Theoretical chemistry in Belgium

Benoît Champagne · Michael S. Deleuze ·
Frank De Proft · Tom Leyssens

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In Belgium, theoretical chemistry began more than 50 years ago, with an initial focus on quantum chemistry, which gradually developed into a general interest in different domains of theoretical chemistry. In the *Florilège des Sciences en Belgique* [1], Louis d'Or cites as founding members of quantum chemistry in Belgium: Jean-Claude Lorquet at the Université de Liège, Georges Leroy at the Université catholique de Louvain (UCL), Georges Verhaegen at Université libre de Bruxelles (ULB), Luc Van-

quickenborne at Katholieke Universiteit Leuven (KUL), and Piet van Leuven at Antwerpen (RUCA).

Nowadays, Belgium counts around 200 theoretical chemists, spread over 10 universities (Fig. 1). This special issue includes contributions from the different theoretical chemistry groups, illustrating the diversity and richness of the field whereas this Editorial is the occasion to sketch some aspects of the evolution of quantum chemistry and theoretical chemistry in our country.

Key elements in the developments of the field have also been the collaborations, the creation of working groups, and the organization of conferences, of which the two-yearly meeting *Quantum Chemistry in Belgium*, that was the stimulus for preparing this special issue. The first issue of the meeting took place in 1995 at the University of Namur, and during the last 17 years (1996 in Leuven, 1997 in ULB, 1999 in Antwerpen, 2001 in Liège, 2003 in Ghent, 2006 in Mons, 2008 in Hasselt, 2010 in Louvain-la-Neuve, 2012 in VUB), it has been organized in all the universities. The second round will start in 2014 in Namur.

Progresses in theoretical chemistry have always been associated with the development of computational resources, from more local architectures to the larger centers recently installed in the two regions of the country, the Vlaams Supercomputer Center and the Consortium des Équipements de Calcul Intensif (CÉCI). Theoretical chemistry in Belgium has over the years largely benefited from funding by scientific agencies such as the *Fonds voor Wetenschappelijk Onderzoek* (FWO-Vlaanderen) and the Instituut voor Wetenschap en Technologie on the Flemish side, the Fonds de la Recherche Scientifique (F.R.S.–FNRS) and the Fonds de la Recherche pour la Formation dans l'Industrie et dans l'Agriculture on the French speaking side, as well as the Belgium Science Policy Office at the national level.

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B. Champagne (✉)

Laboratory of Theoretical Chemistry, Unit of Physical Chemistry, Chemistry Department, University of Namur, Rue de Bruxelles, 61, 5000 Namur, Belgium
e-mail: benoit.champagne@unamur.be

M. S. Deleuze

Research Group of Theoretical Chemistry and Molecular Modeling, Hasselt University, Agoralaan Gebouw D, 3590 Diepenbeek, Belgium
e-mail: michael.deleuze@uhasselt.be

F. De Proft

Faculteit Wetenschappen, Eenheid Algemene Chemie (ALGC), Vrije Universiteit Brussel (VUB), Pleinlaan 2, 1050 Brussels, Belgium
e-mail: fdeproft@vub.ac.be

T. Leyssens

Laboratory of Crystal Engineering, Institute of Condensed Matter and Nanosciences, Université Catholique de Louvain, Place Louis Pasteur 1, bte L4.01.03, 1348 Louvain-La-Neuve, Belgium
e-mail: tom.leysens@uclouvain.be

Fig. 1 Map of Belgium representing the cities where the different universities discussed below are located

1 University of Antwerp

The *Universitaire Instelling Antwerpen* (UIA) was founded in 1971. On October 1, 2003, it became part of the University of Antwerp (UA) which united RUCA (State University Centre Antwerp), UFSIA (University Faculties Saint Ignatius Antwerp) and UIA (University Institution Antwerp). Since the foundation of the UIA, research using quantum chemical methods has been performed in the “Structural Chemistry” group led by H.J. Geise working on electron diffraction (ED) and by A.T.H. Lenstra working on X-ray diffraction. At that time, mainly semi-empirical calculations such as MINDO/3 were performed to assist in the interpretation of the experimental data with these techniques. C. Van Alsenoy joined this group in 1978. During this period, structural chemists became aware of the enormous potential of P. Pulay’s force method in their research field. With this in mind, C. Van Alsenoy went to the USA for two postdoctoral stays, the first one with L. Schäfer at the University of Arkansas and the second one with J. Boggs at the University of Texas (at Austin) where he worked under the guidance of P. Pulay for a period of 6 months. During this period, the basis for the Multiplicative Integral Approximation (MIA) was established, which later evolved into the Multiplicative Integral Approach.

When C. Van Alsenoy returned to the University of Antwerp, research in the group of quantum chemistry was directed mainly along two lines. A first purpose was to make the Molecular Orbital Constrained Electron Diffraction (MOCED) approach routinely available to people doing Electron Diffraction in the group. In the MOCED approach,

differences in bond lengths and vibrational amplitudes from ab initio (HF) gradient calculations were used as constraints in the refinement of ED-experimental data. A code was thereby set up to do geometry refinements, force field, and vibrational frequency calculations, along with normal-mode fitting. The second line of research involved the further development and implementation of the MIA into Pulay’s TEXAS quantum chemical package. This code, after further refinement and optimization, evolved into the program BRABO, a package which besides enabling SCF (HF and DFT) calculations in parallel also contains software to relax the molecular structure in geometry optimization, to construct clusters based on fractional coordinates and space group symmetry, to calculate and plot molecular density (-difference) maps, and to partition molecular quantities using the Hirshfeld approach.

To date, the MIA approach can be applied routinely in HF and DFT calculations as well as CPHF and CPKS calculations of polarizabilities and NMR chemical shifts. These developments were used in numerous studies, among others for unraveling the structure of the crambin peptide. At the time, this achievement was recognized by I. Levine in his book “quantum chemistry” as the largest ever performed quantum chemical calculation. Other studies involved cluster calculations in order to explain structural differences and vibrational frequency shifts in molecules between the gas-phase and the crystal-phase structures, depending on the space group. Another important and more recent line of research in C. Van Alsenoy’s research group is devoted to the study and use of the Hirshfeld approach for partitioning molecular properties such as total charge distributions, molecular polarizabilities

as well as the total molecular energy into atomic contributions, at various levels of theory (HF, DFT, and MP2). A very promising extension along these lines of research is the use of Hirshfeld partitioned quantities of multipole polarizabilities in the study of dispersion effects complementing DFT calculations.

In parallel to the work by C. van Alsenoy, research by Renaat Gijbels and Annemie Bogaerts in Antwerp led over the years to the foundation of an interdisciplinary research group “PLASMANT” (Plasma, Laser Ablation and Surface Modeling—ANTwerp) where theoretical chemistry also forms an important line of research. R. Gijbels started his PhD work at Ghent University in 1961, in the research group of J. Hoste, which later evolved to the Institute of Nuclear Sciences. His topic was the determination of traces of noble metals in other, high-purity noble metals, via neutron activation analysis (NAA). After a few years, D. Desoete and R. Gijbels, together with J. Hoste, embarked on the preparation of a monograph on Neutron Activation Analysis. R. Gijbels took care of the more “fundamental” chapters and realized that NAA practitioners were not enough aware of a number of basic concepts, elastic and inelastic scatterings, excited states and metastable states, among others. So, he started to study and to clarify these concepts in the book. As a consequence, a number of PhD works started in the group, for example, for the determination of average cross-sections of so-called threshold reactions induced by fission neutrons, by J.P. François (see his contribution at the University of Hasselt).

R. Gijbels continued to follow this double track: theory and different practical applications by NAA in a variety of nuclear reactors. Modeling received again a boost with the arrival of a postdoc from the Hungarian Academy of Sciences, A. Vertes who started a 1-D model for laser-solid interaction. Another even more fruitful line of research started with the arrival, in 1986, of Jan M. L. Martin for his master thesis in Antwerp. R. Gijbels had seen a large variety of carbon cluster ions in spark source and laser induced mass spectra, and wondered what their structure could be. J. Martin performed quantum chemical calculations to model these clusters, in close collaboration with J.-P. François, at the University of Hasselt.

In 1993, A. Bogaerts joined the group as a PhD student, and she developed a computer model for a glow discharge plasma, used as an ion source for glow discharge mass spectrometry. After finishing her PhD thesis in 1996, she became an FWO postdoc in the group and started a subgroup on plasma modeling, also for other applications than analytical spectrometry (see below). This group was gradually growing, and new activities started, that is, on classical molecular dynamics simulations for plasma-surface interactions (in 2001) and on modeling for laser ablation (i.e., laser-solid interaction, plume expansion, and

plasma formation) (in 2002). In 2001, A. Bogaerts was prize winner of the Royal Flemish Academy of Belgium for Sciences and Arts. In 2003, she was appointed as a professor. After the retirement of R. Gijbels in 2004, the group was renamed as “PLASMANT”. In 2011, E. Neyts, who made his PhD and postdoctoral work in the group on molecular dynamics (MD) simulations for plasma deposition of coatings and carbon nanotube growth, respectively, started in the group as a tenure track professor.

Currently, the group consists of about 20 people (PhD students and postdoctoral researchers; under the supervision of A. Bogaerts and E. Neyts, and one technical-administrative coworker). As the name says, the group is mainly performing computer modeling for (i) plasmas, (ii) laser ablation (laser-surface interactions), and (iii) plasma-surface interactions. The first two fields are under the supervision of A. Bogaerts, whereas the third topic is under the supervision of E. Neyts, especially the combination of modeling both the plasma itself and its interaction with surfaces gives the group a unique expertise.

Theoretical chemistry activities in the University of Antwerp are discussed in the papers by Geldolf et al. [2] and by Neyts and Bogaerts [3] of the present issue.

2 Free University of Brussels

2.1 Université Libre de Bruxelles (ULB)

Quantum chemical research at the Université Libre de Bruxelles started in the mid-sixties. In 1965, Reginald Colin (RC) and Georges Verhaegen (GV) completed their PhD theses in high-temperature chemistry in the Laboratory of P. Goldfinger. The main characteristic of these studies was the discovery of numerous new molecules by mass spectrometry. It was the urge to learn more about the structure of these new species that determined the fields of postdoctoral studies they both chose: RC went to the Herzberg Institute in Ottawa to work with A.E. Douglas in Molecular Spectroscopy; GV went to the “Centre de Mécanique Ondulatoire Appliquée” (CMOA) of R. Daudel in Paris to work with C. Moser in quantum chemistry. The first publications of GV in this emerging field concerned the molecules BeO and MgO, both treated in his thesis.

Ab initio calculations have always demanded, and still demand, the largest possible computing possibilities, both in terms of speed and capacity. Back from the CMOA, the available computer in the ULB-VUB Center was then an IBM 650, much too small to accomplish anything, but a moderate LCAO-SCF calculation on very small atoms. Therefore, after discussions with the FNRS, GV was able to set up the then well-known SCF diatomic molecular

program of Nesbet on the large computer of the Darmstadt Center. At the time, a quantum chemistry calculation meant sending by post perforated cards and waiting around a week to receive the listings of results back—converged or not! Fortunately, things improved rapidly, and calculations could then be carried out on the CDC computer mainframes newly installed at ULB. Nevertheless, in order to predict theoretically meaningful results, the Hartree–Fock results are insufficient, and for most properties, correlation effects need to be considered. Since the *ab initio* calculations involved were much too extensive for the available computers of the time, GV and his students developed an original atom-in-molecule approach to insert correlation effects in the results. Indirectly, this prompted the beginning of studies in theoretical atomic physics in the laboratory. G. Verhaegen was continuously deeply involved in getting the adequate computer resources to perform state-of-the-art calculations. Rector of ULB (1986–1990), he looked for partners interested in using or promoting the access to the first supercomputer of the country. The FNRS launched a stimulus program, leading to the inauguration of the CRAY X-MP/14 installation at the VUB/ULB Computing Center in 1989.

From the early start, G. Verhaegen's successors Jacques Liévin (JL) and Michel Godefroid (MG) and their students have greatly developed both the quantum chemistry (JL) and atomic physics (MG) fields of research in the laboratory. Later on (1990), Nathalie Vaeck (NV) also contributed to the atomic physics field before opening an original research line in quantum dynamics. The three permanent members (JL, MG, and NV) of the “Quantum Chemistry and Atomic Physics” theoretical group of the “Chimie Quantique et Photophysique” Laboratory developed numerous fruitful international collaborations and networks. Their various research activities are illustrated in Cauët et al.'s [4] contribution to the present issue. The contribution of Brian Sutcliffe, who has been a visiting professor in the group since 1998, focuses on formal aspects related to the concept of rovibrational hamiltonians and potential energy surfaces [5].

2.2 Vrije Universiteit Brussel (VUB)

The VUB offered a compulsory course on basic quantum mechanics and an introduction to quantum chemistry from the start of the chemistry curriculum in the early sixties. These lectures were given by André Bellemans, a former student of Nobel Laureate Ilya Prigogine at the ULB and still one of his collaborators at that time, specialist in statistical mechanics. In 1974 when Bellemans resigned from his VUB charge, Henk Lekkerkerker was appointed for teaching the complete range of theoretical physical chemistry courses (including thermodynamics and quantum

mechanics). His teaching was as excellent as that of his predecessor, but just like André Bellemans the main part of his research was not devoted to quantum chemistry. In 1985, he changed his Full Professorship at the VUB for the position of head of the prestigious Van't Hoff Institute in Utrecht (where he previously obtained his master degree). Since the late sixties, Hubert Figeys, a pupil of the famous ULB organic chemistry Professor Richard Martin, gave a small elective course on Theoretical Organic Chemistry, followed among others by Paul Geerlings and Christian Van Alsenoy during their Master studies. Both of them graduated with him and finished their PhD in 1976 and 1977, respectively, on theoretical aspects of IR and NMR spectroscopy. Whereas C. Van Alsenoy left the VUB soon after to join Herman Geise's structural chemistry group in Antwerp, P. Geerlings stayed at the VUB and was appointed for the Quantum Mechanics and Theoretical Organic Chemistry Courses in 1985. He started a research group, which at the end of the eighties fully concentrated on theoretical and applied aspects of Density Functional Theory, with particular attention to conceptual or “Chemical Reactivity” DFT. Once appointed as full-time professor in 1990, as successor of Louis Van Hove as director of the General Chemistry Laboratory, his group grew quickly, also under the impetus of two young PhD students, Wilfried Langenaeker and Frank De Proft. For more than 20 years, now his group is responsible for all teaching activities around quantum chemistry, molecular modeling ... (besides the basic course in General Chemistry for the Faculty of Science and, until 1997, the Faculty of Medicine). Meanwhile, F. De Proft became professor and codirector of the group, whereas W. Langenaeker left the group for a position in industry. In 2007, F. De Proft was Laureate of the Royal Flemish Academy of Belgium for Sciences and Arts in the division of natural sciences.

The group attracted many pre- and postdoctoral fellows and has collaborated with numerous research groups all over the world. It took care of 30 promotions (six being in progress) and published around 450 papers in international journals or as book chapters. In 2003, the group published an influential review on the field of conceptual DFT (*Chemical Reviews* 2003, 103, 1793–1873), which, at the present moment, has been cited more than 1,050 times. The group has been the nucleus group for more than 15 years of the FWO Research Network “Quantum chemistry: fundamental and applied aspects of density functional theory”, and has been active in the organization of several international meetings around DFT, from which DFT 2003, the Xth International Conference on Applications of Density Functional Theory in Chemistry and Physics, Brussels, Belgium, September 7–12, 2003, is best known.

The present composition of the group varies between 20 and 25 members with various backgrounds (chemistry,

biology, physics, chemical, and bio-engineering), often enabling interdisciplinary research. Research activities are varying from fundamental aspects of DFT to applications in organic chemistry, catalysis, bio systems, and “nano” technology such as fullerenes, nanotubes, and graphene for which G. Van Lier was recently offered a part-time professorship. The contribution of the group in this special issue by De Vleeschouwer et al. [6] focuses on the computation of one of these chemical concepts, the electrophilicity, for radicals and the scrutiny of the effect of the solvent on this quantity.

3 Ghent University

3.1 Ghent quantum chemistry group (GQCG)

The department of chemistry at Ghent University, more specifically the then Laboratory of General and Inorganic Chemistry, already started to use quantum chemical calculations in 1970s mainly to assist in interpreting spectroscopic data although a dedicated quantum chemistry group did not exist. Only limited courses were taught by local spectroscopists. In 2000 Professors A. Goeminne and D. Van de Vondel, respectively, head of department and the spectroscopist lecturing quantum chemistry decided that a dedicated quantum chemistry group that bases its lecturing tasks on research expertise was due. Thanks to their initiative and insight, such a group was eventually founded in 2001 and has become known as the Ghent Quantum Chemistry Group (GQCG).

The group started with one professor (Patrick Bultinck) appointed in October 2001 and one Ph.D. student and started activities over a widespread range of areas including computational medicinal chemistry and chiroptical vibrational spectroscopy. At the beginning, the research was rather application directed with emphasis on conformational analysis, QSAR, and electronegativity equalization in medicinal chemistry and combined experimental/computational studies in Vibrational Circular Dichroism (VCD).

Nowadays, the group, varying in number between 8 and 12, concentrates on two themes, broadly categorized as “Electron density (matrices)” and “chiroptical spectroscopies.” The first category contains both the fundamental study of density matrices, including their (wavefunction free) variational optimization and especially their use and meaning for studying chemical concepts through analysis of their properties. Examples are the study of Domain Averaged Fermi Holes from the study of the exchange–correlation density, delocalization indices and especially Aromaticity. The interest in the last being an obvious consequence of the fact that F.A. Kekulé published his

famous papers on the tetravalence of carbon and the cyclic structure of benzene while being professor at Ghent University. The group introduced the popular density matrix based multicenter indices for aromaticity and scrutinized the meaning of this chemical concept. The meaning and the relevance of the many indices available were critically investigated. A second example of work along these lines is the Hirshfeld-I atom in the molecule, which corrects some issues with the traditional Hirshfeld atom in the molecule. The second line of research concentrates on Chiroptical spectroscopies like VCD and Raman Optical Activity (ROA) where we carry out both experimental studies using own infrastructure and quantum chemical calculations and implement new algorithms with emphasis on using both techniques to establish absolute configurations of molecules and higher-order structures of biomolecules.

3.2 Center for Molecular Modeling (CMM)

Since the end of the eighties, there was a worldwide tendency to break off research activities in low-energy and even intermediate-energy nuclear physics, and policy makers started with emphasizing the necessity of the presence of applied, economical, and utility finalities in the funded research activities. In 1997, Michel Waroquier decided to switch his research field from nuclear many-body problems to *ab initio* methods for tackling molecular systems. He started the new research area with a PhD student, Veronique Van Speybroeck. The first paper in the new field appeared 3 years later in 2000. It was a subject in the Chemical Technology with focus on model development and application to an industrially important chemical reaction. The strategy was the development of new models, new methodologies going beyond state of the art, implementation in computational codes, and application to important processes to validate the model. It was a success, the first and also the only paper in 2000 is currently still the most cited paper of the CMM in the new field. Gradually, the team grew with special attention in maintaining a good balance between physicists, chemists, and engineers with the principal aim to stimulate a strong synergy between the various research cells, encouraging interdisciplinary research that goes beyond the state of the art, and with a special focus to application driven areas.

The current research of the CMM is focused along six major areas. The core activities are situated in the research domains “Nanoporous materials-catalysis,” “Organic Chemistry and Biochemistry,” “Spectroscopy,” “Computational Material Research,” “Model development,” and a more fundamental area “Many Particle Physics.” The six areas define the core business of the main activities, and research in each of them is performed within the frame of a strong network with partners at the UGent, in Flanders and

at an international level. There is a strong synergy between the various research cells, stimulating interdisciplinary research.

Nowadays, the research center has grown to a population of 35 researchers with more than 50 publications per year. The first PhD student in the new field of molecular modeling, V. Van Speybroeck, has become now full professor at the UGent and leads currently the computational division of the CMM. Dimitri Van Neck is head of the more fundamentally oriented area. The other research domains of the CMM are headed by a part-time professor. Currently, the CMM members are author of about 437 papers in ISI journals, among which *Nature Materials*, *Angewandte Chemie*, *Journal of the American Chemical Society*, *Physical Review Letters*, *Journal of Catalysis*, etc., with more than 6,600 citations. The paper by A. Ghysels et al. [7] and A. Cedillo et al. [8] in the present issue gives illustrations of the research carried out in the CMM and the QCG.

4 University of Hasselt

Research in quantum chemistry at the Limburgs University Center (Now: University of Hasselt) started in 1978 under the motivation of Jean-Pierre François (Professor of Chemistry in the period 1975–2008 (JPF)). JPF obtained his PhD in 1971 at the State University of Ghent (Now : Ghent University) in the field of nuclear chemistry under the supervision of the late Prof. J. Hoste. In 1973, J.-P. François left Ghent University and became Chief Assistant at the University of Hasselt (UHasselt). He was promoted Professor of Chemistry in 1975 and switched to quantum chemistry in 1978. The first study that has been undertaken was the computation of an extensive series of monosubstituted pyridines and phenolates in the gas phase using semi-empirical (MINDO/3, MNDO, and AM1) and ab initio methods using a program vectorized in the group for the Cyber 205 vector processor.

In 1987, Jan M.L. Martin (JM) joined the group of quantum chemistry in Hasselt as PhD student. He obtained his PhD degree in Sciences in February 1991 (supervisor: J.-P. François, cosupervisor: R. Gijbels). The main purpose of his research activities was to study extensively neutral and charged carbon and boron-nitride cluster species of relevance in materials science and astrophysics. Combined bond-polarization basis sets were developed for accurate calculations of dissociation energies. In 1991–1995, J. Martin became Postdoctoral Fellow (“Postdoctoraal Onderzoeker”) at the Belgian National Science Foundation (NFWO/FNRS). In this period, anharmonic force fields and thermochemical quantities of a variety of molecular species (including clusters) were computed, starting from ab initio

potential energy surfaces. The methods used were vibrational perturbation theory and vibrational CI. In 1995, J. Martin became Senior Research Associate (“Onderzoekssleider”) at the NFWO/FNRS. He left the research group in 1996, to become Assistant Professor at the Weizmann Institute of Science, Rehovot, Israel. He has been awarded the Dirac medal at the 7th congress of the “*World Association of Theoretically Oriented Chemists*” (WATOC05, President Henry F. Schaefer III—University of Georgia, USA) in Cape Town, South Africa (January 15–21, 2005). The paper by U. R. Fogueri et al. [9] of the present issue illustrates recent research activities by J. Martin and his coworkers in Rehovot and at the University of North Texas in Denton.

In the period 1988–2001, extensive work was done by J.-P. François and his coworkers on the structure and IR spectra of carbon clusters ranging from C_3 and C_3^+ to C_{24} and on a number of boron-nitrogen B_mN_n clusters. Further theoreticians involved in that work were P.R. Taylor (NASA Ames Research Center, Moffett Field, CA), the late Prof. J. Almlöf (University of Minnesota, Minneapolis, MN), Z. Slanina (Heyrovsky Institute of Physical Chemistry and Electrochemistry, Prague), Zhengli Cai (Nanjing University of Science and Technology, China), M.S. Deleuze (MD), and several PhD students. A main purpose of the work on the larger carbon clusters (C_{20} , ...) was to find which species exhibits first a fullerene structure. Results obtained for the vibrational spectra of the lower C_n clusters were of great value for the IR spectroscopic work with Doppler limited resolution of J.R. Heath (University of California, Berkeley, CA), who performed later the historical experiments leading to the discovery of C_{60} with Sir H.W. Kroto, R.E. Smalley, and R.F. Curl as well as S.C. O'Brien. The complex IR spectra of C_n species trapped in noble gas matrices could be analyzed quantitatively with the aid of quantum chemical data obtained using a computer program developed in the group.

The successor of J. Martin, Michael S. Deleuze, obtained his PhD in 1993 at the Facultés Universitaires Notre-Dame de la Paix de Namur in the field of ionization spectroscopy using propagator theory (Supervisor J. Delhalle), prior to undertaking three postdocs on behalf of the FNRS and of the Training and Mobility Research program of the EU, in the groups of Barry T. Pickup (Sheffield University, UK, 1994), Lorenz S. Cederbaum (Heidelberg University, Germany, 1995), and F. Zerbetto (University of Bologna, Italy, 1996). In 1997, Dr. M.S. Deleuze went back to Belgium to join the group of theoretical chemistry at the UHasselt as Postdoctoral Fellow (FWO Vlaanderen). In 1999, he was promoted Senior Research Associate and in 2000 Research Professor. He introduced one-electron Green's function theory and the interpretation of advanced orbital imaging experiments employing Electron

Momentum Spectroscopy or Penning Ionization Electron Spectroscopy into the research activities of the UHasselt. Further research topics developed under his supervision on local ES40, ES45, and ES47 workstations comprise: material sciences and polymer physics (long-range and delocalization effects, hyperconjugation ...); electronic excited states; shake-up and correlation bands in valence ionization spectra, linear response properties; molecular dynamics of supramolecular systems (e.g., catenanes) and clusters of buckminsterfullerenes; conformational analysis, with emphasis on the relationships prevailing between the molecular and electronic structures; electronic and structural properties of carbon and boron-nitrogen clusters, or boranes and carboranes; reaction mechanisms of the conversion of sulfoxide, sulfonyl and xanthate precursors of conjugated polymers; thermal effects on the structural, electronic and optical properties of conjugated chains; nucleation of organic half-conductors on inert surfaces; photochemistry under far-UV-radiation; ring currents and magnetic responses in polycyclic aromatic hydrocarbons; symmetry-breakings and correlation effects in *n*-acenes, graphene nanoislands and nanoribbons. In 2006, M. S. Deleuze was prize winner of the Royal Flemish Academy of Belgium for Sciences and Arts. The paper by B. Hajgató et al. of the present issue [10] gives an illustration of recent research activities of the group of theoretical chemistry at the University of Hasselt on complications inherent to the interpretation of orbital imaging experiments.

5 University of Leuven

Quantum chemistry was introduced at the University of Leuven (KU Leuven) in 1967 by Luc Vanquickenborne, who had obtained a PhD in combustion chemistry in 1964 at that same University. His passion for quantum chemistry was kindled during his 2-year postdoctoral research stay in the US, where he worked in the Laboratory of Sean McGlynn on the theory of molecular spectroscopy. Upon his return to Belgium in 1967, he obtained a FWO postdoctoral fellowship, and he developed a research group focused on theoretical aspects of inorganic chemistry, in close collaboration with the experimental inorganic chemistry groups. Even up to now, the study of inorganic systems remains a major focus of the theoretical chemistry group in Leuven. In the years following his arrival at the KU Leuven, L. Vanquickenborne guided a multitude of PhD students, among which Arnout Ceulemans, Marc Hendrickx, and Kristine Pierloot.

Arnout Ceulemans obtained his PhD working on a ligand field and group theoretical analysis of photochemical reactions of transition metal compounds. Following his

PhD, A. Ceulemans obtained a permanent position at the FWO, becoming the second permanent staff member focusing on quantum chemistry. In 1995, A. Ceulemans switched from the FWO to a permanent position as a full professor at KU Leuven. Although his initial research focused on inorganic compounds, his current interest has shifted to research on clusters, fullerenes, and bioorganic systems.

After having received his PhD in 1985 at KU Leuven, Marc F. A. Hendrickx was the second researcher to join the theoretical chemistry group of this university as a permanent member of the academic staff. Since then, the main focus of his research activities has been on the study of properties of a wide variety of transition metal compounds. His recent research activity is mainly directed toward applying quantum chemical methods on small transition metal-containing clusters. Their frequently complicated open-shell electronic structures are studied in relation to their magnetic and spectroscopic properties.

The theoretical chemistry group at Leuven was expanded further with Kristine Pierloot, who like A. Ceulemans obtained a permanent research (FWO) position prior to joining the KU Leuven academic staff in 2000. The current research area of her group primarily concerns the investigation of the electronic structure of transition metals in a variety of coordination environments, with a special focus on bioinorganic and biomimetic systems, as well as on electronic spectroscopy. For this purpose, she is strongly involved in the development and application of multiconfigurational wave function methods, in collaboration with the MOLCAS developer's team, which has its origin in Lund (Sweden), but has by now spread its wings all over the world.

The fourth member, Minh Tho Nguyen, followed a different path. After surviving the difficult years of the Vietnam war, he obtained, in 1971, a scholarship to study chemistry at UCL. In 1980, he completed his doctoral thesis in Louvain-la-Neuve under G. Leroy, focusing on mechanisms of organic reactions. Subsequently, he did several postdocs (Universität Zürich, ETH Zürich, KU Leuven, University College Dublin, Australian National University Canberra) before joining the University of Groningen, Nederland, in 1988 as an associate professor. In 1985, he was awarded a D. Sc. degree by the National University of Ireland. He then received a phone call from L. Vanquickenborne. Ardently attracted by the charm of the Brabant region he returned to Leuven in 1990, definitively and for good, becoming first a research director of the FWO and later a full professor at KU Leuven. Nguyen was/is visiting scientist and professor at different institutions in France, USA (in particular University of Alabama), Taiwan, Japan, and Vietnam. His study focuses on the discovery

of novel chemical phenomena and concepts by use of quantum chemical computations.

The last member of the group, Liviu Chibotaru, has obtained his Ph.D degree in 1985 in Chişinău (Moldova, that time in the USSR) under the supervision of Isaac Bersuker. After the collapse of Soviet Union he, like many of his colleagues, drifted West to pursue scientific research. In 1995, he became a postdoctoral fellow in the quantum chemistry group at KU Leuven. He joined the permanent staff in 2004 soon after L. Vanquickenborne became professor emeritus in 2003. His research combines expertise from chemical and condensed matter physics and is currently focused on the investigation of novel nanomagnets, mesoscopic superconductors, and carbon materials.

Together, the theoretical chemistry group that was initiated by L. Vanquickenborne has by now published over 1200 articles in international journals or as book chapters. A total number of 66 students have finished their doctoral studies in this group, and 10 are currently working on a PhD. The group has also attracted many pre- and post-doctoral fellows, and research is most often performed in concert with other, often experimental partners. Together, the staff members take care of a vast number of introductory and advanced theoretical courses in the bachelor and master programs offered by KU Leuven (quantum- and computational chemistry, group theory, molecular spectroscopy, reaction kinetics, solid-state methods). These courses also form the core of the KU Leuven contribution to the European master in theoretical chemistry and computational modeling, an Erasmus Mundus master course offered jointly by six European universities, introduced at KU Leuven in 2010 with A. Ceulemans as the local coordinator.

The theoretical chemistry research activities from the KULeuven are illustrated in the contributions by Ceulemans et al. [11], Phung et al. [12], and Tai et al. [13].

6 University of Liège

The story of quantum chemistry at the university of Liège started in November 1956 when, 2 days after receiving his B. Sc. degree, a young researcher knocked at the door of the Centre de Chimie Théorique in Paris, headed by Raymond Daudel. Jean-Claude Lorquet had received permission from his suspicious adviser to start a thesis in quantum chemistry, and he later on was allowed to develop a research group under the strict condition that “useful results” should be derived. In practice, this meant maintaining close cooperation with an experimental team working by mass spectrometric techniques on the chemistry in ion beams. The way to proceed was not at all obvious. For example, methods to perform calculations on

open-shell molecules were largely a matter of debate in the fifties, and each group had to develop techniques of its own. But, even worse, it soon appeared that it also required tackling problems that are not commonly part of the chemist's stock in trade. As a result, in its efforts to study reaction dynamics in electronically excited states, the group specialized in such problems as nonstationary states, time-dependent wave functions, breakdown of the Born–Oppenheimer approximation, potential energy surface crossings, nonadiabatic transitions, and spin–orbit couplings. Among the persons who were most instrumental in developing proper methods is Michèle Desouter who, in her Ph.D. thesis, established symmetry relations between the diabatic and adiabatic representations and showed their complete equivalence. Much later on, she moved to the university of Paris-Sud. But before leaving Liège, she had supervised the thesis of Françoise Remacle (FR), who has since renewed the impetus and who is currently heading the group.

The group of theoretical physical chemistry (TPC) is led by F. Remacle since 2001, after the retirement of J. C. Lorquet. After her PhD in Liège on the role of resonances in unimolecular reactions, F. Remacle made a postdoc with R. D. Levine at the Hebrew University of Jerusalem and maintains a close collaboration with the Jerusalem group since then. The TPC group focuses on controlling the dynamics of the responses of molecular systems to perturbations, mainly pulses of photon and voltage. Early work includes the study of reactivity in a dense set of excited states in polyatomic molecules, the dynamics of high molecular Rydberg states, and transport properties of nanostructures. More recently, the TPC group showed how to use the specificity of molecular responses to selective excitations viewed as inputs to build complex logic circuits at the molecular scale. Molecular states being discrete, they can be used for implementing memory units, which opens the way to realizing finite state machines: at each cycle, the next state and outputs are functions of both the inputs and the present state. This work was supported by several EC FET grants that involved theorists and experimental groups and provided physical realizations of the designed logic schemes by electrical, optical, and chemical addressing. A new EC collaborative project on unconventional multivalued parallel computing called MULTI, coordinated by F. Remacle, is just starting. The project aims at fully harvesting molecular complexity by going beyond two-valued Boolean logic and implementing logic operations in parallel exploring alternative avenues to quantum computing.

The highest speed for logic operations will ultimately be reached by providing inputs with ultrashort atto ($1 \text{ as} = 10^{-18} \text{ s}$) photon pulses. These will allow addressing electrons directly and reach petaHz cycling frequencies.

The TPC group has a strong research activity in attochemistry and investigates the responses of molecular systems to strong ultrashort, subfemtosecond, photon pulses. The aim is to control chemical reactivity on an ultra-short timescale by directly manipulating electrons before subsequent nuclear dynamics has set in. This is also a challenge since it requires describing the coupled electron-nuclear dynamics beyond the realm of validity of the Born–Oppenheimer approximation. Investigating dynamics implies having good knowledge of electronic structure and the TPC group maintained a strong activity in this field. Special emphasis is given on the properties of gold and metallic nanoclusters and their tuning by the chemical nature of the ligand shell.

Another impulse to the development of quantum chemistry in Liège came from Georges Dive, Pharmacist, who started to work in the medicinal chemistry department of Charles Lapière in 1973. The subject of his PhD thesis was the study of anti-inflammatory drugs by quantum chemistry and multivariate statistical analysis. To analyze the conformations of more than 40 atoms molecules, he worked in Georges Leroy's laboratory at Louvain-La-Neuve (LLN) with the CNDO/2 method. It was the starting point of an efficient collaboration between several researchers, particularly with Daniel Peeters. In the eighties, G. Dive joined the microbiological laboratory of Jean-Marie Ghuysen which was devoted to the study of the activity of penicillin-like molecules on isolated enzymes involved in the synthesis of the external membrane of bacteria. Dominique Dehareng, who performed her chemistry thesis in the field of quantum dynamics with J.C. Lorquet at Liège and Xavier Chapuisat at Orsay, joined the group of J.M. Ghuysen in 1987. At that stage, the main research objective was the study of enzymatic reactions pathways, with a particular attention devoted to the electrostatic potential and its usefulness in providing a fast computable value of the electrostatic energy term in small- and medium-sized molecular complexes. Another research interest of the group is the description of the potential energy surfaces, and the location of its critical points and several PhD theses focused on that point. A special interest has been dedicated to valley-ridge inflexion points. A significant work was also devoted to the study of Hartree–Fock instabilities.

In 1990, with the first financial support of IAP (Inter-university Attraction Poles) program, the microbiological laboratory became the «Centre d'ingénierie des protéines» (CIP). It was organized as a consortium between several internal and external laboratories. A significant contribution has been the collaboration between the theoretical group and the organic laboratory of Léon Ghosez and Jacqueline Marchand (LLN) in the design of novel antibiotic molecules. Apart from pure quantum chemistry, the group

extended its research domain to classical molecular modeling based on molecular mechanics force fields and molecular dynamics as well as mixed approaches using both molecular mechanics and quantum chemistry to describe reactions occurring in a large protein-solvent environment. For the study of large molecular systems, the computer power has been very often the rate limiting step. During more than 10 years, powerful computational facilities have been installed at the CIP mainly to run Gaussian program in vectorial/parallel mode. It has been an opportunity to welcome in Liège the Gaussian workshop in December 1996.

The articles by Dive et al. [14] and by Ganesan and Remacle [15] of the present issue describe the theoretical and modeling activities of the theoretical chemistry groups in the University of Liège.

7 University of Louvain-la-Neuve

Georges Leroy (†) can be seen as one of the founders of quantum chemistry research in Belgium. After a PhD in physical chemistry (1959) focusing on crystallization, organic synthesis and UV Spectrometry G. Leroy joined the laboratory of R. Daudel at the CMOA in Paris for a postdoctoral research stay. It was during this period that he developed his passion for quantum chemistry. Upon his return in 1965, he created a physical chemistry research group interested mainly in theoretical chemistry even though experimental chemistry was still going on. The main focus of his research lays on the study of π -electrons systems such as aromatic species, graphite, ..., trying to improve semi-empirical methods such as modified Hückel theory, Pariser Parr Pople approaches, and so on. His research was internationally recognized as illustrated by a contribution to the very first issue of the International Journal of Quantum Chemistry. Among his PhD students was J.M. André, who later on founded a laboratory of quantum chemistry at the University of Namur. Physically located in Leuven, the laboratory of G. Leroy moved to Louvain-la-Neuve in 1973.

In 1974, Daniel Peeters obtained his PhD under the guidance of G. Leroy. The purpose of his thesis was to describe the chemical bond using a depiction of the wave function in terms of localized orbitals. At that time, the Centre Européen de Calcul Atomique et Moléculaire directed by C. Moser was the place to be as it provided the computational facilities unavailable elsewhere. D. Peeters, along with M. Sana (†), spent some time at Orsay (France) working on the description of potential hypersurfaces to understand chemical reactivity. In 1981, M. Sana was promoted to a permanent position as research leader of the FNRS, while D. Peeters obtained a permanent position at the UCL, and both joined the quantum chemistry group as

full members. At this stage of their career, their research was mainly focused on the investigation of the electronic structure of chemical species, with particular emphasis on their thermodynamic stability, or the description of chemical bonds with regard to reactivity issues. M. Sana, D. Peeters, and G. Leroy continued the development of the quantum chemistry group at the UCL by intensifying their collaborations with the organic chemistry groups and in the later stages with the inorganic chemistry groups. R. Robiette, part of the organic medicinal chemistry group, performed part of his PhD under the coguidance of D. Peeters. Currently, holder of a permanent FNRS research position, R. Robiette, still continues to investigate organic reactions using quantum chemistry. After G. Leroy retired in 2000, T. Leyssens performed his PhD under the guidance of D. Peeters. He then moved to UCB Pharma, and after a short postdoctoral research stay in the group of W. Thiel (Max-Planck-Institut für Kohlenforschung, Mülheim, Germany), finally returned to the UCL in 2009. He focuses on the mechanistic understanding of chemical reactions, using experimental as well as theoretical techniques, in collaboration with D. Peeters.

In parallel, in 1992, X. Gonze joined the UCL as a permanent FNRS researcher, in the engineering faculty. He switched from the FNRS to an UCL academic position in 2004. His research focuses on first principles studies of high-technology material properties at the nanoscale (electronic, optical, dynamical properties). Some fundamental aspects of Density Functional Theory have also been central to his activities over the years. At the end of the nineties, he started to develop ABINIT, to which several dozen scientists have since contributed, and which is now used worldwide for calculations on periodic solids.

The contributions by Zanti et al. [16], Vergote et al. [17], and Dive et al. [14] focus on chemical reactivity of inorganic, organometallic, and organic systems, whereas the contribution of Avendaño-Franco et al. [18] illustrates the research currently going on in the group of X. Gonze.

8 University of Mons

The laboratory for Chemistry of Novel Materials at the University of Mons was founded in 1988 by Jean-Luc Brédas. In the early days, the research activities mostly focused on the understanding of the structural and electronic properties of conducting polymers with the help of (correlated) ab initio and semi-empirical Hartree–Fock calculations and models such as the Valence Effective Hamiltonian. Another center of interest that rapidly grew up was the theoretical modeling of the nonlinear optical properties (i.e., hyperpolarizabilities) of π -conjugated molecules. Experimental activities based on scanning probe

microscopies to unravel the solid-state properties of organic materials were grafted on the core research of the laboratory with the arrival of Roberto Lazzaroni in 1990. Over the years, the field of organic electronics blossomed with the exploitation of undoped conjugated molecules and polymers in devices such as light-emitting diodes, solar cells, field-effect transistors, and sensors. Many theoretical studies were then performed to design the best materials for those applications and to understand all key electronic processes (such as energy and charge transport, charge injection, charge recombination, or exciton dissociation). These developments led to the progressive use of Density Functional Theory methods and force-field-based calculations in the research in Mons. J.L. Brédas crossed the Atlantic in 2000 to join first the University of Arizona, then his current position at the Georgia Institute of Technology in Atlanta.

Over the past few years, the theoretical activities have further diversified, with projects revolving around metal/organic interfaces, oxide/organic interfaces, hybrid biomaterials, polymer/nanotube composites, ionic liquids, graphene, and molecular electronics. These research activities are mostly carried out in the framework of national and European projects, allowing the laboratory to establish a wide network of collaborations in Belgium and at the European level. Among the European networks, of particular importance are the STEPLED project, for which the group was awarded the Descartes Prize of the European Commission in 2003, and the FP7 MINOTOR project, which centered on the modeling of interfaces for organic electronics and was coordinated by the University of Mons. A new extension of the research activities took place in 2008 with the opening of electronic device fabrication facilities at the Materia Nova R&D center in Mons; this gives the ability to study materials from the design and modeling to their incorporation in devices, often in joint projects with industrial partners.

Nowadays, the laboratory headed by R. Lazzaroni comprises around 30 researchers, including four permanent FNRS research fellows (David Beljonne, Jérôme Cornil, Philippe Leclère and Mathieu Surin). This laboratory is a founding member of the Interuniversity Scientific Computing Facility located in Namur, the Materia Nova Research Center in Mons, and the Institute for Materials Research recently established at the University of Mons. The article by Van Regemorter et al. in the present issue [19] illustrates the theoretical chemistry activities of the Laboratory for Chemistry of Novel Materials at the University of Mons.

9 University of Namur

In the *Florilège des Sciences en Belgique*, Louis d'Or writes that in 1971, a spreading occurs in the Quantum

Chemistry Laboratory of the Catholic University of Louvain-la-Neuve. Professor J.-M. André, surrounded by several researchers, starts a new laboratory at the Facultés Universitaires Notre-Dame de la Paix de Namur. In agreement with G. Leroy, it is in Namur that from then the research on the quantum chemistry of polymers will take place [20]. The *Laboratoire de Chimie Théorique Appliquée* (CTA) was developed with the help of Marie-Claude Roeland-André, Joseph Fripiat, and Joseph Delhalle. The first doctorate was delivered in 1975 to Simone Vercauteren-Delhalle. International cooperations were extended making profit of the contacts with Per-Olov Löwdin's group and Enrico Clementi's network (J.-M. André having been postdoc at IBM Research San Jose in 1968 and 1969). To improve visibility in the field of quantum chemistry of polymers, a series of NATO summer schools was organized with Janos Ladik: in 1974 (*Electronic Structure of Polymers and Molecular Crystals*), in 1977 (*Quantum Theory of Polymers*) in Namur, and in 1983 (*Quantum chemistry of polymers, solid-state aspects*) in Braunlage (Germany). In the late 1980s, these summer schools were then followed by the annual SCF (Scientific Computing Facility) meetings.

The research on quantum chemistry of polymers has dealt with conceptual aspects, that is, development of specific codes—Polymol, PLH, DJMol, solving difficult technical questions: long-range Coulomb and exchange contributions, band indexing, as well as specific applications to the interpretation of XPS spectra, the (semi)conductivity in conjugated polymers, studies in linear and nonlinear optical properties of polymers In this very quickly evolving period of quantum chemistry, the laboratory has been pioneering new ways of computing starting with the PDP 11/45, followed by the Digital DEC20 to the SCF initiative developed in cooperation between the FNRS, IBM, and FPS.

Several members of the laboratory have now academic or permanent research (FNRS) positions in Belgium or outside: Daniel Vercauteren at the University of Namur, J.-L. Brédas at the Georgia Technical Institute of Technology, M.S. Deleuze at the Hasselt University, Benoît Champagne and Eric Perpète at the University of Namur, and Denis Jacquemin at the University of Nantes. Two personalities issued from the CTA Lab have been awarded the Francqui Prize, the highest Belgian scientific award: Jean-Marie André in 1991 and Jean-Luc Brédas in 1997.

In 1991, the Administration Board of the University of Namur asked for the opening of a second laboratory specialized in theoretical chemistry with the principal aim to foster on the increasingly important aspects of molecular modeling that complemented the already well-established quantum mechanical approaches. A new laboratory, called "Laboratoire de Physico-Chimie Informatique" (PCI for

Computational Physico-Chemistry), was thus started by D. Vercauteren, former Ph.D. student with J.-M. André and PostDoctoral Fellow with E. Clementi at IBM Poughkeepsie in 1982–1983, with the help of Laurence Leherste, also former Ph.D. student with J.-M. André and PostDoctoral Fellow with Suzanne Fortier and Janice Glasgow in the School of Computing at Queen's University in 1992–1993.

Since then, the PCI Laboratory has developed its research activities in the domain of molecular engineering on computers. Over the years, the research work concerned the study of molecular conformations, similarities, interactions, and recognition in mixed environments (supramolecular systems, adsorbed phases, microporous materials, membranes, ...) by molecular modeling (graphics, molecular mechanics, hybrid QM/MM methods, coarse-graining, and multiscale approaches) and statistical mechanics (Monte Carlo, molecular dynamics, ...) methods, as well as by knowledge-based approaches (databases, logic and functional programming, fuzzy logic, expert systems, neural networks, genetic algorithms, hidden-Markov models, ...). Shortly, those analyses have been applied to the characterization and manipulation of "molecular images", like the electron density or the electrostatic potential at different levels of resolution, to zeolites, aluminophosphate frameworks, heterogeneous and homogeneous polymerization catalysts, cyclodextrins and their tubular complexes, proteins, drug-DNA, protein-DNA, protein-lipid domains. Researchers in the laboratory also tackled original aspects in computer-assisted organic chemistry and very recently in the development of reactive force-field approaches in the study of organocatalysis. Several members of the PCI Laboratory now occupy leading positions in academic or research institutions outside Belgium. Let us cite, Andy Becue at the University of Lausanne, Nathalie Meurice and Joachim Petit at Mayo Clinic in Scottsdale, and Thibaud Latour at the Tudor Research Institute in Luxembourg.

After 20 years as FNRS researcher, Benoît Champagne took over the position of J.-M. André when he retired in 2009. After defending his PhD Thesis in 1992 on the elaboration of *polymer band structure methods for evaluating the polarizabilities of polymers*, for which he received in 1994 the IBM Belgium Award of Computer Science, B. Champagne accomplished a postdoctoral stay at the Quantum Theory Project (Gainesville, Florida) with Yngve Öhrn and visited frequently Bernard Kirtman at the University of California in Santa Barbara. In 1995, he obtained a permanent position as Research Associate of the FNRS. In 2001, he presented his Habilitation thesis on the *development of methods for evaluating and interpreting vibrational hyperpolarizabilities*. In 2009, he founded the Laboratoire de Chimie Théorique (LCT). The LCT

develops an expertise in theoretical and quantum chemistry. Its research focuses on the elaboration and application of methods for predicting and interpreting properties responsible for optical and electrical effects in molecules, supramolecules, polymers, and molecular crystals. The main research axes are the development and application of quantum chemistry methods (i) to predict and interpret the linear and nonlinear optical properties of molecules, polymers, and supramolecular systems, (ii) to study the properties of open-shell systems (radicals, diradicals, multiradicals) and in particular the optical properties, (iii) to simulate and interpret vibrational spectra (VROA, SFG, hyper-Raman, resonant Raman, Raman, VCD, IETS), (iv) to calculate the linear and nonlinear optical properties of molecular crystals using methods combining *ab initio* calculations and electrostatic interactions, (v) to unravel the structural, reactive, optical, electronic, and magnetic properties of polymer chains, and (vi) to predict and understand the molecular properties associated with chirality.

Several of these investigations are carried out within an interdisciplinary environment where the theoretical work is intertwined with synthesis and experimental characterizations. Over the years, the group has fostered intensive collaborations with B. Kirtman (University of California in Santa Barbara), D.M. Bishop (†) (University in Ottawa), F. Castet (Institut des Sciences Moléculaires de l'Université de Bordeaux), and M. Nakano (Department of Materials Engineering Science of Osaka University). Moreover, the LCT carries on the tradition of participating to the development of high performance computing facilities, via the initiative of the CÉCI of the Fédération Wallonie Bruxelles, a distributed computer architecture for about 400 users, financed by the F.R.S.-FNRS and the Universities.

The theoretical chemistry research activities from the UNamur are illustrated in the contributions by Fripiat and Harris [21], Hubin et al. [22], Leherte and Vercauteren [23], as well as Liégeois and Champagne [24].

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References

1. Florilège des Sciences en Belgique, II, p 133 (1980). Available for free downloading at http://www2.academieroyale.be/academie/documents/FLORILEGE_VOL0214276.pdf
2. Geldof D, Krishtal A, Blockhuys F, Van Alsenoy C (2012) Quantum chemical study of self-doping PPV oligomers: spin distribution of the radical form. *Theor Chem Acc* 131:1243
3. Neyts EC, Bogaerts A (2013) Combining molecular dynamics with Monte Carlo simulations: implementations and applications. *Theor Chem Acc* 132:1320
4. Cauët E, Carette T, Lauzin C, Li JG, Loreau J, Delsaut M, Nazé C, Verdebout S, Vranckx S, Godefroid M, Liévin J, Vaeck N (2012) From atoms to biomolecules: a fruitful perspective. *Theor Chem Acc* 132:1254
5. Sutcliffe B (2012) Is there an exact potential energy surface? *Theor Chem Acc* 131:1215
6. De Vleeschouwer F, Geerlings P, De Proft P (2012) Radical electrophilicities in solvent. *Theor Chem Acc* 131:1245
7. Ghysels A, Vandichel M, Verstraeten T, van der Veen MA, De Vos DE, Waroquier M, Van Speybroeck V (2012) Host-guest and guest-guest interactions between xylene isomers confined in the MIL-47(V) pore system. *Theor Chem Acc* 131:1324
8. Cedillo A, Van Neck D, Bultinck P (2012) Self-consistent methods constrained to a fixed number of particles in a given fragment and its relation to the electronegativity equalization method. *Theor Chem Acc* 131:1227
9. Fogueri UR, Kozuch S, Karton A, Martin JML (2013) A simple DFT-based diagnostic for nondynamical correlation. *Theor Chem Acc* 132:1291
10. Hajgató B, Morini F, Deleuze MS (2012) Electron Momentum Spectroscopy of metal carbonyls: a reinvestigation of the role of nuclear dynamics. *Theor Chem Acc* 131:1244
11. Ceulemans A, Lijnen E, Fowler PW, Mallion RB, Pisanski T (2012) S5 graphs as model systems for icosahedral Jahn-Teller problems. *Theor Chem Acc* 131:1246
12. Phung QM, Vancoillie S, Delabie A, Pourtois G, Pierloot K (2012) Ruthenocene and cyclopentadienyl pyrrolyl ruthenium as precursors for ruthenium atomic layer deposition: a comparative study of dissociation enthalpies. *Theor Chem Acc* 131:1238
13. Truong BT, Nguyen MT, Nguyen MT (2012) The boron conundrum: the case of cationic clusters B + n with n = 2–20. *Theor Chem Acc* 131:1241
14. Dive G, Robiette R, Chenel A, Ndong M, Meier C, Desouter-Lecomte M (2012) Laser control in open quantum systems: preliminary analysis toward the Cope rearrangement control in methyl-cyclopentadienylcarboxylate dimer. *Theor Chem Acc* 131:1236
15. Ganesan R, Remacle F (2012) Stabilization of merocyanine by protonation, charge, and external electric fields and effects on the isomerization of spiropyran: a computational study. *Theor Chem Acc* 131:1255
16. Zanti G, Peeters D (2013) Electronic structure analysis of small gold clusters Aum ($m \leq 16$) by density functional theory. *Theor Chem Acc* 132:1300
17. Vergote T, Gathy T, Nahra F, Riant O, Peeters D, Leyssens T (2012) Mechanism of ketone hydrosilylation using NHC-Cu(I) catalysts: a computational study. *Theor Chem Acc* 131:1253
18. Avendaño-Franco G, Piraux B, Grüning M, Gonze X (2012) Time-dependent density functional theory study of charge transfer in collisions. *Theor Chem Acc* 131:1289
19. Van Regenmortel T, Guillaume M, Sini G, Sears JS, Geskin V, Brédas JL, Beljonne D, Cornil D (2012) Density functional theory for the description of charge-transfer processes at TTF/TCNQ interfaces. *Theor Chem Acc* 131:1273
20. Original French text: En 1971, un essaimage se produit dans le Laboratoire de Chimie quantique de l'Université catholique de Louvain-la-Neuve. Le professeur J.M. André, entouré de plusieurs chercheurs, fonde aux Facultés universitaires de Namur un nouveau laboratoire. En accord avec le professeur Leroy, c'est à Namur qu'auront lieu désormais les recherches sur la chimie quantique des polymères

21. Fripiat JG, Harris F (2012) Ewald-type formulas for Gaussian-basis studies of one-dimensionally periodic systems. *Theor Chem Acc* 131:1257
22. Hubin PO, Jacquemin D, Leherste L, André JM, van Duin ACT, Vercauteren DP (2012) Ab initio quantum chemical and ReaxFF-based study of the intramolecular iminium-enamine conversion in a proline-catalyzed reaction. *Theor Chem Acc* 131:1261
23. Leherste L, Vercauteren DP (2012) Smoothed Gaussian molecular fields: an evaluation of molecular alignment problems. *Theor Chem Acc* 131:1259
24. Liégeois V, Champagne B (2012) Implementation in the Pyvib2 program of the localized mode method and application to a helicene. *Theor Chem Acc* 131:1284