemical and physical problems. Recent efforts are aimed at describing chemical reactivity in complex environments; the efforts of the past years have advanced the tools to a state at which this is now a realistic, though ambitious, goal. Salahub's group is making noteworthy contributions to a wide spectrum of frontier issues[25] associated with DFT: new improved functionals have been proposed, tested, and implemented, along with others, in the code suite deMon, which was developed in Montreal and is in use at dozens of labs around the world. In the context of the deMon program, a fusion of DFT with other techniques (reaction fields, molecular dynamics, etc.) is under way. These advances in methodology and techniques have vastly extended the scope of DFT, and this is bearing fruit in Salahub's laboratory in the area of transition metal chemistry[26] and biomolecular modeling. Salahub moved to the Steacie Institute for Molecular Sciences of the National Research Council of Canada in July 1999. This will give him expanded opportunities to direct research with the aid of post doctoral fellows.

The University of Montreal made a very significant commitment to theoretical and computational chemistry with the appointments of Greg Corey (1987), Tucker Carrington Jr. (1988), Benoit Roux (1991), and John Gunn (1994). Carrington's research deals primarily with the development of new methods to calculate vibrational and rotational–vibrational energy levels of polyatomic molecules. He and his coworkers have developed a potential optimized discrete variable representation (PODVR) to choose DVR basis functions adapted to the potential.[27] In a series of papers,[28] Carrington's group has shown that if matrix–vector products are evaluated efficiently, the Lanczos algorithm becomes an attractive alternative to standard Householder methods for calculating vibrational spectra. They have also compared carefully the advantages of using contracted or direct product basis sets with the Lanczos algorithm.

Benoit Roux's group is using molecular dynamics (MD) simulations to elucidate the fundamental principles governing the transport of ions with a special interest in constructing detailed atomic models of the gramicidin channel.[29] They also use MD simulations and free energy methodologies to study

multiple occupancy effects on the conformation of the gramicidin channel in phospholipid bilayers. John Gunn is also interested in the statistical mechanics of complex systems. He is especially interested in the development of new algorithms and potential functions for protein structure prediction and refinement.[30]

University of British Columbia (1957)

With Charles McDowell as the new head and a mandate to build a strong Department of Chemistry, the University of British Columbia (UBC) was the second Canadian university to make a serious commitment to theoretical and computational chemistry. John Coope and Bob Snider were appointed in 1957 and 1958, respectively. Coope had an early interest in semiempirical theories of the electronic structure of molecules and the solid state, and particularly in deeper theories of why simple semiempirical theories may work, including effective Hamiltonian, and spin wave interpretations of valence bond theory. He has been concerned with various mathematical aspects of molecular physics, including effective Hamiltonians, the formulation of resonance conditions, perturbation theory based on characteristic equations, and topological aspects of polycrystalline spectra. Much of his work has focused on the effects of electric and magnetic fields on molecules, including the effects of fields on molecular transport properties, and his findings were published in papers often coauthored with R. F. Snider. Their papers on irreducible Cartesian tensors are good examples[31-33] of their research during the 1960s.

Snider is best known for his paper reporting what is now referred to as the Waldmann–Snider equation.[34] (L. Waldmann independently derived the same result via an alternative method.) The novelty of this equation is that it takes into account the consequences of the superposition of quantum wavefunctions. For example, while the usual Boltzmann equation describes the collisionally induced decay of the rotational state probability distribution of a spin system to equilibrium, the modifications allow the effects of magnetic field precession to be simultaneously taken into account. Snider has used this equation to explain a variety of effects including the Senftleben–Beenakker effect (i.e., is, the magnetic and electric field dependence of gas transport coefficients), gas phase NMR relaxation, and gas phase muon spin relaxation.[35]

Other novel work of Snider includes a paper[36] on the kinetic theory of recombination and decay, which covers the whole range of concentrations from pure monomers to pure dimers. Thus it becomes possible to describe, in a uniform and consistent manner, the dynamical properties of a reacting gas of monomers and dimers. As well, the 1974 paper[36] led to an understanding of the differences in predicted kinetic behavior if a dimer is treated as a pair of monomers.

Some of Snider's more recent papers[37] question the validity of earlier approaches to the treatment of the density corrections to ideal gas kinetic

theory. Questions arose because the quantum formulation of the Boltzmann equation emphasizes, by its basic nature, aspects different from those stressed in the classical formulation, pointing to questions of consistency in the earlier treatments.

On the occasion of the Canadian Centennial in 1967, Bob Snider wrote an interesting article on theoretical chemistry for *Chemistry in Canada,* which is no less valid today than it was a third of a century ago. Three paragraphs are reproduced here for their historical significance.[38]

> In its broadest interpretation, theoretical chemistry includes any attempt to devise an explanation, however qualitative or quantitative, of any experimental chemical result. As such, a theory can serve two very different roles. Firstly, a theory provides a set of concepts and a language in which experiments can be discussed; secondly it may provide a tool for the quantitative prediction of experimental results.

> The first use of a theory is by now quite well developed in the sense that any new experimental discovery is fairly rapidly explained qualitatively, in fact, there are often several rival explanations. With such theories, it is sometimes possible to correlate different experimental results quantitatively with the use of a few, hopefully only one or two, empirical parameters. This is always beneficial to science as it organizes and summarizes large bodies of data. However, since we now believe that all of chemistry can be explained in terms of quantum mechanics, one must go further and deduce these empirical relations from basic principles. This either verifies the qualitative theory or replaces it by a more rigorous one and at the same time may give numerical values for the empirical parameters. It may be thought that this is merely a job for a computer and the theoretician is necessary only to write the computer program. In fact, computers are of great assistance in solving specific problems in a short time. But even if computers grow large enough to solve all the chemist's problems, someone will be required to understand the answer so that the answer can be applied. This, of course, can be quite difficult if the result is expressed as a million numbers which could not possibly be memorized much less understood. Thus the result of a calculation must be in a form in which it is useful—which again means in terms of simple equations between experimental quantities. Here then is the real work for the theoretician—to start off with basic equations such as the Schrödinger equation and derive from them *simple* relations which may be compared with experiment.

> Theoretical chemistry as its own specialized field is growing very rapidly throughout the world. One prediction of its future[39] is very easy to see, namely, that a much greater use will be made of abstract mathematics, not

only in the mathematical manipulations that are performed but also in the whole language in which theoretical problems are described. On the other hand, with the increasing elaborateness of experimental methods it will be increasingly important for a theoretician to understand the experimental methods in order to correctly transcribe the experimental problem into theoretical terms. In short, the theoretical chemists of the future must have the best theoretical and experimental background that they can obtain if they are to keep up with this challenging but fascinating field.

Theoretical chemistry at UBC was further strengthened with the arrival of Delano Chong and Keith Mitchell in 1965 and 1966, respectively. Chong's interests in quantum chemistry have spanned the full range from semiempirical to ab initio molecular orbital methods. His long-standing interests in perturbation methods and constrained variations have figured prominently in his publications. He is probably best known for his attempts to calculate the X-ray and UV photoelectron spectra of molecules, often by means of perturbation corrections to Koopmans' theorem.[40] More recently he has shifted his focus to coupled pair functional methods and density functional methods, with a special interest in polarizabilities and hyperpolarizabilities.[41]

Although Keith Mitchell's initial research was in quantum chemistry, in the early 1970s he became interested in the possibility of developing low energy electron diffraction (LEED) to learn about the details of structure at well-characterized single-crystal surfaces. The research of his group has given new insight into chemical bonding at surfaces. In particular, for chemisorption of electronegative atoms on transition metal surfaces, four types of situation have been recognized as being capable of giving ordered commensurate structures: (1) simple chemisorption, where metal atoms relax in the vicinity of the electronegative atom,[42] (2) surface compound formation,[43] (3) an independent reconstruction,[44] and (4) underlayer formation.[45] Although Mitchell and his coworkers note that some initial guidance for the choice of structural type can be obtained from Pauling-type bond length arguments, new theoretical approaches would be advantageous to give deeper insights into the relaxations and reconstructions identified experimentally in the surface structures.

When Bernie Shizgal arrived at UBC in 1970, his research interests were in applications of kinetic theory to nonequilibrium effects in reactive systems. He subsequently applied kinetic theory methods to the study of electron relaxation in atomic and molecular moderators,[46] hot atom chemistry, nucleation,[47] rarefied gas dynamics,[48] gaseous electronics, and other physical systems. An important area of research has been the kinetic theory description of the high altitude portion of planetary atmospheres, and the escape of atmospheric species.[49] An outgrowth of these kinetic theory applications was the development of a spectral method for the solution of differential and integral equations referred to as the quadrature discretization method (QDM), which has been used with considerable success in statistical, quantum, and fluid dynamics.[50]

UBC's strong tradition for statistical mechanics was given another major boost when Gren Patey arrived in 1980. Within a few years he established a reputation as a leading researcher on the theory of liquids and solutions, and more recently interfacial phenomena.[51] His group uses simulation techniques to investigate the equilibrium and dynamical properties of liquids,[52] solutions[53] and molecular clusters.[54] Their specific interests include the dynamics of ion solvation, the forces between immersed macroscopic objects, the coagulation of colloidal suspensions, and the closely related membrane fusion processes of importance in biological systems. In some of their simulations of the phase behavior of liquid crystals, they demonstrated that ferroelectric nematic liquid crystals can exist.[55]

Mark Thachuk joined the UBC Department of Chemistry in 1996. His research program focuses on the study of the dynamics and rates of chemical reactions and processes by mathematical and computational techniques. Typically, such investigations utilize classical, semiclassical, or quantum mechanics, and combine scattering theory with reaction rate and kinetic theories.

University of Alberta (1959)

Theoretical and computational chemistry at the University of Alberta started in 1959 when Fraser Birss arrived in Edmonton. Within a few years he was joined by two excellent graduate students, Bill Laidlaw and Reg Paul, both of whom later started theoretical and computational chemistry at the University of Calgary. Serafin Fraga was a postdoctoral fellow with Birss in 1961–1962 and returned a year later to join the faculty. Fraga has suggested that being a theoretician in Alberta in the late 1950s was probably a difficult experience for Fraser Birss. Fortunately the head of the department was Harry Gunning, who had been brought from the Illinois Institute of Technology to transform the Department of Chemistry into a major research center. The fact that Gunning had been in the city of Chicago (which at the time was a preeminent center for theoretical chemistry) may have accounted for his support of work in that field. In the early 1960s all calculations had to be done with a Marchant electrical calculator. (Somewhere in the Department of Physics there was an LPG-30 computer, but it was not available to the theoretical chemists.) It was during Fraga's postdoctoral year at Alberta that he and Birss developed a general formulation for performing self-consistent field calculations on open-shell configurations.[56]

In 1964 Birss and Fraga finally got a real computer, an IBM 1620, which they operated themselves, first with paper tape and finally with cards. As theoreticians they did not have much of an identity within the Department of Chemistry and therefore with Gunning's approval they formed the Division of Theoretical Chemistry. To make the division better known, they organized a conference in 1965 called the Alberta Symposium on Theoretical Chemistry. Fraga recalls that the budget was really small: about $35 for accommodation

and meals in the university residences! He notes that "altogether it was a terrific experience, although I decided that I would never organize another meeting." With that seminal conference, he and Fraser Birss had put Canada on the theoretical chemistry map by bringing strong contingents from the Chicago group and the Hirschfelder group to Alberta's capital. By the time the Symposium on Theoretical Chemistry returned to Alberta in 1980, it had become the Canadian Symposium on Theoretical Chemistry, a triennial event with an excellent reputation as already mentioned.

In the late 1960s, the University of Alberta, with Harry Gunning as head of the Department of Chemistry, decided to build a center of excellence in theoretical chemistry by hiring Sigeru Huzinaga, Walter Thorson, and Bruce Clarke in a two-year period.

Sigeru Huzinaga, one of the pioneers in the development of Gaussian basis sets for molecular calculations, held the position of associate professor in physics at Kyushu University in the 1960s. During that time he was invited by R. S. Mulliken to join the research group at the Laboratory of Molecular Structure and Spectra at the Department of Physics of the University of Chicago, where, under Mulliken and C. C. J. Roothaan, he participated in the inception of computational quantum chemistry (1959–1961). He was appointed professor of chemistry at the University of Alberta in 1968.

The scientific interests of Huzinaga are numerous. He initially worked in the area of solid-state theory. Soon, however, he became interested in the electronic structure of molecules. He studied the one-center expansion of the molecular wavefunction, developed a formalism for the evaluation of atomic and molecular electron repulsion integrals, expanded Roothaan's self-consistent field theory for open-shell systems, and, building on his own work on the separability of many-electron systems, designed a valence electron method for computational studies on large molecules.

Very early Huzinaga recognized the usefulness of Gaussian-type functions for the evaluation of molecular integrals. Subsequently, for 25 years he was engaged in the development of Gaussian basis sets for molecular calculations. His 1965 paper,[57] which contained the first comprehensive compilation of Gaussian-type functions for the atoms H through Ne, was designated a Citation Classic by *Current Contents* in April 1980. The use of analytical basis sets in molecular calculations led Huzinaga to studies on the nature and manipulation of virtual space and on the role of the continuum in the superposition of configurations. He has written two books on the molecular orbital method and edited a compendium of Gaussian basis sets for molecular calculations.[58]

In February 1992 the *Canadian Journal of Chemistry* honored Sigeru Huzinaga on the occasion of his sixty-fifth birthday with a special issue containing more than 50 papers by his coworkers, students, colleagues, and friends from many countries. The special issue, edited by Russell Boyd and Mariusz Klobukowski, is a fitting tribute to celebrate 40 years of achievement in theoretical chemistry and physics.

Huzinaga was the recipient of the 1994 John C. Polanyi Award of the Canadian Society for Chemistry. In his award lecture he described his model potential method, which deals only with the active electrons in molecular and solid state calculations. An invited review article,[59] based on his 1994 Polanyi Award lecture, chronicles his efforts to develop a sound theoretical framework for the core–valence separation of electrons, a problem Van Vleck and Sherman[60] once referred to as "the nightmare of the inner core."

In 1968, after 10 years on the faculty at MIT, Walter Thorson was appointed professor of chemistry at the University of Alberta. The research problems that attracted his attention and for which he developed insight and skill involved quantum dynamics of few-body systems or systems with a limited number of degrees of freedom, focused especially on cases of motions that were not separable or only poorly separable. The choice of few-body systems was always based on the idea that rigorous critical studies could provide important paradigms valid for application to more complex systems. Many such studies concerned the theory of atomic collisions and focused on interactions of electronic and heavy particle motions. This was the most fruitful area of research for Thorson and his coworkers; the concepts and insights they developed proved useful not only in collision theory itself, but also in applications to problems in the spectroscopy and properties of small molecules. A secondary topic was the vibrational dynamics of hydrogen-bonded systems, where proton motion is highly nonharmonic and strongly coupled to other vibrational motions of the system. His group made some exhaustive studies of the strongly H-bonded bifluoride ion [FHF⁻] as a paradigm that rigorously explored the dynamics and its consequences for the IR spectra of such a coupled system;[61] a later classical dynamics study[62] on the same system showed the relation between such coupled quantum states and classical resonances.

Thorson's interest in fundamental studies of atomic collision processes in the slow collision regime (up to 25 keV collision energies) arose from recognition that while the theoretical context of the problem of electronic states of stable molecules had become well defined by 1960, there was then no corresponding conceptual framework for describing what happens (for example) when a proton collides with a ground state hydrogen atom. The adiabatic or "Born–Oppenheimer" separation of electronic motions from heavy particle motions in a stable molecule is clearly relevant in some way to the physics of collision between two atomic or molecular systems. However, an electron bound to an atom in a colliding system has translational kinetic energy and momentum with respect to the system center of mass merely by being a passenger on the partner to which it is bound, and an explicit account of this is needed to rationalize the dynamics of excitation, charge transfer, or other processes. Thorson made fundamental contributions to the theory of such "electron translation factors" and their effects on collision processes in primary model systems, showing how the theory can be systematized to take such effects into account.[63]

In reflecting on the Division of Theoretical Chemistry at the University of Alberta, Walter Thorson has written to this author:

> Until his untimely death in 1987, a vital ingredient in my own work and experience at Alberta was the presence and fellowship of a kindred spirit in Professor Fraser W. Birss, with whom I had many fruitful discussions and who shared with me a similar outlook on theoretical chemistry, the problems of computational chemistry and the aims of educating students in science. While Fraser and I never actually wrote a paper together, we influenced each other's works and methods, and for me and my students the climate of theoretical and computational chemistry at Alberta was shaped in large measure by that collegiality. I would hazard that every one of the theoretical and computational chemists at Alberta during the period also shared this to some extent; Fraser Birss had the gift of stimulating a sense of value in ideas for their own sake, and it was partly his vision which imagined the possibilities in bringing such an unusual number of theoreticians together in one department.

Nearly four decades after the arrival of Fraser Birss and the recent retirements from active teaching of Serafin Fraga, Sigeru Huzinaga, and Walter Thorson, it appears that the University of Alberta has abandoned its commitment to establishing one of the largest theoretical chemistry groups in Canada. Only Bruce Clarke and Mariusz Klobukowski continue to teach and do research in theoretical and computational chemistry. Some of the aura of the 1965 symposium has been diminished. Hopefully, additional appointments will restore Edmonton to its former glory as one of Canada's major centers for theoretical and computational chemistry.

University of Ottawa (1959)

Richard Bader joined the Department of Chemistry at the University of Ottawa in 1959. He left Ottawa four years later to return to his alma mater, McMaster University. He was immediately replaced in Ottawa by David Bishop. The key theme of Bishop's theoretical and computational research has been its relevance to experiment and its reliability. In recognition of his relatively precise calculations of the properties of small atoms and molecules, Bishop is acknowledged to be the major player in theoretical nonlinear optics in Canada. His results not only have been cited as a benchmark for other calculations but also very often serve in the calibration of the experimental equipment itself.

Initially the properties calculated were energetic in nature and related to IR spectroscopic measurements.[64] Bishop's work was the first serious attempt to calculate the rovibronic energies of the hydrogen molecule and molecular ion without using the Born–Oppenheimer approximation (i.e., three- and four-body calculations). Many years later, this work is still cited and the relativistic

and radiative corrections are still used; in particular, his determination of the quadrupole moment of the deuteron remains the recommended value in the CERN handbook.

Bishop's attention turned to accurate calculations of electrical and magnetic properties, especially those of importance in nonlinear optics. Since most experiments in this field measure ratios, not absolute values, it is necessary to have a calculated value. Universally, Bishop's helium nonlinear optical properties are used. In the same field, he was the first to seriously investigate the effects of electric fields on vibrational motions, with a much-quoted paper.[65] His theory and formulation has now been added to two widely used computational packages: HONDO and SPECTROS. He has also derived a rigorous formula to account for the frequency dependence (dispersion) in nonlinear optical properties.[66] He used this theory to demonstrate that the anomalous dispersion in neon, found experimentally, is an artifact of the measurements.

Bishop has recently been studying the magnetic properties of small systems (including the Cotton–Mouton effect and the Faraday effect), once again providing accurate values with which the experiments can be judged. As well, his concerns with the effects of magnetic fields on vibrations have received widespread attention.

Since joining the University of Ottawa in 1993, Alain St-Amant has focused on developing methods specifically designed to treat large systems, especially biological molecules.[67] Whenever possible, he concentrates on methods that will be of use in both conventional Hartree–Fock and density functional programs. He is also involved in the development of linear scaling methods[68] for electronic structure calculations, an intensely competitive topic in computational chemistry.

University of Saskatchewan (1959)

When Lenore McEwen was appointed in 1959, she became the first woman named as a theoretical chemist on the faculty of a department of chemistry of a Canadian university. She maintained her interest in the electronic structures and spectra of nitrogen–oxygen compounds for several years after her arrival in Saskatoon, although most of her papers resulted from her postdoctoral research in Sandorfy's group.[69]

Following McEwen's resignation in June 1977, Paul Mezey took up the torch of computational chemistry in Saskatchewan. His research activities range from numerous new mathematical theorems on molecular stability, symmetry, chirality, reactivity, and molecular shape–complementarity to new computational methodologies that may be applied in many fields such as computer-aided drug discovery, toxicological risk assessment, and molecular engineering. His numerous papers and books discuss his attempts to describe the shapes of molecules[70] and to model chemical reactions at the molecular level.[71] He is especially interested in using shape analysis[72] of molecules in pharmaceutical compound design and in modeling large molecules such as proteins. He

developed macromolecular density matrix methods to image the electron density of proteins.[73] Related methods show promise for the calculation of various molecular properties, such as the forces acting on the nuclei of macromolecules, and provide computational tools for the study of protein folding. In 1993, Mezey's group reported[74] their thorough analysis of all 20 geometric conformations of β-alanine by means of the shape group method that Mezey had pioneered 10 years earlier. Their shape-similarity analysis of the stable conformations of β-alanine provides an important test case of their methodology and represents a level of complexity approaching that of typical conformational problems in computer-aided drug design.

Laval University (1961)

Laval University was one of the first Canadian universities to hire a theoretical chemist. Wendell Forst arrived in 1961 and developed a research program based on the theory of unimolecular reactions[75] and quantum chemistry. He maintained ties with experimental physical chemistry through a strong interest in mass spectrometry and gas phase kinetics. In many of his papers he sought analytical solutions to fundamental problems.[76] In 1986, after a quarter-century at Laval, he moved to the University of Nancy in France.

T. T. Nguyen-Dang joined Laval University in 1987. His research has concentrated on the nonadiabatic dynamics of simple molecules. In this context, he has contributed to the generalization of the Numerov–Fox–Goodwin algorithm[77] for multichannel Schrödinger equations involving nonadiabatic, kinetic couplings. He has also pioneered the development of high-order adiabatic representations using unitary transformations to resume first- or low-order nonadiabatic interactions.[78] These methods are being used to study the dynamics of laser-driven molecules. The dynamics and structure of molecules in intense laser fields is the second main topic of Nguyen-Dang's research, a theme he has maintained from his time at the University of Sherbrooke where, together with A. D. Bandrauk, he contributed seminal works on the adiabatic separation of dressed molecules within the Bloch–Nordsieck representation.[79] At Laval, continuous efforts in the directions listed above led to the development of original nonperturbative wave packet methods for the simulation of the dynamics of the laser-driven molecules.[80] Returning to the formal theory of dressed molecules treated completely within the a priori quantum electrodynamical representation of Coulomb systems, Nguyen-Dang and his group are seeking to develop symbolic tools for further analytical explorations of the high-order adiabatic representations they developed for these systems.

University of Toronto (1961)

The origins of computational and theoretical chemistry at the University of Toronto can be traced to two key appointments made by Professor Donald J.

Le Roy soon after his appointment in 1960 as head of the Department of Chemistry.

In 1961 John Valleau became the first theoretical chemist to join the faculty at Toronto. The subsequent appointments of Ray Kapral (1969), Stuart Whittington (1970), and Paul Brumer (1975) led to the formation of the Chemical Physics Theory Group, designed to complement the excellent experimental chemical physics group of John Polanyi and his colleagues. The group of theoreticians soon became the strongest group of its kind in Canada and comparable to the best in the world. In the same year that John Valleau joined the faculty at Toronto, Le Roy also appointed Keith Yates, an organic chemist with a keen interest in the kinetics and mechanisms of organic reactions. During the following two decades, Yates and his colleagues built up Canada's leading physical organic chemistry group. This achievement was, in part, due to the appointment of Imre Csizmadia to the faculty in 1964.

Although Csizmadia and Yates both received their Ph.D. degrees in organic chemistry from the University of British Columbia, their paths to joining the faculty of the University of Toronto have little in common. Keith Yates grew up in England and after leaving school joined the Royal Navy. After several years of travel, he enrolled at the University of British Columbia (B.A., 1956, M.Sc., 1957, Ph.D., 1959). Following the completion of his doctorate in physical organic chemistry under the supervision of Prof. Ross Stewart, he went to Oxford and earned a D.Phil. in 1961 as an NRC and NATO postdoctoral fellow. Imre Csizmadia completed his diploma in engineering in Budapest just prior to the Hungarian Uprising in 1956. Like many Hungarians he was granted refugee status in Canada and continued his studies at the University of British Columbia. Following the completion of his Ph.D. in organic chemistry, he joined Prof. John C. Slater's group at MIT in 1962. During his first year at MIT he completed the first ab initio calculation with a Gaussian basis set on an organic molecule (HCOF). Given their complementary interests, it was natural that Csizmadia and Yates would collaborate on many computational projects. The arrival of Tom Tidwell in 1972 brought the number of organic chemistry faculty members with interests in computational chemistry to three, until Keith Yates retired in about 1990.

John Valleau's research has focused on Monte Carlo simulations for liquids and solutions. Many of his papers deal with the application of Monte Carlo methods to the structure of electrolyte solutions,[81] the structure of double layers,[82] polyelectrolytes,[83] and the phase transition in Coulombic systems.[84] He made a major contribution with the invention of umbrella sampling.[85] He noticed that one can sample from distributions other than the Boltzmann distribution, and then reweight. The original idea was to use this insight to estimate free energy differences at different temperatures, but it is also extremely helpful for solving, or helping with, quasi-ergodic problems. These come from slow convergence of the Markov chain along which the sampling is being carried out. Often, using umbrella sampling will help speed up con-

vergence of the Markov chain to its limit distribution. It is now a widely used method. Valleau has extended the method so that it works simultaneously at several densities as well as at several temperatures (essentially a two-dimensional umbrella).

Imre Csizmadia is generally acknowledged to be the pioneer of ab initio theoretical organic chemistry in Canada. His interest in the optimization of Gaussian basis sets goes back to his postdoctoral research. The work of Csizmadia and his coworkers has been summarized in a book.[86] Protonation and deprotonation in both ground and excited states have also constituted a major theme in Csizmadia's work. His publications in this area reflect his strong ties to organic chemistry.[87] Many of the papers from his group are associated with stereochemistry and his interest in the classification of the topology of potential energy hypersurfaces. This research has led to an ongoing interest in the development of peptide models by means of ab initio calculations.[88]

Ray Kapral came to Toronto from the United States in 1969. His research interests center on theories of rate processes both in systems close to equilibrium, where the goal is the development of a microscopic theory of condensed phase reaction rates,[89] and in systems far from chemical equilibrium, where descriptions of the complex spatial and temporal reactive dynamics that these systems exhibit have been developed.[90] He and his collaborators have carried out research on the dynamics of phase transitions and critical phenomena, the dynamics of colloidal suspensions, the kinetic theory of chemical reactions in liquids, nonequilibrium statistical mechanics of liquids and mode coupling theory, mechanisms for the onset of chaos in nonlinear dynamical systems, the stochastic theory of chemical rate processes, studies of pattern formation in chemically reacting systems, and the development of molecular dynamics simulation methods for activated chemical rate processes. His recent research activities center on the theory of quantum and classical rate processes in the condensed phase[91] and in clusters, and studies of chemical waves and patterns in reacting systems at both the macroscopic and mesoscopic levels.

The mechanisms of reactions that occur in condensed phases involve the participation of solvent degrees of freedom. In some cases, such as in certain ion association reactions involving solvent-separated ion pairs, even the very existence of reactant or product states depends on the presence of the solvent. Traditionally the solvent is described in a continuum approximation by reaction–diffusion equations. Kapral's group is interested in microscopic theories that, by treating the solvent at a molecular level, allow one to investigate the origin and range of validity of conventional continuum theories and to understand in a detailed way how solvent motions influence reaction dynamics.

While many aspects of the theory of classical condensed phase rate processes are well developed, no computationally tractable scheme based on a theory with controlled approximations exists for quantum systems, or for mixed quantum–classical systems, where some degrees of freedom, typically those of the solvent, may be treated classically. Kapral's research in this area has

focused on the development of constrained MD schemes for classical and quantum reaction rates, path integral methods, and theories for mixed classical–quantum systems. These theoretical methods are used to study quantum reaction rates, such as proton transfer, in the condensed phase as well as in large molecular clusters,[92] where new chemical mechanisms arise. The cluster environment differs radically from the bulk, and therefore Kapral is interested in exploring the distinctive chemistry that occurs in solvent-influenced reactions in these small model systems. This research forms part of a larger study of reaction rates and mechanisms in the mesoscale domain.[93]

Stu Whittington's initial interests in statistical mechanics were focused on the use of Monte Carlo methods, especially for polymer problems.[94] Minor early interests included solution dimensions and shapes of polysaccharides, as a function of linkage type, and so on. He was also concerned with how solution dimensions of copolymers (such as alginate) are affected by the comonomer sequence statistics. He then moved into percolation theory[95] and its connection with gelation. In particular, his group showed that gels should have the same critical exponents as percolation, and not Flory–Stockmayer exponents. More recently he has worked on rigorous treatments of models of polymer adsorption, polymers in confined geometries, polymer collapse, knots and links in polymers, ribbon models of double-stranded polymers,[96] lattice animals as models of branched polymers, and related topics.[97]

The research of Paul Brumer and his colleagues addresses several fundamental problems in theoretical chemical physics. These include studies of the control of molecular dynamics with lasers.[98] In particular, the group has demonstrated that quantum interference effects can be used to control the motion of molecules, opening up a vast new area of research. For example, one can alter the rate and yield of production of desirable molecules in chemical reactions, alter the direction of motion of electrons in semiconductors, and change the refractive indices of materials etc. by creating and manipulating quantum interferences. In essence, this approach, called coherent control, provides a method for manipulating chemistry at its most fundamental level.[99]

Another interest of the Brumer group comes under the heading of classical and quantum chaos. The dynamics of most molecular systems is nonlinear and, if the nonlinearity is sufficient, the motion is chaotic (i.e., extremely sensitive to initial conditions). Brumer and his colleagues have explored the role of chaotic motion in chemical reactions and in isolated molecule dynamics as it relates to statistical theories of dissociation. Deep insights into isolated molecule dynamics have emerged. Their research program continues to focus on this area by exploring the role of chaos in quantum mechanics and in understanding quantum–classical correspondence in chaotic systems. This work also provides a general framework for the understanding of correspondence in all conservative systems.[100]

Theoretical computations on molecular dynamics require new developments in semiclassical mechanics to treat many degrees of freedom. Thus,

Brumer's group is examining numerous methods, based on the semiclassical initial value representation, to develop a highly effective and general scheme for studying molecular motion. They are also interested in time- and frequency-resolved laser experiments[101] that provide insight into the dynamics of atoms and molecules. Ongoing research in the group is dedicated toward developing a general computer-based approach to understanding these experiments and to learning what they tell us about molecular motion.

University of Waterloo (1961)

Quantum chemistry at the University of Waterloo began with the appointment of John Moffat in 1961, only 2 years after the university received its charter. While Moffat's primary research interests have centered on heterogeneous catalysis and surface chemistry, he had an interest in quantum chemistry since his time at DuPont (1956–1961). With the arrival of a mainframe computer (likely an IBM 650) at DuPont, he was asked if he would like to learn how to program it. He was thus initiated into basic machine language and began doing some simple molecular orbital calculations on nitriles. He continued this work at Waterloo. Among other topics, he worked on energy partitioning studies on nitriles.[102] A few years later Moffat and Popkie showed that the sharing interference energies of various bonds correlated with the dissociation energies.[103] The calculations were subsequently extended to surfaces.

The Soviet Union's military invasion of Czechoslovakia in August 1968 had a significant impact on the development of computational and theoretical chemistry in Canada. The troubles in Czechoslovakia led to Jiri Cizek and Joe Paldus accepting appointments at the University of Waterloo in 1968. Their many achievements include the first ab initio study of the coupled cluster method.[104]

Jiri Cizek's research program centers on the quantum theory of molecular electronic structure and related developments in quantum chemical methodology, coupled-cluster approaches to many-electron correlation problems,[105] large-order perturbation theory,[106] dynamical groups and exactly solvable models, lower bounds, and the use of symbolic computation language in physics and in chemistry.

For the treatment of electron correlation, Cizek uses classical techniques as well as techniques based on mathematical methods of quantum field theory, namely, a coupled-cluster approach. A rapid development and deployment of these methods during the past decade was stimulated by the realization of the importance of size consistency or size extensivity in the studies of reactive chemical processes. Although truly remarkable accuracy and development have been achieved for ground states of closed-shell systems, an extension to quasi-degenerate and general open-shell systems is most challenging. Cizek also works on the exploitation of these approaches to study the electronic structure of extended systems (molecular crystals, polymers[107]). His many interests in-

clude the development of methods for the estimation of upper bounds and the use of symbolic computations in quantum chemical calculations.[108]

In the fall of 1958, Josef Paldus started a CSc. (Ph.D.) program at the Institute of Physical Chemistry of the Czechoslovak Academy of Sciences in Prague, working under the supervision of Dr. J. Koutecky. He was Koutecky's first student to tackle the field of quantum chemistry and subsequently became one of the founders of the Prague theoretical school. In his CSc. thesis, Paldus succeeded in providing the first interpretation of electronic spectra of systems with nonbonding interactions, which were very puzzling to organic chemists at the time. His most significant contribution in this regard was a semiquantitative explanation of the optical spectra of [m, n]-paracyclophanes, barrelene, germacrol, and other systems involving so-called transannular interactions.

In 1962 Paldus was awarded a National Research Council of Canada Postdoctoral Fellowship and joined the Division of Pure Physics at NRC to work in the Larger Molecules Section (LMS) under the supervision of Dr. D. A. Ramsay. This was just at the time when new, high resolution spectrographs were being obtained at LMS and computers were beginning to help with the analysis of complex molecular spectra. Aided by these new tools, Paldus embarked on an analysis of the vibrational and rotational fine structure of glyoxal electronic spectra.

Paldus resumed his position in Prague in August 1964 to continue his work in quantum chemistry and the analysis of glyoxal spectra. He subsequently returned to Ottawa twice (1966 and 1968) as an NRCC Visiting Scientist, to complete work on the 0–0 vibrational band of the first singlet $n \to \pi^*$ transition in glyoxal, as well as to collaborate on the rotational analysis of vibrationally excited bands. This work laid the foundation for extensive studies of glyoxal spectra by Ramsay's LMS group.

During his 1968 stay at the NRCC in Ottawa, following the Soviet invasion of Czechoslovakia, Paldus decided to settle permanently in Canada. In December 1968, he accepted an offer of a visiting associate professorship in the Department of Applied Mathematics at the University of Waterloo, where he has remained to the present day; he obtained a regular appointment in September 1969 and tenure in July 1970. He was cross-appointed to the Department of Chemistry in June 1973.

Since his appointment at the University of Waterloo, Paldus has fully devoted himself to theoretical and methodological aspects of atomic and molecular electronic structure, while keeping in close contact with actual applications of these methods in computational quantum chemistry. His contributions include the examination of stability conditions and symmetry breaking in the independent particle models,[109] many-body perturbation theory and Green's function approaches to the many-electron correlation problem,[110] the development of graphical methods for the time-independent many-fermion problem,[111] and the development of various algebraic approaches and an exploration of convergence properties of perturbative methods. His most important

and influential work produced during the past two decades involves the development of the unitary group[112] and coupled-cluster[113] approaches to the many-electron correlation problem. These papers include several seminal contributions, which enabled the highly successful development of large-scale configuration interaction and both single-reference and multireference coupled cluster calculations; these works nowadays form a basis of many state-of-the-art methods of post-Hartree–Fock quantum chemistry.

Fred McCourt arrived at the University of Waterloo in 1970. His Ph.D. research had led to the first theoretical description of external magnetic field effects on the transport properties of gases of diamagnetic polyatomic molecules. These field effect phenomena were called Senftleben–Beenakker effects, after Herman Senftleben, who discovered such effects for gases of paramagnetic molecules, and Jan Beenakker, who first measured them for gases of diamagnetic molecules. The next few years involved a careful systematic extension of the perturbation expansion of the distribution function/density matrix for a polyatomic gas. This allowed the introduction of the now nearly universal "telephone number" notation for the identification of the many expansion terms involved.[114] The methods of analysis were extended to include electric field effects in gases of polar molecules (mainly symmetric or near symmetric top molecules) and effects associated with second gradients of flow velocity and temperature (the so-called Burnett effects). An essentially complete understanding of the thermomagnetic torque in rarefied gases was achieved in this way. These developments led to a unified description of transport properties and relaxation phenomena along lines first suggested by Roy Gordon (Harvard). Key relaxation phenomena considered were the relaxation of the bulk magnetization of diatomic gases (as probed by NMR) and the scattering of light, especially depolarized Rayleigh scattering, by such gases. An ease of comparison between the various transport and relaxation phenomena was achieved by expressing all experimentally measurable quantities in terms of the "telephone number," or effective cross sections, introduced in 1970.[115]

A second stage in the systematic development of transport and relaxation phenomena required an ability to evaluate the effective cross sections for a given potential energy surface describing the interaction between a pair of molecules. The extensive research of McCourt and his coworkers on these topics culminated in a two-volume monograph.[116] Once a sound theoretical description of transport and relaxation phenomena was in place and methods for calculating the effective cross sections were available, McCourt turned his attention to the problem of obtaining accurate potential energy surfaces for the interactions between the atoms and molecules. In many cases a number of empirical potential surfaces were available from inversions of various sets of experimental data, none of them particularly definitive, with the exception of the interaction of molecular hydrogen with the rare gases, for which Bob Le Roy had obtained reliable results. Higher level ab initio calculations have been employed to give

reliable potential energy surfaces for a series of second-row diatomic species with He, Ne, and Ar.[117]

Bob Le Roy, son of D. J. Le Roy, came to Waterloo in 1972. His research focused on the properties and dynamical behavior of small molecules and molecular clusters, and on the intermolecular forces that govern them. The techniques used include analytic derivations, detailed quantum calculations for dynamics on multidimensional potential energy surfaces, classical Monte Carlo or molecular dynamics simulations, and nonlinear least-squares fits to experimental data of various types.[118] Le Roy has a long-standing interest in the inversion of bound state or photodissociation data, and in the exploitation of the theoretically predicted characteristic near-dissociation behavior of simple molecules.[119] He uses the spectra of van der Waals dimers to obtain information about intermolecular forces, and in particular, about their anisotropy and dependence on internal coordinates. His group is developing accurate new methods for calculating the properties of these very floppy molecules and applying them to new spectroscopic data[120] and other types of measurement to determine accurate multidimensional potential energy surfaces[121] for systems such as He–HF and H_2–CO.

The structures and dynamical behavior of larger clusters consisting of a few to a few dozen atoms or molecules is of great interest because the properties of such aggregates are intermediate between bulk and isolated-molecule behavior, and their study promises insight into the molecular nature of solvation and phase transitions. Le Roy's classical simulations of these systems have been combined with computer visualization techniques and quantum predictions of the spectral perturbation of a chromophore "solute" molecule in such clusters, to relate their microscopic dynamics to experimental observables.

McGill University (1962)

Since his arrival at McGill University in 1962, Tony Whitehead has been known as one of the most colorful and entertaining members of the Canadian theoretical chemistry community. His appointment coincided with the establishment of *Theoretica Chimica Acta,* the first journal devoted specifically to the subjects of theoretical and computational chemistry. At the time, *TCA,* which was established in Germany, encouraged the publication of manuscripts in English, French, and German. The instructions to the authors indicated that Latin was also acceptable, presumably in keeping with the name of the journal. No articles written in Latin appeared in the first six volumes. However, the invitation to publish in Latin was a challenge to Whitehead and so with the help of a classicist he published a short note,[122] *Modus Computandi Eigenvectores et Eigenaestimationes e Matrice Densitatis,* in 1967. To assist readers lacking a knowledge of Latin, the abstract was also printed in the other three official languages of the journal. *TCA* was renamed *Theoretical Chemistry Accounts:*

Theory, Computation, and Modelling in 1997 and now publishes exclusively in English.

During the early years at McGill University, Whitehead's group concentrated on experimental nuclear quadrupole resonance spectroscopy[123] and a variety of π- and all-valence electron semiempirical molecular orbital methods.[124] His recent interests have included topics as diverse as density functional theory[125] and related topics,[126] and molecular models of surfactants.

The general themes of B. C. Eu's research are theoretical studies in nonequilibrium statistical mechanics, extended irreversible thermodynamics for systems far removed from equilibrium, generalized hydrodynamics, and related topics in transport properties in both simple and complex fluids.[127] Since Eu's arrival at McGill in 1967, his group has developed and studied kinetic theories of irreversible processes in fluids (liquids and gases), the kinetic theory foundations of irreversible thermodynamics,[128] and the kinetic theory of chemical reactions in solution, fluid dynamics, and rheology of complex liquids. In particular, chemical oscillations and waves in excitable media have been studied as part of a research program on nonlinear dynamics and generalized hydrodynamics. Mechanisms for pattern formations and selections and their irreversible thermodynamic bases have been investigated. Also, his group has studied extensively the equilibrium and nonequilibrium statistical mechanics of polymeric liquids.

The Eu group apply their theories of nonlinear transport processes and related irreversible thermodynamics to transport properties[129] of semiconductors, rheology of polymeric liquids and polyatomic liquids, and gas dynamics problems. Specifically, they calculate pair correlation functions and structure factors to correlate with experimental data on nonsimple liquids; various conductivities of charge carriers in semiconductors subjected to high external field gradients, and non-Newtonian viscosities of complex liquids subjected to high shear rates. In addition, kinetic theories of quantum gases and gases interacting with radiation are being investigated as part of the research program on irreversible thermodynamics and its kinetic foundations.

Bryan Sanctuary's research is based on nonequilibrium statistical mechanics, with emphasis on the applications. Before coming to McGill in 1976, he worked to extend the Boltzmann equation to a quantum form valid for nondegenerate internal states. There are a few applications for this, such as pressure broadening, the effects of electric and magnetic fields on the transport properties of gases (the Senftleben–Beenakker effect), and NMR. Thus Sanctuary entered into a general study of NMR at a time when the experimental field was rapidly advancing. His present research is aimed at providing a theoretical basis for nuclear magnetic resonance and nuclear quadrupole resonance spectroscopies by means of general relaxation theory. His group has developed numerical methods for the assignment of multidimensional proton spectra of amino acid residues in proteins.[130] The Sanctuary group studied the line shapes of the NMR spectra of solid H_2, D_2, and HD, and their dynamics. Analytical

solutions for a variety of spin systems were obtained[131] as part of research on the theory and application of NMR spin dynamics. The development of pulse sequences in NMR to understand echo formation and structure has also been investigated. Research on multipole NMR was summarized in a 1990 review.[132]

After eight years on the faculty at Harvard, David Ronis established in 1988 at McGill University a research program based on the nonequilibrium and equilibrium statistical mechanics in condensed complex systems. Specific examples include transport in membrane and zeolite channels, and the static and dynamic properties of suspensions of highly charged colloidal particles.[133] In many respects, dilute colloidal suspensions mimic atomic systems and can be used to study static and dynamic processes such as solidification, rheology, shear-induced melting, and shear-induced order or pattern formation. Ronis's group has developed several simple, but successful, theoretical models that explain many features of experiments on these systems.[134]

The interests of the Ronis group include correlations and conformations in polymer-coated colloids.[135] In particular, they are developing theories to describe the coupled intra- and interparticle interactions in systems comprising colloidal particles coated with charged or neutral, polymeric chains. This research is designed to answer several fundamental questions, including: What is the distribution of counterions in the polymer layer and in the space between the colloidal particles? They have developed a theory to explain the surface texturing observed on the surfaces of extruded plastics.[136] Their current research includes a study of turbulent fluid flow and its effects on the thermodynamics and kinetics of phase transitions in binary liquid mixtures using renormalization group methods.

Queen's University (1962)

In one sense, research in theoretical chemistry at Queen's University at Kingston originated outside the Department of Chemistry when A. John Coleman came in 1960 as head of the Department of Mathematics. Coleman took up Charles Coulson's challenge[150] to make the use of reduced density matrices (RDM) a viable approach to the N-electron problem. RDMs had been introduced earlier by Husimi (1940), Löwdin (1955), and McWeeny (1955). The great attraction was that their use could reduce the $4N$ space–spin coordinates of the wavefunctions in the variational principle to only 16 such coordinates. But for the RDMs to be of value, one must first solve the celebrated N-representability problem formulated by Coleman, namely, that the RDMs employed must be derivable from an N-electron wavefunction.[151] This constraint has since been a topic of much research at Queen's University, in the Departments of Chemistry and Mathematics as well as elsewhere. A number of workshops and conferences about RDMs have been held, including one in honor of John Coleman in 1985.[152] Two chemists, Hans Kummer [Ph.D. Swiss Federal Technical

Institute (ETH), Zurich, with Prof. H. Primas] and Robert Erdahl (Ph.D. Princeton with Prof. L. C. Allen), arrived in 1965 as postdoctoral fellow's with Coleman and stayed to become professors of mathematics.

In 1962, when Prof. R. L. McIntosh came to Queen's as the new head of the Department of Chemistry, one of the items on his agenda was to build up a strong group in theoretical chemistry. The first member was R. Julian C. Brown, who joined Queen's University in September 1962 and began research and teaching in theoretical chemistry. He had just completed his Ph.D. at the University of Illinois under the supervision of H. S. Gutowsky; his thesis work was the theoretical analysis of spin-relaxation measurements in a liquid. His research at Queen's continued in the areas of molecular dynamics in fluids and nuclear spin relaxation.[153,154] He established a graduate course in quantum theory, in which, in those early days of NMR, the emphasis was placed on the quantum theory of spin systems in addition to the theory of electrons in atoms and molecules. At Queen's it was natural for him to begin research in density matrix theory.[155] In 1966, he returned to his native Australia; but he came back[156] to Queen's in 1969 and began experimental research on crystals, particularly disordered ammonium salts. Recently he has turned to computational studies of molecular dynamics in crystals.[157]

In 1966 Neil Snider joined Queen's Department of Chemistry. He was followed by Douglas Hutchinson in 1964, Vedene Smith in 1967, and Hans Colpa in 1969 (see Table 2). Hutchinson left in 1984, while Colpa and Snider retired in 1991 and 1995, respectively. Meanwhile, Axel Becke and David Wardlaw arrived in 1984, Donald Weaver in 1989 and Natalie Cann in 1997.

During his time at Queen's University, Neil Snider was involved in theoretical research in two main areas: reaction dynamics and kinetics and equilibrium statistical mechanics of fluids. In the area of reaction kinetics, Snider's work dealt in large part with understanding the effect of the details of the energy transfer probability on the kinetics of gas phase reactions wherein the rates are determined either in part or entirely by energy transfer.[158] His work in the area of reaction dynamics was most notably directed toward understanding the dynamics of recrossing of reactant–product dividing surfaces by reactive and nonreactive classical trajectories.[159] In the area of equilibrium statistical mechanics of fluids, Snider devoted effort to extending the scaling theory of critical phenomena. Most of his work in this area, however, dealt with the theory of classical simple dense fluids. In particular, he sought ways in which generalized van der Waals equations of state might be applicable to real liquids,[160] and he looked to the justification of such equations on the basis of statistical mechanical perturbation theory.[161]

Although Doug Hutchinson's original interest was in semiempirical methods, some of his research at Queen's was concerned with the theory of interaction of electromagnetic radiation with matter,[162,163] an interest aroused during his postdoctoral time with Hank Hameka at Penn and stimulated further

by a sabbatical year with C. Mavroyannis at NRC. As well, Hutchinson had an interesting and fruitful collaboration on various problems in electron spin resonance theory with his colleague at Queen's, Jeff Wan.[164]

Vedene Smith came to Queen's in 1967 for one of the Conferences on Reduced Density Matrices and Their Applications and returned later that year to join the Department of Chemistry. It should be no surprise that density matrices and electron density functions in position and momentum space have played a large role in the research program of Smith and his group. Although people had previously considered momentum densities and the related experimental quantity, the Compton profile, Smith was the first to point out and implement the use of the Fourier transform of the one-electron density matrix as the route to the momentum density, instead of the procedure of transforming the entire wavefunction and then reducing the N-electron momentum density to the one-electron momentum density. Benesch and Singh[165] of the Smith group also showed the nature of the relationship between the momentum density and the charge density and introduced a function, today called $B(r)$, which was the 3D-Fourier transform of the momentum density and the 1D-Fourier transform of the Compton profile. For these reasons, it has proven very useful in the analysis and interpretation of experimental Compton profile data. Schmider and Smith[166] used $B(r)$ to develop a very neat treatment of the modulation of the Compton profile by a weak laser field.

Smith and his group have made a number of careful studies of the electron charge and momentum densities for atoms, molecules, and solids with respect to the role of electron correlation and bonding, topology, concentration regions, and shell structure. They have examined the intracule (relative electron–electron) and extracule (electron pair center-of-mass) distributions in both position and momentum space for ground and excited states at both SCF and CI levels. They were the first to do this at the CI level for more than two electrons. In addition to information about bonding and other properties, these pair densities are useful for developing exchange-correlation potentials with allowance for the kinetic energy contribution.[167] These researchers have also calculated elastic, inelastic, and total scattering of X-rays and fast electrons, efforts that have been useful for the calibration of experiment, elucidation of bonding effects, and assessing the effects of basis set quality and electron correlation. Recently they have investigated the role of relativity.[168]

The Smith group has also developed the methodology for making high precision calculations for small systems without invoking the Born–Oppenheimer approximation and have made calculations for two-electron atomic ions, small muonic molecules, and potentials of the screened Coulomb form. Their method for determining nonlinear parameters is now referred to as random tempering.[169]

Hans Colpa, known for his work on the pressure-induced rotational absorption spectra of hydrogen[170] and on hyperfine coupling constants, came to Queen's in 1969. The interpretation of Hund's rules for energy differences in

atomic and molecular systems, and their common misinterpretation, became one of his major initial research interests.

For more than 20 years, Colpa and his group had a very fruitful collaboration with the Max Planck Institute in Heidelberg and a successor research group in the Free University of Berlin. A primary interest of the work was in the mechanism of optical nuclear polarization (ONP), as observed in single molecular crystals at low temperatures. The particular importance of the level anticrossing (noncrossing) rule was stressed, and the impact of relaxation effects[171] was considered in some detail. In addition, a formal theory for microwave-induced ONP was derived.[172] The role of tunneling in the formation and decay kinetics of photochemical proton transfer in aromatic single crystals was studied by means of time-resolved ONP and optically detected (spin resonance) methods.[173]

Returning to his alma mater, Queen's University, Axel Becke has made significant advances in two research areas of quantum chemistry: the design of new computer algorithms[174] for the computation of molecular energies and structures, and development of new density functional theories[175] of the electronic structure of atoms, molecules, and condensed systems.

Becke's work on algorithms focuses on high numerical precision. Conventional quantum chemistry employs finite basis sets to represent electronic orbitals in molecules. Accuracy is limited by the nature and the size of the basis set, and systematic basis set extension is difficult. His approach, however, employs grids of several thousand points per atom to represent molecular functions.[176] In addition to superior flexibility, the precision of this grid-based method is straightforwardly controlled by adjusting the number of grid points. Becke's methodology is unique (though aspects of his procedure have been adopted by others) and is invaluable as a generator of benchmark results for the calibration of basis sets and theories. The product of this work is a full-featured molecular structure program called NUMOL, which he hopes to make available for general distribution.

About 14 years ago, Becke discovered that a certain class of DFTs gives remarkably good molecular energies[177] at significantly less cost than conventional non-DFT methods (i.e., correlated wavefunction methods). Indeed, density functional theory now holds great promise as the theory of choice for first-principles simulation of chemical reactions involving dozens or even hundreds of atoms, and of materials as well. Becke's goal has been, and still is, to refine the accuracy and reliability of DFT as far as possible. He has shown the way to density functional theories with precision approaching that of the best available quantum chemical methods (typical thermochemical errors of a few kcal/mol). Data extremely difficult to obtain experimentally (e.g., accurate reaction energy profiles) now lie within the reach of a highly economical computational/theoretical methodology.[178] Becke's recent theories have been incorporated into virtually every molecular structure program in the world, including major commercial packages.

David Wardlaw's research in theoretical chemical physics focuses on the dynamics of chemical reactions and related molecular processes. His research program has three predominant themes: fundamental development, design and testing of simple models and interpretation and simulation of selected experiments. His postdoctoral research with Marcus led to flexible transition state theory (FTST), which predicts rate constants for barrierless association reactions (e.g., two radicals, or ion + molecule), and the reverse dissociation reactions.[179] FTST forms a "new" branch of transition state theory parallel to the famous RRKM theory for reactions with barriers developed by Rice, Ramsperger, Kassel, and Marcus (RRKM). FTST remains a topic of active research at Queen's and elsewhere, and has been applied to numerous experimentally studied reactions. In the early 1990s Wardlaw and coworkers entered a relatively young but rapidly expanding field, the dynamics of chemical and physical processes under the influence of electric fields associated with very short, very intense laser pulses. Strong fields open up a domain of pathways to chemical products that cannot be explored by traditional chemical methods. This suggests the possibility of precise control over reactions, a long-pursued goal in chemistry. The Wardlaw group contributions to date[180] in this field are twofold: development and refinement of simulation methods (both quantum and semiclassical), and creation of simplified interpretive models. Other topics that have been investigated include quantitative descriptions of classical and quantum chaotic dynamics,[181] extensive contributions to the theory of time delay in scattering systems,[182] and developments in the modeling and interpretation of fall-off behavior (pressure dependence) of rate constants.[183]

The modeling and simulation components of research in the Wardlaw group are inextricably linked to various aspects of computational chemistry. At one end of the spectrum is the effort to render exact dynamics simulations feasible for systems with more than a few degrees of freedom. This requires fundamental developments in either the form or application of the equations of motion, yielding "new" or "improved" methodologies. A noteworthy example is their recently formulated "semiclassical" method (classical nuclei and quantum electrons) for treating the dynamics of molecules in intense laser pulses. At the other end of the spectrum is the development of computer programs or a suite of programs (software package). A pertinent example of the latter is the VARIFLEX software for the calculation of flexible transition state theory rate constants.

Don Weaver joined the Department of Chemistry in 1989. He holds the distinction of being the only computational chemist in Canada who is also a practicing physician; he is a part-time clinical neurologist and professor of medicine at the Queen's teaching hospitals. His research is at the chemistry–neuroscience interface, and he has concentrated on macromolecular modeling of brain molecules using empirical force field methods.[184] He has worked on new methods for searching conformational space, including genetic algorithms (GA)[185] and variable basis Monte Carlo (VBMC).

Weaver has developed force field parameterizations for phospholipids and various other biomolecules using ab initio molecular orbital methods. He has used semiempirical techniques to study amino acids, neurotransmitters, and various neuroactive drugs.[186] From an applied perspective, he has performed computational quantitative structure–activity relationship studies (QSARs) on a wide range of anticonvulsant drug molecules. His work has led to the design and patenting of new chemical entities for treating epilepsy and Alzheimer's disease. In conjunction with this design work, he cofounded Neurochem Inc. and Neuroceptor Inc. for the commercialization of his drug designs.[187]

Natalie Cann joined the Department of Chemistry at Queen's in 1997. She is developing a research program focused on the study of separations of mixed systems. In particular, she is using simulations and analytic theories to model simple chiral mixtures.

University of New Brunswick (1962)

Theoretical chemistry began at the University of New Brunswick in 1962 with the appointment of Fritz Grein. His early work focused on the development of methods to include a correlation factor $(1 + \alpha r_{12})$ in one-center wavefunctions in order to improve the ground state for two-electron molecules.[137] Next he turned his attention to the development of configuration interaction (CI) programs for the calculation of the excited states of diatomic molecules.[138] As noted earlier, he became interested in multiconfiguration self-consistent field methods (MCSCF) in 1965 as a consequence of his participation in the Edmonton conference. He developed the first MCSCF method with second-order convergence and later extended the method to excited states. One of his MCSCF papers[139] was selected by Fritz Schaefer[140] as one of the landmark papers of quantum chemistry for the period 1928 to 1983. For more than two decades, Grein has carried out many careful studies of the excited states of small molecules by means of multireference CI calculations. Many of the computations for these studies were completed during his annual sojourns in Bonn.

Some the best-known work of Grein and his coworkers involves the development of methods for the calculation of hyperfine coupling constants.[141] More recently the focus has shifted to calculating magnetic g-tensors from highly correlated wavefunctions. Grein's current interests include the study of stereoelectronic effects (such as the anomeric and reverse anomeric effects in acetal-like systems) in organic chemistry, a topic to which he has made important contributions.[142]

After four years at the University of Waterloo, Ajit Thakkar joined the Department of Chemistry at the University of New Brunswick in 1984, where he has maintained a very dynamic and productive research program centered around the quantum mechanical calculation of molecular properties and interactions. Some of his best known work is concerned with van der Waals forces and potential energy surfaces. His method for a generalized expansion for the

potential energy curves of diatomic molecules[143] is cited by spectroscopists as the Thakkar expansion. Thakkar and his coworkers have also published important papers on polarizabilities, hyperpolarizabilities, and related properties.[144]

Thakkar and his group have completed many careful studies of intracules, extracules, Coulomb holes, and related topics.[145] These position–space results are complemented by Thakkar's many studies of momentum densities and related quantities. He also has a long-standing interest in electron and X-ray scattering.[146] His current interests include the relationship[147] between the aromaticity of heterocyclic compounds and their polarizabilities, and the prediction of "push–pull" molecules that have a large nonlinear optical response and are thus candidates for materials to be used in optical computers.

Saba Mattar came to the University of New Brunswick in 1986. His research program is split between experimental and theoretical studies of the electronic structures and bonding in clusters and organometallic intermediates. Several experimental techniques are used to study matrix-isolated transient species, and the results are interpreted with the assistance of multireference CI calculations.[148] He also uses local density functional methods.[149]

McMaster University (1963)

Richard Bader was among the earliest of workers to realize the importance of electron density in providing an understanding of chemistry. Early on he was led to formulate the first symmetry rule governing a chemical reaction in answer to the question of how the electron density changes in response to a motion of the nuclei. This rule, termed the pseudo- or second-order Jahn–Teller effect, provides the theoretical underpinnings of frontier molecular orbital theory and is still widely used in discussions of reaction mechanisms and molecular geometries.

Beginning in the 1960s, Richard Bader initiated a systematic study of molecular electron density distributions and their relation to chemical bonding using the Hellmann–Feynman theorem.[188] This work was made possible through a collaboration with the research group of Professors Mulliken and Roothaan at the University of Chicago, who made available their wavefunctions for diatomic molecules, functions that approached the Hartree–Fock limit and were of unsurpassed accuracy.

The continuing study of molecular electron distributions led to the realization in 1972 that an atom, and the functional groups of chemistry, could be defined in terms of the fundamental topology exhibited by the electron density.[189] This work culminated in the demonstration that the predictions of quantum mechanics could be extended to the topological atom, using the new formulation of quantum mechanics afforded by the work of Feynman and Schwinger. In particular, Schwinger's principle of stationary action is used to identify the chemical atom with a proper open quantum system, one whose observables obey the correct equations of motion. Thus the functional groups

of chemistry are bounded space-filling objects whose properties are defined by the quantum mechanics of a proper open system, properties that faithfully recover the role of the functional group in chemistry.

The topology of the electron density also leads to the identification of a chemical bond with a line linking neighboring nuclei along which the electron density is a maximum.[190] This identification leads to a definition of molecular structure that is remarkable in its ability to recover all chemical structures. The dynamics of the density, as occasioned by nuclear displacements and analyzed by means of the mathematics of qualitative dynamics, leads to a complete theory of structural stability, one that clarifies the meaning of the making and breaking of a chemical bond.

The Laplacian of the electron density plays a dominant role throughout the theory.[191] In addition, Bader has shown that the topology of the Laplacian recovers the Lewis model of the electron pair, a model that is not evident in the topology of the electron density itself. The Laplacian of the density thus provides a physical valence-shell electron pair repulsion (VSEPR) basis for the model of molecular geometry and for the prediction of the reaction sites and their relative alignment in acid–base reactions. This work is closely tied to earlier studies by Bader of the electron pair density, demonstrating that the spatial localization of electrons is a result of a corresponding localization of the Fermi correlation hole.

The theory of atoms in molecules[192] recovers all the fundamental concepts of chemistry, of atoms and functional groups with characteristic properties, of bonds, of molecular structure and structural stability, and of electron pairs and their role in molecular geometry and reactivity. The atomic principle of stationary action extends the predictions of quantum mechanics to the atomic constituents of all matter, the proper open systems of quantum mechanics. All facets of the theory are predictive and, as a consequence, the theory can be employed in many fields of research at the atomic level, from the design and synthesis of new drugs and catalysts, to the understanding and prediction of the properties of alloys.

Richard Bader's seminal contributions to the field are beautifully documented in his most recent book[193] and celebrated in the June 1996 issue of the *Canadian Journal of Chemistry*. The special issue of nearly 60 papers, edited by Russell Boyd and Nick Werstiuk, was published on the occasion of Bader's sixty-fifth birthday.

David Santry arrived at McMaster in 1967 following a postdoctoral fellowship with John Pople during the semiempirical, all-valence-electron period at Carnegie-Mellon University. He pursued a variety of research topics relating to electronic structures but is probably best known for his long-standing interest in infinite three-dimensional networks of hydrogen molecules[194] and time-dependent Hartree–Fock theory.[195] Many of his papers are concerned with molecular polarizabilities and hyperpolarizabilities.[196]

Since his arrival at McMaster in 1988, Randall Dumont has focused on statistical theories and their origin in quantum and classical mechanics. His interests include the development of Monte Carlo implementations of statistical theory wherein dynamical processes are simulated by random walks on potential energy surfaces. The breakdown of statistical theory and the appearance of nonexponential population decay are also topics of his ongoing investigations. Other questions of interest are the incorporation of quantum effects into statistical theory and the effects of collisions on reaction processes. He has a special interest in argon cluster evaporation in vacuum[197] and in the description of simple isomerization reactions.[198] His other interests include the semiclassical description of classically unallowed processes such as tunneling.[199]

University of Calgary (1964)

Theoretical chemistry at the University of Calgary began with the arrival of Bill Laidlaw in 1964, two years before the university gained full autonomy from the University of Alberta. In his early work, Laidlaw focused on the applications of quantum chemistry to spectroscopic and structural problems. With teaching responsibilities in physical and theoretical chemistry, he wrote a textbook[200] designed to introduce students to the world of quantum mechanics through spectroscopy. Later he became interested in symmetry breaking and molecular orbital instabilities in sulfur–nitrogen compounds.[201] His continuing interest in the transition of open thermodynamic systems from a state with one symmetry to another state with a new symmetry is reflected in several of his papers. His many contributions include an important paper on light scattering.[202] In recent years he has shifted the focus to applications of theoretical chemistry to a myriad of problems such as flow in porous media, enhanced recovery of trapped fluids in oil reservoirs, flow of heat in fruit, enzyme degradation in fruit, flight patterns of insects in forest environments, and mortality of insects in quarantine treatments. His work is truly a basket of delight, but nearly all of it linked to fluid mechanics modeling[203] and simple models for kinetic processes.

Laidlaw was joined by Reginald Paul, his former colleague from the University of Alberta, in 1966. Paul initially worked on the application of field theory to chemical physics. His many papers during the 1970s culminated in the publication of a book on the subject.[204] Subsequently, he became interested in studying the interaction of electromagnetic fields with biological cells, and also in theoretical electrochemistry.

The number of theoreticians at the University of Calgary doubled in 1970 with the arrival of Arvi Rauk and Rod Truax. Rauk has made many important contributions to theoretical organic chemistry. Perhaps his most significant contribution arises from his long-standing interest in the chiroptical properties of molecules, that is, properties connected with the handedness and optical

activity of molecules. These properties are important for many applications including asymmetric synthesis, drug design, and even optical recording devices. The work of the Rauk group has played a significant part in the analysis of vibrational circular dichroism spectroscopy,[205] the only spectroscopic tool for the determination of absolute configurations of molecules.

In the early 1970s Rauk and his coworkers derived the formalism to obtain optical rotatory strengths from CI wavefunctions and wrote the first ab initio computer program to calculate them. Subsequently the origins of optical activity in chiral carbonyl compounds and several other compounds were explained, and in the 1980s, Rauk's interest turned to vibrational optical activity.[206] He adapted the vibronic coupling theory of Nafie and Freedman for ab initio calculation and wrote a computer program (VCT90) that has been widely used to calculate the vibrational rotatory strengths of compounds from first principles. His group derived sum rules for the phenomenon,[207] and elucidated the role of electron correlation and the molecular force field in determining the signs and intensities of the circular dichroism of all vibrational transitions. Collaboration with experimentalists permitted comparison of theory and experiment, and also enabled the Rauk group to demonstrate the usefulness of theory for understanding conformational and compositional properties of synthetic polymers, proteins, and nucleic acids.

Although all his research applications are in the realm of ab initio electronic structure theory and computations, Rauk has taught the theory of structure and bonding from a frontier orbital point of view. He is the author of a graduate-level textbook on the orbital interaction theory of organic chemistry.[208]

The research interests of Rod Truax fall under the general heading of symmetry and supersymmetry and their applications to problems of chemical and physical interest. He is especially interested in finding the symmetry associated with time-dependent models and exploiting this symmetry to compute solutions to the quantum mechanical equations of motion.

The number of theoretical chemists at the University of Calgary grew to five with the appointment of Tom Ziegler in 1981. He was no stranger to the Department of Chemistry, having received his Ph.D. in 1978 for research on new computational methods based on density functional theory carried out under the supervision of Arvi Rauk. Ziegler quickly established a major research group in the general area of theoretical inorganic chemistry and the development of new computational methods based on DFT.

Ziegler is best known as an early proponent of DFT[209] and its practical applications. His 1994 Alcan Award lecture[210] gives a masterful account of approximate density functional theory as a practical tool in studies on organometallic energetics and kinetics. He discusses electronic excitations and ionizations, electron capture, conformational changes, molecular vibrations, bond energies, and reaction profiles.

His group implemented some of the first DFT methods for the calculation of bond energies, molecular structures, and reaction paths, and developed together with the group of Evert Jan Baerends (the Free University in Amsterdam) the Amsterdam density functional package (ADF). Ziegler's group also implemented DFT methods for the calculations of NMR and ESR parameters.[211] Early on Ziegler demonstrated the ability of nonlocal DFT to provide accurate structures and bond energies in transition metal complexes. He has applied DFT extensively to elementary reaction steps in homogeneous catalysis. More recently the Ziegler group has included steric bulk, solvation, and first-principles molecular dynamics[212] in their study of elementary reaction steps.

University of Western Ontario (1965)

The University of Western Ontario built up a strong theoretical chemistry group in the short span of four years with the appointments of Bill Meath and Patrick Jacobs in 1965, Colin Baird in 1968, and Alan Allnatt the following year.

After 14 years on the faculty of Imperial College, Jacobs moved from London, England, to London, Ontario, where his research program focused on the optical and electrical properties of ionic crystals, as well as on the experimental and theoretical determination of thermodynamic and kinetic properties of crystal defects.[213] Over the years his research interests have expanded to include several aspects of computer simulations of condensed matter.[214] He has developed algorithms[215] for molecular dynamics studies of non-ionic and ionic systems, and he has carried out simulations on systems as diverse as metals, solid ionic conductors, and ceramics. The simulation of the effects of radiation damage is a special interest. His recent interests include the study of perfect and imperfect crystals by means of quantum chemical methods. The corrosion of metals is being studied by both quantum chemical and molecular dynamics techniques.

As noted elsewhere in this chapter, 1965 was a very significant year for the development of theoretical and computational chemistry in Canada. After completing his Ph.D. under the supervision of J. O. Hirschfelder at the University of Wisconsin, Bill Meath immediately took up an appointment at the University of Western Ontario, where he developed a strong research program in three, often interconnected, areas.

His early work on atomic and molecular properties and dispersion energies involved the development and application of ab initio pseudostate techniques for the reliable evaluation of atomic and molecular multipolar properties and dispersion energies for small species.[216] This was followed by the development and application of practical constrained dipole oscillator strength (DOSD) techniques, based on a combination of experimental and theoretical input, for the reliable evaluation (errors < 1–2%) of the dominant dipolar

properties and dispersion energies of a wide variety of large atoms and, in particular, molecules. These results, which have found application in a variety of areas from radiation research to the study of the additivity of molecular properties, furnish input for the construction of potential energy surfaces for atomic and molecular interactions. More recently the constrained DOSD methods have been extended to the reliable determination of anisotropic dipolar molecular properties and dispersion energies.

A major contribution of Meath's work on intermolecular forces and interaction energies has involved investigation of charge overlap effects in atomic and molecular interactions leading to representations of the correction terms ("damping functions") needed to extrapolate the well-known results for long-range interaction energies to small values of the intermolecular distance. This, together with reliable results for the long-range energies, in particular the dipolar dispersion energies, has led to the development of successful models, including an exchange-Coulomb model, for both isotropic and anisotropic intermolecular potential energies.

In his research on laser–molecule interactions, Meath has centered his efforts around the development of efficient numerical computational methods for dealing with the interaction of continuous wave and pulsed lasers with molecules.[217] More recently the research has focused on the effects of molecular permanent dipoles and molecular structure, on laser–molecule interactions, and on the use of two-color laser–molecule interactions to control the population of molecular states.[218] The derivation and use of analytical rotating wave approximation results for both one- and two-color laser–molecule couplings, which include the effects of permanent dipoles, has played an important role in this work.[219] For example, for one-photon transitions the molecule–laser couplings actually decrease with increasing laser field strengths if the molecule has permanent dipole moments relative to the expected increase with increasing field strength for atoms. This leads to drastic molecular effects in the resonance profiles, and in the associated molecular state dynamics, for such transitions. For multiphoton transitions, both one- and two-color, the effects of permanent dipoles are also pronounced.

After participating in the development of the MINDO method in Michael Dewar's group, Colin Baird initiated a research program at Western Ontario based on ab initio calculations on the ground and excited states of molecules.[220,221] Over the years his interests shifted more to chemical education, with a special interest in developing concepts and materials for environmental chemistry.

Alan Allnatt's research interests at Western Ontario have been concerned with the statistical mechanics of the transport of matter through crystals. His earliest work centered on obtaining methods for calculating the equilibrium distributions and thermodynamic properties of the point defects (vacancies, interstitials, solutes) that make transport possible. He first studied dilute systems, so the methods could be largely analytical. The methods for ionic crystals,

Allnatt's main interest, were based in part on extensions to lattice models of Mayer's cluster methods for electrolyte solutions. For solids with low dielectric constants, these methods were best combined with a new exact formulation of the mass action equations for defect equilibria.[222] At higher concentrations, purely numerical methods are required. Between 1980 and 1986 Allnatt's group developed inexpensive methods for calculating equilibrium radial distribution functions for defects such as vacancies and solute ions in ionic crystals using lattice versions of the hypernetted chain equations.[223] At the same time, they set up a general linear response theory for use with the particle-hopping models used to describe matter transport in crystals. Formal expressions for the Onsager phenomenological coefficients in this theory have been a fruitful starting point for both analytical and simulation studies, some of which are described in a monograph.[224]

Allnatt's group has also developed a method for using Monte Carlo simulation to calculate the Onsager phenomenological coefficients. The method has been used to provide detailed numerical information on interacting lattice gas models of transport in concentrated alloys and highly defective systems. Furthermore, these workers have developed a second simulation method,[225] which gives not only the transport coefficients but also the underlying time correlation functions.

York University (1965)

Many contributors to the field of computational chemistry were initially trained as experimentalists. Often their research interests were shaped by the circumstances in which they worked. Huw Pritchard is a case in point. After completing a Ph.D. in experimental thermochemistry and establishing a successful career as a theoretical chemist at the University of Manchester, he emigrated across the Atlantic Ocean in 1965 to then newly established York University in suburban Toronto. At York, he began to reestablish a program in computer applications in traditional theoretical chemistry. The promised computing facilities were a long time coming, and so he gave up wavefunctions for reactions and developed a research program with the emphasis on kinetics and relaxation phenomena, an appropriate choice given the emphasis on interdisciplinary topics at York University.

In a long series of papers on the master equation, Pritchard and his coworkers elucidated for the first time the effects of rotational and vibrational disequilibrium on the dissociation and recombination of a dilute diatomic gas. Ultrasonic dispersion in a diatomic gas was analyzed by similar computational experiments, and the first example of the breakdown of the linear mixture rule in chemical kinetics was demonstrated. A major difficulty in these calculations is that the eigenvalue of the reaction matrix (corresponding to the rate constant) differs from the zero eigenvalue (required by species conservation) by less than

the rounding error of the machine, but this problem was subsequently resolved by a matrix perturbation approach.[226]

On a different front, one of the first successful modelings of a practical combustion system was achieved, that of the formation of NO in a jet engine combustor.[227] The method proposed for nitric oxide abatement is being incorporated into the next generation of aircraft jet engines. Later, essentially complete agreement between experiment and numerical modeling was achieved for the thermal explosions of methyl isocyanide in spherical vessels.[228]

Recent work by Pritchard has concentrated on a state-to-state description of unimolecular reactions[229] and an examination by classical trajectory methods of the effects of overall molecular rotation on the unimolecular rate. The latter calculations have revealed a most interesting aspect of computing in chaotic systems, namely, that the same algorithm gives different results on different machines for a trajectory with identical initial conditions, or even on the same machine with different releases of the same compiler. However, the ensemble average behavior, with an ensemble comprising 100 or more trajectories, is acceptably the same each time.[230]

Geoffrey Hunter, a former Ph.D. student of Pritchard's at the University of Manchester, joined the faculty at York University in 1966. His research interests have often dealt with the enigmas of atomic and molecular quantum mechanics. This life-long interest can be traced back to his Ph.D. thesis, in which he set out to compute the ground state energy of the hydride ion using a wavefunction similar to the Born–Oppenheimer wavefunction of H_2^+, except that the large and small masses were interchanged. However, he developed the theory in a general form that made it applicable to any three-particle system interacting by Coulomb forces. Computational results were published for several systems.[231]

A notable result of this work was a scaling of the electronic wavefunction to make the separated-atom limit of the adiabatic electronic energy exact, and in 1965, while in Texas, Hunter applied this scaling to a calculation of the hydrogen molecule, thereby showing that the published experimental dissociation energy of H_2 must be in error by about 4 cm^{-1}. This result motivated Gerhard Herzberg to remeasure the dissociation energy by a more precise method.

Hunter's unique expertise and experience enabled him to realize that the square root of the electron density satisfies a one-electron Schrödinger equation and that the effective potential in this Schrödinger equation is, in principle, an exact representation of the motion of a single electron within a many-electron system.[232] He subsequently showed that there is a surface enclosing a molecule outside of which the electron's kinetic energy would be negative. He has proposed that this molecular envelope provides a nonarbitrary definition of molecular size and shape that is suitable for implementation in computer graphics.

The arrival in 1969 of Alan Hopkinson from the University of Toronto, where he had worked with Yates and Csizmadia, broadened the theoretical interests at York to include theoretical organic chemistry. Hopkinson went to Toronto with a background in strong acid physical chemistry. Yates initially put

him on an extended Hückel project involving amide hydrolysis, but Hopkinson quickly found that extended Hückel theory could not satisfactorily answer their questions, so he quietly became more interested in the gas phase property of proton affinities. He persuaded Mike Robb and Roy Kari to allow him to use the Polyatom program that Csizmadia had brought from Slater's group. He proceeded to show for the first time that as the size of the basis set is increased, the enthalpies of reaction, in this case the calculated proton affinities, converge on experimental values.[233]

Hopkinson was hired by York to teach theoretical organic chemistry (the Woodward–Hoffmann rules were then a hot topic) and to carry out experimental chemistry. Despite the limited computing capacity at York at the time, he managed to complete some work on the electrophilic addition to alkenes. He is probably best known, however, for his work on proton affinities, destabilized carbocations,[234] organosilicon compounds, silyl anions and cations, and more recently, on the calculation of potential energy surfaces and thermodynamic properties. He has had a particularly fruitful collaboration with Diethard Bohme.[235]

Theoretical chemistry at York University was strengthened in the 1990s with the appointments of Bill Pietro in 1991 and René Fournier in 1996. Pietro wrote part of the Gaussian code as a graduate student and several modules of SPARTAN while an assistant professor at the University of Wisconsin. While he was in Madison he developed a research program based on molecular electronic devices.[236] He expanded his interests to several facets of molecular electronics, including molecular electroluminescent materials, molecular electronic devices (diodes, switches, and sensors), and functionalized semiconductor nanoclusters.[237] These new materials not only are scientifically very exciting, but they offer the possibility of revolutionary impact on the future of the electronics industry.

René Fournier is studying atomic clusters[238] and transition metal complexes.[239] He is using a combination of density functional methods, tight-binding models, and molecular simulations with empirical interaction potentials, as part of a research program designed to study materials by computations on simple model systems.

Simon Fraser University (1966)

Simon Fraser University, founded in 1964, was one of many new universities established in Canada in the 1960s. It appears that Natalia Solony (a student of Fraser Birss) was the first theoretician at SFU, but she left after a year or two. In 1966 Margaret Benston, Gulzari Malli, and John Walkley joined the Department of Chemistry. Benston carried out research on the electronic structures of atoms and molecules for a few years, often in collaboration with Chong at UBC, before moving to the Department of Computing Science and then later to the Department of Women's Studies. She died prematurely in 1981. John

Walkley's research interests over the years have focused on the thermodynamics and statistical mechanics of dilute solutions,[248] intermolecular potential calculations, and Monte Carlo calculations.

Gulzari Malli has maintained his interest in ab initio relativistic quantum chemistry since he joined SFU in 1966. In fact, the earliest work in Canada on relativistic quantum chemistry appears to have been carried out in 1965 by Malli and Serafin Fraga at the University of Alberta. Their earliest results were documented in a compact book.[249]

Malli's interest in treating relativistic effects for many-electron systems containing heavy elements ($Z = 75-112$) led to the development of the first relativistic self-consistent field theory for closed-shell molecules.[250] The formalism was tested on light diatomics. Later for heavy element molecular systems, Malli introduced the use of Gaussian basis sets in relativistic quantum chemistry. Next he and his coworkers developed a relativistic SCF theory for open-shell molecules and a multiconfiguration relativistic SCF theory. They showed that relativistic effects predict a bond shortening of about 0.45 bohr and a doubling of the binding energy for AuH. Malli and Pyper reported the first relativistic configuration interaction (RCI) calculations for AuH taking an all-electron, fully relativistic Dirac–Fock wavefunction as the reference.[251] Ab initio all-electron, fully relativistic wavefunctions were also reported for a number of diatomics involving heavy elements; the effects of relativity on the dipole moments of various diatomics were reported for the first time.[252] During the last decade, Malli and coworkers have investigated the effects of relativity and electron correlation using the fully relativistic Dirac–Fock SCF treatment and MP2, and most recently the relativistic coupled-cluster (REL-CCSD) and relativistic many-body perturbation theory (RMBPT) for atoms and molecules.

Computational chemistry at Simon Fraser University received a major boost in 1990 when Saul Wolfe joined the Department of Chemistry. After obtaining his B.A. and M.A. degrees at the University of Toronto, Wolfe attended the University of Ottawa, where he was the first graduate student of Prof. Ray Lemieux and the first Ph.D. graduate in the Faculty of Science. Following postdoctoral work with Prof. Franz Sondheimer at the Weizmann Institute of Science, he joined Bristol Laboratories in 1959 as an organic chemist. In 1961 he moved to Queen's University at the instigation of R. L. McIntosh who, as noted earlier, built up a strong theoretical group during his term as head of the Department of Chemistry. Wolfe remained at Queen's until 1990, when he hung up his snow shovel and moved to Vancouver.

In a review based on his 1992 Lemieux Award lecture,[253] Wolfe presents a highly personal account of his research career and interests over a period of 33 years. His review provides an overview of his studies on the relative antibacterial activities of penicillins and cephalosporins and their differing abilities to complex to and react with penicillin-binding proteins. He describes how the insights from his analysis could be used to attempt the design of new structures targeted to the penicillin receptor. Besides providing glimpses of Wolfe's signifi-

cant contributions to organic chemistry, his review describes how he became interested in computational chemistry through the work of Arvi Rauk,[254] one of his first graduate students.

Saul Wolfe has made important contributions on a broad range of topics in bioorganic chemistry and many aspects of theoretical organic chemistry. His special interests include the gauche effect,[255] β-lactam compounds,[256] and isotope effects.[257]

University of Manitoba (1966)

The University of Manitoba appointed its first theoretical chemist in 1966. With postdoctoral experience in the groups of P.-O. Löwdin and A. D. Buckingham, Bob Wallace established a research program focused on the basic quantum theory of molecules, electromagnetic interactions, and the theory of molecular collisions and molecular processes. His interest in molecular collisions gave way with the passage of time to an emphasis on molecular processes.[240] During the past decade he has had a special interest in large amplitude nuclear motion,[241] including new methods for deriving internal coordinates.[242]

Bryan Henry joined the faculty of the University of Manitoba in 1969 and developed a research program based on the experimental and theoretical study of highly vibrationally excited molecules. At high enough energies, light interacts with molecules containing X—H oscillators to prepare states that are more localized than those expected on the basis of the traditionally accepted normal mode description of molecular vibrations. Henry and his group have developed the local mode description of such vibrational states, and this description has now gained general acceptance.[243] States with overtone spectra, because of the localization, are extremely sensitive to the properties of X—H bonds. The spectra are measured with a variety of sophisticated spectroscopic techniques, including intracavity laser photoacoustic spectroscopy, and are used to study molecular structure and conformation. The time scale of the overtone experiment allows study of conformational processes that are much too fast to be observed by conventional spectroscopic techniques like NMR. The spectra also allow determination of intramolecular vibrational energy redistribution.

Henry's group is also involved in theoretical studies to determine sources of local mode overtone intensity. These investigators have developed a very successful approach that uses their harmonically coupled anharmonic oscillator local mode model to obtain the vibrational wavefunctions, and ab initio calculations to obtain the dipole moment functions. The researchers have applied these calculations to relatively large molecules with different types of X–H oscillator. Recently they have compared intensities from their simple model to intensities from sophisticated variational calculations for the small molecules H_2O and H_2CO. For example, for H_2CO they generated a dipole moment function in terms of all six vibrational degrees of freedom.[244] This comparison has allowed them to determine the quality of basis set needed to calculate dipole moment

functions that lead to reasonable values for fundamental and overtone intensities. Henry's group also calculated intensities at different levels of ab initio theory and found a very surprising result. The calculations consistently show that although electron correlation in the calculation of dipole moment functions can be important for fundamental intensities, it is apparently unimportant for overtone intensities. Henry moved to the University of Guelph in 1987.

Since joining the University of Manitoba in 1985, John Cullen has concentrated on quantum chemical methods for large molecules. He is interested in treating biological molecules by semiempirical methods.[245]

The research of Kathleen Gough, appointed in 1995, is concerned with theoretical and experimental studies of vibrational intensities and electronic charge flow. Gough and her coworkers use ab initio electron density distributions[246] and calculate Raman scattering intensities to assist in the interpretation of observed spectra. One of the goals of their research is to quantify relationships between observed intensities in vibrational spectra, molecular structure and electronic charge flow in vibrating molecules.[247]

Carleton University (1970)

Carleton University was one of many Canadian universities to appoint its first theoretical chemist in 1970. Since his arrival in Ottawa, Jim Wright has had a strong interest in unusual chemical structures[258] that arose from his postdoctoral research with Lionel Salem on diradical mechanisms in the ring opening of cyclobutane. The C_4H_8 system was too big to explore using ab initio techniques at the time (even with a minimum basis set), and therefore Wright used the simpler isoelectronic model, cyclic O_4. Much to the researcher's amazement the O_4 ring was puckered, like cyclobutane. The lone pairs were acting as if they were localized, a representation that is hard to prove theoretically but which has great predictive value. Further work in this area led to predictions on cyclic ozone, nitrogen rings including the quasi-aromatic N_6, and quasi-aromatic H_6 as a transition state for the $H_2 + D_2$ exchange reaction. Wright returned to this theme later when he examined the structure and stability of hydrogen rings.[259] Recently Wright's group has been examining polymeric nitrogen and oxygen analogs of saturated hydrocarbon chains: for example, how stable is HO—$(O)_n$—OH or H_2N—$(NH)_n$—NH_2? The nitrogen chains turn out to be much more stable than expected (in the gas phase) and to adopt a helical conformation. It is interesting to note that Paul Giguère at Laval identified H_2O_3 and H_2O_4 by matrix IR spectroscopy.[260,261]

Wright and his collaborators have studied extensively the doubly excited states of molecules.[262] They have shown that promotion of electrons from antibonding MOs in diatomic molecules can lead to a drastic shortening of the bond length with an accompanying huge increase in the harmonic vibrational frequency. This new type of strongly bound, doubly excited state has now been observed experimentally in compounds such as B_2, where the ground state

single bond is converted into what is essentially a triple bond in the doubly excited state. An offshoot of this work on heteronuclear diatomics led to the interesting discovery that some diatomics can be thermodynamically stable, even with a +2 charge.[263] This can occur when charge-induced polarization overcomes the Coulomb repulsion.

Wright is also interested in modifying potential surfaces for chemical reactions by the use of intense infrared laser fields. This interesting new area of research shows that not only is it possible to lower a barrier to chemical reaction, but the barrier can be caused to pulsate up and down with the frequency of the laser. Wright's group looked at the $H + H_2$ exchange reaction as a first application and showed that the reactivity can be enhanced significantly with intense fields.[264]

Dalhousie University (1970)

Despite being one of Canada's oldest universities (founded in 1818), Dalhousie University did not formally begin a program of theoretical chemistry until Charles Warren was appointed in 1970. Initially Warren's research interests were divided between spectroscopy and semiempirical MO calculations. By the end of the 1970s, he was primarily interested in laser Raman spectroscopy and related theoretical topics, including the Jahn–Teller effect. In the 1980s he turned his attention to computed-aided instruction, especially for undergraduates. He has written many interactive programs to illustrate the principles of quantum mechanics. Most of his programs feature excellent graphics, a consequence of his research on algorithms for computer graphics.

Since arriving at Dalhousie University in 1975, Russell Boyd has established a broadly based research program. Boyd's earliest work on electron correlation was concerned with developing physical pictures for the Coulomb and Fermi holes, initially in atoms and later in molecules; the research had interesting implications for many topics, including the interpretation of Hund's multiplicity rule.[265] Later the emphasis shifted from two-electron density distributions to one-electron density distributions. Boyd and his coworkers have made extensive comparisons between electron densities calculated from density functional methods and high level configuration interaction calculations.[266] The long-term goal of this research is to use an accurate representation of the electron density as the basis for the development of new functionals for use in DFT.

Boyd and his coworkers have studied several reaction mechanisms in detail including S_N2 reactions.[267] The group has published many papers on oxygen radicals ($RO^•$ and $ROO^•$), which are of vital interest in biological systems because their attack on lipid biomembranes is related to many pathological events. Their interest in peroxyl radicals arose initially from the uncertainty surrounding the mechanism of the 1,3-migration in allylperoxyl radicals. Calculations[268] showed that the experimentally observed stereoselectivity of

the rearrangement must result from a cage effect wherein diffusion of the allyl–oxygen pair is prevented because of caging by the solvent. Subsequent oxygen labeling and stereochemical studies of the rearrangement as a function of solvent stability and viscosity support the theoretical conclusions.

The long-standing interest of Boyd and his coworkers in radicals and radical ions has led to many papers since 1993 on hyperfine structures. These papers have pushed the conventional multireference configuration interaction methods to the limits of the available computers, tested the predictive ability of various functionals commonly used in DFT calculations, and, among other topics, modeled the effect of a noble gas matrix on the hyperfine structures of radicals. Recent research focused primarily on radicals formed as a consequence of radiation damage to DNA.

In the mid-1990s the Boyd group broadened their interests to include molecular dynamics simulations. One of the objectives of their research is to include quantum mechanical force fields directly or indirectly in MD simulations. One of the key issues underlying the success of a hybrid approach is an appropriate description of the coupling between the quantum mechanical and molecular mechanical force fields. A paper that reports the first ab initio MD simulation of liquid methanol[269] showed somewhat surprisingly that the best overall agreement between the experimental and simulated results for vibrational frequency shifts is obtained with the weakest quantum mechanical/molecular mechanical coupling.

Peter Kusalik took up an appointment at Dalhousie University in 1989 and developed a research program focused on computer simulation studies of molecular liquids, solids, and solutions. As well as standard simulation approaches, he has explored nonequilibrium molecular dynamics techniques and applied field simulations, the development of new models and methodologies being one aim of his research. A common focus throughout his work has been the examination of the interplay between microscopic structure and dynamics in condensed matter and their relationship to bulk properties.

Kusalik's group has published several key studies[270] examining the local structure in liquid water. Using a unique spatial approach, their work has served to clarify greatly the understanding of the local molecular environment in aqueous systems. These investigators have also made exciting advances in modeling nucleation and crystal growth processes. Kusalik's group reported the first successful computer simulation studies of the crystallization of molecular liquids, first with the electrofreezing of water[271] and later with the crystallization of carbon dioxide[272] under field-free conditions. The development of these techniques also allowed them to discover a possible new polymorph for ice.[273]

University of Guelph (1970)

Theoretical chemistry began at the University of Guelph in 1970 with the appointment of Mike Zerner from the United States. It was a period of rapid

expansion of the relatively new university, which had been created in 1964 when the Ontario Veterinary College, the Ontario Agricultural College, and the MacDonald Institute joined with a new college of arts and science. Zerner quickly established a research program based on semiempirical treatments of large molecules, especially porphyrins and related systems.[274] By the end of the decade Zerner and his coworkers had become well known for their work on the spectra of iron, cobalt, and copper complexes.[275] The widely used ZINDO method was developed during the Guelph era of Zerner's career and was applied to many systems.[276]

In 1980 Zerner made the first application to NSERC for a computer dedicated to computational chemistry. Zerner and his coapplicants (Jiri Cizek, Saul Goldman, Bob Le Roy, Fred McCourt and Joe Paldus) made a favorable impression on the site visit team but were told that computers were the domain of the university and that NSERC equipment grants were for experimental equipment only. The following year Zerner accepted a position at the University of Florida, and the NSERC application was resubmitted through the University of Waterloo. Bob Le Roy and his coapplicants were awarded $215,000 in 1982 and took delivery of a VAX 11/750 in the summer months. It is interesting to speculate about the extent to which Zerner's resignation led to a reversal of the NSERC policy on funding computers. Although Zerner moved to the Sunshine State in 1982, his 12 years in Guelph had a lasting impact on the development of theoretical and computational chemistry in Canada. Indeed, his inspiring research and personality have had a large impact on computation chemists everywhere.

After completing a Ph.D. degree and postdoctoral research in analytical chemistry, Saul Goldman joined the University of Guelph in 1972. As a reformed experimentalist, he developed a research program based on statistical mechanics,[277] with special interests in supercritical fluid extraction, spectra of endohedral fullerenes,[278] the transport of ions through biological channels, and the molecular basis for the properties of liquids and solutions.[279]

Upon joining the University of Guelph in 1983 John Goddard built up an applied quantum chemistry program spanning many aspects of main group inorganic chemistry and organic chemistry, with special interests in reaction intermediates and transition structures in excited state chemistry. Many of his papers in the 1980s resulted from a fruitful collaboration[280] with two experimental colleagues, Richard Oakley and Nick Westwood, who joined the department at about the same time. This talented trio of chemists combined forces to synthesize, characterize, and study many interesting new compounds.

Goddard and his coworkers have had a continuing interest in electronic, vibrational, and rotational spectra. This is evident from their papers on carbenes[281] and other highly reactive species. Many of Goddard's papers have studied the potential energy surfaces of thioformaldehyde[282] and related species. Much of this research has resulted from his willingness to carry out high level ab initio calculations of interest to spectroscopists. The Goddard group

recently expanded the scope of their research to include effective core potentials and density functional methods for main group inorganic compounds.[283]

As described in connection with the University of Manitoba, Bryan Henry moved to Guelph in 1987 and has continued with his experimental studies of overtone spectra involving X—H stretching and the analysis of the results in terms of the local mode model.[284] Indeed, most of his research on the intensities of overtone spectra was completed at Guelph.

University of Sherbrooke (1970)

André Bandrauk joined the University of Sherbrooke in 1970 following a NATO postdoctoral fellowship with C. A. Coulson and M. S. Child at Oxford, where he developed his interest in the theory of predissociation. His paper with Child reported the first application of scattering theory to spectroscopy.[285]

Bandrauk's long-term research interests include the dressed-state representation of molecular spectroscopy. His contributions to the nonperturbative treatment of molecular spectroscopy from the weak field to strong field limits have been summarized in two chapters in a book he edited in 1993.[286] Bandrauk and his coworkers published the first theoretical demonstration of the use of chirped pulses to effect laser bond breaking in less than a picosecond.[287] His other firsts include the first prediction of molecular stabilization in intense laser fields[288] and the first complete non-Born–Oppenheimer calculation of dissociative ionization of molecules in intense femtosecond laser pulses.[289]

COMPUTATIONAL CHEMISTRY IN CANADIAN INDUSTRY

Canadian industry employs far fewer computational chemists than do companies in the United States, the United Kingdom, Japan, and many European countries. Nevertheless, there are a few successful cases, which are summarized briefly in this section.

Hypercube, Inc.

Hypercube's HyperChem is the best-known computational chemistry software package developed in Canada and marketed worldwide. Hypercube, Inc., the brainchild of Neil Ostlund, was incorporated in February 1985. The company has a headquarters in Waterloo, Ontario, adjacent to the University of Waterloo, but moved most of its operations to Gainesville, Florida, in 1997.

The first phase of Hypercube's existence involved contracting for Intel in association with Intel's development of the first commercial highly parallel computer, the Intel iPSC hypercube computer. This is the origin of the com-

pany's name. Subsequently, Intel and Hypercube cooperated in the development and marketing of molecular modeling software for the commercial Intel iPSC. Later Hypercube focused on the internal development of a turn-key molecular modeling "instrument" called a Chemputer, consisting of a PC integrated with a parallel processing subsystem of transputers running Hyper-Chem, Release 1. The market for such a product was small, and in 1990 Hypercube formed a joint venture by which Autodesk acquired an exclusive licence to manufacture, market, and sell HyperChem, with Hypercube doing all development and technical support for the products. The first Autodesk product, HyperChem Release 2, was shipped in March 1992. Release 3 was shipped in April 1993.

The desktop computational chemistry software market turned out to be smaller than desired. In January 1994, Hypercube recovered its licence from Autodesk and brought all control for HyperChem and related products in-house. Hypercube has since shipped Releases 4 and 4.5 of HyperChem and has delivered other new products such as ChemPlus and HyperNMR. The company is developing a range of new molecular modeling products for a range of platforms, although its dominant product remains HyperChem for Windows. The latest product is HyperChem Release 5 for Windows 95 and Windows NT, which includes many new computational features and new graphical abilities for the presentation of the results of calculations. HyperChem continues its wide popularity among PC users.

Ayerst Laboratories

In 1982 Ayerst Laboratories in Montreal became the first company in Canada to install a commercial software tool (the SYBYL suite from Tripos Associates) to help in the development of pharmacophoric models from structure–activity relationships. The installation of the software was the second ever, worldwide, by a company and is a testimonial to the foresight of the director of medicinal chemistry, Dr. Leslie Humber, for having championed its installation. Dr. Adi M. Treasurywala, then an organic chemist with some experience in medicinal chemistry, became the first industrial computational chemist in Canada that year. The use of modeling approaches contributed in a minor but significant way to the discovery of the compound known as Tolrestat, which was an inhibitor of lens aldose reductase. This led to the acknowledgment of Treasurywala as a coinventor of the drug on several patents that were filed in this research area. Approximately in 1983, Ayerst closed down its discovery effort in Canada and moved to Princeton, New Jersey, where an expanded effort in the area of computational chemistry continues.

Merck Frosst Canada Inc.

Merck & Company became involved in the early 1970s in applying computational chemistry to pharmaceutical research, an effort that continued and

expanded. Although Merck Frosst, the Canadian subsidiary of Merck & Company situated in Montreal, had these resources available to draw upon, there was no on-site expertise to facilitate collaborations.

In 1992 Merck Frosst created a permanent senior-level position for a computational chemist. Christopher Bayly accepted the position, leaving a postdoctoral appointment in Peter Kollman's group (University of California, San Francisco), where he had been doing force field development and free energy calculations. (Prior to that, Bayley had obtained his Ph.D. under the supervision of Fritz Grein at University of New Brunswick, having received an M.Sc. in synthetic organic chemistry with Pierre Deslongchamps at University of Sherbrooke in Quebec.) Recently, an additional junior-level position at Merck Frosst was filled by Daniel McKay (an M.Sc. graduate from Jim Wright's group at Carleton University). Currently, the modeling effort involves the usual spectrum of methods: macromolecular simulation, small molecule force field and electronic structure calculations, with 2-D and 3-D database searching. Software resources consist primarily of proprietary Merck & Company modeling code, some of which has been published [e.g., the Merck Molecular Force Field (MMFF), SEAL, for fitting molecules together, and FLOG, a 3-D database docking program]. Other commercial codes are used, as well as academic code (e.g., AMBER, DeFT). The modeling group is integrated with the overall medicinal chemistry department from both a functional and organizational standpoint.

Xerox Research Centre of Canada

During its relatively brief existence, the Materials Modeling and Simulation group at the Xerox Research Centre of Canada (XRCC) applied quantum mechanics and molecular dynamics simulations to the design of new materials for use in xerography. The group was headed by Tom Kavassalis, a theoretical chemist by training with a B.Sc. from the University of Toronto and a Ph.D. from MIT. The group disbanded a few years ago.

In a 1995 overview of their research interests at the XRCC, Kavassalis[290] summarized the role that computational chemistry plays in an industrial research laboratory. His overview describes the application of quantum mechanical methods to polymerization reactions and the role of crystal packing in determining the bond structure of organic photogenerators. He also shows how simulation methods have been incorporated into the treatment of problems that were until recently primarily in the experimental domain. Thus it is now possible to study polymer thermal and molecular properties and to estimate solvent diffusion coefficients by molecular dynamics simulations. The same methods are providing insight into polymer and surfactant solubility parameters, as well phase transitions. In the same article, Kavassalis summarizes the challenges and opportunities for the application of theoretical methods in materials science. It

is clear that methods that were once the purview of academic researchers are making a significant impact on the development of new technologies.

ORTECH, Inc.

ORTECH is a relatively new Canadian company in the computer-aided drug design field. From their Mississauga, Ontario, location, they have developed and marketed a program called MolScan, which does conformational analysis of ligands to identify pharmacophores. The resulting pharmacophores can be used to search databases of three-dimensional molecular structures for compounds that fit the criteria for potential biological activity. The group is headed by Robert Kirby, a 1990 Ph.D. graduate in organic chemistry from Carleton University in Ottawa. Kirby joined ORTECH after holding molecular modeling positions at BioChem Therapeutic, Inc., and Allelix Biopharmaceuticals, Inc.

BioChem Therapeutic

In 1997 Alan Cameron, formerly with Eastman Kodak Company of Rochester, New York, and SynPhar Laboratories, Inc., of Edmonton, Alberta, joined BioChem Therapeutic, Inc., Laval, Quebec. He has been involved at BioChem in new compound identification and optimization of existing lead compounds directed toward various therapeutic agents including antithrombotics, opioid analgesics, and anticancer and antiviral compounds. He obtained his Ph.D. under the supervision of Mike Baird and Vedene Smith at Queen's University and was a postdoctoral fellow with Mike Zerner at Florida.

Dr. Miguel Quimpère joined BioChem Therapeutic in 1990 as a synthetic organic chemist and has since moved on to data management and molecular modeling, with a particular view to diverse library design and combinatorial chemistry.

Advanced Chemistry Development, Inc.

Advanced Chemistry Development, Inc., is also relatively new to the Canadian scene. The idea behind ACD originated with a small team of postdoctoral researchers working together at Moscow State University in Moscow, Russia. In 1990 these scientists brought together their talents in the areas of chemistry, spectroscopy, and computer science to develop chemical software tools focusing on accuracy and simplicity for the analysis of physicochemical properties. ACD continues to grow rapidly to keep pace with the demand for its products and innovations worldwide. From the original group of seven at Moscow State University, ACD grew to a staff of 70 chemists, algorithm specialists, programmers, and database compilation experts in Moscow. The Toronto headquarters has a staff of 20, providing sales, graphic design, market-

ing, product development, and technical support for the ACD product line. In addition to full-time staff, the Toronto office hires freelance programmers and scientific consultants.

Broadly stated, ACD expertise lies in the area of data compilation (and critical analysis of these data), verification, correlation, prediction, and systematization of common scientific knowledge. The systematization falls into two categories: well-defined rules (e.g., nomenclature), which can be applied more uniformly on a computer than manually, and less rigorous "rules of thumb," which must be devised, tested and parameterized, refined and then retested. ACD algorithms are proprietary and mainly based on a heuristic structure-fragment approach. The product line includes software for predicting NMR spectra, ionization coefficients, solubility, octanol–water partition coefficients, vapor pressure, Hammett-type constants, and related physicochemical phenomena, as well as software for chemical structure drawing and for assigning chemical nomenclature.

SynPhar Labs, Inc.

SynPhar, a small-sized pharmaceutical R&D company located in Edmonton, is involved in the research and development of drugs against several therapeutic targets including infectious diseases and cancer. The company has its own internal research programs as well as collaborations (in various stages) with Japanese, European, and American multinational firms. The Molecular Modeling/Computational Chemistry group in SynPhar Laboratories, Inc., was set up in 1993 as part of a collaborative project involving molecular design with the Biotechnology Research Institute of the National Research Council in Montreal. At that time, the company had one computational chemist at SynPhar (Edmonton) and another at NRC (Montreal). In the molecular modeling area is one senior scientist, Dr. Sanjay Srivastava, who joined the company in 1993.

Bio-Méga

The computational chemistry group at Bio-Méga is mainly involved in aiding the company's medicinal chemistry efforts in the design of new antiviral agents. A variety of modeling techniques are used to help elucidate and understand the structure and conformation of inhibitors and how they might be acting in the target enzymes. The firm's computational chemist, Dale Cameron (B.Sc., 1989, Ph.D., 1994, Queen's University), was hired in 1995.

Astra

Dr. Shi-Yi Yue joined the Astra Research Centre in Montreal in 1995 as a computational chemist. The Astra computational chemistry effort started with pharmacophore analysis and G-protein coupled receptor modeling and has

recently expanded to include library design, quantitative structure–activity re-lationships, and bioinformatics.

Other Examples

It is impossible to make this account inclusive, but a few other examples of computational chemists working in Canadian industry should be mentioned. Liangyou Fan (Ph.D. Calgary 1992, under T. Ziegler) has been working for Nova Chemicals in Calgary since 1993 as a staff researcher conducting theoreti-cal calculations on single-site olefin polymerization catalysis using density func-tional theory. Also Danya Yang (Ph.D. Calgary 1994, under A. Rauk) has been working since 1996 with Travis Chemicals in Calgary simulating corrosion with molecular mechanics and quantum mechanics methods.

HISTORY OF THEORETICAL CHEMISTRY AT THE NATIONAL RESEARCH COUNCIL OF CANADA

Although the research of Gerhard Herzberg and his colleagues at the National Research Council of Canada needs no introduction to computational and theoretical chemists, the emergence of theoretical and computational chemistry at the NRC is a relatively recent development.

In the 1950s, the Division of (Pure) Chemistry needed theoretical as-sistance in areas such as NMR and solid state chemistry. It acquired this as-sistance from long-term visitors, including John Pople (1998 winner of the Nobel Prize in Chemistry), David Buckingham, and Hugh Barron. Attempts to hire any of these visitors on a permanent basis failed. Therefore it was decided to start hiring recent PhDs to fill the gap. The first theorists hired were Willem Siebrand and Constantine Mavroyannis in 1963, soon to be followed by Ray-mond Somorjai and Michael Klein. Initially each theorist was assigned to an experimental group. After a few years, postdoctoral fellows joined NRC, and a new permanent member was added, David Peat. It was then decided that the theorists should form their own group, with Siebrand as group leader. The main areas of activity were molecular crystals and radiationless transitions (W.S.), elementary excitations in solids (C.M.), condensed phase structural simulations (M.K.), and quantum chemistry and biology (R.S.). A succession of postdoc-toral fellows and (later) research associates (RA) joined the group, some of whom found permanent positions in other parts of NRC, while Marek Zgierski and Roger Impey were appointed to continuing positions in the group. An experimentalist turned theorist, John Tse, did not join the group but became a frequent collaborator.

The changing political climate of the early 1980s, which ran against "pure" science and especially theory, coincided with the need of theorists to

obtain new and costly computational facilities. For these reasons, Michael Klein left for the University of Pennsylvania, and Roger Impey, Ray Somorjai, and David Peat went to other parts of NRC. The decimated theory group thus lost its group status and was "hidden" inside an experimental group. The restructuring of NRC in the early 1990s resulted in the formation of the Steacie Institute for Molecular Sciences and led to the resurrection of the theory group, as the Theory and Computational Program, consisting of Willem Siebrand and Marek Zgierski and two RAs. In recent years the group acquired four more staff members: Philip Bunker, who joined the group from the Herzberg Institute of Astrophysics; John Tse and Dennis Klug, who transferred from an experimental group in the institute; and Tamar Seideman, an RA appointed to the continuing staff. The group then consisted of six continuing staff members with three RAs and three postdoctoral fellows and was led by John Tse, Willem Siebrand having become director of research of the Steacie Institute. The research programs are focused on material sciences (J.T. and D.K.), spectroscopy (P.B.), quantum chemistry and biology (M.Z. and W.S.), and chemical dynamics (T.S.). Most recently, Dennis Salahub has taken over the reins of Steacie Institute. This augurs well for the status of computational chemistry in Canada.

HIGH-PERFORMANCE COMPUTING IN CANADA

Canada's computational chemistry community has not had the level of support for high performance computing that has been commonplace in the other leading industrialized nations for the past 15 years. Throughout the 1960s and 1970s, Canadian computational chemists worked on central mainframe computers, not unlike those used by other university-based researchers throughout the world. With the introduction of the VAX 11/780 in 1978 and similar minicomputers in the following few years, there was a strong migration to smaller dedicated computers. As discussed above, the Natural Sciences and Engineering Research Council of Canada started providing equipment grants to researchers for the purchase of computers in the early 1980s. It soon became apparent that university administrations no longer saw the provision of high performance computing to be a high priority.

A recent study of high performance computing (HPC) in Canada[291] included a detailed assessment of the Canadian situation and a comparison of the Canadian environment with that of other countries. The study showed that the United States, Europe, and Japan have multi-billion-dollar government programs in place to support the infrastructure requirements of high performance computing and communications, whereas Canada has not adopted a national strategy to support and develop such activity. This situation is dramatically illustrated by the fact that in August 1994 only one Canadian facility, the

Atmospheric Environment Service computer center in Dorval, Quebec, ranked in the top 176 HPC sites in the world. Given Canada's large land area (9.2 million km²) and low population (30 million), it is not surprising that weather forecasting is a national priority. Indeed, Canada is a leader in numerical weather prediction research, and many other nations have adopted methods developed in Canada. The dominant industrial application for high performance computing in Canada is the processing of seismic data for the oil and gas industry, which is centered in Calgary.

As the new millenium commences, an infusion of funds from the Canada Foundation for Innovation has led to a dramatic increase in HPC facilities in Canada. Moreover, the formation of networks of researchers and facilities has virtually eliminated the problem of accessibility.

MAJOR CONFERENCES

The distinguished history of the Canadian Symposia on Theoretical Chemistry (CSTC) was summarized earlier in this chapter. Several other significant conferences should be mentioned.

Fifth International Congress on Quantum Chemistry

The awarding of the Fifth International Congress on Quantum Chemistry (ICQC) to Canada in 1985, a mere 20 years after the historic conference convened by Fraser Birss and Serafin Fraga in Edmonton, signaled that Canada had arrived in the field of computational and theoretical chemistry. The first four ICQCs were held in France (Menton, 1973), the United States (New Orleans, 1976), Japan (Kyoto, 1979), and Sweden (Uppsala, 1982).

The Fifth ICQC was held at the University of Montreal in August 1985 with Camille Sandorfy as president and Willem Siebrand and Vedene Smith as vice presidents. The congress was the largest scientific meeting devoted exclusively to theoretical chemistry held in Canada up to that time. More than 450 participants attended the congress, which was a great success. The proceedings were published in the *International Journal of Quantum Chemistry* in four parts, spanning more than 1500 printed pages.[292] The proceedings concluded with a general survey of the current (1985) state of the art of quantum chemistry by Per-Olov Löwdin.

Second World Congress of Theoretical Organic Chemists

An even larger conference took place in Toronto in July 1990, when more than 550 participants attended the Second World Congress of Theoretical

Organic Chemists at the University of Toronto. The First WATOC Congress had been held in Budapest in August 1987. Both congresses were chaired by Imre Csizmadia.

The Toronto Congress, like the earlier one in Budapest, attracted many quantum chemists and computational chemists whose interests were not restricted to compounds containing carbon. At a general discussion relating to the focus of future congresses, it was decided that the name should be modified to better reflect the actual interests of the participants. The original name was, however, retained for the Third WATOC Congress, which was chaired by Keiji Morokuma in July 1993 in Toyohashi, Japan. The name was officially changed to the Fourth World Congress of Theoretically Oriented Chemists (WATOC 96) starting with the July 1996 meeting in Jerusalem.

Canadian Computational Chemistry Conference

In recognition of the rapid expansion of computational chemistry in the 1980s, André Bandrauk and André Michel of the University of Sherbrooke organized the First Canadian Symposium on Computational Chemistry in May 1991 in Orford, Quebec. The conference included invited papers on dynamics, density functional methods, molecular modeling, Monte Carlo methods, and topics in quantum chemistry and statistical mechanics. About half of the invited speakers were from abroad (mostly from the United States).

A subsequent conference was convened in May 1994 at Queen's University in Kingston, Ontario. The name was changed to the Second Canadian Computational Chemistry Conference. The conference was organized by Ken Edgecombe and Vedene Smith with programming assistance by Axel Becke, Dave Wardlaw, and Don Weaver. The Third CCCC was held in July 1997 at the University of Alberta in Edmonton. The Organizing Committee was chaired by Mariusz Klobukowski. A preliminary presentation of the material in this chapter at the Edmonton CCCC meeting led to the recollection of many anecdotes from earlier conferences in Canada. At least one story is worth recalling. At the CSTC conference in Banff in 1989, a prominent quantum chemist was asked when one of the major computer manufacturers would introduce parallel computers. With hardly a pause, the speaker replied that he would rather have one ox to plough a field than 10,000 chickens!

SPREADING THEIR WINGS

Numerous references have been made throughout this chapter to the large number of outstanding computational and theoretical chemists who have been attracted to Canada. This history would not be complete without at least some

coverage of the equally remarkable Canadian computational and theoretical chemists who have pursued their careers in other countries. We include only a few notable examples.

Rudolph A. Marcus is perhaps the most famous theoretician to be raised in Canada. He has received many awards, most notably the 1992 Nobel Prize in chemistry. Marcus was born in Montreal. He received a B.Sc. degree in chemistry from McGill University in 1943, and a Ph.D. degree from the same institution in 1946. After doing postdoctoral research at the National Research Council of Canada and at the University of North Carolina, Chapel Hill, he became a professor at the Polytechnic Institute of Brooklyn from 1951 to 1964 and at the University of Illinois from 1964 to 1978, when he was named the Arthur Amos Noyes Professor of Chemistry at California Institute of Technology. His seminal contributions to the realms of electron transfer theory and intramolecular dynamics continue to earn him honors, including the 1997 ACS Award in Theoretical Chemistry.

Marcus is a coauthor of the Rice–Ramsperger–Kassel–Marcus theory of molecular reactions. However, his theoretical work in the 1950s on electron transfer reactions started him on his pathway to eminence. Back then, chemists knew electron-transfer processes were occurring during chemical reactions, but they had no way to predict the speed of a reaction or to develop strategic chemical experiments. In addition, some reactions that were predicted to proceed rapidly instead poked along at a snail's pace.

In a series of papers between 1956 and 1965, Marcus solved much of the mystery by outlining a description of the probability of fluctuations in the geometry of reactants and their solvents. These fluctuations lead to changes in the energy barriers that the reactants must surmount before an electron can be transferred from one molecule to another. Marcus extended the theory to other systems, such as electrochemical rate constants at electrodes, and to chemiluminescent electron transfer reactions. The by-now famous "inverted effect" is a consequence of his theory: after a certain point, adding more energy to an electron transfer reaction actually slows the process. Scientists believe photosynthesis can occur because of the inverted effect.

The RRKM theory is a ubiquitous tool for studying dissociation or isomerization rates of molecules as a function of their vibrational energy. Still highly active in the theoretical field, Marcus has tackled such issues as the semiclassical theory for inelastic and reactive collisions, devising reaction coordinates, new tunneling paths, and exploring solvent dynamics effects on unimolecular reactions in clusters.

Vincent McKoy, a long-time colleague of Marcus at Caltech, was also educated in Canada. McKoy graduated from the Nova Scotia Technical College (now part of Dalhousie University) and went on to earn his Ph.D. degree at Yale University.

Attila Szabo and Neil Ostlund, authors of a standard quantum chemistry text,[293] graduated from Canadian universities. After receiving his undergradu-

ate education at the University of Saskatchewan, Neil Ostlund earned his Ph.D. under the guidance of John Pople at Carnegie-Mellon University in 1968. Attila Szabo graduated from Montreal's McGill University in 1968 and completed his Ph.D. at Harvard University in 1973.

Several of the authors of the Gaussian software package were educated in Canada. Berny Schlegel graduated from the University of Waterloo and received his Ph.D. from Queen's University (Saul Wolfe), while a few years earlier Mike Robb[294] completed his Ph.D. at the University of Toronto (Imre Csizmadia). Schlegel is a long-time faculty member across the border at Wayne State University in Detroit whereas Robb is at King's College London.

There are many other examples of individuals who are not computational chemists but who are well known for their scientific contributions. Walter Kohn, a pioneer of density functional theory, graduated from the University of Toronto (B.A., 1945; M.A., 1946) and received his Ph.D. in physics from Harvard in 1948. He has received many awards (including the 1998 Nobel Prize in Chemistry) for his research in the theory of solids during his long association with the Department of Physics at the University of California, Santa Barbara. Charlotte Froese Fischer, well known for her computations on atoms,[295] graduated from the University of British Columbia (B.A., 1952; M.A., 1954) and earned her Ph.D. in 1957 for research carried out under the supervision of D. R. Hartree at Cambridge. She returned to UBC in 1957 and rose through the ranks to become a professor of mathematics. She later became a professor of computer science, briefly at Waterloo, then at Penn State, and more recently at Vanderbilt University.

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