

**Figure 1.** Jan Erik Almlöf, 1 April 1945–16 January 1996.

Accounts published a Memorial Issue edited by Odd Gropen, Björn Roos, Per Siegbahn, Peter R. Taylor and Donald Truhlar [4]. This document lists all of his students and colleagues, and also shows his complete bibliography.

## 2. The scientist

### 2.1. The early days: computation as an emerging tool in chemistry

‘In quantum chemical research, a considerable fraction of the effort is devoted to improving the computational strategy. The work in this field can basically be characterised as aiming at a single goal: how can larger systems be made accessible to increasingly sophisticated methods?’ is the opening statement in an article contributed by Jan Almlöf and Peter R. Taylor to the monograph ‘Advanced Theories and Computational Approaches to the Electronic Structure of Molecules’, edited by Clifford E. Dykstra and published by the D. Reidel Publishing Company in 1984 [5]. Born into the world of third-generation computers, with the ambition to explore chemistry on the basis of quantum-chemical methods, Jan Almlöf recognised the tremendous potential of current and future computing machineries early on. However, given the size and the complexity of the chemical problems that interested him, he also understood that, in order to make these systems accessible to an increasingly sophisticated computational treatment, a considerable effort would have to be spent on implementation of a method – that is, mapping it on a given compute resource (CPU, memory, external storage).

One of Almlöf’s early papers as a single author, published in 1974 [6], reports the first *ab initio* calculation on the molecular and electronic structure of porphine (Figure 2), a topic he would pursue throughout his career. The focus in his 1974 article is on the position of the ‘inner’ hydrogen atoms, with a comparison of computed N-H vibrational frequencies and computed ionisation potentials with experiment. Regarding the

computation, Almlöf writes: ‘The calculations [136 contracted Gaussian functions] were performed with the *ab initio* MO-LCGO-SCF method, using the formalism described by Roothaan’. Using his MOLECULE code [1,2] on the 360/195 computer of the San Jose IBM Research Laboratory, with a theoretical peak performance of approximately 5 Million Floating Point Operations per Second (MFLOPS), a single-point calculation took as little as 31 minutes for the electron repulsion integrals and 6 seconds for each self-consistent field (SCF) cycle. This performance was achieved by a rigorous exploitation of molecular symmetry within a supermatrix formalism, reducing the number of integrals to be stored from 170(unscreened) to 6 million, bringing the disk-storage requirements to tractable size (in the order of 100 MBytes).

### 2.2. Hitting the I/O wall and a change of paradigm

While CPU power was steadily increasing, it became apparent that input/output (I/O) would remain a bottleneck in *ab initio* calculations. Thus, when trying to reproduce the experimental structure of ferrocene and other metallocenes using the Hartree–Fock method, the I/O became the ultimate obstacle. In an attempt to harness every single MByte of ‘scratch space’ available, including those on the system disk, to store the 11 million, two-electron integrals on the University of Oslo CDC Cyber computer, a severe system crash ensued. Denied further access to the machine, the team was forced to reconsider the situation [7].

Indeed, inspired by a comment made by Enrico Clementi, his host at the San Jose IBM Research Laboratory, Jan Almlöf and co-workers Knut Fægri and Knut Korsell were at the time developing a prototype Hartree–Fock code that would build the Fock matrix from two-electron integrals as they were evaluated, avoiding any external storage [8]. Running on a departmental Norwegian-built Norsk Data mini-computer (ND500) with a powerful CPU but limited I/O capabilities, the code needed to recompute the integrals in every SCF iteration, putting the entire workload onto the CPU. Porting the code, later named Direct Self Consistent Optimization (DISCO) on the IBM 3033 main-frame computer of the University of Zürich, it became possible to perform Hartree–Fock calculations on ferrocene to the basis-set limit. In the largest computations, the wave function was expanded by more than 500 contracted basis functions [9], breaking the barrier of 255 functions set by eight-bit integral addressing.

The direct SCF method was a breakthrough – an egg of Columbus, marking the entry into a new paradigm and making new problems amenable to *ab initio* studies.

## Ab Initio Calculations on Porphin

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### Abstracts

*Ab initio* SCF calculations are reported for the porphin molecule. The positions of the central protons have been optimized, and the equilibrium geometry is found to be a linear  $\text{NH} \cdots \text{HN}$  arrangement. The NH vibrational frequencies have been computed and are compared to experimentally measured quantities. Several low ionized states have also been studied in separate spin-restricted SCF calculations. The lowest state is found to have  $B_{1u}$  symmetry with an ionization potential of 8.0 eV.

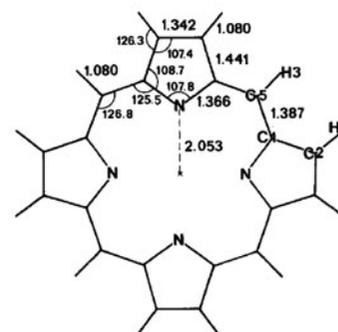


Figure 1. The molecular geometry used in the calculations. Bond distances in Å, angles in degrees.

Figure 2. The siren call of real chemistry: looking at porphin, the (unsubstituted) parent structure of all porphyrins.

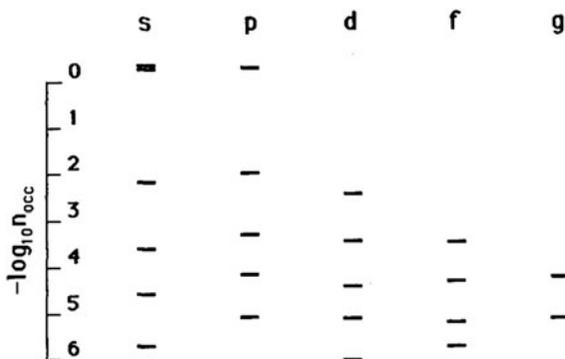


FIG. 1. NO occupation numbers for Ne in (13s 8p 6d 4f 2g) basis.

Figure 3. The occupation numbers of the natural orbitals (NO) of the neon atom showing a regular pattern which permits a systematic truncation of the NO space: a successive expansion of a minimal basis from [2s 1p] to [3s 2p 1d], [4s 3p 2d 1f] and [5s 4p 3d 2f 1g] includes all NOs within a given occupation number threshold (quote from [26]).

At the latest, with the massive direct SCF calculations on the thermodynamic stability of  $\text{C}_{60}$  performed on the Minnesota Supercomputer Institute Cray-2 in 1986 involving over 1000 contracted basis functions [10], the integral-direct approach was readily accepted by the community. One reason for the success of the integral-direct approach was the fact that ‘a few requirements that are rather obvious’ were successfully addressed (a quote from [5]):

- the integral evaluation must be as fast as possible;
- the number of SCF iterations must be minimal;
- in any iteration, the amount of data to be recomputed must be minimal.

The same requirements apply also in traditional calculations but then carry considerably less weight. Given that the density matrix is known at each iteration, a better

screening of unimportant integrals became possible with the direct SCF method. For large applications in extended basis sets, the observed effective scaling was as low as  $O(n^{2.2})$ , much lower than the formal fourth-order scaling. With the advent of larger memory systems, it made sense to store two-electron integrals in memory, to reduce the operation count further. Early adapters, such as the developers of TURBOMOLE [11], lead by Reinhart Ahlrichs, developed a *semi-direct* code where ‘expensive’ integrals were stored on disk rather than recomputed.

In the integral-direct SCF scheme, it becomes attractive to develop more involved methods to accelerate convergence: a number of authors [12–15] had shown the benefit of second-order methods using an exponential parametrisation of the orbital rotations between occupied and virtual space to converge the Hartree–Fock energy calculation. In a 1992 article, Thomas Fischer and Jan Almlöf presented a method that allows to recursively update the inverse Hessian from gradient and displacement information collected at each iteration, thus avoiding the storage of the full matrix [16]. This same scheme can also be used for geometry optimisation and be combined with Pulay’s direct inversion of the iterative subspace algorithm (DIIS [17,18] and GDIIS for geometry optimisation [19]) for further acceleration.

In 1988, Martin Head-Gordon, John A. Pople and Michael J. Frisch [20] argued that ‘it is desirable to extend the direct approach pioneered by Almlöf and co-workers to correlated methods’ presenting a CPU-bound scheme for the second-order Møller–Plesset (MP2) energy calculation implemented in their GAUSSIAN88 program [21]. In fact, only a few months before, S. Sæbø and Jan Almlöf had presented their own approach [22], ‘in which the correlation energy can be built directly from atomic integrals’. The early applications of the scheme, where the calculated integrals are transformed directly to the molecular orbital (MO) basis without resorting to external

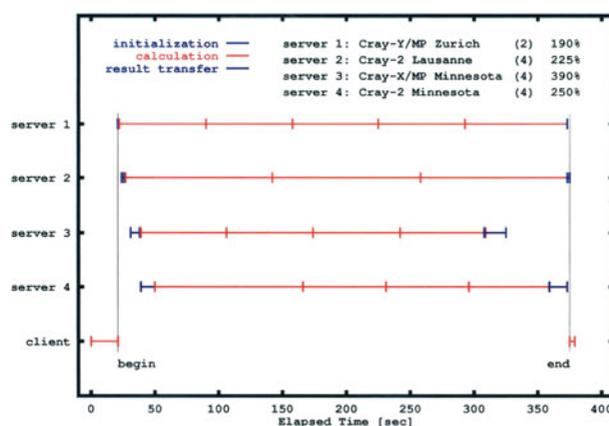
storage, showed that the CPU time for the MP2 calculation is dominated by the integral evaluation. The authors concluded that ‘if the resources allow a direct SCF calculation for a given system, then the second-order energy can be obtained for an additional fraction of the cost’.

### 2.3. From vector computing to the basis-set limit

With the advent of vector and shared-memory multiprocessor computers in the early 1980s, new challenges and opportunities emerged. To take advantage of the new architectures, algorithms had to be redesigned and codes rewritten. Segmented orbital contraction, in which each primitive basis function contributes to one contracted function only, was at the time by far the most popular choice for one-electron basis sets but turned out to inhibit efficient vectorisation. Adapting his MOLECULE integral code to vector processing in the mid 1980s, Almlöf implemented general contraction, in which each primitive orbital may contribute to several contracted functions, as proposed by Raffennetti in 1973 [23]. General contraction has a higher floating-point operation count than does segmented contraction, but this additional cost is offset by an efficient vectorisation of the transformation from primitive to contracted orbitals. As it turned out, this minor technical change in the MOLECULE integral code in response to changes in computer architecture had far-reaching consequences for how we design and perform quantum-chemical calculations.

In Fritz Schaefer’s words from 1971 [24], quoted by Jan Almlöf in 1988 [25], the selection of an appropriate atomic-orbital basis is ‘more art than science’. Specifically, in post-Hartree–Fock calculations, there was at the time no systematic way to improve a basis set, making it difficult to estimate the deviation from the basis-set limit.

Exploiting general contraction, Almlöf and Taylor introduced the atomic natural orbital (ANO) basis sets in 1987 [26], making the important observation that the occupation numbers of the ANOs in atomic calculations (Figure 3) provide an excellent criterion for their inclusion in molecular calculations of dynamical correlation. Their work provided the inspiration for Dunning’s hugely successful correlation-consistent polarised basis sets in 1989 [27] (in which basis-set inclusion is based on the related energy criterion in atomic systems) and later to the two-point basis-set extrapolation scheme of Helgaker and co-workers in 1999 [28]. Exploiting the underlying principal (rather than partial-wave) expansion of the ANO and correlation-consistent basis sets, such extrapolation schemes reduce the error in the correlation energy by an order of magnitude at little or no cost, thereby reaching chemical accuracy for the dynamical correlation energy.



**Figure 4.** (Colour online) The graphical event log-file of the first globally distributed – locally parallel Hartree-Fock calculation involving servers on both sides of the Atlantic returning GFLOPS performance. The availability of the servers (number of CPUs in parenthesis) is given in terms of per cent of processors. The evaluation of the Fock matrix was split into 16 batches. The transfer of the partial results happened to be fairly synchronous. The transfer times across the Atlantic are visibly larger. The processing of the one-electron integrals and the diagonalisation are performed by the client running on server 1.

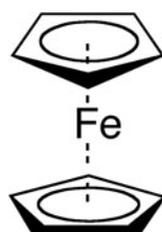
### 2.4. Parallel and network computing for large SCF calculations

The integral-driven Hartree–Fock method lends itself to parallelisation much more easily than the conventional disk-based implementation. The most straightforward approach is to have each slave process a batch of integrals and to build from these a private copy of the Fock matrix. Subsequently, the master process collects the partial Fock matrices from its slaves to determine the energy and the new density matrix, which then is broadcast to the slaves in case another SCF cycle has to be performed. With this simple scheme of parallelisation, DISCO was among the first application codes to reach Billion Floating Point Operations per second (GFLOPS) performances on an eight-processor Cray YMP computer [29].

Obviously, the slaves do not need to be running on the same machine, but can be distributed over a network of multiprocessor computers at geographically different locations connected via the Internet [30,31]. In September 1991, in a Hartree-Fock calculation using a wide-area network with servers on both sides of the Atlantic, GFLOPS performance during regular service hours was observed (see Figure 4 or [30] for more details). Later, speedups of close to 30 and a performance of 16.2 GFLOPS were recorded for a network of two (dedicated) 16-processor Cray C90 computers of the Cray Corporate Network and the Pittsburgh Supercomputer Center (see Table 3 in [31]). The communication between master and slaves was through asynchronous remote procedure calls



**Figure 5.** Per Siegbahn, Jan Almlöf and Ulf Wahlgren at their host's (Björn Roos' (leftmost)) cottage in Smedjebacken near Lund around 1980.



**Figure 6.** Ferrocene, next to the porphyrins (see Figure 2), accompanied Jan Almlöf over his entire career: from 1975, Hartree–Fock calculations on the Mössbauer shift in ferrocene and ferricinium with Ulf I. Wahlgren and Paul S. Bagus [67], via the first direct SCF calculation at the Hartree–Fock basis-set limit [9], to giving the final hint to the solution of the metal to ligand distance issue using a precursor of coupled cluster theory [68].

via the Internet using a self-written package [32]. This all happened at a time well before grid computing became a major topic in computing.

Even though it received much attention by the community of computational scientists, the limitations of network computing soon became evident. Issues such as fault-resistance, fault-tolerance or balancing the load on a (heterogenous) network of computers running under different schedulers would require a substantial effort. More importantly, there was the insight that the linear increase in compute power generated by adding more and more computers to the network would not make much larger systems accessible as long as the underlying computational algorithms did not scale linearly with system size.

## 2.5. Integral approximations and the Laplace transform

So far, Jan Almlöf's research had focused on accurate first principles methods, strictly avoiding approaches that would introduce uncontrollable or even unpredictable errors. Inspired by the work of Christian van Alsenoy [33], who thoroughly explored the multiplicative integral approximation, an approach suggested by Peter Pulay to accelerate two-electron integral calculation by expanding products of Gaussians in auxiliary basis set, Almlöf, Vahtras and Feyereisen showed that significant gains in computer time can be obtained, at no loss of accuracy, if a suitable expansion basis is chosen [34]. Similar ideas were used by Dunlap in DFT [35,36], often referred to by Jan Almlöf as Density Functional Technology (see e.g. Introduction in [34]). Such expansions of orbital products are today heavily used in all of quantum chemistry and were essential for the MP2-R12 method as developed by Wim Klopper, who was a post-doctoral fellow of Jan Almlöf's in the early 1990s [37].

The presence of the denominator in the conventional expression for the MP2 energy,  $E^{(2)} = -\frac{1}{4} \sum_{ijab} \langle ab || ij \rangle^2 / (\epsilon_a + \epsilon_b - \epsilon_i - \epsilon_j)$  was an obstacle to the parallelisation of this method. In 1991 [38], Almlöf showed that the Laplace transformation of the energy denominator  $(\epsilon_a + \epsilon_b - \epsilon_i - \epsilon_j)^{-1} = \int_0^\infty \exp[-(\epsilon_a + \epsilon_b - \epsilon_i - \epsilon_j)t] dt$  allows the orbital energies to be transferred into the respective MOs,

leading to transformed orbitals  $\psi_i(t) = \psi(0)\exp(\varepsilon_i t/2)$ . The resulting Laplace-transformed energy expression  $E^{(2)} = \int_0^\infty \sum_{ij,ab} \langle a(t)b(t) | i(t)j(t) \rangle^2 dt$  becomes embarrassingly simple to parallelise. The numerical integration can be efficiently and robustly calculated using standard numerical quadrature schemes.

Almlöf pointed out that ‘much of the significance of the above lies in the fact that a canonical representation [of the molecular orbitals] is no longer needed’, which ‘leaves some additional flexibility in defining different types of orbital rotations for computational convenience’. Here, computational convenience implies orbital localisation to reduce the orbital space and cost of integral transformation. Together with Marco Häser [39] and PhD-student Angela K. Wilson [40], he showed that the quadrature error can be controlled and that different localisation methods can be employed to increase the sparsity of the integral arrays, thus substantially reducing the memory storage requirements. Today, the Laplace transform method is his most frequently cited work.

Computational performance and controllable trade-offs between speed and accuracy are also the focus of one of Almlöf’s last scientific ventures: the separate calculation of Coulomb and exchange contributions. Inspired by the quest for linear scaling in the DFT community, the evaluation of the Coulomb and exchange matrices was decoupled, thereby *providing the freedom to pursue different specialised algorithms for the evaluation of each*. Together with his students Eric Schwegler and Matt Challacombe, he developed a method for the fast construction of the Coulomb matrix [41,42]. Using a combination of multipole expansion and explicit integral evaluation, the authors calculated the Coulomb contributions to the Fock matrix at sub-quadratic scaling for 5–60 membered water clusters and up to 16-membered polyglycine  $\alpha$ -helices.

The follow-up article on the linearly scaling computation of the Hartree–Fock exchange matrix, published in 1996 [43], does not carry Jan Almlöf’s name; Eric Schwegler and Matt Challacombe dedicate the extension of this research to his memory.

## 2.6. Method development and molecular property evaluation

As a young professor in Oslo, next to his focus on ‘faster’ and ‘larger’ calculations, Jan Almlöf also had a strong ambition towards ‘more sophisticated’ methods. Together with his colleagues Roos and Siegbahn (Figure 5), his co-workers Anders Heiberg and Peter Ruoff, along with his frequent visitors Peter R. Taylor (NASA Ames) and Alexander Nemukhin (Moscow State University), he further developed and applied multi-configuration SCF, in particular CASSCF (complete active space SCF)

methods introduced by Roos, Siegbahn and co-workers [44,45]. The most notable contribution of the Oslo group was to the Newton–Raphson formulation of the method [46], allowing to reach convergence faster, also for excited state calculations. The CASSCF method was applied to explore the full potential surfaces of small molecules with a large number of valence states within a comparatively narrow energy range (such as *BO*), highly polar species (such as *LiB*) [47–53] or chemical reactions [54,55].

In the 1990s, Jan Almlöf’s friendship with Odd Gropen (1941–2005) at the University of Tromsø (now the Arctic University of Norway) brought him back to relativistic quantum chemistry, after initial work in the field with Knut Fægri and Hans H. Grelland at the University of Oslo in the early 1980s. With Odd Gropen, he co-authored six publications on relativity [56–61]. Much of the focus of this research was the accurate evaluation of spin–orbit coupling in PdH (needed for correct state ordering) using two- and four-component multi-reference configuration interaction (CI) methods. Today, the annual Almlöf–Gropen lectures (<http://www.ctcc.no/events/almlof-gropen-lecture>) honour the memory of two central scientists in quantum-chemical research in Norway.

## 2.7. Challenges from ‘real chemistry’

Jan Almlöf did not have a specific chemical agenda – rather, his interest in chemistry was focused on the understanding of electronic structure and molecular properties at large. Still, ferrocene (Figure 6) as well as porphins and porphyrins (with or without transition-metal centres) received much attention throughout his entire career – indeed, it was a study of ferrocene that gave the impetus to the development of the direct SCF method. From a chemical perspective, being able to reach the one-particle basis-set limit gave proof that the Hartree–Fock method fails to reproduce the molecular structure of ferrocene and other transition-metal compounds [62]. In the case of ferrocene, the metal–ligand bond length at the Hartree–Fock limit was 1.89 Å, in poor agreement with the (unchallenged) experimental value of 1.65 Å. In the case of iron pentacarbonyl ( $\text{Fe}(\text{CO})_5$ ), the axial iron to carbon bond length is again much longer than the experimental value (2.05 vs. 1.81 Å), whereas the equatorial bond distance is only marginally longer (1.88 vs. 1.83 Å) [63].

The failure of the Hartree–Fock model to describe bonding in transition-metal compounds caused major irritation in the community. On the other hand, early DFT calculations did not encounter this problem to the same degree [64–66] – on the contrary, the success of DFT

**Table 1.** The most frequently cited articles of Jan Almlöf's. Even today, 20 years after he passed away, his work is being cited more than 300 times a year.

Title	Authors	Source	Year	Ref.	Cit.	'15
Atomic Natural Orbitals and the Calculation of Atomic and Molecular Properties	Almlöf, Taylor	JCP	1987	[26]	974	17
Integral Approximations for LCAO-SCF Calculations	Vahtras, Almlöf, Feyereisen	CPL	1993	[34]	851	67
General Methods for Geometry and Wave-Function Optimization	Fischer, Almlöf	JCP	1992	[16]	819	84
The Complete Active Space SCF (CAS-SCF) Method in a Newton-Raphson Formulations with Application to the HNO Molecule	Siegbahn, Almlöf, Heiberg, Roos	JCP	1981	[46]	662	26
Principles for a Direct SCF Approach to LCAO-MO ab initio Calculations	Almlöf, Faegri, Korsell	JCC	1982	[8]	571	9
Avoiding the Integral Storage Bottleneck in LCAO Calculations of Electron Correlation	Sæbø, Almlöf	CPL	1989	[22]	519	24
A Theoretical Study of Multidimensional Nuclear Tunneling in Malonaldehyde	Shida, Barbara, Almlöf	JCP	1989	[87]	231	1
Ab initio Studies on the Thermodynamic Stability of the Icosahedral C-60 Molecule Buckminsterfullerene	Lüthi, Almlöf	CPL	1987	[71]	182	1
Elimination of Energy Denominators in Moller-Plesset Perturbation-Theory by a Laplace Transform Approach	Almlöf	CPL	1991	[38]	181	10
The Equilibrium Geometry of C60 as Predicted by 2nd-Order (MP2) Perturbation-Theory	Häser, Almlöf, Scuseria	CPL	1991	[73]	175	3
Publications	187					
Sum of the times cited	11'418					
Average citations per item	61.06					
<i>h</i> -index	54					

with transition-metal systems opened the doors to acceptance of the method in chemistry.

At the time, it was clear that electron correlation would need to be addressed in calculations on transition-metal compounds, but it would take years before the problem was finally solved. While early multiconfiguration self consistent field (MCSCF) and 18-electron CI calculations on ferrocene clearly showed an improvement, the metal-ligand distance was still 0.07 Å longer than the experimental reference. Correlating the entire valence space of 66 electron was at the time difficult and would call for size-consistent methods. Early MP2 calculations including the entire valence space resulted in too short metal-ligand distances. In a 1991 article, Almlöf and his student Changyok Park, using modified coupled-pair functional calculations with dispersion corrections derived from accompanying MP2 calculations, correctly reproduced the molecular structure of ferrocene, highlighting the importance of dispersion interactions [68]. In 1996, a Coupled Cluster calculation with full treatment of Single and Double excitations, plus perturbative correction for Triple excitations [CCSD(T)] study of Koch, Jørgensen and Helgaker agreed fully with experiment, demonstrating that ferrocene is not a multi-reference system [69].

The controversy regarding the existence of 'Buckminsterfullerene', proposed in 1985 by Richard Smalley and co-workers [70], triggered Jan Almlöf's interest. Based on preliminary Hartree-Fock calculations performed on the MSI Cray-2 computer, using a version of the direct SCF code that would keep part of the two-electron integrals in the 256 million double precision word central memory

of the machine, Almlöf and co-workers came to the conclusion that 'comparison of the energy per carbon atom computed for  $C_{54}$  and "Buckminsterfullerene" shows that the soccerball shaped  $C_{60}$  molecule is indeed more stable than the single-layer sheet of graphite' [71]. Later, this claim was confirmed by computations on much larger sheets of graphite (with up to 150 carbon atoms) and by comparison to diamond-like structures (with up to 84 carbon atoms) [10].

The fact that 'Buckminsterfullerene' may well be thermodynamically stable was an encouraging message to its discoverers (personal communication). Later, after the new form of elemental carbon was isolated and characterised by Krätschmer, Huffman and co-workers [72], the calculations of the equilibrium geometry of  $C_{60}$  and derivatives reported by Almlöf, Häser and Scuseria [73], and by Scuseria [74], marked some of the largest MP2 calculations performed to that date (well beyond 1000 contracted basis functions). These calculations, involving codes other than DISCO, showed the acceptance and the power of the integral-direct methods.

Trained as a crystallographer, hydrogen bonds and hydrogen (proton) transfer were phenomena that fascinated Jan Almlöf since the early days of his career. In the 1970s, he published nearly 20 papers that carry the term 'hydrogen' in their title, some of which focus of porphyrins or their building blocks. In the 1990s, with tools much more advanced than the semi-empirical methods applied in the early days, an extensive collaboration with colleagues of the University of Minnesota faculty, Paul G. Gassman and Abhik Ghosh, on the electronic structure of porphyrins and their response to peripheral substituents

[75–84] evolved. A collaboration with Paul F. Barbara and post-doctoral student Norihiro Shida concerned proton tunnelling [85–88]. Of particular interest was malonaldehyde, an experimentally well explored but challenging system to theory, known to yield a  $21\text{ cm}^{-1}$  tunnelling splitting due to the intramolecular keto-enol tautomerism. Using a reaction surface Hamiltonian and a combination of *ab initio* SCF and Modified Coupled Pair Functions (SCF-MCPF) calculations to – first time – construct a three-dimensional potential energy surface, they obtained an effective tunnelling path that strongly deviated from the minimal energy path. Even today, this work gets considerable attention (see also Table 1).

Other collaborations with University of Minnesota faculty involved the work on degeneracy-corrected perturbation theory for electronic structure calculations with Don Truhlar [89]. Don Truhlar and Jan Almlöf also were an important and well-heard voice for supercomputing in chemistry [90,91].

### 3. Scientific legacy

Among Jan Almlöf's contributions to computational quantum chemistry, three items stand out. Perhaps the most visible are *integral-direct schemes*: they truly made larger systems accessible, also for post-Hartree–Fock methods. The *ANO basis sets* allowed basis-set error in correlated calculations to be controlled and opened the door to extrapolation schemes. Finally, The *Laplace-transform* extended the efficiency and applicability of MP2 theory and still finds new uses. The corresponding publications are among his most frequently referenced and still frequently cited today; see his top-10 articles in Table 1.

Most of his research was on the development of methods and algorithms that would perform well on the most advanced computer architectures. Often, he took up existing methods, some of which had not gotten the attention they deserved, and transformed them to deliver the best possible performance. He used to say that he would rather be running the most recent code on older hardware, than running older code on the most recent hardware. Given his record and his influence on method and code development, Jan Almlöf stands out as one of the pioneers of computational quantum chemistry.

At the same time, Almlöf was among the scientists who helped turn computational chemistry into a well-accepted tool in chemical research. His work on hydrogen bonding, on the transition-metal–ligand interactions and on the molecular and electronic structure of allotropic carbon compounds carried a strong message and served as inspiration and motivation for many other chemists.

### Notes

1. Usually referred to as the 1963–1974 generation of computers, such as the CDC Cyber 6000 series, the Univac 10/11 or the IBM 360/195.
2. Reference [2] is a translation of the original MOLECULE technical report [1] provided by Peter R. Taylor and published in the Jan Almlöf memorial issue of Theoretical Chemistry Accounts [4].
3. these results were published only much later; see also reference 23 in [68]
4. Which at the time was a huge memory, comparable to the total external storage available in an academic compute-centre, and often difficult to explain to colleagues who thought that this can't possibly be true

### Acknowledgments

This article is dedicated to Elisabeth Almlöf. First as graduate students, then as post-docs and later as frequent visitors, we both enjoyed the great hospitality of the Almlöf family.

### Disclosure statement

No potential conflict of interest was reported by the authors.

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