

# Annual Report 2022



UNIVERSITY  
OF OSLO



UiT The Arctic  
University of Norway



Norwegian  
Centre of  
Excellence

The Research Council of Norway



# Hylleraas Centre for Quantum Molecular Sciences

The Hylleraas Centre is a Norwegian Centre of Excellence (CoE) shared equally between the University of Oslo (UiO) and UiT The Arctic University of Norway (UiT), with UiO as project owner. It receives an annual funding of about 15 million NOK from the Research Council of Norway. In addition, it receives substantial financial support from UiO and UiT. The centre was established on October 1, 2017 for a period of six years, which in 2022 was extended by four years, to September 30, 2027.

The Hylleraas Centre is one of 23 national CoEs in Norway. The goal of the CoE program is to stimulate Norwegian research groups to establish larger units focusing on frontier research at a high international level and to raise the quality of Norwegian research.

The Hylleraas Centre aims to develop and apply computational methods to understand, interpret, and predict new chemistry, physics, and biology of molecules in complex and extreme environments. It has an extensive visitors' program for scientists from around the world, as well as for PhD candidates and postdocs from other research groups who wish to benefit from the expertise at the centre.

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# From the Directors

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**Having successfully coped with the pandemic, activities at the Hylleraas Centre were back to full capacity in 2022, with many new externally financed projects started. Our research plans and leadership team were revised during the year, in preparation for the second period of operation. Towards the end of year, the centre was prolonged by the Research Council of Norway to 2027, following a cancellation of the planned midterm evaluation in 2023.**

After two years defined by the COVID-19 pandemic, activities at the Hylleraas Centre in 2022 were almost back to normal, with minimal external restrictions on our scientific and social undertakings. Having surpassed its initial five-year milestone on October 1, 2022, the centre was once again operating at full capacity. Our productivity is on an upward trajectory following a decline in 2021, and numerous new projects have either recently commenced or are set to begin soon, revitalizing the centre.

A reliable gauge of our activities is the turnout at the Annual Hylleraas Meeting. Following a historic low attendance of only 35 participants at Sundvollen in August 2020 amid the pandemic, the numbers rebounded significantly, with a robust attendance of 53 at Hamn in 2021 and a record-breaking high of 63 at Klækken in 2022. The organization of the Klækken meeting was spearheaded by Mahika Luthra and Samiran Sen, the Oslo representatives of the Young Researcher Parliament (YRP). Our sincere thanks go to them for ensuring the success of the Klækken meeting.

Several other noteworthy events took place at the centre in 2022, including the Hylleraas Hackathon at Skibotn, organized by Dr. Magnus Ringholm, a Norway–Japan symposium coordinated by Michiko Atsumi and Erik Tellgren, two Young CAS workshops orchestrated by Erik Tellgren, and a Global Women's Breakfast locally organized by Ainara Nova. We extend our gratitude to all the organizers for their dedicated efforts in making these events a success.

Outreach activities fully resumed in 2022. A notable event was *Hvorfor Det?*, a poster competition for children aged 8–12 from Ila skole in Oslo. This initiative was organized by the school's parent board and held at the Department of Chemistry in Oslo. Our appreciation goes out to all members of the centre who contributed to this event, with special recognition for Thomas Bondo Pedersen and his wife Anne-Dorothee Pedersen, who hosted the event on behalf of the Hylleraas Centre. We also extend our thanks to Audun Skau Hansen, André Laestadius, and Erik Tellgren, who took the lead in several outreach events throughout 2022.

In January 2022, Luca Frediani assumed the role of deputy director, succeeding Kenneth Ruud. Additionally, Dr. Michal Repisky took over as principal investigator from Ruud. Following an internal process overseen by the Hylleraas Board, Thomas Bondo Pedersen was officially confirmed in October 2022 as the new director of the Hylleraas Centre, effective from July 1, 2023, succeeding Trygve Helgaker. We are confident that Thomas will effectively lead the centre forward and extend our congratulations on his appointment.

In 2022, we initiated preparations for the midterm evaluation of the Hylleraas Centre, scheduled for 2023. As part of these preparations, we undertook a comprehensive review of our research plans and the team of principal investigators for the upcoming second centre period spanning 2023–2027. This process took into account the achievements of the centre during its first period, global developments in the field, and changes in personnel at the centre.

The revised plans were positively received by our Scientific Advisory Committee (SAC), who conducted a thorough four-day visit to the Hylleraas Centre in September 2022. We extend our gratitude to the SAC for their detailed report, offering valuable feedback on centre activities and providing insightful advice for the future.

Towards the end of 2022, the Research Council of Norway made the decision to cancel the midterm evaluation,

– We are pleased and enthusiastic that the Hylleraas Centre will continue its work for another five years, with revised research plans and a new leadership team taking over in July 2023.

resulting in the prolongation of all Centres of Excellence established in 2017 through 2027. We are pleased and enthusiastic that the Hylleraas Centre will continue its work for another five years, with revised research plans and a new leadership team taking over in July 2023.

Following the many research grants secured by the centre in previous years, the year 2022 was meagre in terms of new external projects but we are delighted to congratulate Dr. Marco Bortoli on winning a Marie Skłodowska-Curie Action Individual Fellowship to work with Michele Cascella.

Additionally, the commencement of several external projects in 2022 introduced many new faces to the centre, and we welcome them warmly as they play a vital role in shaping the centre's future. We are also looking forward to working with our new communication officer Cathrine Strøm, who began her work with us in March 2022, and extend our appreciation to Elina Melteig, who left the centre to take up a position as journalist and communication officer at the Faculty of Mathematics and Natural Sciences, University of Oslo, in 2021.

We congratulate Dr. André Laestadius for his achievement in securing an ERC Starting Grant in 2021 and for his subsequent appointment as an associate professor at Oslo Metropolitan University (OsloMet) in 2022. We wish him the best for the future and are pleased that he will keep a 20% position at the

centre for the next three years. Our congratulations and best wishes are also extended to the four young centre members who defended their PhD theses in 2022: Anders Brakestad, Håkon Emil Kristiansen, Morten Ledum, and Karolina Di Remigio Eikås.

In 2022, Kajsa Ryttberg-Wallgren made the decision to step down as a member of the Hylleraas Board. We express our appreciation for her contributions to the Board and extend our sincere thanks. We are pleased to welcome Prof. Aslak Tveito, who will replace her from

2023. Tveito served as the managing director of Simula Research Laboratory from 2002 to 2022 and was a member of the Board of the Centre for Theoretical and Computational Chemistry (CTCC) from 2007 to 2010. His extensive experience brings valuable insights to the Hylleraas Board as the centre embarks on its second period of operation.

Finally, we would like to thank all members and affiliates of the Hylleraas Centre for their good work in 2022, as detailed in this report.



*Trygve Helgaker*  
**Prof. Trygve Helgaker**  
*Director*



*Luca Frediani*  
**Prof. Luca Frediani**  
*Deputy Director*



# From the Board of Directors

**Apart from science and research, the primary tasks of the Hylleraas Centre at the beginning of 2022 were to revise its research plans for the second centre period, with an eye toward the midterm evaluation, and to identify new leadership and principal investigators for the second centre period. All tasks have been completed to the full satisfaction of the Board of Directors.**

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On January 1st, 2022, Prof. Kenneth Ruud assumed the role of Director General at the Norwegian Defence Research Establishment (FFI), while Prof. Luca Frediani stepped in as the Deputy Director of the Hylleraas Centre. With Ruud's departure from the Hylleraas Centre, the search for a new director to succeed Prof. Trygve Helgaker, set to retire in 2023, began. Working closely with the director, the Board of Directors selected Prof. Thomas Bondo Pedersen, who will officially assume the directorship on July 1st, 2023. We are confident that the centre will continue to thrive under the new leadership of Pedersen as director and Frediani as deputy director. Additionally, we express our gratitude to Helgaker, who extended his directorship until July 1st, 2023, to facilitate a smooth transition and prepare for the scheduled midterm evaluation in 2023.

We are pleased that the Hylleraas Centre approached the important task of preparing new research plans and reorganizing its research themes in 2022 with great dedication, commencing its efforts early in the year. A significant milestone in this process was the internal review conducted by the Scientific Advisory Committee (SAC) in September

2022. Their report not only affirmed the centre's excellence in science and research but also provided valuable feedback and comments for the development of new research plans.

The newly formulated plans build upon the foundation of the old plans in crucial ways, while also introducing new research lines such as machine learning and the Hylleraas Software Platform. These additions are aimed at enhancing synergies within the centre. We are particularly pleased that many young researchers have assumed the role of principal investigators for the second centre period, attesting to the centre's success in promoting and supporting the development of young talent. The achievement of Dr. André Laestadius, securing an associate professorship at OsloMET after winning an ERC Starting Grant at the Hylleraas Centre, also stands as a testament to this success.

The unexpected cancellation of the midterm evaluation by the Research Council of Norway in December 2022 resulted in the centre defaulting to a full ten-year-period and that new research plans could be implemented with less delay. On the other hand, the ongoing reorganization of the Research Council

We are particularly pleased that many young researchers have assumed the role of principal investigators for the second centre period, attesting to the centre's success in promoting and supporting the development of young talent.

that led to the cancellation of the mid-term evaluation also introduces grave uncertainties regarding future research funding in Norway, particularly affecting fundamental research. Fortunately, there is no immediate threat to the Hylleraas Centre – a large number of externally funded projects began in 2021 and 2022, ensuring that the research at the centre will remain at full throttle for the next few years.

A parallel external uncertainty surrounds the national computing infrastructure, with Sigma2 negotiating funding arrangements with the BOTT universities (i. e., University of Bergen, University of Oslo, UiT The Arctic University, and Norwegian University of Science and Technology). The Board underscores the importance of robust computing resources for the Hylleraas Centre and urges a solution that ensures afford-

bility and accessibility, recognizing the positive impact of computation on research and innovation throughout Norway.

Overall, the Board of Directors is very pleased with the developments of the Hylleraas Centre in 2022 and looks forward to guiding and supporting the centre in its second operational period.



**Unni Olsbye (chair)**  
*Professor  
Dept. Chemistry  
UiO*



**Annette Bayer**  
*Professor  
Dept. Chemistry  
UiT*



**Atle Jensen**  
*Professor  
Dept. Mathematics  
UiO*



**Nathalie Reuter**  
*Professor  
Dept. Bio. Sciences  
UiB*



**Kajsa Rytberg-Wallgren**  
*Vice President  
Vacuum Conveying Division  
Piab Group, Sweden*



# From the Young Researchers

**The Young Researcher Parliament (YRP) is a platform for supporting young researchers in their career development. Two YRP student representatives elected from each node are members of the Management Team of the Hylleraas Centre, bringing forward their insights and perspectives. Through the YRP visitor program, young researchers have the opportunity to invite scientists and former members of the centre to give seminars focused on their career paths.**

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The YRP visitor program, which was conceptualized in 2021 and implemented in 2022, aims at the organization of talks and workshops by and for the young researchers. Lucas Lang, a post-doctoral fellow at the centre, invited and organized with the YRP in Oslo a two-day meeting (October 13–14) with Prof. Pascal Friederich, from the Karlsruhe Institute of Technology (KIT, Germany). During his visit, Prof. Friederich had informal discussions with the young researchers, sharing insights on navigating career paths. Additionally, he held a scientific seminar about recent developments of self-explaining graph neural networks. Gabriel Gerez, PhD candidate and representative for YRP at UiT, arranged for a visit to the high-performance computing cluster Fram at UiT, to see the equipment we all use day-to-day.

In 2022, the YRP student representatives of the Oslo node, Samiran Sen and Mahika Luthra, organized the Annual Hylleraas Meeting. Held over three days at Klækken (September 5–7), the meet-

ing consisted of several talks from internal and external speakers focusing on different crucial aspects of modern-day scientific practices, raising awareness among early career researchers about some of the promising avenues they can take, should they choose to further their roles in science. A round of 5-Minute Thesis Presentations by the master's and PhD students was organized as in previous years. The YRP annual assembly, where both nodes of the YRP met to discuss ways to improve representation and work towards its vision, was also held at the Klækken meeting. Different developments at the centre, both recent and upcoming, were presented and debated to keep all its members au courant and involved.

Beyond the ambit of science, the YRP arranged monthly pizza meetings in Oslo and social events in Tromsø, providing an informal platform for the members to discuss issues and ideas for betterment of the Hylleraas Centre. Matters related to career development are often brought up in these meetings and events.



YRP session during the Hylleraas Annual Meeting at Klækken Hotel in September 2022: young researchers brainstorming ideas for collaborations between the Oslo and Tromsø nodes.

The Scientific Advisory Committee (SAC) visited the Hylleraas Centre this year. They met with the YRP both in Tromsø (September 13) and Oslo (September 14) over lunch, discussing its engagement and contributions to the administration of the centre and to the social life and welfare of its members.

In Oslo, YRP representative Samiran Sen organized an exposition of various posters that its members presented in different recent conferences and schools, which were framed and put on display along the corridors of the Hylleraas Centre.

Later in the year, new representatives of both nodes were elected. In Oslo, Samiran Sen and Mahika Luthra stepped down and their places were taken by Marinella de'Giovannetti and Hannes Kneiding. In Tromsø, Ryan Wilkins and Bente Barge chose to step down and Gabriel Gerez and Tonje Haugen were elected as the new representatives. We thank the previous representatives for their contributions to the YRP and to the Hylleraas Centre as a whole.



**Marinella  
de'Giovannetti**



**Gabriel  
Gerez**



**Tonje Reinholdt  
Haugen**



**Hannes  
Kneiding**

Elected YRP representatives 2022

# 2022

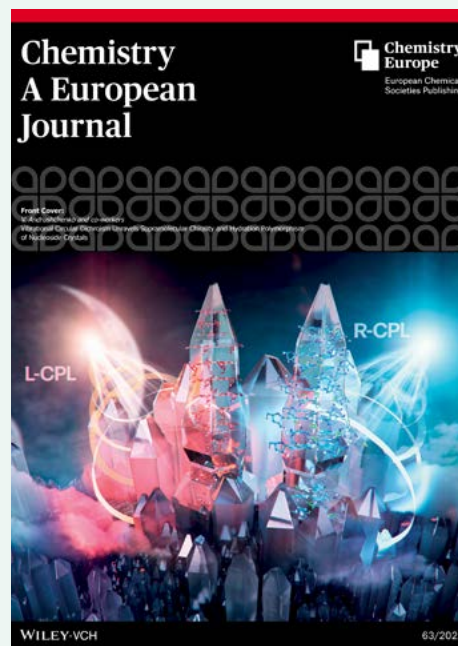
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## in Brief

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**Figure 1:** Cover of *Physical Chemistry Chemical Physics* issue 47, promoting the perspective article *DFT exchange: sharing perspectives on the workhorse of quantum chemistry and materials science*, *Physical Chemistry Chemical Physics* 2022, **24**, 28700–28781. Reproduced with permission from the Royal Society of Chemistry.



**Figure 2:** “Crystal City in a chiral world”, cover of *Chemistry – A European Journal*, issue 63, promoting the paper *Vibrational circular dichroism unravels supramolecular chirality and hydration polymorphism of nucleoside crystals*, *Chem. Eur. J.*, 2022, **28**. Reproduced with permission from John Wiley and Sons.

## Publications

Members of the Hylleraas Centre published 49 articles in 2022, bringing our total scientific production to nearly 300 articles. Among these articles, 18 were published in journals with an impact factor five or greater and four in journals with an impact factor greater than ten: three articles in *ACS Catalysis* and one in the *Journal of American Chemical Society*. At the end of 2022, the articles published by the Hylleraas Centre since its opening in 2017 had been cited about 2700 times (excluding self-citations), with an H index of 28. About one half of these articles are devoted to theoretical and methodological developments, the other half being computational studies, several carried out in collaboration with experimentalists.

Together with Andreas Savin of UPMC Sorbonne Université in Paris, Trygve Helgaker and Andrew Teale were the initiators and corresponding authors of the perspective article *DFT exchange: sharing perspectives on the workhorse of quantum chemistry and materials science*, published as a HOT Article in *Physical Chemistry Chemical Physics (PCCP)* in August 2022 (Figure 1). The format of this perspective article is that of a round-table discussion, in which the 70 authors (including Hylleraas members Trygve Helgaker, Simen Kvaal, André Laestadius, Andrew Teale, and Erik Tellgren) exchange views on the past, present, and future of density-functional theory (DFT) in the form of 302 individual contributions, formulated as responses to 26 set questions over 82 pages. With

a bibliography of 777 references, the article gives a lively snapshot of DFT anno 2022. According to the PCCP Blog *Celebrating our Perspective ‘DFT exchange: sharing perspectives on the workhorse of quantum chemistry and materials science’*, published on December 7, 2022, the perspective article, with “its unique round-table discussion format” is “the first of its kind in PCCP”.

In 2022, Dr. Monika Krupová published in *Chemistry – A European Journal* the cover paper *Vibrational circular dichroism unravels supramolecular chirality and hydration polymorphism of nucleoside crystals* with her former colleagues at the Institute of Organic Chemistry and Biochemistry (IOCB) of the Czech Academy of Sciences in Prague (Figure 2).



**Figure 3:** Participants of the Hylleraas Hackathon 2022.



**Figure 4:** Participants of the Hylleraas Annual Meeting 2022.



**Figure 5:** Michiko Atsumi closing the Norway–Japan symposium at the Hylleraas Centre in Oslo on March 16, 2022. Photo: Trygve Helgaker.



## Meetings and conferences

With the worst of the pandemic behind us, more in-person meetings and conferences were held in 2022 than in the two preceding years. The Hylleraas Hackathon was held at Skibotn near Tromsø on April 25–29, organized by Dr. Magnus Ringholm, with about 25 participants (Figure 3). The Annual Hylleraas Meeting took place at Klækken near Oslo on September 5–7 (Figure 4). It was organized by the YRP representatives Samiran Sen and Mahika Luthra and attracted a record number of 63 participants.

On February 16, many members of the Hylleraas Centre participated in the *IUPAC 2022 Global Women's Breakfast: Empowering Diversity in Science*, organized by Ainara Nova as part of the NordCO<sub>2</sub>

and CO<sub>2</sub>PERATE projects. A month later, on March 15–16, the *4th Norway–Japan Symposium on Theoretical and Experimental Chemistry of Complex Systems* was organized by Dr. Michiko Atsumi, Dr. Erik Tellgren, and Trygve Helgaker, with 70 participants (Figure 5). Both events were held in a hybrid fashion, from the Hylleraas Centre in Oslo.

The annual National Meeting of the Division of Quantum Chemistry and Modelling of the Norwegian Chemical Society took place at Zander K Hotel in Bergen on November 28–29 2022, organized by Dr. Marco Foscato, University of Bergen. The Hylleraas Centre sponsored the event, which was attended by many of its members.

As part of the Young Cas project *Global Optimization in Electronic Energy Landscapes: Finding a Needle in a Haystack* at the Centre for Advanced Study (CAS), Dr. Erik Tellgren organized two workshops in 2022, held at the Hylleraas Centre on April 11–14 and at the Norwegian Academy of Science and Letters on June 20–23. The first workshop of this project was held on September 13–17 2021.

Professor Clémence Corminboeuf from EPFL, Switzerland gave the 2022 Almlöf–Gropen Lecture, *Going further by leveraging data for solving quantum/computational chemistry problems* on June 1 in Oslo and June 2 in Tromsø. At both events, the Almlöf–Gropen Young Speakers were Sahil Gahlawat from Tromsø and Samiran Sen from Oslo.



**Figure 6:** Trygve Helgaker delivering his plenary lecture *Molecular bonding, structure, and dynamics in a strong magnetic field* at WATOC 2022 on July 5, 2022. Photo: Chemical Institute of Canada.



**Figure 7:** Laurens Peters giving a Friday Seminar at the Hylleraas Centre in Oslo on November 25, 2022. Photo: Trygve Helgaker.

## Dissemination

The members of the Hylleraas Centre gave a total of 60 oral presentations in 2022. Of these, 14 presentations were given by young members as Friday seminars at the centre, while 33 presentations were given by Hylleraas members at international meetings and conferences. Thomas Bondo Pedersen and Kenneth Ruud gave plenary lectures at the *Molecular Quantum Mechanics Conference (MQM 2022)* at Virginia Tech. Trygve Helgaker gave a plenary lecture at the *12th Triennial Congress of the World Association of Theoretical and Computational Chemists (WATOC 2022)* in Vancouver (Figure 6), while Luca Frediani and Trygve Helgaker gave plenary talks at the *19th International Conference on Density Functional Theory and its Applications (DFT 2022)* in Brussels.

A total of 23 Friday seminars (Figure 7) were given at the Hylleraas Centre in 2022, of which nine were given by guests to the centre.

At the CECAM school *Multiscale Molecular Dynamics in MiMic* held in Lausanne, Switzerland July 18–22 2022, PhD candidate Marinella de'Giovanetti (UiO) was awarded a poster prize for her poster *Towards an understanding of Turbo-Grignard reagents: structural information from AIMD studies*. Dr. Lukas Konecny (UiT) won a prize for his poster *Relativistic TDDFT allows prediction and interpretation of XAS spectra near heavy metal L- and M-edges*, at the *9th Workshop on Time-Dependent Density-Functional Theory* at the Centro de Ciencias de Benasque Pedro Pascual, October 18–28 2022.



**Figure 8:** Summer students at the Hylleraas Centre. From left to right: Audun Skau Hansen (Hylleraas Centre), Ayla Coder, Elias Dalan, Hanan Gharayba, and Andreas Alstad. Photo by Andreas Haraldsrud.

## Outreach

On Saturday November 26 2022, the Hylleraas Centre hosted *Hvorfor Det?*, a science poster competition for school children organized by the parents' board of Ila Skole, a primary school in Oslo. About 60 children aged 8–12 participated together with their parents and grandparents.

The Hylleraas Centre was visited by Talentsenteret i realfag (part of Oslo vitensenter at the Norwegian Museum of Science and Technology) on November 9, 2022. The visiting students carried out exercises related to quantum chemistry and attended talks by Trygve Helgaker, André Laestadius, and Audun Skau Hansen.

During the summer 2022, four chemistry students spent several days at the Hylleraas Centre, supported by the Centre for Computing in Science Education (CCSE) at the Faculty of Mathematics and Sciences, University of Oslo.

The four students were given programming tasks related to chemistry and chemistry education, supervised by Audun Skau Hansen (Figure 8). Two of the students gave a presentation of their work at the CCSE Christmas Seminar 2022.

As part of the UNGFORSK event at the University of Oslo on September 28–29 2022, groups of high-school students attended the workshop *A Journey from the Large to the Small*, organized by Audun Skau Hansen, Hanan Gharayba, and Elise Mohr Skogan at the Hylleraas Centre. The students carried out interactive simulations of gases, fluids, and molecules all the way down to the quantum level.

In 2022, Audun Skau Hansen wrote a popular science article *Shuffleboard og kjemi* for *Kjemi*, the journal of the Norwegian Chemical Society. He also published a blog post *Kan man ta på en*

*orbital?* on the web page of the Department of Chemistry, University of Oslo.

On October 25, Erik Tellgren together with three other panellists from academic research and the medical industry participated in a panel discussion at Ullern Senior High School in Oslo. The audience consisted of students from Forskerlinjen, an education program oriented towards scientific research. The panellists presented their own research and thoughts about career paths and answered questions from the students. On December 12, the panellists visited the school again to continue the discussions in the more informal setting of a Christmas dinner.

Throughout 2022, the Hylleraas Centre ran Hylleraas Math Help for students at the Department of Chemistry in Oslo, helping with mathematics at all levels, led by Dr. Erik Tellgren.

## Training and career support

Four members of the Hylleraas Centre defended their PhD thesis in 2022: Anders Brakestad (UiT), Håkon Emil Kristiansen (UiO), Morten Ledum (UiO), and Karolina Di Remigio Eikås (UiT). At the end of the year, a total of 18 PhD candidates had completed their degrees at the Hylleraas Centre.

David Balcells organized the NordCO<sub>2</sub> Summer School *Computational Molecular Design* at the University of Bergen on May 9–11 2022.

In 2022, Simone Meloni (University of Ferrara), Hylleraas adjunct Jógvan M. H. Olsen, and Ursula Röthlisberger (EPFL) organized *Multiscale Molecular Dynamics with MiMiC*, a flagship school of the Centre Européen de Calcul Atomique et Moléculaire (CECAM). The school was held at the CECAM headquarters in Lausanne on July 18–22 with 55 participants, of whom 23 attended online.

Five present and three previous members of the Hylleraas Centre participated at the 18<sup>th</sup> *European Summer School of Quantum Chemistry (ESQC)* in Sicily, September 11–24 2022 – one as lecturer, two as tutors, and five as students.

## Young Researcher Parliament

The Young Researcher Parliament (YRP) hosted a series of events tailored to the younger members of the Hylleraas Centre in 2022. They invited Pascal Friedrich, KIT (Karlsruhe, Germany) to Oslo on October 13–14. During his visit, Friedrich delivered a talk on *Machine learning for accelerated materials discovery* and engaged in a discussion on academic careers with young researchers at the centre. Additionally, the YRP continued its Alumni Talks series, featuring contributions from Dr. Roberto Di Remigio Eikås and Dr. Arne Bunkan on December 9.

The Hylleraas Annual Meeting in September 2022 was organized by the YRP representatives, Samiran Sen and Mahika Luthra, at Klækken Hotel.

In 2022, Gabriel Gerez and Tonje Reinholdt Haugen assumed the roles of YRP representatives succeeding Ryan Wilkins and Bente Barge in Tromsø, while Marinella de'Giovanetti and Hannes Kneiding replaced Samiran Sen and Mahika Luthra in Oslo.

Since 2019, the young researchers at the Hylleraas Centre have organized all our Annual Meetings: Bardufoss 2019, Sundvolden 2020, Hamn 2021, and Klækken 2022.

## Management

The Management Team met 11 times in 2022 (all months except July), while the Board of Directors met four times (January 7, March 25, June 1, and October 14). The Scientific Advisory Committee visited the centre in Tromsø and Oslo September 12–15, 2022.

## Visitors

In 2022, a total of 23 researchers visited the Hylleraas Centre, 15 in Oslo and 8 in Tromsø. This count includes the Almlöf-Gropen speaker but excludes inter-node visits and visits by affiliate members.



**Figure 9:** Ceremony in honour of grant winners at the Faculty of Mathematics and Natural Sciences in Oslo on April 7, 2022. To the left: all grant winners present at the ceremony, with Abril Castro and André Laestadius in the first row; to the right: Abril Castro with Rector of the University of Oslo Svein Stølen and Hylleraas Director Trygve Helgaker.

## External projects and funding

Dr. Marco Bortoli, a postdoctoral researcher from the University of Girona, Spain, was awarded a Marie Skłodowska-Curie Action (MSCA) Individual Fellowship for his project *Rationally-Designed Turbo Reagents for Innovative Organometallics (RATIO)*. He will be conducting his research at the Hylleraas Centre for a two-year period, starting in 2023.

Apart from the MSCA project of Bortoli, the Hylleraas Centre did not secure any new external projects in 2022, marking a notable departure from the trend observed in recent years. Since 2017, a total of 27 research proposals from the centre have received funding from external agencies: seven in 2017, two in 2018, three in 2019, six in 2020, and seven again in 2021.

Annually, the Faculty of Mathematics and Natural Sciences in Oslo hosts a ceremony to celebrate recipients of Young Research Talents (YRT) grants from the Research Council of Norway and grants from the European Research Council (ERC). In 2022, the honourees included YRT grant winner Abril Castro and ERC Starting Grant winner André Laestadius (Figure 9).

Since 2017, the members of the Hylleraas Centre have secured a total of 27 research grants from national and international funding agencies, greatly boosting research at the centre.



**Figure 10:** In 2022, Odile Eisenstein was inducted to the American Academy of Arts & Sciences (left) and to the US National Academy of Sciences (right).

## Personnel

Kenneth Ruud became Director General of the Norwegian Defence Research Establishment (FFI) on January 1, 2022, but remains in a 20% position at the centre; he was replaced by Luca Frediani as Deputy Director and by Dr. Michal Repisky as Principal Investigator of RT3. Dr. André Laestadius was appointed associate professor at Oslo Metropolitan University (OsloMet) on September 1, but continues in a 20% adjunct position at the Hylleraas Centre.

Odile Eisenstein was inducted into the American Academy of Arts & Sciences and the National Academy of Sciences in 2022, following her election into these academies in 2021 (Figure 10).

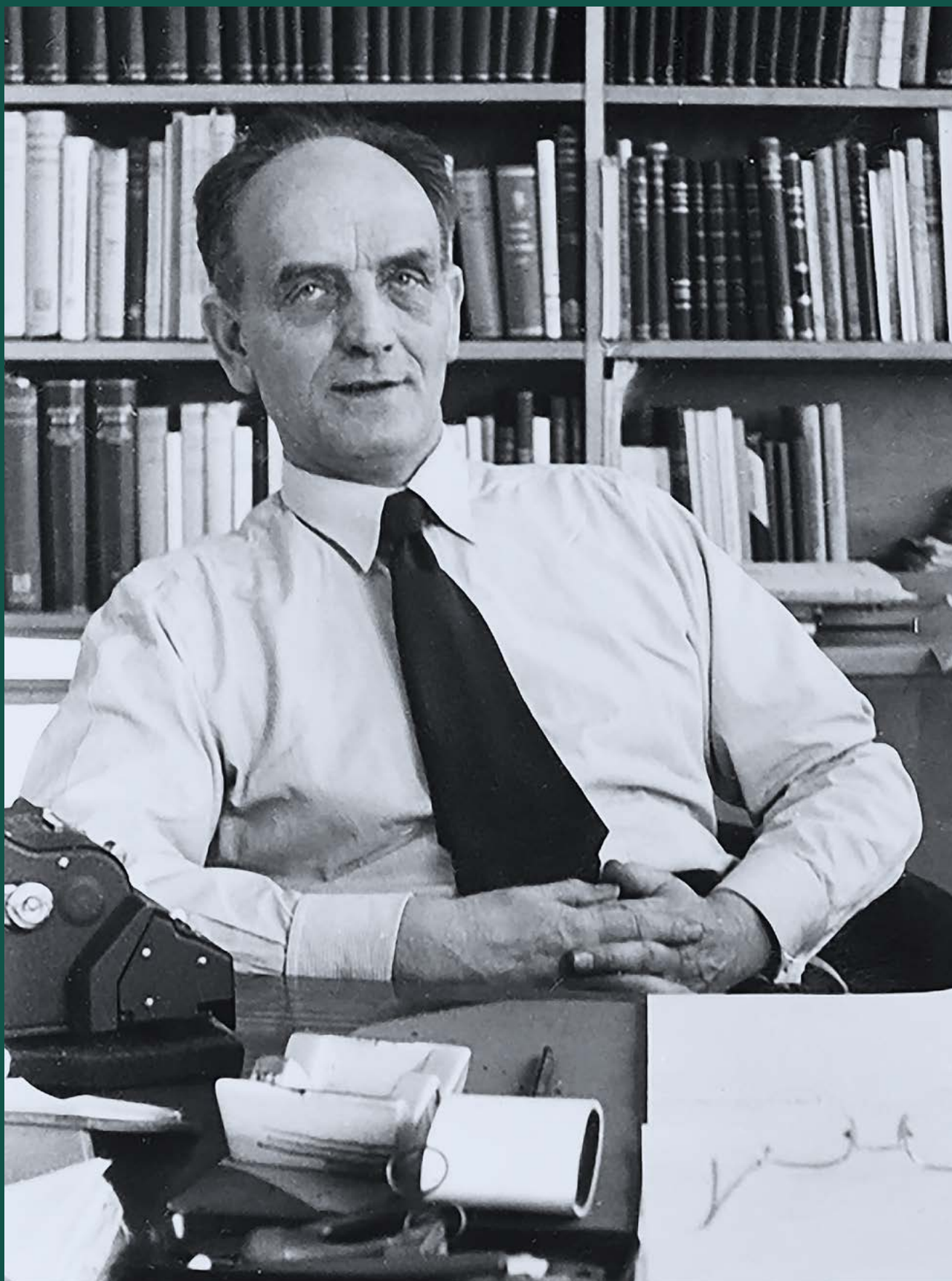
At a ceremony at Grand Hotel in Oslo on March 28, Trygve Helgaker, Jan Ingar Johnsen and Simen Reine were



appointed Oslo Ambassadors by Oslo Convention Bureau, for attracting the 13th WATOC congress to the city. The Oslo Conference Ambassador network "consists of key figures within various academic and research communities in Oslo, who want to highlight their field of research internationally by hosting a conference". The diplomas were awarded by the Mayor of Oslo, Marianne Borgen (Figure 11).



**Figure 11:** Oslo Ambassador Ceremony at Hotel Bristol in Oslo on March 28. From left to right: Simen Reine, Trygve Helgaker, Jan Ingar Johnsen, and Mayor of Oslo Marianne Borgen.



# Egil A. Hylleraas

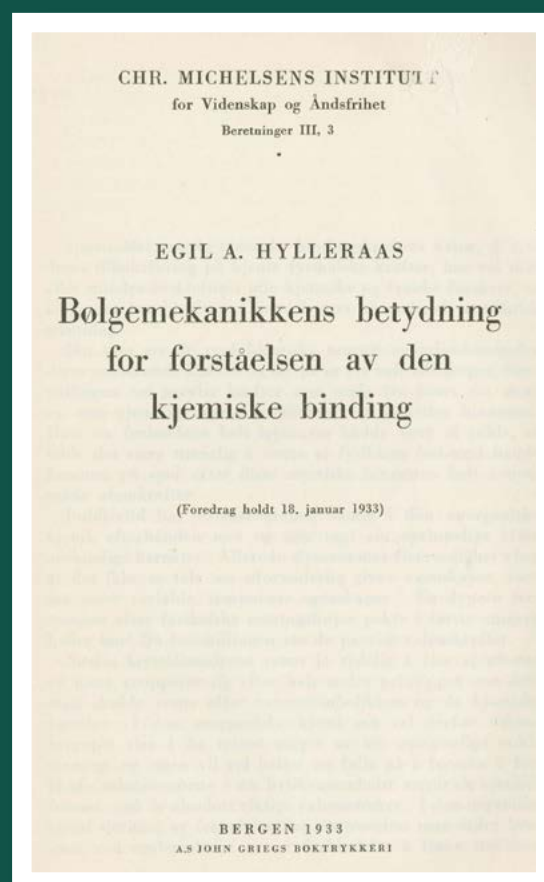
The Norwegian physicist Egil A. Hylleraas (1898–1965) helped usher in the era of scientific computing by carrying out accurate calculations on helium, thereby confirming the validity of quantum mechanics for more than one particle (1929), by predicting the stability of the hydrogen anion, later detected in the Sun's atmosphere (1930), and by performing the first calculation of the cohesive energy of a molecular crystal, LiH (1930). In 1933, he introduced the term “kvantekjemi” into the Norwegian language:

*“Idet jeg avslutter den utredning håper jeg at tilhørerne vil ha fått et inntrykk av at der nu foreligger ganske vidtrekkende muligheter for å bygge op en teoretisk kjemi, en kvantekjemi, på samme grunnlag som den fysikalske kvanteteori.”*

*English translation:*

*“In closing my presentation, I hope to have convinced the audience of the far-reaching opportunities that now exist for establishing a theoretical chemistry, a quantum chemistry, on the same footing as the physical quantum theory.”*

*“Importance of wave mechanics for understanding the chemical bond.”*  
(1933)





# Research Highlight



The Hylleraas Centre carries out research on a broad range of topics in theoretical chemistry, with relevance not only to chemistry but also to physics, biology, and other related fields of science. In this section of the Annual Report, we highlight recent and ongoing work at the centre, including work in fundamental theoretical chemistry, the development of new computational methods and techniques, and the use of computational chemistry to address challenging questions in modern science, often in collaboration with experimentalists. For 2022, we present recent research carried out in Research Theme 4, Extreme Environments.



## Molecular Dynamics in an Ultrastrong Magnetic Field

Tanner Culpitt, Trygve Helgaker,  
Laurens Peters and Erik Tellgren

Within the Born–Oppenheimer approximation, it is often useful to regard the atoms that make up molecules as classical particles whose motion is determined by the forces acting on the nuclei. By calculating these forces quantum-mechanically and solving the classical equations of motion for the nuclei, we may determine the motion of molecules and in this way study molecular reactivity, rovibrational spectra, and so on. The quality of such studies depends critically on the accuracy of the forces used for the dynamics. Many techniques are available for their calculation, ranging from moderately accurate but inexpensive empirical methods to accurate but expensive *ab initio* methods. Such direct *ab initio* molecular dynamics (AIMD) studies were pioneered by Helgaker and Uggerud in the early 1990s, when they calculated the translational energy release upon dissociation of molecular cations using multiconfigurational self-consistent-field (MCSCF) wave functions [1,2]. For a recent review of AIMD, see Ref. [3].

An intriguing question that has not yet been addressed concerns the effect of a magnetic field on molecular dynamics – how does a magnetic field affect the motion of molecules? We are in particular interested in fields that are sufficiently strong to change the electronic structure of the molecules. At field strengths of the order of one atomic unit,  $B_0 = 235$  kT, the magnetic interactions with the electrons are as strong as the Coulombic interactions. Such field strengths cannot be generated on Earth but exist in the atmospheres of magnetic white dwarf stars. Over the last decade, we have studied chemistry under such conditions extensively and discovered new phenomena such as paramagnetic bonding [4], a bonding mechanism fundamentally different from covalent and ionic bonding, giving rise to molecules that do not exist on Earth, such as molecules consisting of strongly bound noble gas atoms [5,6]. The study of molecules in an ultrastrong magnetic field is not only fascinating in itself but needed, for example, to

calculate spectra of molecules on magnetic white dwarfs and in this way help their experimental detection.

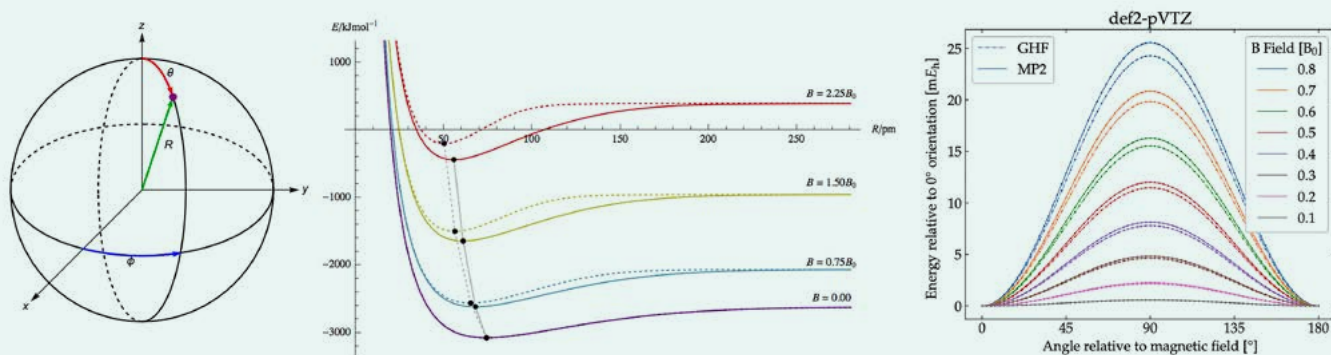
The inclusion of a magnetic field requires fundamental changes to the existing AIMD schemes. In the absence of a magnetic field, the force on a given nucleus  $I$  is simply the negative derivative of the Born–Oppenheimer potential-energy surface  $U(\mathbf{R})$  with respect to the coordinates  $\mathbf{R}_I$  of that nucleus,

$$\mathbf{F}_I(\mathbf{R}) = -\nabla_I U(\mathbf{R}),$$

and may be calculated using various well-established *ab initio* gradient techniques [7]. In a magnetic field  $\mathbf{B}$ , by contrast, the force on nucleus  $I$  depends not only on the positions of the nuclei  $\mathbf{R}$  but also on their velocities  $\mathbf{v}$  [8–12]:

$$\mathbf{F}_I(\mathbf{R}, \mathbf{B}, \mathbf{v}) = -\nabla_I U(\mathbf{R}, \mathbf{B}) - Z_I \mathbf{e} \mathbf{B} \times \mathbf{v}_I + \sum_j \Omega_{IJ}(\mathbf{R}, \mathbf{B}) \mathbf{v}_j.$$

The first term is the Born–Oppenheimer gradient force – it is the same as in the field-free case except that it is calculated from a potential-energy surface  $U(\mathbf{R}, \mathbf{B})$  that depends on the field strength.



⬆ **Figure 1:** Left: rovibrational coordinates of  $\text{H}_2$  with the field vector  $\mathbf{B}$  along the  $z$  axis; middle: bond-dissociation curves of  $\text{H}_2$  at different field strengths in parallel (full lines) and perpendicular (dashed lines) field orientations [4]; right: potential energy of  $\text{H}_2$  as a function of the polar angle  $\theta$  at different field strengths, calculated at the equilibrium bond distance for each field strength [15]. Figure in the middle from Ref. [4], reprinted with permission from AAAS. Figure to the right reproduced from Ref. [15] under CC-BY 4.0 license.

The remaining two terms represent the Lorentz force on nucleus  $I$ , with both a nuclear contribution and an electronic contribution. The nuclear contribution is trivial – it is the *bare Lorentz force* that the nucleus would experience in the field in the absence of electrons. The electronic contribution, known as the *Berry force*, is more complicated. To understand how it arises, we may think of the electrons as screening the nuclei from the externally applied field, generating at the position of each nucleus a local field that differs from the externally applied field. The Berry force provides this screening by the electrons and must be included for the dynamics to be meaningful. For a neutral atom, for example, the Berry force cancels the bare Lorentz force exactly. The total force on the nucleus then vanishes and the atom moves with a constant velocity in a magnetic field (rather than undergoing a cyclotron motion), as expected for a neutral system. For a molecule, the screening of the individual atoms is not perfect, and the (screened)

Lorentz force will affect the motion of the molecule in subtle ways, as illustrated later.

The Berry force is obtained from the *Berry curvature*, which for a molecule of  $N$  atoms is a  $3N$ -by- $3N$  real-valued antisymmetric matrix, whose  $IJ$  block is given by the expression

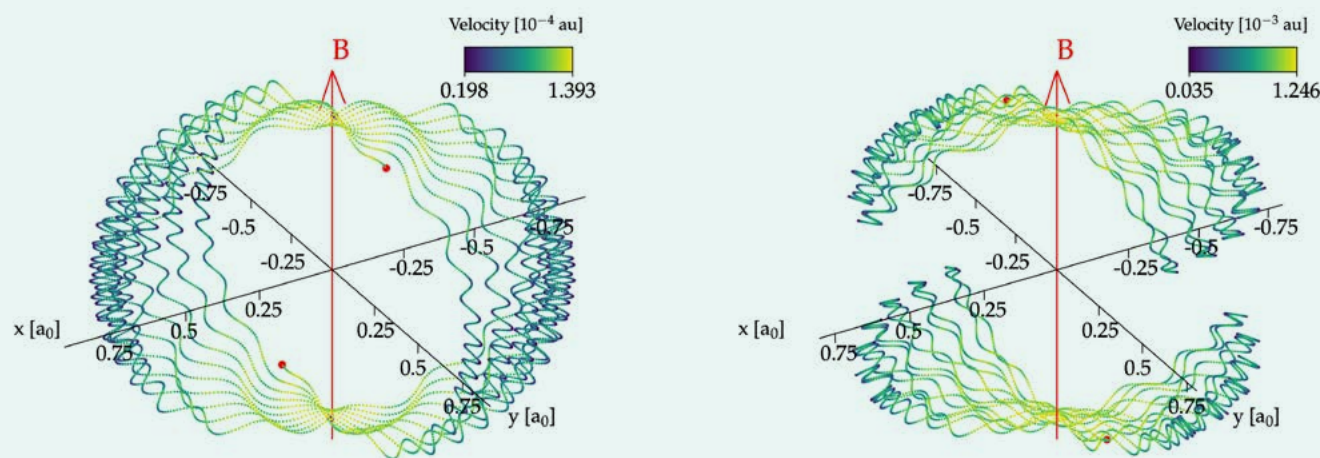
$$\Omega_{IJ}(\mathbf{R}, \mathbf{B}) = -2\hbar \text{Im} \langle \nabla_I \Psi(\mathbf{R}, \mathbf{B}) | \nabla_J^\dagger \Psi(\mathbf{R}, \mathbf{B}) \rangle,$$

where  $\Psi(\mathbf{R}, \mathbf{B})$  is the electronic wave function at nuclear positions  $\mathbf{R}$  and magnetic field strength  $\mathbf{B}$  (omitting electronic coordinates),  $\hbar$  is Planck's constant divided by  $2\pi$ , and  $\text{Im } z$  denotes the imaginary part of the complex number  $z$ . The evaluation of the Berry curvature is similar to that of the molecular Hessian (the second derivatives of the Born–Oppenheimer potential-energy surface), requiring the calculation of first derivatives of the electronic wave function with respect to nuclear displacements  $\nabla_I \Psi(\mathbf{R}, \mathbf{B})$ . Force calculations are therefore considerably more expensive in the presence of a magnetic

field than in the absence of a magnetic field.

In Refs. [11–12], we present the first gauge-invariant *ab initio* molecular dynamics calculations for general molecules; previous studies were restricted to  $\text{H}_2$  in a perpendicular field orientation and in a minimal atomic-orbital basis [10]. The analytic calculation of the Berry curvature is described in Ref. [13], while Ref. [14] discusses some properties of the Berry curvature. In particular, we demonstrate that the magnetic-translational sum rule, which ensures a correct translational motion in a magnetic field, is satisfied not only for exact electronic wave functions but also for approximate wave functions constructed from London atomic orbitals.

Having calculated the forces, the next step is the integration of Newton's equations of motion within the Born–Oppenheimer approximation, to obtain the trajectories that molecules trace out in the presence of a magnetic field.



⌚ **Figure 2:** Trajectories of  $\text{H}_2$  in a magnetic field of strength  $B = 0.2B_0$  along the  $z$  axis [15]. The nuclear velocity is indicated by different colour shades, with a darker colour corresponding to a slower motion. For the depicted total simulation time of 435 fs, the molecule rotates 90 degrees about the magnetic field axis. The trajectory plotted to the left represents a situation of hindered rotation, with an initial energy in the polar mode of 3.5 millihartree (sufficient to overcome the rotational barrier); the trajectory plotted to the right represents a situation of libration (polar vibration), with an initial polar energy of 2.8 millihartree (insufficient to overcome the rotational barrier). Reproduced from Ref. [15] under CC-BY 4.0 license.

The standard propagators developed for molecular dynamics cannot be used since the classical Hamiltonian is no longer “separable”, with the kinetic energy depending both on the momentum and the position of each nucleus. The challenge is to come up with propagators that are both stable and efficient, with good conservation of the total energy, for molecules subject to velocity-dependent forces. Our best propagators achieve this goal [12,15]. The higher cost of calculating molecular trajectories in the presence of a magnetic field therefore arises primarily from the higher cost of force evaluation than from their more complicated integration.

From the calculated Born–Oppenheimer trajectories, we can next extract the rovibrational spectra of molecules by taking the Fourier transform of the velocity autocorrelation function obtained from the nuclear kinetic momenta cal-

culated along the trajectories. We have calculated the rovibrational spectra of several small molecules in a magnetic field in this manner [12,15].

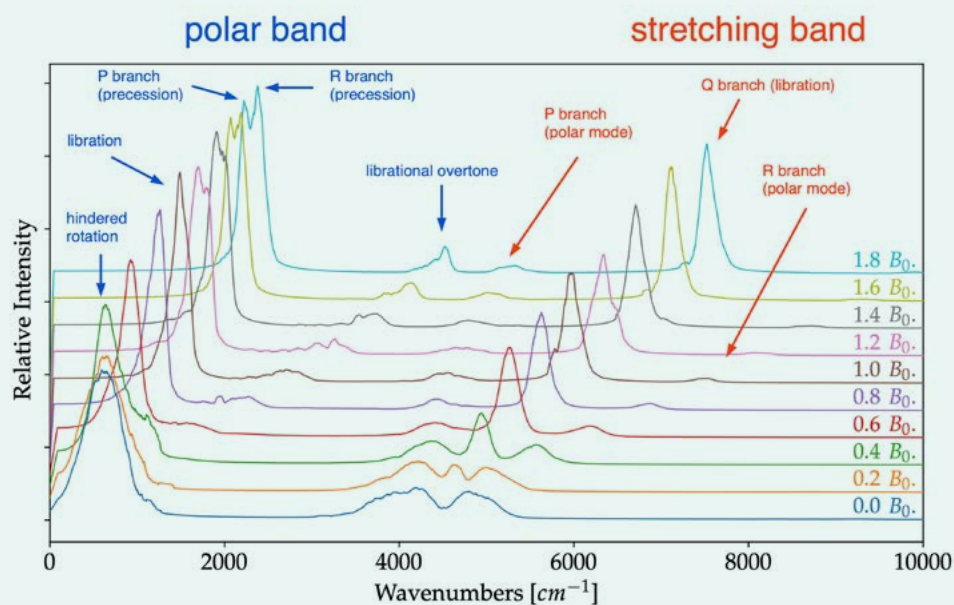
Let us consider the rovibrational spectrum of  $\text{H}_2$  in the lowest singlet state. There are six degrees of freedom – three translational, two rotational, and one vibrational degree of freedom. We can ignore translations and describe the molecule using spherical coordinates: the radial coordinate  $r$  representing the bond distance and the azimuthal angle  $\phi$  and polar angle  $\theta$  describing the orientation of the molecule; see Figure 1. In the absence of a magnetic field, the electronic energy depends only on  $r$ ; in a magnetic field along the  $z$  axis, it depends also on  $\theta$  but not on  $\phi$ .

As shown in Figure 1, the equilibrium bond distance of  $\text{H}_2$  contracts and the dissociation energy increases with increasing magnetic field strength. For a

given bond distance, singlet  $\text{H}_2$  has its lowest energy in the parallel orientation to the field and its highest energy in the perpendicular orientation. Furthermore, the barrier to polar rotation increases with increasing field strength, reaching 66 kJ/mol at one atomic unit field strength,  $B = B_0$ .

In Figure 2, we have plotted two trajectories of  $\text{H}_2$ , both initiated with the molecule in a parallel field orientation, with an initial energy in the polar mode of 3.5 (left) and 2.8 (right) millihartree. Some initial energy was also put in the stretching mode [15].

The polar motion of  $\text{H}_2$  is fundamentally different in the two cases depicted in Figure 2. To the left, there is sufficient energy in the polar mode to overcome the rotational barrier in the  $xy$  plane, although the motion slows down considerably as the molecule passes through the barrier. To the right,



⬆ **Figure 3:** Rovibrational spectra of singlet  $H_2$  up to a field strength of  $1.8B_0$  [15]. For the canonical ensemble, a set of 2000 spectra were calculated using a target temperature of 1500 K. Each individual spectrum was calculated from 1 ps simulations with initial velocities from a Maxwell–Boltzmann distribution. To smooth the averaged spectrum, all frequencies were averaged in an interval of  $\omega \pm 20 \text{ cm}^{-1}$ . Adapted from Ref. [15] under CC-BY 4.0 license.

the molecule does not have sufficient energy in the polar mode to cross the barrier; instead, it performs a pendulum motion or libration relative to the  $z$  axis.

In addition to undergoing a polar motion, the molecule vibrates, with a frequency higher than that of the hindered rotation or libration. In both cases, more vibrations occur when the molecule is close to the perpendicular orientation, which is simply a reflection of the fact that the polar motion is slower for large polar angles than for small ones, implying that the molecule spends most of its time in a (near) perpendicular orientation.

We also notice that the molecule undergoes a slow azimuthal rotation – this happens even though the molecule began its motion in a parallel field orientation, for which no energy can be deposited in this mode. Instead, the azimuthal motion is generated by the

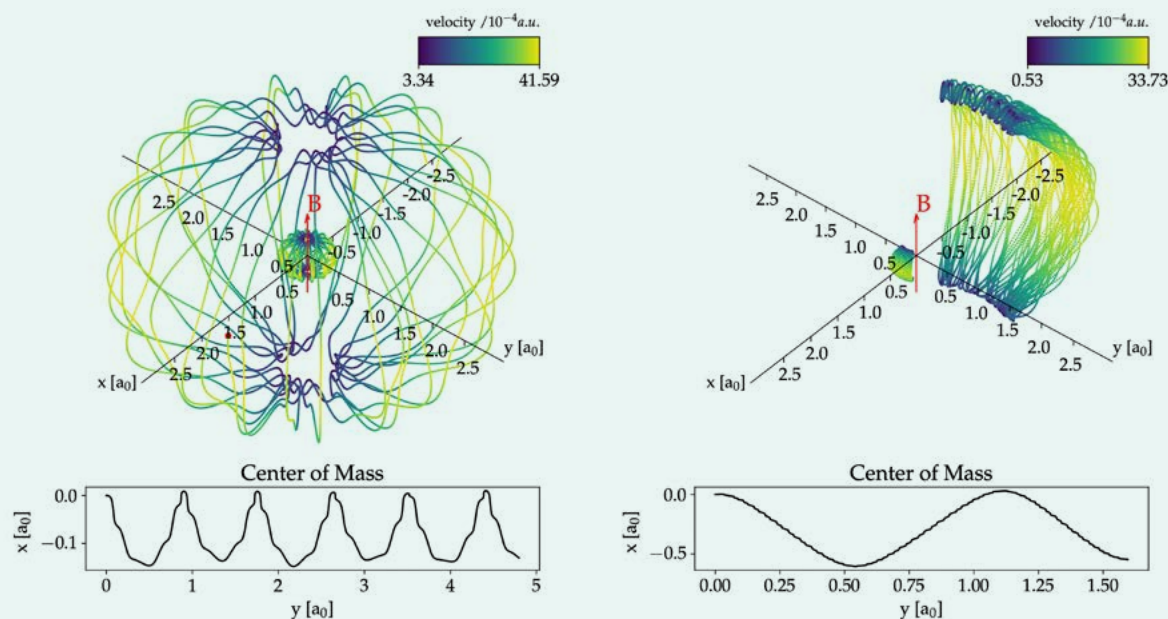
Lorentz force. Since screening of the external field is not perfect, a small residual Lorentz force acts on each nucleus, inducing a cyclotron motion of each atom. As a result, the molecule precesses slowly about the field axis, with the centre of mass at rest by symmetry.

Let us now consider the rovibrational spectrum of singlet  $H_2$  in a magnetic field. In Figure 3, we have plotted the rovibrational spectrum of  $H_2$  at ten different field strengths up to  $1.8B_0$  [15]. Each spectrum has been obtained from an NVT ensemble averaged over 2000 individual spectra calculated from 1 ps trajectories, with initial conditions determined by the Maxwell–Boltzmann distribution.

The rovibrational  $H_2$  spectra are dominated by two bands – a polar band at low frequencies and a stretching band at high frequencies. The polar band

undergoes a transition from hindered rotation to libration at about  $0.4B_0$ ; in weaker fields, the polar band is very slightly red shifted by the field; in stronger fields, the band is strongly blue shifted. The red shift occurs as the barrier to hindered rotation increases with increasing field strength; the blue shift occurs as the potential for libration becomes steeper with increasing field strength; see Figure 1.

As seen in the spectrum, libration behaves as expected for a vibrational mode. For example, it couples to the precessional motion, splitting into P and R branches in the usual manner, although the two branches can be distinguished only for very high field strengths. We also observe a librational overtone, which is strongly blue shifted with increasing field strength, approaching the P branch of the stretching band.



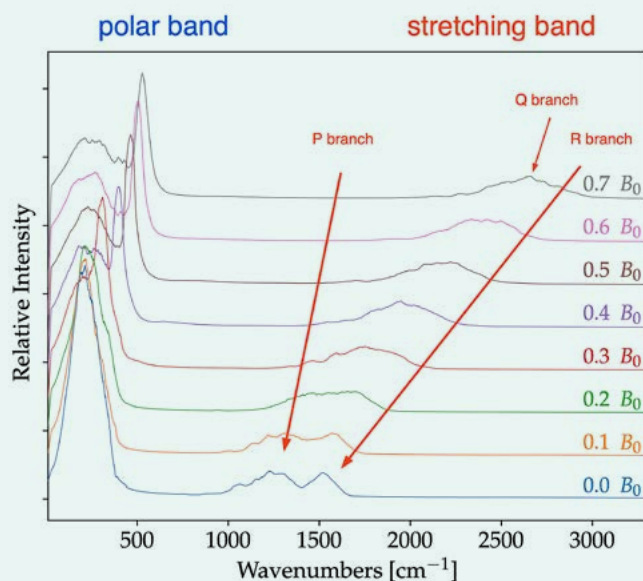
⌚ **Figure 4:** More rotation-like trajectory (a) and more libration-like trajectory (b) of LiH at a magnetic field strength of  $0.2B_0$ . The centre-of-mass motion has been eliminated here and is shown separately below the respective trajectories. Both simulations were initiated from the equilibrium geometry with LiH parallel to the x axis. Only the rotation around the y axis and the stretching vibration were given initial velocities. *Reproduced from Ref. [15] under CC-BY 4.0 license.*

Turning our attention to the stretching band, we note that it is strongly blue shifted (by several thousand wave numbers) as the radial potential becomes deeper and steeper with increasing field strength; see Figure 1. At weak field strengths, the stretching band is split in the usual manner into P and R branches by coupling to the (hindered) rotation; at higher field strengths, a Q branch appears and eventually dominates the spectrum, as in a polyatomic molecule.

We have also calculated vibrational spectra of number of other small systems, each yielding additional insight. In LiH, for example, the magnetic field induces a periodic centre-of-mass motion in addition to the precession observed in singlet  $H_2$ . The centre-of-mass motion of LiH arises since the induced cyclotron motion of the two partially screened atoms are not related by symmetry, unlike in  $H_2$ ; see Figure 4.

Another difference relative to singlet  $H_2$  is that LiH has a preferred perpendicular field orientation, with a rotational barrier in the parallel orientation. As a result, LiH never passes through the parallel orientation even though sufficient energy is present to accomplish this. The resulting rovibrational spectrum is depicted in Figure 5.

Triplet  $H_2$  is not bound on Earth and therefore has no vibrational spectrum. In a strong magnetic field, however, this molecule becomes bound by the paramagnetic bonding mechanism. Our calculations revealed a stretching band at about  $940\text{ cm}^{-1}$  at  $B = B_0$ , much lower than in singlet  $H_2$ .



⬆ **Figure 5:** Boltzmann averaged spectra calculated from a set of 2000 LiH spectra at different field strengths. The initial velocities were chosen to yield a target temperature of 1500 K. The spectra were smoothed by averaging all frequencies within the interval  $\omega \pm 20 \text{ cm}^{-1}$ . Adapted from Ref. [15] under CC-BY 4.0 license.

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# Almlöf–Gropen Lecture Series

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The Almlöf–Gropen Lecture Series was established by the Centre for Theoretical and Computational Chemistry in 2008 to honour the memory of two pioneers of quantum chemistry in Norway: Prof. Jan Almlöf (1945–1996) at the University of Oslo and Prof. Odd Gropen (1941–2005) at UiT The Arctic University of Norway. Each year, a prominent theoretical chemist is invited to deliver the Almlöf–Gropen lecture at the Hylleraas Centre in Oslo and Tromsø, targeting a broad audience. Since 2020, the Almlöf–Gropen Lecturer has been preceded by two Almlöf–Gropen Young Speakers, selected among the young members of the Hylleraas Centre.

# Previous Almlöf–Gropen Lecturers

- 2008 Prof. Björn Roos**  
University of Lund, Sweden  
*Multiconfigurational quantum chemical methods and heavy element chemistry*
- 2009 Prof. Tom Ziegler**  
University of Calgary, Canada  
*Analyzing complex electronic structure calculations on large molecules in simple chemical terms*
- 2010 Prof. Michele Parrinello**  
ETH Zürich, Switzerland  
*Through mountains and valleys with metadynamics*
- 2011 Prof. Pekka Pyykkö**  
University of Helsinki, Finland  
*Relativity and chemistry: some recent results*
- 2012 Prof. Harry B. Gray**  
California Institute of Technology, USA  
*The 21st century solar army*
- 2013 Prof. Henry F. Schaefer**  
University of Georgia, Athens, USA  
*From donor-acceptor complexes to Gallium Nitride nanorods*
- 2014 Prof. Leo Radom**  
University of Sydney, Australia  
*Adventures in free radical chemistry: a computational approach*

- 2015 Prof. Arie Warshel**  
University of Southern California, USA  
*How to model the action of complex biological systems on a molecular level*
- 2016 Prof. Emily Carter**  
Princeton University, USA  
*Quantum solutions for a sustainable energy future*
- 2017 Prof. Jack Simons**  
University of Utah, USA  
*The wonderful world of molecular anions*
- 2018 Prof. Walter Thiel**  
MPI Mühlheim, Germany  
*Chemistry with the computer*
- 2019 Prof. Sharon Hammes-Schiffer**  
Yale University, Connecticut, USA  
*Proton-coupled electron transfer in catalysis and energy conversion*
- 2020 Prof. Leticia Gonzalez**  
University of Vienna, Austria  
*Light and shadows on the quantum simulation of molecular electronic excited states*
- 2021 Prof. Alán Aspuru-Guzik**  
University of Toronto, Canada  
*There is no time for science as usual: materials acceleration platforms*

# Almlöf-Gropen Lecturer 2022



## Professor Clémence Corminboeuf

École Polytechnique Fédérale de  
Lausanne (EPFL), Switzerland

Going further by leveraging  
data for solving quantum/  
computational chemistry  
problems

June 1 2022, University of Oslo  
June 2 2022, UiT The Arctic  
University of Norway

Prof. Clémence Corminboeuf earned her PhD degree from the University of Geneva. After postdoctoral work at New York University and University of Georgia, she began her independent career as an assistant professor at the EPFL in 2017, where she was promoted to associate professor in 2014 and full professor in 2019. She is the recipient of the Silver Medal at the European Young Chemists Award (2010), the Werner Prize of the Swiss Chemical Society (2014), and the Theoretical Chemistry Award from the Physical Chemistry Division of the American Chemical Society (2018). She has also been awarded two ERC grants (Starting Grant in 2012 and Consolidator Grant in 2018). She has co-authored over 200 publications on quantum-chemical methods and on conceptual tools targeting homogeneous catalysis and organic functional molecules.



Prof. Clémence Corminboeuf giving her lecture at the Hylleraas Centre in Oslo on June 1 2022. *Photo: Trygve Helgaker.*

In her Almlöf-Gropen lecture, Corminboeuf presented a collection of tools and concepts that aim to accelerate homogeneous catalyst discovery, mixing proof-of-principle examples with real-life applications and highlighting the benefits and challenges of the use of statistical models to predict catalytic properties. Focusing on a handful of challenging properties, she showed how quantum-chemical techniques can be leveraged to improve the accuracy and broaden the field of applicability of kernel-based models.

# Almlöf–Gropen Young Speakers 2022



Sahil Gahlawat

Hylleraas Centre,  
UiT The Arctic University of Norway

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## Mechanistic insights into the reduction of CO<sub>2</sub> using molecular catalysis

Sahil Gahlawat received his bachelor's degree in chemistry from Delhi University in 2018 and his master's degree from the Indian Institute of Technology, Roorkee in 2020, where he was introduced to computational chemistry during his last term. His research interests then changed to quantum-chemical aspects of chemistry including molecular modelling, computational catalysis, and the application of theoretical techniques to the field of organometallic and organic chemistry. He began his PhD study at UiT in 2021, applying theoretical methods to study the mechanisms of homogeneous transition-metal catalysed reactions like the conversion of carbon dioxide to products of higher value.



Samiran Sen

Hylleraas Centre,  
University of Oslo

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## Pressure in lipid membranes

Samiran Sen graduated from St. Xavier's College, Kolkata with a bachelor's degree in Physics Honours from the University of Calcutta in 2017. He completed his master's course in theoretical and computational physics with a specialization in condensed matter from the University of Calcutta in 2019. He is currently working as a PhD research fellow at the Hylleraas Centre at the University of Oslo, supervised by Michele Cascella.



# Meetings and Events

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## Attosecond Quantum Dynamics Beyond the Born–Oppenheimer Approximation

Simen Kvaal and  
Thomas Bondo Pedersen

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Centre for Advanced Study, Oslo  
2021–2023

The project *Attosecond Quantum Dynamics Beyond the Born–Oppenheimer Approximation (ATTODYN)*, led by Hylleraas members Simen Kvaal and Thomas Bondo Pedersen at the Centre for Advanced Study during the academic year 2021/22, was granted an extension to the academic year 2022/23 because of the COVID-19 pandemic.


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The Centre for Advanced Study (CAS) at the Norwegian Academy of Science and Letters is an independent foundation funded by the Norwegian Ministry of Education and Research that furthers fundamental, curiosity-driven research in humanities, social sciences, and natural sciences. Every year, three research groups work at CAS, having been selected in an application process two years earlier. The project *Attosecond Quantum Dynamics Beyond the Born–Oppenheimer Approximation (ATTODYN)* is led by Thomas Bondo Pedersen and Simen Kvaal.

In the past decade, a rapid development of novel experimental techniques producing laser pulses with a duration on the time scale of the electron – the attosecond ( $10^{-18}$  s) time scale – has opened new, exciting opportunities in chemistry. Attosecond laser pulses

allow us to create and monitor electronic wave packets within molecules in a controlled fashion. These electronic wave packets radically change the forces driving the motion of the nuclei. Hence, by controlling the wave packets by means of laser-pulse parameters, we may control the motion of the nuclei. In plain words: we can potentially make molecules do what we want them to do by means of attosecond laser pulses, including detailed control of chemical reactions.

The advanced experiments carried out with attosecond laser pulses are, of course, subject to interpretation by means of theoretical models. Therefore, it is of fundamental importance to develop computational models that do not inadvertently lead to misinterpretation or even miss important phenomena. The main objective of the ATTO-



Banner of the CAS project *Attosecond Quantum Dynamics Beyond the Born–Oppenheimer Approximation*.  
Design: Camilla Kottum Elmar | CAS.

DYN project is to investigate coupled electronic–nuclear dynamics induced by attosecond and femtosecond laser pulses. The starting point is the molecular time-dependent Schrödinger equation for both nuclear and electronic degrees of freedom, which we seek to solve numerically without assuming the Born–Oppenheimer approximation.

Imposing the concept of molecular structure on the quantum theory of matter, the Born–Oppenheimer approximation is omnipresent in theoretical chemistry and solid-state physics. Developing computationally models without it, we face fundamental conceptual difficulties. Our initial efforts aim at a simplified computational non-Born–Oppenheimer model that can correctly predict laser-induced molecular alignment, including the post-pulse periodic revival structure arising from quantum interference, which is a key phenomenon routinely exploited in attosecond experiments.

The short duration of attosecond laser pulses implies a broad energy distribution which, almost invariably, leads

to nonvanishing one- and many-electron ionization probabilities and, in turn, a nonzero probability of molecular dissociation and fragmentation. Consequently, another research line pursued in the ATTODYN project is the mathematical representation of ionization and dissociation continua using computationally efficient, fully flexible complex Gaussians as basis functions. The key challenge is the development of numerically stable integrators with the required adaptivity in both space and time.

The ATTODYN group is interdisciplinary, consisting of researchers from chemistry, physics, and mathematics. In the fall 2022, the stay at the CAS premises at the Norwegian Academy of Science and Letters in Oslo had ended and the members of the project worked individually from their home institutions. Activity on the CAS project was partially put on hiatus. However, future collaborations in the wake of the project were planned, and an ERC Synergy Grant application was submitted. Long-term visits by Prof. Caroline Lasser (Technical University of Munich) and

Prof. Ludwik Adamowicz (University of Arizona) were planned for spring 2023, financed by CAS. An international workshop financed by CAS was scheduled, also for spring 2023.

Apart from the core members Ludwik Adamowicz and Caroline Lasser, CAS fellows Prof. Sonia Coriani (Technical University of Denmark), Prof. Reinhold Schneider (Technische Universität Berlin), and Prof. Takeshi Sato (University of Tokyo) worked with Kvaal and Pedersen in Oslo in 2022. In addition, Prof. Daniel Crawford (Virginia Tech) visited CAS in the spring of 2022.

Several scientists from the Hylleraas Centre also contributed to the CAS project in 2022: Trygve Helgaker, Erik Tellgren, André Laestadius, Mihály Csirik, Håkon Kristiansen, Einar Aurbakken, and Benedicte Ofstad. Finally, PhD candidates Øyvind Schøyen and Jonas Flaten from the Department of Physics at the University of Oslo (both co-supervised by Pedersen) worked at CAS one day per week.

For details on the ATTODYN research, see the RT3 Activity report.



Young CAS Workshops:  
Global Optimization in Electronic Energy Landscapes:

## Finding a Needle in a Haystack

Hylleraas Centre, Oslo  
September 13–17 2021

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Hylleraas Centre, Oslo  
April 11–14 2022

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Norwegian Academy of Science  
and Letters, Oslo  
June 20–23 2022

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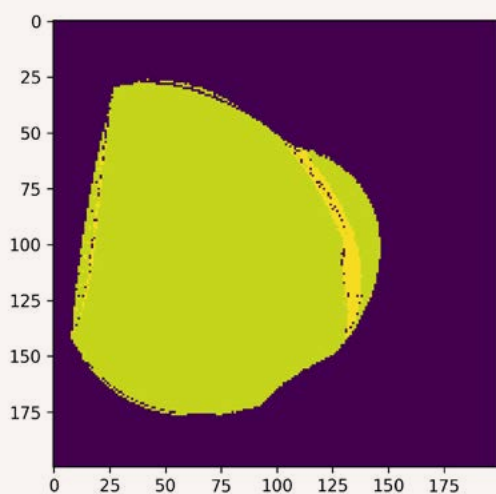
**In 2020, Dr. Erik Tellgren was awarded a Young CAS grant devoted to global optimization in electronic-structure theory. As part of his Young CAS project, three workshops for young researchers have been organized by Tellgren – one in 2021 and two in 2022.**

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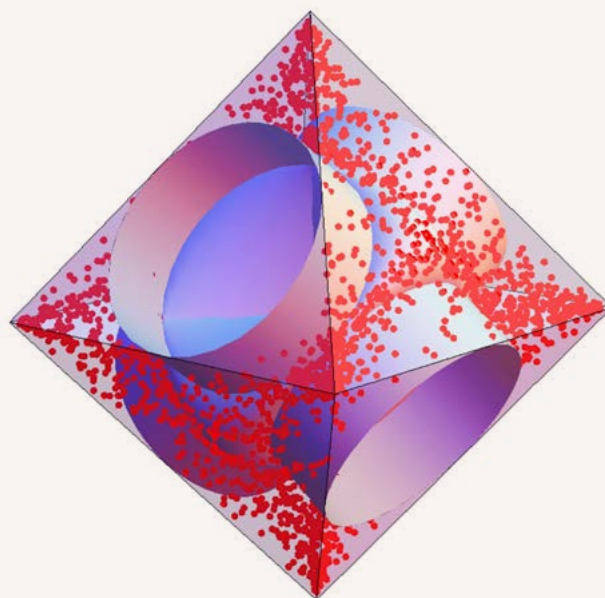
Most quantum-chemical calculations rely on our ability to find the electronic ground state – that is, the state of lowest possible electronic energy. But how do we know that the output of a quantum-chemical software is in fact the lowest energy state and that no state of lower energy has been overlooked? Erik Tellgren's Young CAS project, *Global Optimization in Electronic Landscapes: Finding a Needle in a Haystack*, is devoted to this and related questions.

The project consisted of three workshops, with the first held at the Hylleraas Centre on September 13–17 in 2021; see last year's Annual Report. Its participants – Erik Tellgren, André Laestadius, Mihaly Csirik, Audun Skau Hansen, Tanner Culpitt, and Laurens Peters from the Hylleraas Centre and Markus Penz and Michael Herbst from abroad – discussed the general topic of the project from a number of angles, ranging from ground-state convergence in exact density-functional theory to the problem of multiple energy minima when both electrons and nuclei are treated quantum mechanically.

The second workshop was held at the Hylleraas Centre on April 11–14, 2022. Here the five centre participants together with Markus Penz, Klaas Giesbertz, Sarina Sutter, Nicholas Cartier, and Mathias Oster continued the discussion on global optimization. Oral contributions to the workshop suggested proofs of convergence for the pop-



⤴ **Figure 1:** Dr. Penz discussed a simple lattice model where the set of states that are difficult to reach can be given a precise geometrical characterization.



⤴ **Figure 2:** Numerical example due to Dr. Skau Hansen of how different initial states for the optimization lead a standard algorithm to find different final states. The colour represents the energy of the final, optimized state. Standard algorithms do not lead to a connected basis of attraction.

ular Hartree–Fock method, gave examples where multiple solutions are found in Hartree–Fock theory, and discussed new approaches such as density-matrix embedding theory. The participants also considered density-matrix functional theory and how convexity may be exploited in practical approximations within this framework. A session of the workshop considered situations where even the exact Kohn–Sham method fails to converge due to degeneracy and examined a new scheme for global optimization for nonconvex problems using regularization. Finally, the participants discussed what can be inferred about the global optimum of the Hartree–Fock model from the properties of a local optimum.

The third and final Young CAS workshop was held at the Norwegian Academy of Science and Letters on June 20–23, 2022. Continuing where the second workshop had left off, the five participants from the Hylleraas Centre together with Markus Penz, Tibor Györi, and Louis Garrigue discussed topics ranging from more mathematical to more practical. For example, Penz presented a geometrical perspective on degeneracy in a lattice formulation of density-functional theory (Figure 1), Tellgren considered how the mathematical notion of difference convexity can be exploited in Hartree–Fock and density-functional calculations, while Györi discussed how to avoid local minima in practical applications. Skau Hansen showed how

different initial states for the optimization can lead a standard algorithm to find different final states (Figure 2).

While global optimization remains an outstanding challenge for electronic structure, the three workshops gave a wealth of ideas to pursue in the future.



↑ Dr. Ainara Nova led the Global Women's Breakfast event at the Hylleraas Centre in Oslo, on February 16, 2022.  
Photo: Trygve Helgaker.

## 2022 Global Women's Breakfast

Hylleraas Centre, Oslo  
February 16, 2022

**The Hylleraas Centre in Oslo hosted the 2022 Global Women's Breakfast organized by the NordCO<sub>2</sub> and CO<sub>2</sub>PERATE consortia on February 16, 2022.**

On February 16, 2022, the Hylleraas Centre in Oslo hosted the IUPAC 2022 *Global Women's Breakfast: Empowering Diversity in Science* event, which was organized by the Nordic Consortium for CO<sub>2</sub> Conversion (NordCO<sub>2</sub>) and the consortium Cooperation Towards a Sustainable Chemical Industry (CO<sub>2</sub>PERATE). At the breakfast, Prof. Emer. Christina Moberg from KTH Royal Institute of Technology in Stockholm, Sweden, gave a lecture entitled *Empowering diversity in science – strength lies in differences, not similarities*. After her presentation, a discussion including Prof. Adela Muñoz Páez from the University of Seville, Spain, followed.

PhD candidate Inga Schmidtke at the Hylleraas Centre and Dr. Raquel Jiménez Rama at the Centre for Materials Science and Nanotechnology (SMN) at the University of Oslo helped with the organization.

The Global Women's Breakfast is an annual event organized by the International Union of Pure and Applied Chemistry (IUPAC) on the United Nations Day of Women in Science. According to the IUPAC web pages, the goal of the series is "to establish an active network of people of all genders to overcome the barriers to gender equality in science".



⬆ Skibotn Field Station in glorious weather.  
Photo: Tilmann Bodenstein.



⬆ Unwinding at Skibotn Field Station. Left to right: Marc Joosten, Ryan Wilkins, Tonje Reinholdt Haugen and Sahil Gahlawat. Photo: Bente Barge.

## Hylleraas Hackathon

Skibotn Field Station  
April 25–29, 2022

The Hylleraas Hackathons serve as a well-established meeting place for members of the Hylleraas Centre. Originally an event in Tromsø dating back to the days of the Centre for Theoretical and Computational Chemistry (CTCC), it now draws participants from both Oslo and Tromsø. The purpose of the gathering is to coordinate various activities at the centre, provide training on diverse topics, and foster informal networking.

The first hackathon following the COVID-19 pandemic took place from April 25 to 29, 2022, at Skibotn Field Station, organized by Dr. Magnus Ringholm. This station belongs to UiT The Arctic University of Norway and is situated in a stunning location near Lyngfjorden, approximately 120 km from Tromsø and 35 km from Finland. During the Hackathon, attended by about 25 participants, individuals had the opportunity to engage in focused work on their projects, either independently or collaboratively, away from the usual distractions. Dr. David Balcells conducted an informal workshop on machine learning, and participants also had ample opportunities to socialize around the fireplace or sauna. The accommodations, though simple, are more than sufficient and conducive to productive work.



⌂ Director of JSPS Stockholm  
Tadaharu Tsumoto presenting  
JSPS at the symposium.  
Photo: Trygve Helgaker.

# Norway–Japan Symposium on Theoretical and Experimental Chemistry of Complex Systems

Hylleraas Centre, Oslo  
March 15–16, 2022

**The fourth Norway–Japan Symposium on Theoretical and Experimental Chemistry of Complex Systems was held at the Hylleraas Centre March 15–16, 2022, with more than 70 participants.**

As part of an ongoing collaboration between the Hylleraas Centre and three Japanese research groups led by Professor Masahiro Ehara at the Institute of Molecular Science (IMS), Professor Shigeyoshi Sakaki at Kyoto University, and Professor Susumu Kitagawa at Kyoto University, the fourth Norway–Japan symposium was held at the Hylleraas Centre in Oslo on March 15–16, 2022, attracting more than 70 participants from Norway, Japan, Finland, Germany, France, Poland, and USA. The symposium was organized by Trygve Helgaker, Erik Tellgren and

Michiko Atsumi, with assistance from Simen Reine, all at the Hylleraas Centre in Oslo.

Because of the COVID-19 pandemic, the symposium was held in a hybrid manner, with several online presentations. It was supported by the Hylleraas Centre, the Japan Society for the Promotion of Science (JSPS), the JSPS Alumni Club in Norway (ACN), and the Embassy of Japan in Oslo. The first and third symposia in this series were hosted by IMS, while the second symposium was hosted by the Hylleraas Centre.



⬆ Reception and dinner at the Residence of the Japanese Ambassador to Norway in the evening of March 15, 2022. From left to right: Dr. Michiko Atsumi, Ms. Kaoru Mochizuki, Prof. Unni Olsbye, Prof. Trygve Helgaker, Director of JSPS Stockholm Tadaharu Tsumoto, Ambassador of Japan to Norway Hiroshi Kawamura, Chair of JSPS Alumni Club in Norway Anders Øverby, and Mr. Takenari Yamamori.

The first day of the 2022 symposium featured talks by prominent scientists in the field: Prof. Susumu Kitagawa (Kyoto University), Prof. Pekka Pyykkö (University of Helsinki), Prof. Mitsuhiro Shionoya (University of Tokyo), and Prof. Unni Olsbye (University of Oslo). After these scientific talks, short addresses were given by representatives of the Japanese Embassy, JSPS and the ACN.

The second day of the symposium was devoted to shorter scientific talks, given by three Japanese theoretical chemists, Assoc. Prof. Minoru Abe (Hiroshima University), Assoc. Prof. Naoki Nakatani (Tokyo Metropolitan University), and Prof. Masataka Nagaoka (Nagoya University), by the two Hylleraas members, Dr. Abril Castro and Dr. Michel Repisky, and by two experimental scientists from

Norwegian institutions, Chief Research Scientist Richard Blom (SINTEF Industry) and Prof. Pascal Dietzel (University of Bergen).

After the closing of the symposium, Tadaharu Tsumoto, Director of JSPS Stockholm, Trygve Helgaker, Erik Tellgren, and Michiko Atsumi paid a courtesy call to University of Oslo's central management, meeting with Pro-Rector Åse Gornitzka, Vice-Rector for Education, Bjørn Stensaker, and Senior Advisor Karen Crawshaw Johansen.

In the evening of March 15, Trygve Helgaker, Unni Olsbye, Michiko Atsumi and Tadaharu Tsumoto were invited to dinner at the residence of the Japanese Ambassador to Norway, Hiroshi Kawamura.



↑ Participants of the summer school *Computational Molecular Design* at the University of Bergen.

# Computational Molecular Design

University of Bergen  
May 9–11, 2022

Over three days in May 2022, the Nordic Consortium for CO<sub>2</sub> Conversion (NordCO<sub>2</sub>), organized a summer school in computational molecular design at the University of Bergen.

Within the framework of the NordCO<sub>2</sub> consortium, Ainara Nova together with Prof. Vidar Jensen (UiB) and Dr. Marie-Josée Haglund Halsør (UiT) organized the summer school *Computational Molecular Design* in Bergen on May 9–11, 2022. The school was attended by seventeen students from nine different research institutions in the Nordic countries.

The Hylleraas Centre contributed not only to the organization but also to the teaching of the meeting, including theory lectures (David Balcells) and tutorials (Simen Reine, with contributions from Hannes Kneiding) on the topic of machine learning for chemistry. The school included a dinner, which was an excellent opportunity for the five Hylleraas students who attended the school to socialize and extend their international network.



↑ Participants at the CECAM Flagship School Multiscale Molecular Dynamics with MiMiC in Lausanne, July 18–22 2022. From left to right: Prof. Simone Meloni (University of Ferrara, organizer), Paolo Carloni (Forschungszentrum Jülich), Marinella de'Giovanetti (Hylleraas Centre, poster-prize winner), Aparna G Nair (IIT-PKD, poster-prize winner), Assoc. Prof. Jógvan M. H. Olsen (DTU and Hylleraas Centre, organizer), and Prof. Ursula Röthlisberger (EPFL, organizer).

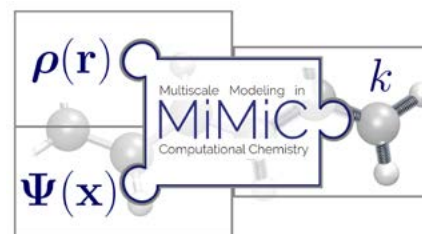
## Multiscale Molecular Dynamics with MiMiC

CECAM-HQ-EPFL, Lausanne, Switzerland  
July 18–22, 2022

**Hylleraas adjunct Assoc. Prof. Jógvan M. H. Olsen was one of the organizers and teachers of the summer school *Multiscale Molecular Dynamics with MiMiC* in Lausanne in July 2022, with 55 participants.**

The *Multiscale Molecular Dynamics with MiMiC* is a flagship school of the Centre Européen de Calcul Atomique et Moléculaire (CECAM), organized by Prof. Simone Meloni (University of Ferrara), Ass. Prof. Jógvan M. H. Olsen (DTU and the Hylleraas Centre), and Prof. Ursula Röthlisberger (EPFL) at the CECAM headquarters in Lausanne July 18–22, 2022. The school focused on the modelling of large and complex (bio) chemical systems, ranging from molecules in solution to membrane-embedded proteins, using multiscale methods. The school, which was aimed at young as well as more experienced researchers, combined lectures with practical sessions using the multiscale modelling framework MiMiC, of which Jógvan Olsen is a principal developer.

PhD candidate Marinella de'Giovanetti at the Hylleraas Centre in Oslo participated in the school and was awarded a prize for her poster *Towards an understanding of turbo-Grignard reagents: structural information from AIMD studies*.





↑ Participants of the NordCO<sub>2</sub> Annual Meeting 2022 in Iceland.

## NordCO<sub>2</sub> Annual Meeting 2022

Hótel Örk, Hveragerði, Iceland  
August 15–19, 2022

**The 2022 Annual Meeting of the Nordic Consortium for CO<sub>2</sub> Conversion (NordCO<sub>2</sub>) took place in Iceland, with 41 participants from seven Nordic universities.**

The NordCO<sub>2</sub> annual meeting, organized by Ainara Nova and Prof. Egill Skúlason (University of Iceland) with the assistance of Dr Marie-Josée Haglund Halsør (UiT), took place in the small town of Hveragerði near Reykjavík in August 2022. The event was attended by 41 participants from seven Nordic universities: Aarhus University, Stockholm University, KTH Royal Institute of Technology, University of

Helsinki, University of Iceland, UiT The Arctic University of Norway, and University of Oslo.

At the meeting, NordCO<sub>2</sub> PhD candidates, postdocs and researchers presented their work in oral presentations or in posters. In addition, Nova gave an introductory talk on the NordCO<sub>2</sub> events in 2022 and 2023, Prof. Troels Skrydstrup presented the newly funded *The Novo Nordisk Foundation CO<sub>2</sub> Research Center (CORC)*, and Prof. Kim Daasbjerg talked about new CO<sub>2</sub> capture strategies.

To promote scientific discussion among students from different universities, a special session, *The Big Challenge*, was organized, in which the World Cafe

Method was used to define new projects on CO<sub>2</sub> conversion. The feedback on this session was very positive, and it proved to be an excellent exercise in how to build a project from scratch.

The Hveragerði meeting included several social activities, such as a visit to Carbon Recycling International, a company that produces methanol from CO<sub>2</sub> and H<sub>2</sub>, the Blue Lagoon, and the Golden Circle Tour, including the Þingvellir National Park, the Geysir Geothermal Area, and the Gullfoss Waterfall. In addition to great weather, most participants could enjoy the spectacle of the erupting Fagradalsfjall Volcano, which had started on August 3.



↑ Organizers of the Hylleraas Annual Meeting at Klækken, PhD candidates Mahika Luthra and Samiran Sen.



↑ Students enjoying a stroll around Storetjern, 10 km from Klækken Hotel.

## Hylleraas Annual Meeting 2022

Klækken Hotell, Ringerike  
September 5–7, 2022

**The Hylleraas Annual Meeting at Klækken Hotel, September 5–7 2022, was organized by PhD candidates Mahika Luthra and Samiran Sen and attracted a record number of 63 participants.**

The Hylleraas Annual Meeting 2022 was held at Klækken Hotell, Ringerike, about 60 km from the University of Oslo and 50 km from Gardermoen Airport. The meeting was organized by the representatives of the Young Researcher Parliament Samiran Sen and Mahika Luthra. With 63 participants, the attendance at the annual meeting was record high, up from 35 at the Sundvollen meeting in 2020 and from 53 at the Hamn meeting in 2021.

The meeting was opened by Samiran Sen, Mahika Luthra, and Trygve Helgaker after lunch on Monday, September 5, 2022. The opening was followed by a session on the Hylleraas School chaired by Simen Reine. The remainder of the day was devoted first to a talk on open and reproducible science by Agata Bachyńska and Matthew Good, senior academic librarians at the University of Oslo, and a talk on entrepreneurship by Vegar Lein Ausrød, senior advisor of 'The Kitchen', Aarhus University.

The second day at Klækken began with the 5-Minute Thesis Competition, where all PhD and master's students presented their research projects in a nutshell. Sverre Løyland and Marinella de'Giovannetti, both doctoral fellows at the



↑ Audun Skau Hansen at Klækken Hotel demonstrating molecular orbitals using virtual reality software.

Oslo node, bagged the first and the second positions, respectively. This competition was followed by a talk on popular science by Hylleraas Communication Advisor Cathrine Strøm, a talk on the Hylleraas Software Platform by Simen Reine and Tilmann Bodenstein, and a talk on virtual reality and molecular visualization by Audun Skau Hansen. In the evening, a banquet dinner was served.

The third day was devoted to meetings of the Management Team and of the Young Researcher Parliament.



⬆ Dr. Simen Kvaal gave five lectures on Mathematical Methods at ESQC 2022.



⬆ Current and previous members of the Hylleraas Centre and CTCC at ESQC 2022. From left to right: Elke Fasshauer (Universität Tübingen, Germany), Roberto Di Remigio (EuroCC National Competence Centre Sweden, Stockholm), Abril Castro, Lisa Rebolini (Institut Laue Langevin, Grenoble, France), Gabrile Geréz, Lucas Lang, and Simen Reine.

## The 18<sup>th</sup> European Summerschool of Quantum Chemistry (ESQC)

Torre Normanna, Sicily, Italy  
September 11–24, 2022

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**The 18th European Summerschool of Quantum Chemistry was held in Sicily, Italy, with strong participation of current and previous members of the Hylleraas Centre.**  
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The European Summerschool in Quantum Chemistry (ESQC) took place for the 18th time September 11–24, 2022. The school attracted 81 students, among whom were Abril Castro and Gabriel Geréz from the Hylleraas Centre in Oslo and Tromsø, respectively. In addition, several members of the centre taught at the school. Simen Kvaal gave lectures on mathematical methods, while Simen Reine and Lucas Lang were exercise tutors. Among the tutors were also Roberto Di Remigio, previous member of the Hylleraas Centre, as well as Elisa Rebolini and Elke Fasshauer, previous members of the Centre for Theoretical and Computational Chemistry (CTCC).

The purpose of ESQC is to introduce the students to modern methods and computational techniques of quantum chemistry. The school aims at developing critical thinking and much time is therefore devoted to the discussion of different electronic-structure problems and the choice of appropriate methods for their solution.

The first ESQC school was held in August 1989 in Sweden, under the directorship of Prof. Björn Roos. Hylleraas Director Trygve Helgaker lectured at the school from 1991 to 2019.



⬆ Dinner for the SAC at Cru Restaurant in Oslo on September 14, 2022. From left to right: Michele Cascella, David Balcells, Markus Reiher, Trygve Helgaker, Samiran Sen, Mahika Luthra, Thomas Bondo Pedersen, Einar Uggerud, Chantal Daniel, Serena DeBeer, Jan Ingar Johnsen, and Kenneth Ruud.

## Visit of Scientific Advisory Committee

UIT The Arctic University of Norway  
September 12–13 2022

University of Oslo  
September 14–15 2022

**The Scientific Advisory Committee (SAC) of the Hylleraas Centre conducted their second visit to the centre on September 12–15, 2022. This visit aimed to assess the centre's developments and achievements while providing guidance for its future endeavours. The focus during their visit was on the new research plans and the preparations for the mid-term evaluation.**

The SAC had previously visited the Hylleraas Centre in November 2019, offering valuable advice for the centre's future development after two years of operation. Nearly all the advice provided at that time has been diligently implemented at the centre.

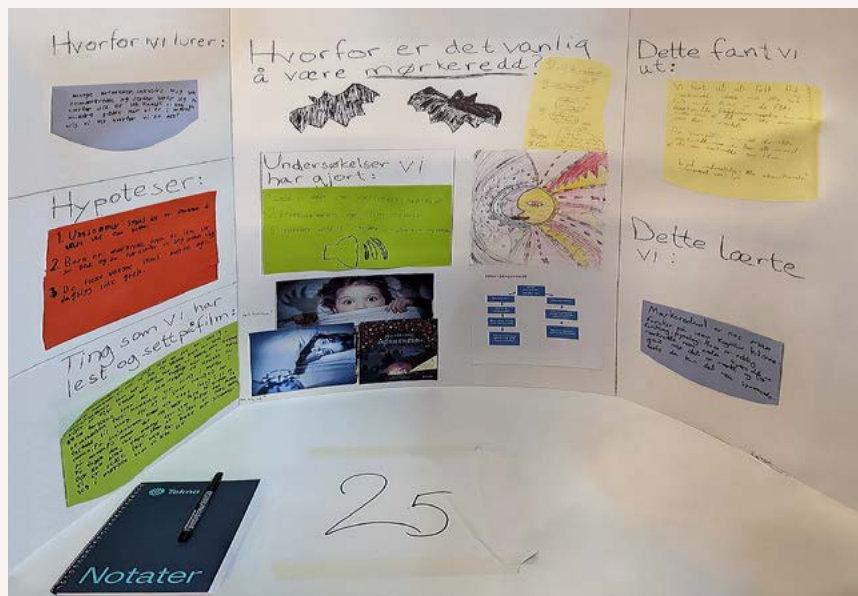
The 2022 visit of three of the SAC members, Prof. Chantal Daniel (chair), Prof. Senera DeBeer, and Prof. Markus Reier, was strategically timed to align with the preparation for the midterm evaluation, then scheduled for 2023 but later cancelled by the Research Council of Norway. The committee paid particular attention to the new research plans, the organization of the centre, and the introduction of new leadership in 2022 and 2023. The revised research plans are described elsewhere in this Annual Report.

The SAC engaged with all members of the centre during their visit, including special sessions with young members and the Young Researcher Parliament

(YRP). Additionally, meetings were held with the Heads of Departments in Oslo (Prof. Einar Uggerud) and Tromsø (Prof. Annette Bayer), along with Prof. Unni Olsbye, who serves as the chair of the Board of Directors.

Overall, the SAC provided an affirmative assessment of the Hylleraas Centre, its research, and the research environment; see page 126. The committee offered constructive criticism on specific aspects. Positive highlights from the SAC report encompassed the recognition of “truly remarkable” research, effective management of the pandemic, the enthusiasm of young researchers, and a well-constructed new research plan.

Areas identified for improvement included career development beyond academia, the exit strategy, collaboration with experimentalists, synergy between research themes, and the corporate identity model for presentations and report.



- ⬆ **Hvorfor er det vanlig å være mørkeredd?** ("Why do many people fear the dark?")  
— one of the 26 posters showcased at the poster competition during *Hvorfor Det?* on November 26, 2022.



- ⬆ Hanan Gharayba, a student at the Department of Chemistry, University of Oslo, captivated the audience with a thrilling display of explosions, flashes and bangs.
- ⬆ Audun Skau Hansen presented live simulations that allowed the audience to envision the gradual shrinking down to the quantum world.

## Hvorfor Det?

Department of Chemistry, University of Oslo  
November 26, 2022

**The Hylleraas Centre hosted an event devoted to the presentation of curiosity-driven research carried out by school children from Ila Primary School in Oslo.**

On Saturday, November 26, 2022, the Hylleraas Centre hosted the event *Hvorfor Det?*, organized by Anne Pedersen, Sook Berge Buer and the parents of Ila Primary School in Oslo. *Hvorfor Det?* is a Norwegian non-governmental, non-profit organization dedicated to promoting scientific research by piquing

the curiosity of children. The concept encourages children to employ the scientific method to explore any questions they may have.

This year, more than 60 school children, aged 8 to 12 years, presented 36 posters describing their research at the Department of Chemistry, University of Oslo. Some worked in pairs, while others worked individually. Awards were presented for "Excellent Research" and "Most Entertaining Project" based on evaluations by professional researchers, including Audun Skau Hansen, Håkon Emil Kristiansen, and Thomas Bondo

Pedersen from the Hylleraas Centre. The "Audience Award" was determined by votes more than 150 participants.

Director Trygve Helgaker addressed an engaged audience on quantum mechanics and chemistry. Audun Skau Hansen guided the children and their families through a computer-simulated journey from the macroscopic world to the realm of individual atoms and molecules.

The event was supported by Sparebankstiftelsen, Utdanningsdirektoratet, Tekna, and the Hylleraas Centre.



↑ Prof. Vidar R. Jensen (left) and Dr. Marco Foscato (right)  
at the closing the 2022 NKS meeting at Zander K Hotel.  
*Photo: Trygve Helgaker.*

## 2022 National Meeting of the NKS Division of Quantum Chemistry and Modelling

Zander K Hotel, Bergen  
November 28–29, 2022

**In November 2022, the theory group at the University of Bergen was host to the first National Meeting of the Division of Quantum Chemistry and Modelling of the Norwegian Chemical Society (NKS) after the COVID-19 pandemic.**

The National Meeting of the Division of Quantum Chemistry and Modelling is an important annual event for all theoretical chemists in Norway. The meeting at Zander K Hotel in Bergen November 28–29, 2022 was the first opportunity for the Division to meet in person after the pandemic – the two previous NKS meetings were organized on-line by the Hylleraas Centre, from Tromsø in 2020 and Oslo in 2021.

Apart from three 40-minute keynote lectures delivered by Prof. Carles Bo (ICIQ, Spain), Assoc. Prof. Heather K. Kulik (MIT Chemical Engineering, USA), and

Prof. Veronique Van Speybroeck (University of Ghent, Belgium), there were four 30-minute invited lectures, nine 20-minute oral presentations, and five 10-minute flash presentations. Of the 21 presentations, 9 were given by members of the Hylleraas Centre. More than 35 participants attended the meeting.

The meeting was organized by Prof. Vidar R. Jensen, Dr. Giovanni Occhipinti, and Dr. Marco Foscato at the University of Bergen. The Hylleraas Centre sponsored the event by NOK 20 000.

## Social events in 2022



Hylleraas members enjoying a beer at Valkyrien Restaurant (Valka) after work on Friday, November 25, 2022. From left to right: Håkon Emil Kristiansen, Marco Bortoli, and Xinqing Li.



In a tradition going back to 2009 and the Centre for Theoretical and Computational Chemistry (CTCC), Trygve Helgaker has held an annual summer party with BBQ at his house at Jar, outside Oslo. After a two-year break caused by the pandemic, the 2022 summer party was attended by about 40 centre members and their family, in driving rain. Here André Laestadius is attending to his BBQ sausages.



The Christmas party is a popular event at the Hylleraas Centre in Oslo. After a two-year break due to the pandemic, the party was back on December 9, 2022.



Hylleraas members Audun Skau Hansen and Laurens Peters dancing at RealMoro, a social event organized by the Faculty of Mathematics and Natural Sciences at University of Oslo on November 4, 2022.



# Alumni Talks

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To inspire and help the young researchers at the Hylleraas Centre in planning their future career, the Young Researcher Parliament (YRP) of the centre invites previous members and collaborators to give a short seminar about their career choices and the position they hold today, whether in academia or in industry. During the Hylleraas Friday Seminar on December 9, two alumni presented themselves: dr. Roberto Di Remigio Eikås and dr. Arne Bunkan.



## Dr. Roberto Di Remigio Eikås

EuroCC National Competence Centre Sweden

### Life after Hylleraas

Online seminar from the Hylleraas Centre, UiT  
The Arctic University of Norway

—  
December 9, 2022

Dr. Roberto Di Remigio Eikås carried out his PhD work under the supervision of Luca Frediani at the Hylleraas Centre in Tromsø and defended his thesis *The Polarizable Continuum Model Goes Viral! Extensible, Modular and Sustainable Development of Quantum Mechanical Continuum Solvation Models* in 2017.



## Dr. Arne Bunkan

Norwegian Defence Research Establishment (FFI)

### From Physical Chemistry to an Effective Defence

Online seminar from the Hylleraas Centre,  
University of Oslo

—  
December 9, 2022

Dr. Arne Bunkan was supervised by Prof. Claus Jørgen Nielsen at the predecessor of the Hylleraas Centre, the Centre for Theoretical and Computational Chemistry (CTCC) in Oslo, where he defended his thesis *A Theoretical and Experimental Study of Atmospheric Reactions of Amines and Their Degradation Products* in 2015.



# PhD Defences 2022

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**Candidate:**

**Anders Brakestad**

**University:**

UiT The Arctic University of Norway

**Date:**

February 18, 2022

**Thesis:**

*Applications of Multiwavelets to Energies and Properties*

**Trial Lecture:**

*Simulating the Electronic Structure of Molecules under High Pressures*

**Supervisors:**

Prof. Kathrin Hoppman, UiT The Arctic University of Norway  
Assoc. Prof. Ingar Leiros, UiT The Arctic University of Norway  
Prof. Bjørn Olav Brandsdal, UiT The Arctic University of Norway  
Prof. Luca Fredianai, UiT The Arctic University of Norway

**Committee:**

Prof. Dage Sundholm, University of Helsinki, Finland  
Dr. Florian Bischoff, Humboldt University of Berlin, Germany  
Dr. Magnus Ringholm, UiT The Arctic University of Norway  
Prof. Annette Bayer, UiT The Arctic University of Norway

**Chair:**

**Format:**

hybrid



**Candidate:**

**Håkon Emil Kristiansen**

**University:**

University of Oslo

**Date:**

May 8, 2022

**Thesis:**

*Coupled-cluster Theory for Electron Dynamics*

**Trial Lecture:**

*Machine Learning for Wavefunction-Based Electronic-Structure Methods*

**Supervisors:**

Prof. Thomas Bondo Pedersen, University of Oslo  
Dr. Simen Kvaal, University of Oslo

**Committee:**

Prof. Kenneth Ruud, UiT The Arctic University of Norway  
Assoc. Prof. Eugene DePrince, Florida State University, USA  
Prof. Sandra Luber, University of Zurich, Switzerland  
Assoc. Prof. Jonathan Polfus, University of Oslo

**Chair:**

**Format:**

Prof. Harald Walderhaug, University of Oslo  
online



Candidate: **Morten Ledum**

University: University of Oslo  
 Date: June 3, 2022  
 Thesis: *Hamiltonian Hybrid Particle-Field Method for Biological Matter: Efficient Simulation and Machine Learning Approaches*  
 Trial Lecture: *The Application of Artificial Intelligence in Biochemistry and Chemical Physics*  
 Supervisors: Prof. Michele Cascella, University of Oslo  
 Prof. Jürgen Gauss, University of Mainz, Germany  
 Committee: Assoc. Prof. Raffaello Potestio, University of Trento, Italy  
 Assoc. Prof. Matteo Dal Peraro, EPFL, Switzerland  
 Assoc. Prof. Hanne Røberg-Larsen, University of Oslo  
 Chair: Prof. Harald Walderaug, University of Oslo  
 Format: physical



Candidate: **Karolina Di Remigio Eikås**

University: UiT The Arctic University of Norway  
 Date: December 16, 2022  
 Thesis: *A Computational Study of Cyclic Peptides with Vibrational Circular Dichroism Techniques for the Determination of Absolute Configurations*  
 Trial Lecture:  
 Supervisors: Prof. Kenneth Ruud, UiT The Arctic University of Norway  
 Professor Bjørn Olav Brandsdal, UiT The Arctic University of Norway  
 Senior Lecturer Maarten Beerepoot, UiT The Arctic University of Norway  
 Prof. John Sigurd Mjøen Svendsen, UiT The Arctic University of Norway  
 Prof. Petr Bour, Czech Academy of Sciences  
 Committee: Prof. Malgorzata Biczysko, Shanghai University, China  
 Prof. Christian Merten, Ruhr University Bochum, Germany  
 Associate Prof. Johan M. Isaksson, UiT The Arctic University of Norway  
 Chair: Prof. Annette Bayer, UiT The Arctic University of Norway  
 Format: hybrid



⬆ Celebration at the Hylleraas Centre after the defence of Håkon Kristiansen (with tie) on May 8, 2022.  
 Photo: Trygve Helgaker.



# Visits and Mobility

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There were in total 28 visits and 23 unique visitors to the Hylleraas Centre in 2022.

## Incoming Visits

### Visitors to Oslo

BSc Manuela Leal Nader	University of Sao Paulo, Brazil	January 28 – April 13
MSc Katrin Gugeler	University of Stuttgart, Germany	January 31 – April 29
BSc Aurore Denjean	University of Montpellier, France	February 1 – July 31
MSc Asier Urriolabeitia Rodrigo	University of Zaragoza, Spain	February 2–12
BSc Emiel Vanden Berghe	Ghent University, Belgium	February 21 – June 20
MSc Juliane Heitkämper	University of Stuttgart, Germany	March 21– June 17
MSc Ruslan Lukin	Kazan Federal University, Russia	April 1 – June 30
MSc Yunfei Bai	Haldor Topsoe A/S, Denmark	April 4 – July 8
Dr Markus Penz	MPI Hamburg, Germany	April 8–20
MSc Sri Harsha Pulumati	University of Iceland, Iceland	April 22 – May 11
BSc Maximillian Asbach	University of Würzburg, Germany	May 2 – June 30
Prof Clémence Corminboeuf	EPFL, Switzerland	May 31 – June 1
MSc Lucía Morán González	ICIQ, Tarragona, Spain	August 8 – November 22
Prof Peter Taylor	Tianjin University, China	September 9
MSc Bastian Bjerkem Skjelstad	Hokkaido University, Japan	September 20–27
Prof Pascal Friederich	KIT, Karlsruhe, Germany	October 13–14
Prof Egill Skúlason	University of Iceland, Iceland	December 12–17

### Visitors to Tromsø

Dr Roberto Di Remigio Eikås	ENCCS, Sweden	January 8–30
BSc Morten Lehmann	TUB, Berlin, Germany	February 7 – June 10
Dr Roberto Di Remigio Eikås	ENCCS, Sweden	February 20 – March 13
MSc Jiri Zdrahala	IOCB Prague, Czech Republic	April 1 – June 30
Prof. Johan Åqvist	Uppsala University, Sweden	April 19–22
Dr Roberto Di Remigio Eikås	ENCCS, Sweden	April 20 – May 10
MSc Moumita Das	IOCB Prague, Czech Republic	May 5 – August 9
Prof Clémence Corminboeuf	EPFL, Switzerland	June 1–2
Dr Roberto Di Remigio Eikås	ENCCS, Sweden	June 13–27
MSc Jonas Vester	DTU, Denmark	November 7–11
Prof Johan Åqvist	Uppsala University, Sweden	November 7–11



Visiting Researcher:

# Lucía Morán González

Lucía Morán González is a PhD candidate from the Institute of Chemical Research of Catalonia (ICIQ) in Spain. She visited the Hylleraas Centre in Oslo for three months in 2022, working in the research group of David Balcells.

In Spain, Lucía Morán González works at the Institute of Chemical Research of Catalonia (ICIQ) in Tarragona, where she uses computational modelling to study catalytic processes with transition metals, supervised by Prof. Feliu Maseras. She visited the Hylleraas Centre in Oslo from August 20 to November 23, 2022, working in the group of David Balcells. The purpose of her visit was to expand her knowledge in machine learning – in particular, graph auto-correlations and their use in deep neural networks.

## Why did you choose to visit the Hylleraas Centre and how did you learn about us?

I decided to join the Hylleraas Centre because I knew about some of the projects carried out at the centre and I was keen on learning from these projects. My PhD supervisor had worked with some of the group leaders at the Hylleraas Centre, so I was aware of their research. I have always followed the Twitter account of the Hylleraas Centre and the work performed there.

## What was the impact of your research stay at the Hylleraas Centre on your scientific activities?

During my research stay, I worked on a project employing computational tools that I had hardly ever used before. I would like to transfer this rewarding experience to future research projects. Moreover, the seminars and group meetings inspired me to learn about different topics.

## What do you think about the social and scientific environment of the Hylleraas Centre? Were you able to participate in any activities?

Friendly and easy. Everyone is willing to help with kindness and respect. I participated in several activities, such as the Hylleraas Annual Meeting at Klækken and the dinner with the visiting speaker Dr. Pascal Friederich and colleagues. I would like to emphasize the waffle day for the whole chemistry department.

## How did you like Norway in general and Oslo in particular?

Norway has always caught my attention, and this was the perfect opportunity to visit it. During my months in Oslo, I was impressed by the organization and the convenience of the city. I loved taking public transport and being in the countryside in a matter of minutes.

## Can you mention a highlight of your stay?

I met amazing people and enjoyed the culture, but if there is one thing that I must highlight it is the proximity of the forest to the city. I loved hiking and it took me only 15 minutes to get lost in nature. I usually hiked up Vettakollen hill to enjoy the incredible views of the city and the fjord.

➔ Visiting PhD candidate Lucía Morán González at Holmenkollen in Oslo in late September 2022.





Visiting Researcher:

# Morten Lehmann

Morten Lehmann is a PhD candidate from the Berlin Institute of Technology (TU Berlin) in Germany. He visited the Hylleraas Centre in Tromsø from February to June 2022, working in the group of Michal Repisky.

Morten Lehmann carried out his ERASMUS+ Traineeship Programme in the group of Dr. Michal Repisky at the Hylleraas Centre from February to June 2022. He is a group member of Prof. Martin Kaupp at TU Berlin, who is a long-term collaborator with members of the Hylleraas Centre. In Tromsø, Morten applied a newly developed atomic-mean-field exact two-component (amfX2C) Hamiltonian to the calculation of EPR and paramagnetic-NMR parameters.

## Why did you choose to visit the Hylleraas Centre and how did you learn about us?

I always wanted to come to Norway for a research stay. The Hylleraas Centre offered me the perfect opportunity to fulfil this wish and to combine it with an excellent research environment in quantum chemistry. My master's degree supervisor knew one PI of the Hylleraas Centre and established the cooperation.

## What was the impact of your research stay at the Hylleraas Centre on your scientific activities?

Since the research stay was part of my master's project, it had a huge impact on my academic education. I learnt a lot about working in an international environment and had the opportunity to work on an interesting topic. Finally, that led to my decision to continue as a PhD student.

## What do you think about the social and scientific environment of the Hylleraas Centre? Were you able to participate in any activities?

It is a great advantage of the centre to have different groups with diverse research interests that are, nevertheless, all within the field of theoretical chemistry. The social environment was very pleasant. There were a lot of social activities.

## How did you like Norway in general and Tromsø in particular?

I know Norway from many summer holidays I spent there, and I was always fascinated by it. Living in Norway confirmed that. Having grown up in Berlin, Tromsø is a nice contrast. You never need much time to reach any place and you often have a good view of the surrounding nature. Experiencing the city from winter with ice and snow to summer was fantastic.

## Can you mention a highlight of your stay?

Walking through a completely empty and quiet city at 2:30 am after having seen the midnight sun from Fjellstua with a beautiful view of Tromsøya.

➔ PhD candidate Morten Lehmann of Berlin Institute of Technology (TU Berlin), Germany, visited the Hylleraas Centre from February to June 2022.





# Externally Funded Projects

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The financial support the Hylleraas Centre receives as a Centre of Excellence from the Research Council of Norway and from its host institutions constitutes a long-term secure funding allowing us to develop and pursue research projects that require a sustained effort over years. However, to reach the ambitious goals of the centre, additional funding is needed—to focus on particular research challenges and to pursue promising new research directions that may arise in the course of the work at the centre. In the following, we present the external research grant proposals that were approved for funding in 2022. Progress in these projects will in the future be reported as part of the activities of the research themes.



## Rationally-Designed Turbo Reagents for Innovative Organometallics (RATIO)

Dr. Marco Bortoli (UiO)

—

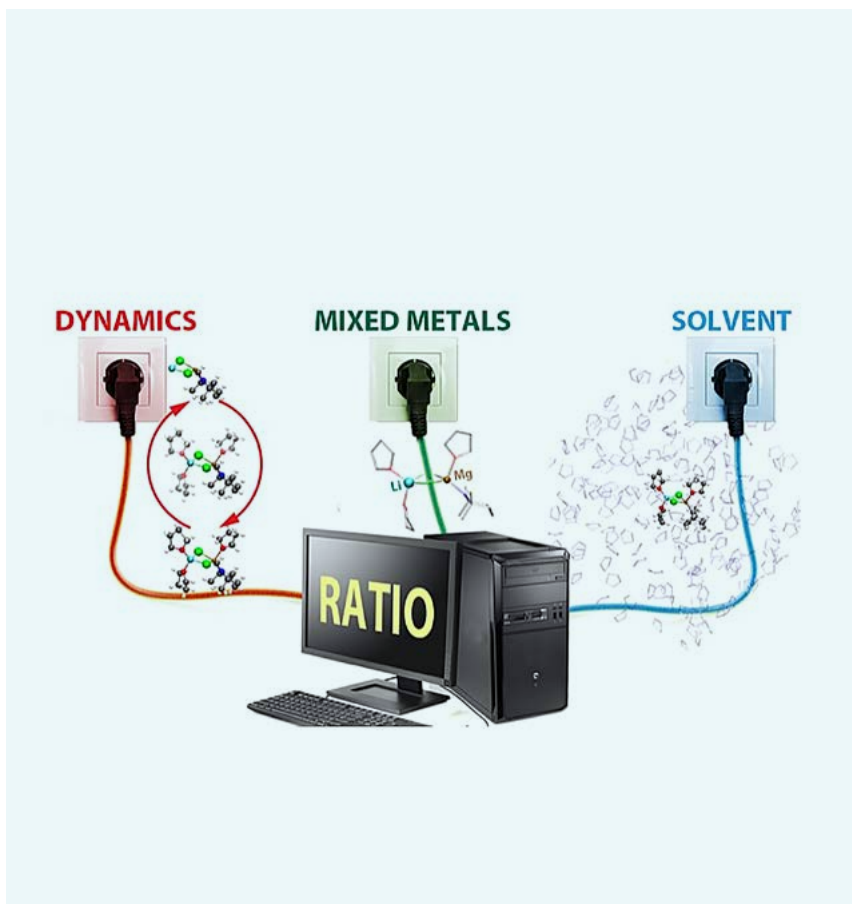
Horizon Europe Marie Skłodowska-Curie Action (MSCA)  
Individual Fellowship  
2022–2025

Dr. Marco Bortoli from the University of Girona, Spain, secured in 2022 a Horizon Europe MSCA-Individual Fellowship grant with the project *Rationally-designed Turbo reagents for Innovative Organometallics (RATIO)* to work at the Hylleraas Centre in Oslo.

—

The RATIO project will explore the chemistry of magnesium-based organometallic compounds and their synergy with lithium salts using *ab initio* molecular dynamics and multi-scale modelling. The aim of the project is to understand the chemistry of main-group organometallic compounds, providing a route towards cheap, green, nontoxic reagents.

The project will be hosted at the Hylleraas Centre under the shared supervision of Michele Cascella and Odile Eisenstein, in collaboration with the experimental group of Prof. Eva Hevia at the University of Bern, Switzerland.





# Research Theme Activity Reports

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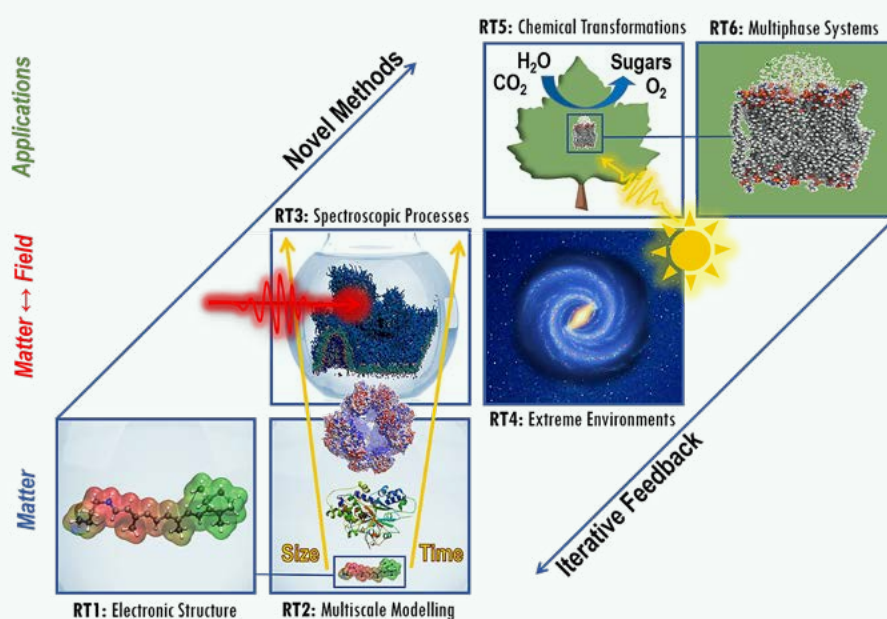


At the Hylleraas Centre, we develop and apply computational methods to understand, interpret, and predict new chemistry, physics, and biology of molecules in complex and extreme environments. Our work is organized into six research themes (RTs):

- RT1 Electronic Structure
- RT2 Multiscale Modelling
- RT3 Spectroscopic Processes
- RT4 Extreme Environments
- RT5 Chemical Transformations
- RT6 Multiphase Systems

Each RT has its own deliverables, directed towards the common goal of the centre. The first two themes, RT1 on electronic structure and RT2 on multiscale modelling, describe matter in isolation, providing the backbone for our work. The next two themes, RT3 and RT4, introduce fields and photons. They are at the heart of the centre, dealing with spectroscopic processes and extreme environments. The tools developed in these four RTs will enable our application themes to address problems that are today beyond the reach of computation. We focus on chemical transformations in RT5 and on multiphase systems in RT6, areas that, among many other things, aim to secure clean energy and to combat antimicrobial drug resistance.

In the following, the 2022 report is provided for each RT.





RT1

## Electronic-Structure Theory

Principal investigators:  
Thomas Bondo Pedersen and  
Luca Frediani

RT1 is the research theme most directly connected to the laws of quantum physics and its implications for the electronic structure of atoms and molecules and their properties. Although those laws are reasonably well understood at a fundamental level, their application can be challenging for molecular systems because of the large number of degrees of freedom of a molecule. The task of RT1 is to develop efficient methods to obtain an accurate and precise representation of the electronic structure of atoms and molecules, which in turn serve as a basis for the work in the more applied RTs.

In 2022, work in RT1 involved multi-reference methods, density-functional theory (DFT), multiwavelet methods, partitioning strategies, and relativity.

### Multireference coupled-cluster theory

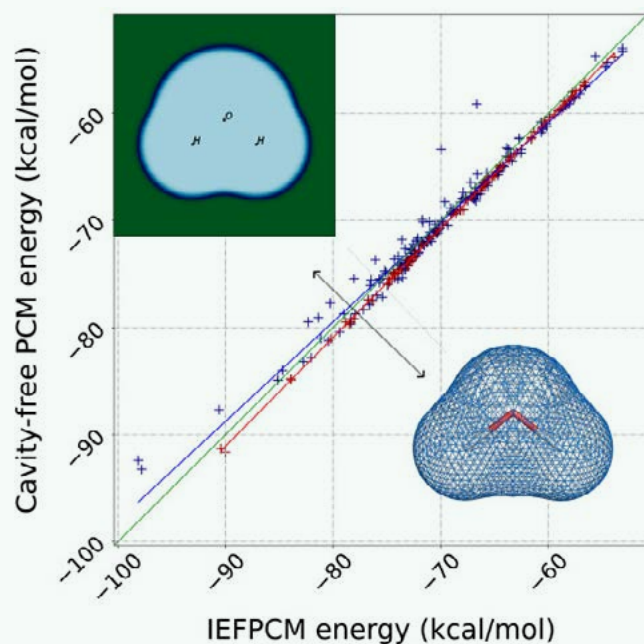
A grand challenge of electronic-structure theory is the multireference problem. In 2022, Dr. Simen Kvaal initiated a collaboration with Prof. Reinhold Schneider (TU Berlin) on a state-universal multi-reference coupled-cluster method, in

which a state-universal bivariational principle guides the development of the working equations. A pilot code will be implemented in the Hylleraas Software Platform.

### Density-functional theory

In 2022, Andrew Teale and Trygve Helgaker, together with Prof. Andreas Savin (UPMC Sorbonne Université), initiated a highly unconventional collaborative writing effort, involving 70 international experts on DFT, including Simen Kvaal, Andre Laestadius, and Erik Tellgren at the Hylleraas Centre, in addition to Teale and Helgaker. The round-table discussion paper *DFT exchange: sharing perspectives on the workhorse of quantum chemistry and materials science* was published in the journal *Physical Chemistry Chemical Physics*, volume 22 and chosen as a HOT article [1]; see section 2023 in Brief.

Members of the Hylleraas Centre have pioneered Moreau-Yosida regulariza-



⬆ **Figure 1:** Correlation between solvation energies computed with the standard surface polarization method and the volume polarization model implemented in MRChem. Reproduced with permission from Ref. [6].

tion of exact DFT. Moreau–Yosida regularization is a mathematical technique to transform convex optimization problems, introducing beneficial smoothness properties, while at the same time being reversible, meaning that no information is lost. Simen Kvaal was invited to contribute a chapter on this topic to a Springer volume on mathematical DFT [2], which describes the state-of-the-art in a mathematically rigorous manner, including novel developments.

Likewise, the use of the Lieb variation principle for *ab initio* calculation of the universal density functional for molecules has been pioneered by members of the Hylleraas Centre. In recognition of their work in this area, Trygve Helgaker and Andrew Teale were invited to contribute a chapter, *Lieb variation principle in density-functional theory*, to *The Physics and Mathematics of Elliot Lieb*, a two-volume Festschrift dedicated to Elliot Lieb on the occasion of his 90<sup>th</sup> birthday [3].

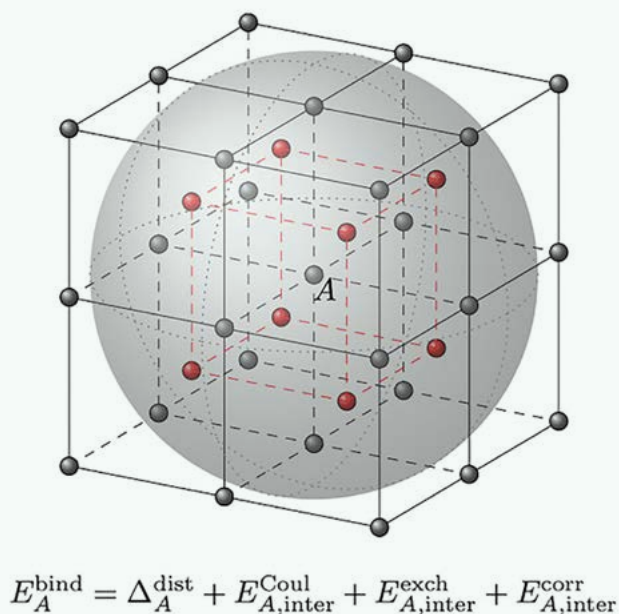
### Multiwavelets

The Tromsø node has been active in the development and application of multiwavelet methods for DFT. Magnar Bjørgve, Luca Frediani and Dr. Stig Rune Jensen have extended the functionality of the VAMPyR code leading to the publication of the kinetic-energy-free implementation of DFT: we have shown that the explicit evaluation of the kinetic energy, which can be problematic in a multiwavelet implementation, can be altogether avoided in a self-consistent-field implementation [4]. Dr. Peter Wind and Jensen have finalized the parallel implementation of MRChem [5], which shows excellent scaling combined with unmatched precision for large molecules. Gabriel Gerez, Dr. Roberto Di Remigio and Frediani have published the implementation of the continuum solvent effect, based on the generalized Poisson equation, which fully accounts for volume polarization effects [6]; see Figure 1. We have officially released the version 1.1 of MRChem, which include, among

the other things, a parallel implementation.

Several new activities have started or continued in 2022. Among them are a study of intramolecular basis-set superposition error in collaboration with Prof. Frank Jensen (Århus University, Denmark), implementation of an excited-state solver together with Prof. Patrick Norman (KTH, Sweden), by making use of the OpenRSP formalism, further development of the relativistic ZORA implementation with Prof. Kathrin Hopmann (UiT) and Dr. Andris Gulans (Univ of Latvia), and a prototype implementation of the four-component relativistic formalism using our Python front-end VAMPyR, originally developed by Magnar Bjørgve.

The group has acquired two new members: Quentin Pitteloud, who started as PhD candidate in October, and Dr. Christian Tantardini, who began a post-doctoral fellowship in April.



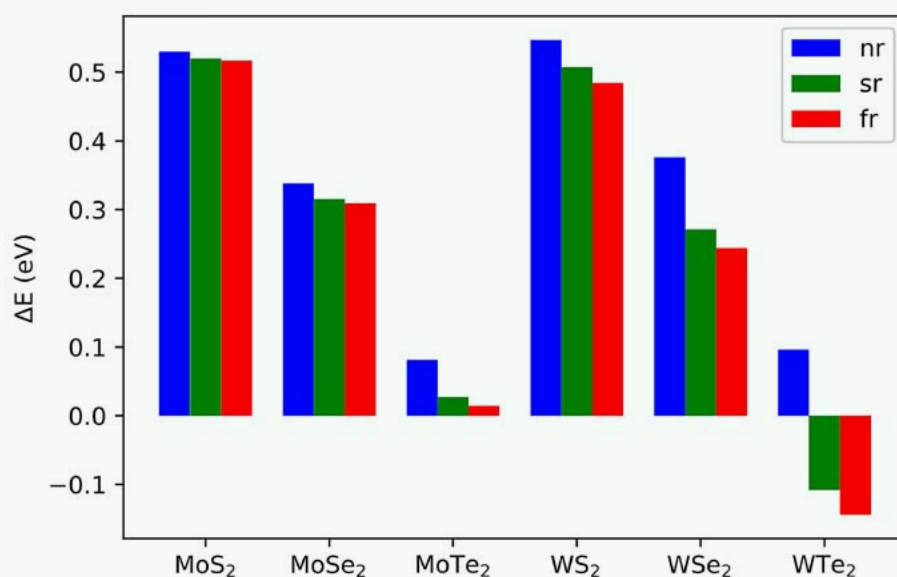
⬆ **Figure 2:** The PATMOS model and its description of bonding in terms of energy contributions. *Reproduced with permission from Ref. [7].*

### PATMOS model

Hylleraas members Prof. Inge Røeggen and Dr. Bin Gao have developed a computationally tractable model, the PATMOS (Perturbed AToms in MOlecules and Solids) model, for the study of electronic structure of pure metals using localized orbitals [7]; see Figure 2. In the PATMOS model, an unrestricted Hartree-Fock wave function is required as a root function in order to have a proper localized picture of bonding. For all except the shortest distances, the inter-atomic correlation energy is the most important component of the calculated binding energy. The calculated cohesive energy for lithium is in fair agreement with the experimental result.

### Two-dimensional materials

Two-dimensional (2D) materials showcase a plethora of interesting properties, many of which are attributed to relativistic spin-orbit coupling. Hylleraas members Marius Kadek, Marc Joosten, Michal Repisky, and Kenneth Ruud, together with colleagues from Northeastern University in Boston (USA), have examined the relativistic band structure as well as Rashba splitting and  $Z_2$  topological invariant of transition-metal dichalcogenides (TMDs), using a fully relativistic all-electron solid-state DFT code based on the Dirac Hamiltonian and Gaussian-type orbitals (GTOs) [8]. Being based on GTOs, the presented methodology can eliminate the need for pseudopotentials and



**Figure 3:** Relative total energy of the 1T' phase of TMD monolayers calculated with respect to the 2H phase. The results were obtained for nonrelativistic (nr), scalar relativistic (sr), and four-component relativistic (fr) Hamiltonians and show the relativistic effects on the structural stability. Reproduced with permission from Ref. [8].

facilitate exploration of all electronic states within a unified framework. Additionally, the authors offer the first systematic studies on the convergence of GTO basis sets for periodic solids at both nonrelativistic and relativistic theoretical levels. For  $\text{WTe}_2$ , the authors demonstrate that the change in what is the most stable phase (distorted tetragonal, 1T') rather than the hexagonal phase (the most stable for other similar 2D materials) is a relativity-driven scalar relativistic effect (Figure 3). Whereas other methods can find the 1T' phase to be the most stable, most of these would not be able to pin-point it to be due to relativity.

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RT2

## Multiscale Modelling

Principal investigators:

Michele Cascella and Bjørn Olav Brandsdal

RT2 is devoted to the development of computational methods from quantum-mechanical studies of small systems to simulations on the mesoscale and to build algorithmic bridges across multiple resolutions. Work in RT2 is in particular focused on development of *ab initio* molecular dynamics (MD) and quantum-mechanics/molecular-mechanics (QM/MM) embedding protocols, as well as coarse-grained methods coupling particle resolution to density-functional-based potentials.

Past and ongoing computational development in RT2 aims to set the basis for the multiscale modelling component of the Hylleraas Software Platform. In this respect, we describe here two computational modules that were initiated/implemented in 2022: REACT and HylleraasMD.

### Multiscale modelling for the Hylleraas Software Platform: REACT

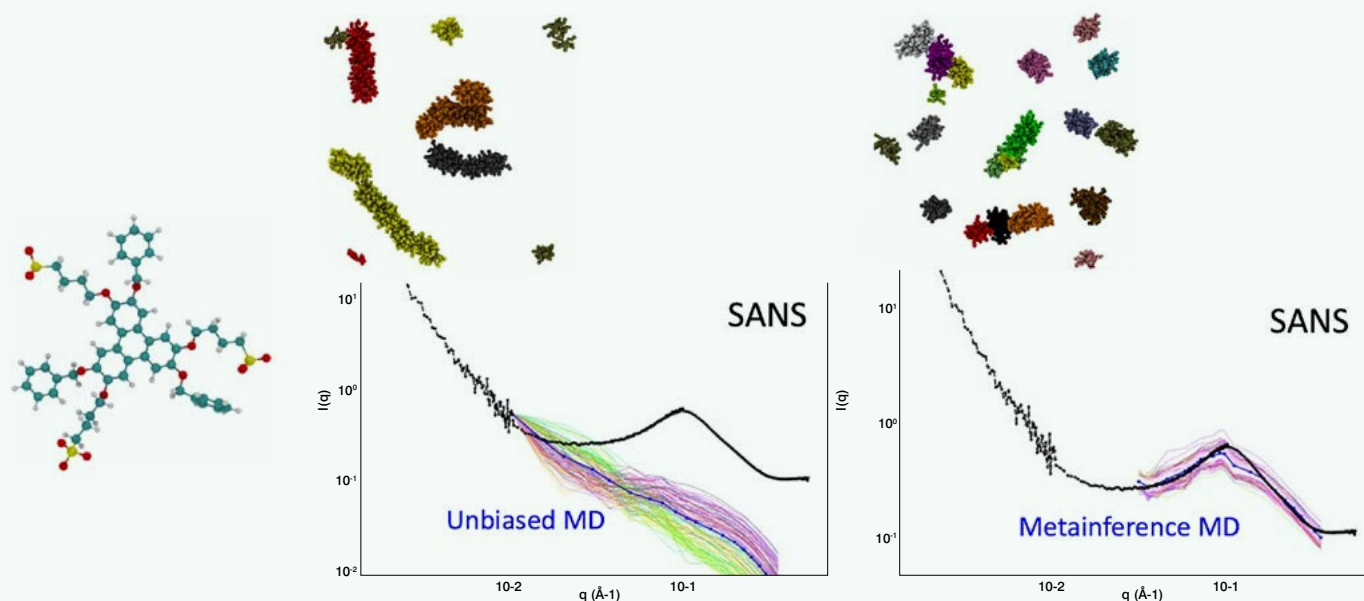
The REACT code [1] has been developed to aid the parameterization of empirical-valence-bond (EVB) simulations. EVB calculations require that we have a suitable reference reaction (uncatalysed reaction) for which the reaction free-energy difference and activation free energy is known. These values are typically obtained from density-functional-theory (DFT) calculations on cluster models.

REACT is primarily aimed at relative-energy calculations using the quantum-mechanical/DFT (QM/DFT) approach. It offers its users a new way of managing

computational projects in a structured manner, with great flexibility and efficiency. REACT keeps track of the many different geometry, input and output files generated for every stationary point of a given reaction pathway.

Creating cluster models is not straightforward – it is time consuming and prone to human error. REACT uses the open source PyMol code to simplify the preparation of protein cluster models of any size for DFT calculations. Analysis of results is enabled through automatic relative-energy calculations with options to include typical correctional terms for solvation, dispersion and vibrations. Furthermore, results can be graphically represented through modern plotting functionalities, including energy diagrams, self-consistent-field convergence plots, and frequency IR spectrum plots. REACT connects all its project files to open source PyMol where not only stationary points can be visualized, but also animations of vibrations from frequency calculations





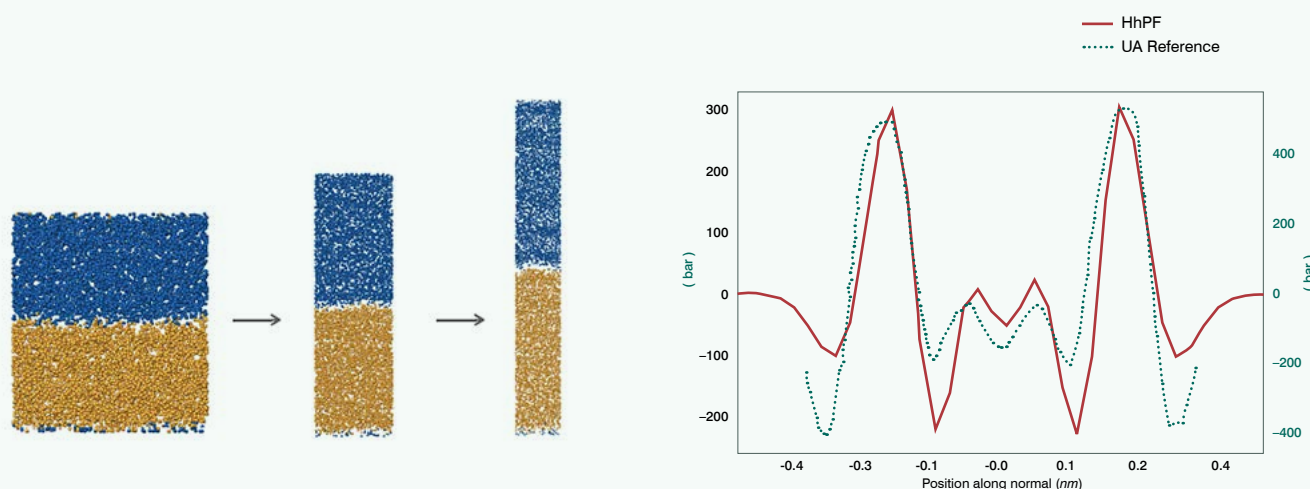
⬆ **Figure 2:** Unbiased MD of a coarse-grained model of anionic surfactant (left) promotes excessive molecular aggregation, in disagreement with the experiment (centre). Metainference simulations (right) reproduce the main feature of the SANS spectrum indicating less aggregation than originally predicted. *Illustration: Dr. Henrique Musseli Cezar.*

### Pressure calculations in the HhPF formalism

The determination of a full anisotropic pressure tensor is not possible in standard hPF simulations. Such simulations are therefore incapable of determining surface tension and hence cannot predict interfacial phenomena under mechanical stress. In 2020, we introduced into the interaction-energy functional a square-gradient term that accounts for the presence of the interface between two species and showed that, by a suitable calibration, it became possible to reproduce surface properties in a qualitative manner – in particular, we obtained tensionless lipid bilayers with the correct area per lipid. Unfortunately, calibration to reproduce the correct area per lipid of a DPPC bilayer gave a too stiff membrane, with an area compressibility two orders of magnitude higher than in all-atom models.

Having developed the HhPF model and implemented it in the HyMD code, we returned to the problem of pressure calculations and obtained an explicit tensorial contribution to the pressure without introducing a square-gradient term. With the use of a Gaussian filter, the tensorial contribution becomes proportional to the Laplacian of the densities, intrinsically producing non-negligible anisotropic pressure contributions at interfaces (Figure 3).

Having implemented the HhPF pressure tensor in the HyMD code, we tested if it was able to reproduce the structural properties of DPPC at 1 bar pressure. We first performed a recalibration of the parameters by our Bayesian optimization protocol [4], in particular introducing the target area per lipid into the penalty function as well as the lateral density profiles. The best parameter sets were then analysed with respect to their ability to predict some not-learned properties – namely, lat-



**Figure 3:** Left: In HhPF simulations at NPT conditions, a bi-phase system tries to reduce its surface, favouring a capillary setup. Right: comparison of lateral pressure profiles in a DPPC bilayer, compared to reference united-atom simulations by Lindahl and Edholm (*J. Chem. Phys.*, 2000, **113**, 3882), indicating less aggregation than originally predicted. Adapted from Ref. [5], under CC 4.0 license.

eral pressure profiles and area compressibility and bending modulus. We found that, within the HhPF scheme, it is possible to obtain parameters that both fit the learned properties excellently and show very satisfactory transferability to other properties.

This striking improvement in the behaviour at constant pressure, without having modified the energy functional, is the clearest example to date of the superiority of the HhPF model vis-a-vis the standard hPF model in describing the correct physics of soft systems. A manuscript describing the constant-pressure calculations with the HhPF models is in preparation [5].

#### Funding / Personnel

In February 2022, Dr. Henrique Musseli Cezar started his contract as postdoc to work on further development of the Hylleraas MD and its interfacing with the Hylleraas Platform.

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## RT3

# Spectroscopic Processes

Principal investigators:  
Thomas Bondo Pedersen  
and Michal Repisky

**Laser-driven quantum dynamics with and without the Born–Oppenheimer approximation and the development and application of (relativistic) density-functional theory (DFT) have been central research topics in RT3 in 2022.**

In 2022, we have in RT3 studied the emergence of molecular alignment in a strong laser pulse, developed a new approach to solving the time-dependent Schrödinger equation, proposed accurate yet efficient algorithms for extracting high-order hyperpolarizabilities and linear absorption spectra from electron dynamics simulations, and formulated a theory for attosecond transient absorption spectroscopy without multipole expansions. In addition, we have developed simple yet accurate two-component Hamiltonian models that offer  $10^{-5}$  hartree accuracy compared with reference four-component data and extended those models to conventional X-ray absorption and transient

absorption spectroscopies. Finally, investigations of molecular properties in complex environments resulted in the first vibrational circular dichroism experiment on nucleoside crystals with two-orders-of-magnitude signal enhancement.

Part of the work in RT3 was carried out within the framework of the project *Attosecond Quantum Dynamics Beyond the Born–Oppenheimer Approximation* at the Centre for Advanced Study (CAS), as described elsewhere in this Annual Report.

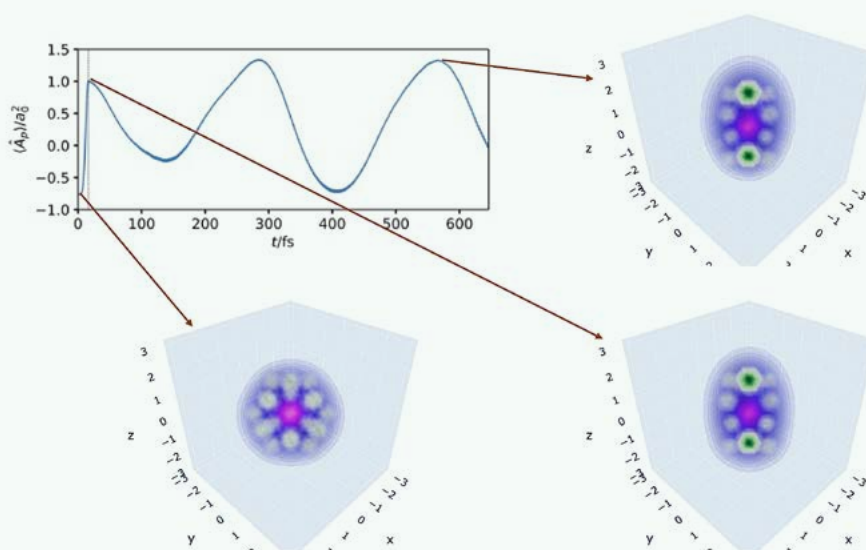
### Molecular alignment in a strong laser pulse

The important chemical concept of molecular structure is not well-defined in quantum mechanics without the Born–Oppenheimer approximation. Laser-induced molecular alignment, however, is very well understood within Born–Oppenheimer-based molecular quantum mechanics [1] and is a widely used technique to lift the veil of random orientation in high-accuracy experimen-

tal attosecond spectroscopy. As described in the Annual Report for 2021, the CAS project investigated the alignment phenomenon using non-Born–Oppenheimer quantum dynamics, where the fully coupled molecular wave function is expanded in explicitly correlated Gaussian basis functions [2]. Finalized in spring 2022 and published in *The Journal of Chemical Physics* [3], this work shows that alignment indeed emerges without the Born–Oppenheimer approximation, including the experimentally observed field-free (post-pulse) periodic alignment pattern caused by quantum interference; see Figure 1.

### Ionization and dissociation continua

Another major component of the CAS project in 2022 has been the theoretical description of the continuous part of the spectrum of the full molecular Hamiltonian operator—that is, ionization and dissociation continua. A physically correct description of these continua is mandatory for theoretical modelling of attosecond experiments, where ionization processes are virtu-



**Figure 1:** Alignment of the HD molecule using non-Born–Oppenheimer simulations with a 16 fs laser pulse. The internal quantum dynamics are described in a coordinate system with the deuteron at the origin. The curve (top left) shows the time evolution of the expectation value of the protonic operator  $z_p^2 - z_p^2 - y_p^2$ , which measures the degree of alignment of the deuteron–proton axis relative to the laser polarization axis, the z-axis, such that maxima correspond to alignment. The contour plots show the proton and electron densities at different times, using blue–purple colours for the electron density and green for the proton density. It is evident that the initial, roughly spherical shape of the molecule turns into an essentially aligned linear shape at the end of the laser pulse, at 16 fs. After the pulse is turned off, alignment periodically revives due to quantum interference. This phenomenon is well known experimentally and has now, for the first time, been confirmed theoretically using the full nonrelativistic molecular Schrödinger equation.

*Illustration: Thomas Bondo Pedersen.*

ally unavoidable due to the high laser intensity and the broadness of the laser frequency distribution. While mesh-based methods can provide an essentially exact description, they are too computationally demanding for routine use for larger systems. Complex Gaussians, with all nonlinear parameters (width, centre, and momentum) treated as time-dependent variables, form an overcomplete set with exceptional adaptivity and, thus, provide an attractive alternative to mesh-based methods. Unfortunately, the equations of motion derived from the Dirac–Frenkel variation principle for the nonlinear parameters quickly become numerically unstable as the number of Gaussians is increased. Much effort has been spent on (1) an in-depth numerical analysis

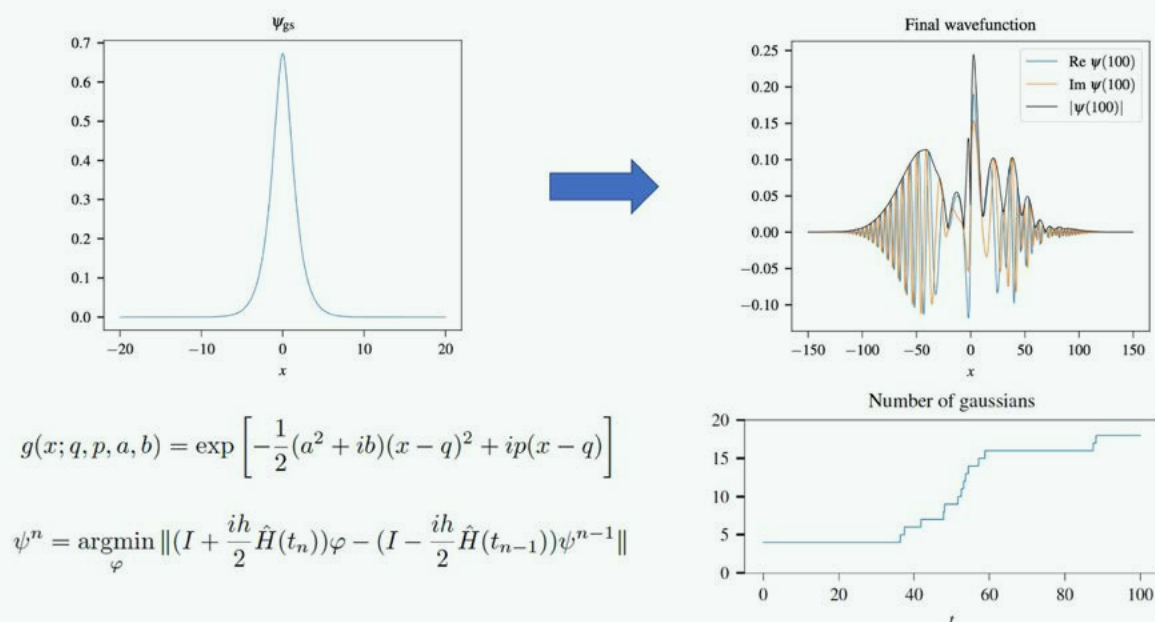
of the observed instabilities, and (2) an alternative discretization of the time-dependent variation principle that provides numerical stability. Using a one-dimensional hydrogenic model system exposed to an extreme laser pulse that rapidly induce ionization, very promising results have been achieved [4]; see Figure 2.

#### Laser-driven many-electron dynamics

The development of coupled-cluster methods for the description of laser-driven many-electron dynamics within the Born–Oppenheimer approximation remains a central goal of RT3. The CAS work on complex Gaussians will be useful also in this context, paving the way for accurate yet affordable simulations of attosecond spectroscopies.

In 2022, however, we have focused on the extraction of linear and nonlinear optical properties from simulations using artificial laser fields and on the theory of attosecond transient-absorption spectroscopy using the exact semi-classical interaction operator without multipole expansions.

Linear absorption spectra can be obtained from simulations of molecular electronic systems exposed to an infinitely short electric-field kick by Fourier transformation of the induced electric-dipole moment. Such electric-field kicks are artificial, since they do not correspond to actual experimental setups, but the resulting spectrum contains *all* electric-dipole allowed transitions from the ground state, including



⬆ **Figure 2:** One-dimensional regularized model of the hydrogen atom exposed to an extreme laser pulse using adaptive, fully-flexible Gaussians. The initial wave function represents the ground state and is well localized at the origin, the position of the proton. After interaction with the laser pulse, the wave function spreads over several hundred bohrs, showing rapid oscillations characteristic of ionization processes. While the initial wave function can be represented accurately with a few Gaussians, the final wave function can be described with just 18 Gaussians. Throughout the dynamics, the wave function is virtually indistinguishable from the essentially exact wave function computed on a large, dense spatial grid using the split-step Fourier method.  
*Illustrations: Simen Kvaal and Thomas Bondo Pedersen.*

the high-lying core excitations studied in X-ray absorption spectroscopy. This approach has the potential to be more efficient than traditional time-independent methods such as response theory. One of the main computational challenges is the long simulation times required to obtain sufficient resolution in the resulting absorption spectra. As described in the 2021 Annual Report, the master thesis of Eirill Strand Hauge [5] contains an algorithm for extrapolation of the induced dipole moment, accelerating simulations by roughly an order of magnitude. Although Hauge joined the Simula Research Laboratory as a PhD student after obtaining her MSc degree in late 2021, she has in 2022 improved the extrapolation algorithm using low-pass filtering of the dipole signal in collaboration with Pedersen, Repisky, Dr. Håkon Kristiansen, Dr. Marius Kadek, and Dr. Lukas Konecny.

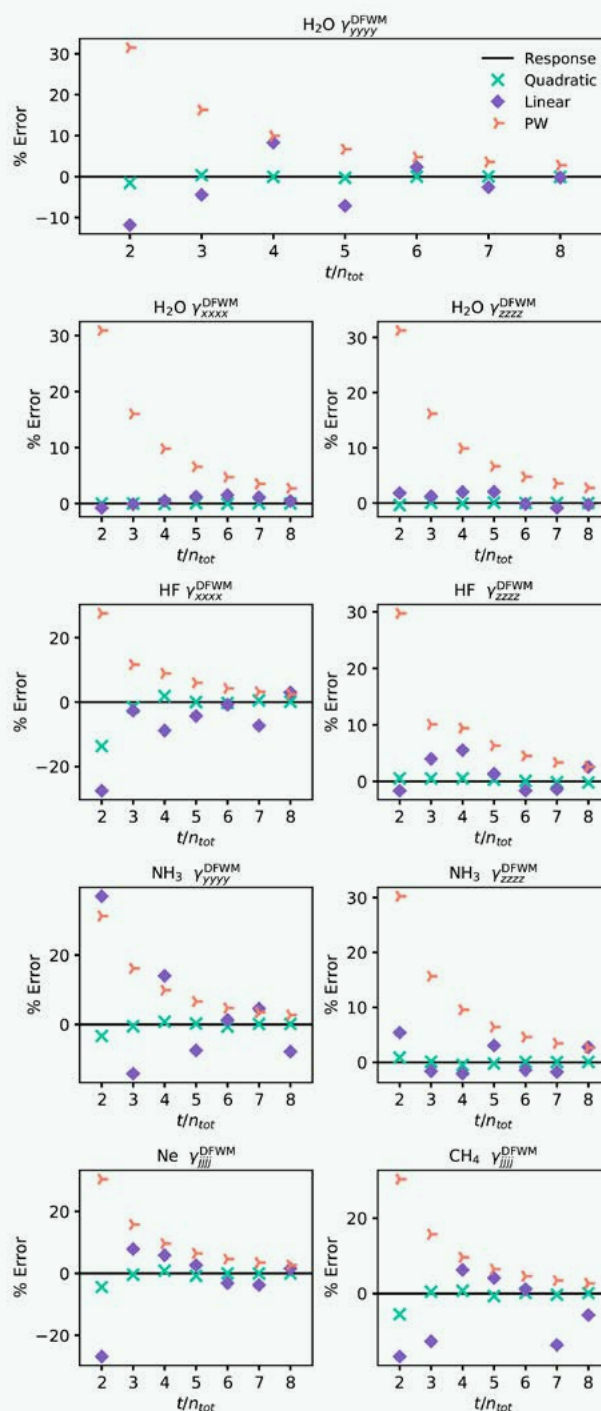
A manuscript on the dipole extrapolation algorithm is in preparation and will be submitted in early 2023.

Frequency-dependent polarizabilities and hyperpolarizabilities can also be extracted from the dipole moment induced by laser fields, either continuous monochromatic waves or finite laser pulses. For the continuous wave approach, one needs to ramp the electric field from zero to full strength in a manner that emulates the adiabatic switching-on assumption of perturbation theory. Traditionally, this has been done using a simple, linear ramp lasting for a single optical cycle [6]. This linear-ramp continuous-wave (LRCW) approach gives pronounced nonadiabatic effects in the induced dipole responses in the time domain, leading to large relative errors in the predicted hyperpolarizabilities that increase by an order

of magnitude at each order. For the pulsed-wave (PW) approach, one needs to filter the time-domain dipole responses around the frequency components of interest, which requires relatively long simulation times to converge to the correct hyperpolarizability. In 2022, in collaboration with Pedersen, Kvaal, and Kristiansen, PhD student Benedicte Ofstad developed an alternative to the LRCW approach based on a quadratic ramping function—the quadratic-ramp continuous-wave (QRCW) approach, which very efficiently eliminates non-adiabatic effects and produces accurate hyperpolarizabilities. An example is shown in Figure 3 for the second hyperpolarizability component corresponding to degenerate four-wave mixing (DFWM). It has been demonstrated that the QRCW approach is more efficient than the LRCW and PW approaches and that it can be used to reliably



→ **Figure 3:** Second hyperpolarizabilities extracted from time-dependent second-order coupled-cluster (TDCC2) simulations using the LRCW and QRCW approaches with  $n_{\text{tot}}-1$  ramping cycles and using the PW approach. The quadratic ramping yields more accurate results at a significantly lower computational cost. *Illustration: Benedicte Ofstad.*

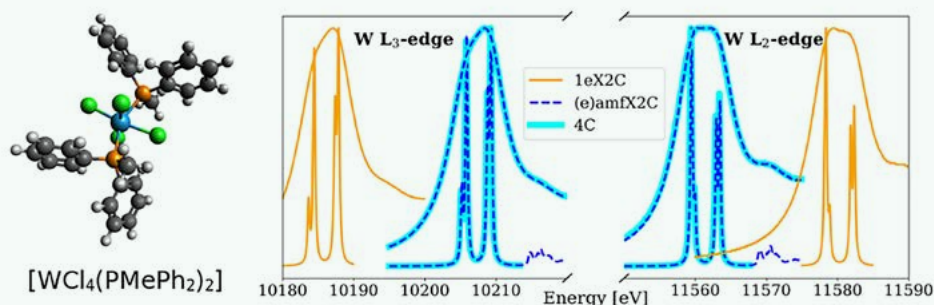


extract up to the fourth hyperpolarizability, which describes five-photon processes governed by quintic response theory. A manuscript is in preparation and will be submitted for publication shortly.

The work described above is based on the electric-dipole approximation for the semi-classical matter-field interaction operator. In a time-dependent, nonperturbative theory, however, there is no computational penalty associated with going beyond the electric-dipole approximation, thus including weak but potentially important dynamics induced by dipole-forbidden interactions among stationary states. As discussed in some detail in the 2021 Annual Report, the exact semi-classical interaction operator introduces several theoretical challenges related, among other things, to gauge invariance. In 2022, PhD stu-

**(e)amfX2C TDDFT for XAS**

- 7-fold speed-up wrt 4c reference
  - includes scalar relativistic effects, spin-orbit coupling
  - includes 2-electron picture-change corrections
  - reproduces 4c reference
- 1eX2C ✗ (e)amfX2C ✓



⬆ **Figure 4:** Tungsten L2,3-edge X-ray absorption spectra of  $[\text{WCl}_4(\text{PMePh}_2)_2]$  calculated using TDDFT at various relativistic levels of theory: conventional one-electron X2C (1eX2C) in orange, (extended) atomic mean-field X2C ((e)amfX2C) in dashed blue, and full four-component (4c) in cyan. *Illustration: Lukas Konecny.*

dent Einar Aurbakken, in collaboration with Pedersen, Kvaal, and Kristiansen, has reformulated the theory of attosecond transient-absorption spectroscopy [7] in terms of the power operator arising from classical electrodynamics, and implemented it for coupled-cluster simulations of time-resolved X-ray absorption spectroscopy and circular dichroism spectroscopy.

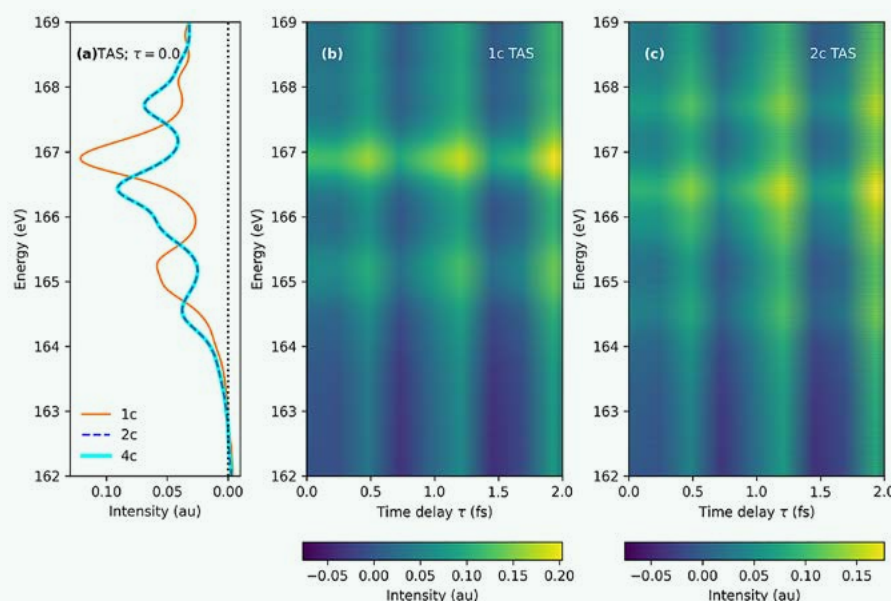
During fall 2022, we began preparing a full open-source release of our software for many-electron dynamics simulations. The software package, Hylleraas Quantum Dynamics (HyQD), will be made public in early 2023 and will be incorporated into the Hylleraas Software Platform, which will provide tools for job management, data management, and visualization.

**Relativistic DFT for laser-driven electron dynamics and molecular properties**

Based on self-consistent-field atomic-mean-field quantities, Michael Repisky, in collaboration with Dr. Stefan Knecht (GSI, Darmstadt), Prof. Hans Jørgen Aa. Jensen (SDU, Odense) and Prof. Trond Saue (CNRS, Toulouse), introduced in 2022 two simple yet computationally efficient and numerically accurate matrix-algebraic approaches to correct scalar-relativistic and spin-orbit two-electron picture-change effects (PCEs) arising within an exact two-component (X2C) Hamiltonian framework [8]. These atomic-mean-field X2C (amfX2C) and extended atomic-mean-field X2C (eamfX2C) models allowed the authors to tailor PCE corrections to the Hartree-Fock and Kohn-Sham mean-field models, in the latter case, also avoiding the need for a point-wise calculation of exchange-correlation PCE corrections.

They assessed the numerical performance of these PCE correction models on spinor energies of group-18 (closed-shell) and group-16 (open-shell) diatomic molecules, achieving a consistent  $10^{-5}$  hartree accuracy relative to four-component reference data. The presented data suggest that the (e)amfX2C PCE models constitute a fundamental milestone toward a universal and reliable relativistic two-component quantum-chemical approach, maintaining the accuracy of the parent four-component approach at a fraction of its computational cost.

X-ray absorption spectroscopy (XAS) has in recent years gained popularity as it probes matter with high spatial and elemental sensitivity. However, the modelling of XAS is difficult since XAS spectra feature a fine structure arising from scalar and spin-orbit relativistic effects – in particular, near



⬆ **Figure 5:** Sulphur L2,3-edge transient absorption spectra (TAS) of thiophene. Pane (a): Spectra with no time delay between pump and probe pulses, employing one-component nonrelativistic Hamiltonian (orange), two-component amfX2C Hamiltonian (blue), and four-component Dirac–Coulomb Hamiltonian (cyan). Panes (b) and (c): Dependence of TAS spectra on the time delay  $\tau$  as obtained with one-component nonrelativistic Hamiltonian (b) and the two-component amfX2C Hamiltonian (c). All spectra were obtained with a damping factor  $\Gamma = 0.01$  atomic units. *Illustration: Torsha Moitra and Michal Repisky.*

L and M absorption edges. While four-component calculations of XAS are nowadays feasible, there is also a need to develop more efficient two-component XAS methods that retain as much as possible the accuracy of the parent four-component approach. To this end, Dr. Lukas Konecny, in collaboration with Repisky, Dr. Stanislav Komorovsky (SAS, Bratislava), Dr. Jan Vicha (Tomas Bata University, Zlin) and Kenneth Ruud extended the (e)amfX2C models to XAS using linear eigenvalue and damped-response time-dependent density-functional theory (TDDFT) [9]. Unlike the commonly used one-electron X2C (1eX2C) Hamiltonian, the (e)amfX2C Hamiltonians account for the scalar and spin-orbit two-electron and exchange-correlation PCEs that arise from the X2C transformation. As demonstrated for L- and M-edge XAS spectra of transition-metal and actinide compounds,

the absence of PCE corrections in the 1eX2C approximation results in a substantial overestimation of spin-orbit splittings, whereas the (e)amfX2C Hamiltonians provided results in excellent agreement with the four-component references for spectral features such as shape, position and spin-orbit splitting, while offering significant computational savings; see Figure 4.

First-principles theoretical modelling of out-of-equilibrium processes observed in attosecond pump-probe transient-absorption spectroscopy (TAS) triggering pure electron dynamics remains difficult, especially for heavy elements and core excitations containing fingerprints of scalar and spin-orbit relativistic effects. To address this, Dr. Torsha Moitra, in collaboration with Repisky, Dr. Lukas Konecny, Dr. Marius Kadek and Prof. Angel Rubio (MPI Hamburg),

formulated and implemented into the ReSpect program a methodology for simulating TAS within a relativistic real-time TDDFT (RT-TDDFT) framework, for core as well as valence energy regimes [10]. In view of the high cost of four-component calculations, efficient but accurate two-component models were also considered. The resulting methodology was applied to study valence and near L2,3-edge TAS processes of experimentally relevant systems. An example is shown in Figure 5 for the TAS spectra of thiophene in the L2,3-edge absorption region, showcasing the need to include relativistic effects for core spectroscopies and the remarkable agreement between the amfX2C two-component and full four-component spectra. In addition, nonequilibrium response theory was extended to the relativistic regime to gain theoretical insights into the



peculiar features of pump-probe TAS [10].

Four-component relativistic treatments of the electron-paramagnetic-resonance (EPR)  $g$ -tensor have so far been limited to the use of a common gauge origin and a restricted-kinetically-balanced basis for the small components. In a work by Michael Repisky, in collaboration with colleagues from the Slovak Academy of Sciences, Debora Misenkov, Florian Lemken, Prof. Jozef Noga, Prof. Olga L. Malkina, and Dr. Stanislav Komorovsky, these limitations were lifted using a distributed-origin scheme based on the use of London atomic orbitals (for gauge-origin independence) and a restricted-magnetically-balanced basis, both proven to be practical approaches for relativistic calculations of other magnetic properties [11]. The authors showed that the gauge dependence of the results is connected to the non-vanishing integral of the current density in a finite orbital basis, which explains why the results of cluster calculations exhibit surprisingly low gauge dependence and why the gauge problem disappears for systems with certain point-group symmetries.

In 2022, Kenneth Ruud, in collaboration with colleagues from University of Warsaw, Katarzyna Jakubowska and Prof. Magdalena Pecul, studied the influence of relativity on the zero-point vibrational (ZPV) corrections to nuclear spin-spin coupling constants by means of the four-component Dirac-Kohn-Sham DFT approach [12]. While the effects of relativity become notable for the ZPV corrections for compounds with lighter

elements such as Se and Ge, for compounds containing heavier atoms such as Bi and Pb, inclusion of relativity becomes crucial and approximating ZPV corrections by the nonrelativistic results may lead to larger errors than omitting the ZPV corrections altogether.

### Molecular spectroscopic processes in complex environments

As a continuation of Kenneth Ruud's collaboration with Dr. Swapan Chakrabarti and co-workers from the University of Calcutta on the study of photo-physical processes, they presented in 2022 a framework based on the time-dependent correlation function for computing the rate of reverse intersystem crossing (RISC) [13]. Using the TXO-TPA and TXO-PhCz molecules in toluene and chloroform solvents as example systems, their work highlights the significant role played by spin-orbit coupling, spin-vibronic (SV) coupling and the influence of the medium in the process. They demonstrated the role of the solvent-assisted SV mechanism towards the net RISC rate constant, which in turn maximizes the efficiency of thermally activated delayed fluorescence.

Postdoctoral fellow Dr. Monika Krupová, in collaboration with Patrycja Leszczenko, Ewa Sierka, Sára Emma Hamplová, Dr. Radek Pelc and Dr. Valery Andrushchenko from IOCB Prague, presented a fast and efficient tool to study chiral crystalline samples and reported the first vibrational-circular-dichroism (VCD) study of nucleoside crystals [14]. The authors observed a two-orders-of-magnitude enhancement of the VCD signal due to the high molecular order

in the crystals leading to supramolecular chirality and high-quality VCD spectra within minutes using small amounts of samples. They showed that VCD is capable of near-instantaneous detection of hydration polymorphs and crystal degradation, which is of substantial interest in pharmaceutical industry.

Dr. Karolina Di Remigio Eikas, who defended her PhD thesis at the end of 2022, presented together with Dr. Maarten T. P. Beerepoot and her supervisor Kenneth Ruud a computational protocol for generating VCD spectra of a series of flexible cyclic oligopeptides of biological/pharmaceutical interest [15]. The method builds on the Conformer-Rotamer Ensemble Sampling Tool (CREST) method in combination with post-optimizations using the B3LYP functional and moderately sized basis sets. Their recommended scheme consists of three steps: (1) conformational sampling with CREST and tight-binding DFT (xTB); (2) energy ranking based on single-point energy calculations as well as geometry optimization and VCD calculations of conformers that are within 2.5 kcal/mol of the most stable conformer using B3LYP/6-31+G\*/CPCM; and (3) VCD spectra generation based on Boltzmann weighting with Gibbs free energies. Somewhat surprisingly, the inclusion of dispersion corrections in the optimization of the conformers led to a poorer agreement with experiment. Further work on the absolute configuration of cyclic peptides using VCD was submitted, in which the limitation of combining experimental and theoretical VCD spectra alone to determine the absolute configuration of cyclic peptide



with multiple stereocentres was explored [16].

One of the most widely studied non-linear light-matter interaction processes is the two-photon-absorption (2PA) phenomenon, in which two photons are simultaneously absorbed, leading to an excitation to a different state (rotational, vibrational, electronic, etc.) of the molecule. Technological advances associated with the phenomenon has triggered the quest for new materials with large 2PA cross sections. Together with international co-workers, Kenneth Ruud and Maarten T. P. Beerepoot have published an extensive benchmark study of the performance of various DFT functionals in predicting 2PAs in dipolar pi-conjugated molecules containing electron-donating/-accepting moieties [17]. The results show that global hybrids are best suited to reproduce the absolute values of 2PA strengths but that they may fail to capture trends in the 2PA cross section across molecules, whereas the range-separated CAM-B3LYP and LC-BLYP functionals correlate best with the reference RI-CC2 results and better reproduce qualitative trends.

#### Personnel and funding

Research in RT3 has in 2022 been supported by the CAS project *Attosecond Quantum Dynamics Beyond the Born-Oppenheimer Approximation* led by Simen Kvaal and Thomas Bondo Pedersen. The CAS project was extended by one year because of the pandemic, and core members Prof. Ludwik Adamowicz, University of Arizona, and Prof. Caroline Lasser, Technische Universität München,

will be in Oslo during spring 2023 to continue work on the project.

Research in RT3 has been supported by three RCN grants: the FRIPRO grant *EPR and Paramagnetic NMR of Solids from Relativistic Two- and Four-Component Density-Functional Theory* led by Kenneth Ruud and Michal Repisky, the mobility grant *First-Principles Modeling of Magnetic Topological Materials from Relativistic Hybrid Density Functional Theory* led by Marius Kadek, and the mobility grant *Linear and Non-linear Response Properties and Spectroscopy of Solids from Relativistic TDDFT* led by Lukas Konecny.

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## RT4

# Extreme Environments

Principal investigators:  
Trygve Helgaker  
and Erik Tellgren

In RT4, we explore the exotic, squeezed chemistry of ultrastrong magnetic fields and high pressure. In 2022, our emphasis was on molecular dynamics and molecular structure in ultrastrong magnetic fields. In addition, we studied the generalization of conceptual density-functional theory to deal with finite magnetic fields, using the tools developed in RT4.

The eight papers published in RT4 in 2022 concern molecular dynamics [1–3], molecular structure [4–6], and conceptual density-functional theory (DFT) [7–8] in a magnetic field and are in the following discussed in that order.

### Molecular dynamics in a magnetic field

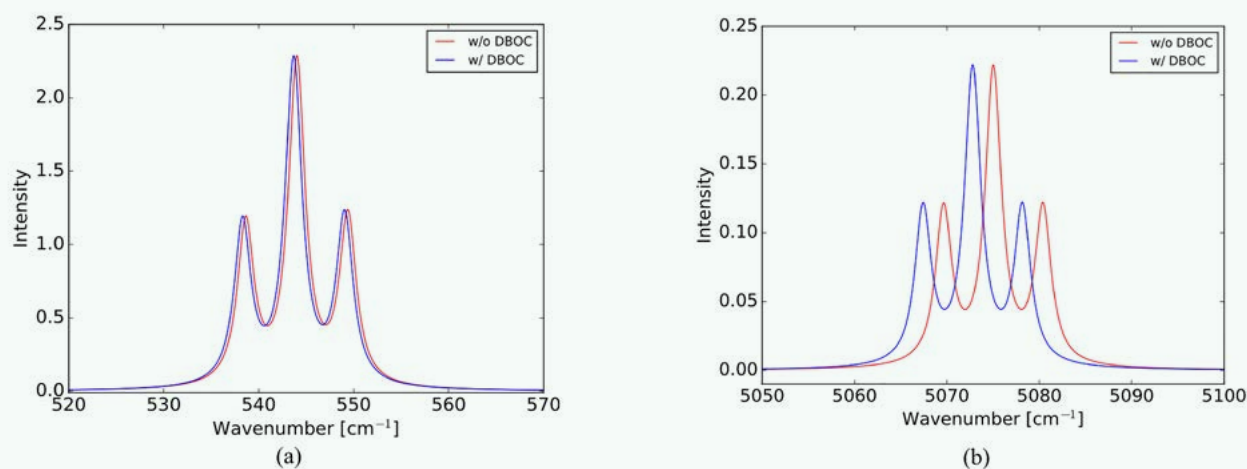
Molecular dynamics continued to be an important research direction within RT4 in 2022, with the publication of three papers on the subject. In the first published paper, *Analytic calculation of the Berry curvature and diagonal Born–Oppenheimer correction for molecular systems in uniform magnetic fields* [1], we

presented the analytical calculation of the Berry curvature and diagonal Born–Oppenheimer correction (DBOC) of a molecule in a uniform magnetic field at the Hartree–Fock level of theory; previously, the Berry curvature had been obtained by a finite-difference approach [9]. The DBOC and Berry curvature are both second-order non-adiabatic coupling matrix elements, calculated from the first derivative of the electronic wave function with respect to nuclear displacements. The corresponding coupled-perturbed Hartree–Fock equations are in Ref. [1] derived in two different manners, using first- and second-quantization formalisms. The second-quantization approach is particularly well suited for post-Hartree–Fock many-body theories and has previously been presented in Ref. [10], but without the use of the numerically stable natural connection [11] as done here.

While the Berry curvature is needed to calculate the local magnetic field experienced by the nuclei in molecular-

dynamics simulations, the DBOC provides a field-dependent correction to the Born–Oppenheimer potential-energy surface. As in the field-free case, the DBOC is small and can be neglected except in high-precision work; see Figure 1. By contrast, the Berry curvature is essential for a correct centre-of-mass motion in a magnetic field, while also providing a sometimes sizeable correction to the internal motion of molecules.

In our second paper on molecular dynamics published in 2022, entitled *Molecular dynamics of linear molecules in strong magnetic fields* [2], we presented a detailed analysis of *ab initio* molecular dynamics simulations of linear molecules in a strong magnetic field and determined, in particular, the rovibrational spectra of H<sub>2</sub> and LiH. This paper, which is a continuation of the work presented in Ref. [12], is a result of a collaboration with prof. Wim Klopper's research group at Karlsruhe Institute of Technology and goes back to the visit of his bachelor student Laurenz



⬆ **Figure 1:** Low energy (a) and high energy (b) vibrational fine structures of  $\text{H}_2$  with (blue) and without (red) the inclusion of the DBOC in the PES. Calculations are performed with the analytic Berry curvature and the Lu-cc-pVTZ basis set for a magnetic field strength  $0.1B_0$ , where  $B_0 = 235$  kT (one atomic unit). Reproduced from Ref. [1] under CC-BY 4.0 licence.

Monzel in Oslo in 2020. The molecular trajectory and rovibrational spectrum of  $\text{H}_2$  and LiH are discussed in the Research Highlight of this Annual Report.

In Ref. [2], we also introduced a new propagator for molecular dynamics in a magnetic field – the Tajima propagator, adapted from plasma physics. Compared with the auxiliary-coordinates-and-momenta (ACM) algorithm developed Ref. [12], the Tajima propagator is significantly more efficient without loss of accuracy or stability. In addition, no empirical coupling constant needs to be adjusted for the Tajima propagator.

In Ref. [3], entitled *Magnetic-translational sum rule and approximate models of the molecular Berry curvature*, we study the mathematical properties of the Berry connection and curvature. In particular, we prove the magnetic-translational sum rule of the Berry curvature, which is crucial for the correct centre-of-mass motion in a magnetic field. Consider a molecule consisting of  $N_{\text{nuc}}$  nuclei at Cartesian positions  $\mathbf{R}$  and  $N_{\text{el}}$  electrons

in the presence of a magnetic field with Cartesian components  $B_x$ ,  $B_y$ , and  $B_z$ . The translational-magnetic sum rule then states that

$$\sum_{I,J=1}^{N_{\text{nuc}}} \Omega_{IJ}(\mathbf{R}) = eN_{\text{el}} \begin{pmatrix} 0 & -B_z & B_y \\ B_z & 0 & -B_x \\ -B_y & B_x & 0 \end{pmatrix}$$

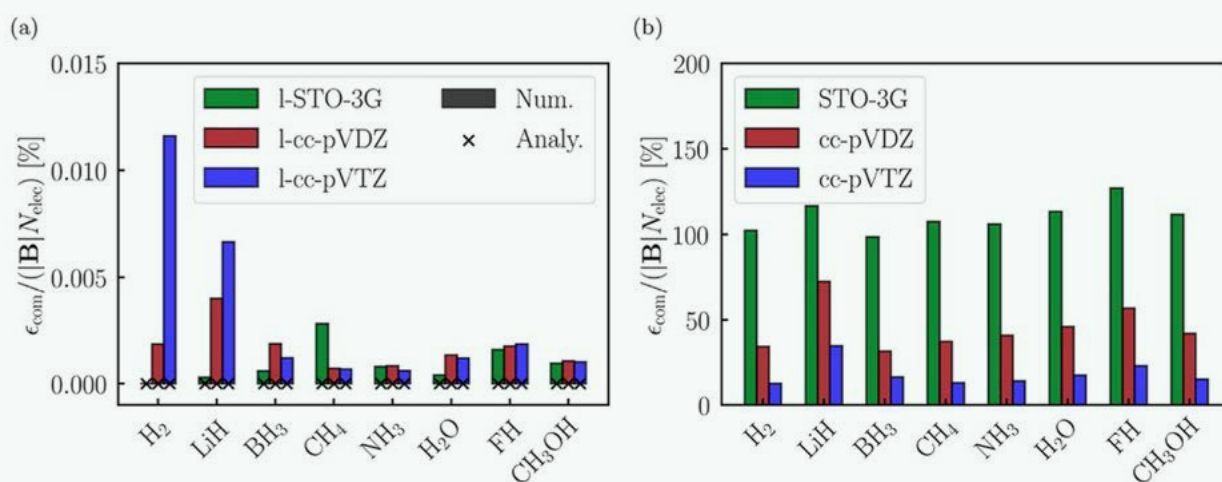
where the three-by-three  $IJ$ th block of the Berry curvature  $\Omega_{IJ}(\mathbf{R})$  is calculated from the derivatives of the electronic ground-state wave function  $\Psi(\mathbf{R})$  with respect to nuclear displacements as

$$\Omega_{IJ}(\mathbf{R}) = -(\hbar/\pi) \text{Im} \langle \nabla_I \Psi(\mathbf{R}) | \nabla_J \Psi(\mathbf{R}) \rangle.$$

In these expressions,  $e$  is the elementary charge and  $\hbar$  Planck's constant. Thus, according to the magnetic-translational sum rule, the ground-state electronic wave function carries information about the magnetic field. Unless this relation holds also in approximate calculations, a neutral atom will unphysically experience an overall Lorentz force in a magnetic field and perform cyclotron rather than translational motion in the field, for example.

We show in Ref. [3] that the sum rule holds not only for exact electronic ground-state wave functions, but also for exact Hartree-Fock wave functions and from finite-basis Hartree-Fock wave functions constructed from London atomic orbitals. As shown in Figure 2, wave functions calculated from standard Gaussian basis sets without London phase factors do not satisfy the magnetic-translational sum rule, whereas wave functions calculated using London orbitals satisfy the sum rule to numerical precision.

Finally, in Ref. [3], we also examine a series of approximations to the Berry curvature that are based on the Mulliken partitioning of the density and satisfy the magnetic-translational sum rule.



⬆ **Figure 2:** Error in the centre-of-mass motion of the Berry curvature calculated for different molecules using London orbital basis sets (a) and regular Gaussian basis sets (b). All calculations were performed at the Hartree–Fock level of theory at field strength  $10^3 B_0$ . We have divided the individual error by the magnetic field strength and the number of electrons to get an estimate for the screening error per electron. In Pane (a), we show errors resulting from the numerical [bars] as well as from the analytical [crosses] implementation of the Berry curvature. In Pane (b) only the former is shown. Note the different scale of the y-axis in Panes (a) and (b). Reproduced from Ref. [3] under CC-BY 4.0 license.

### Molecular structure in a magnetic field

It has been known for a long time that an ultrastrong magnetic field dramatically affects molecular bonding and therefore electronic structure, eventually forcing all electrons into beta spin, breaking covalent bonds and setting up paramagnetic bonds—see, for example, Refs. [13,14]. In 2022, Teale and coworkers made important further contributions to the elucidation and understanding of molecular structure in an ultrastrong magnetic field, as described in this section.

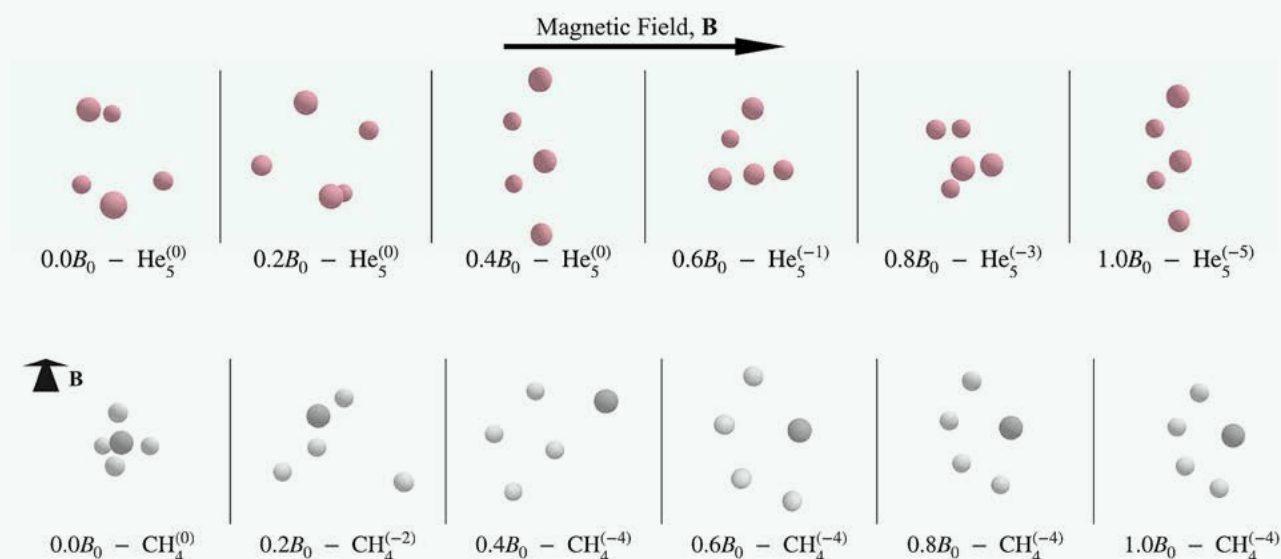
In Ref. [4], *Revealing the exotic structure of molecules in strong magnetic fields*, a new implementation of molecular gradients in a strong magnetic field is used at the level of density-functional theory (DFT) to optimize molecular structure

at different field strengths. In particular, an analogue of the *ab initio* random-structure search method was utilized to determine the ground-state equilibrium geometries for He<sub>n</sub> and CH<sub>n</sub> systems in a strong magnetic field, revealing the most stable structures to be those in high-spin states with a planar geometry aligned perpendicular to the field. The electron and current densities of these systems have also been investigated so as to develop an explanation of chemical bonding in the strong field regime, providing new insight into the exotic chemistry present in these extreme environments.

As seen in Figure 3, with increasing magnetic-field strength, more and more electron pairs are uncoupled by the field as covalent bonds are broken and

replaced by paramagnetic ones. As paramagnetic bonds are strongest in a perpendicular field orientation, the systems become planar in the field, with a perpendicular orientation relative to the magnetic field vector.

In Ref [5], *An embedded fragment method for molecules in strong magnetic fields*, an extension of the embedded fragment method for calculations on molecular clusters is presented. The approach is flexible, allowing for calculations using Hartree–Fock theory, current-density-functional theory (CDFT), second-order Møller–Plesset (MP2) perturbation theory, and coupled-cluster theory in a magnetic field using London atomic orbitals. For systems consisting of discrete molecular subunits, calculations using London orbitals can be performed



⬆ **Figure 3:** Top: Ground-state structures for  $\text{He}_5$  at field strengths up to one atomic units  $B_0 = 235$  kT in increments of  $0.2B_0$  with the direction of the magnetic field relative to the molecular plane indicated by the arrow. Bottom: Ground-state structures for  $\text{CH}_4$  at the same field strengths with the direction of the magnetic field relative to the molecular plane indicated by the arrow (pointing into the plane of the image). In both cases, we use the superscript notation ( $M_s$ ) for the total spin projection of the system. Reproduced from Ref. [4] under CC-BY 4.0 licence.

in a computationally tractable manner for systems beyond the reach of conventional calculations, even those accelerated by resolution-of-the-identity or Cholesky decomposition methods.

To assess the applicability of the approach, applications to water clusters were presented, showing how strong magnetic fields enhance binding within the clusters; see Figure 4. Our calculations suggest that this enhanced binding may not be directly attributable to strengthening of hydrogen bonding, as previously thought. Instead, these results suggest that the enhanced binding arises for larger field strengths as a response of the system to the presence of the external field, which induces a charge density build-up between the monomer units.

The maximum overlap method (MOM) provides a simple yet powerful approach for performing calculations on excited states by targeting solutions with non-Aufbau occupations from a reference set of molecular orbitals. In Ref. [6], *Understanding ground and excited-state molecular structure in strong magnetic fields using the maximum overlap method*, the MOM was used to study the excited states of  $\text{H}_3^+$  and  $\text{H}_3$  in strong magnetic fields, comparing the lowest  $^1\text{A}'_1$ ,  $^1\text{E}'$  and  $^3\text{E}'$  states of  $\text{H}_3^+$  in the absence of a field with the corresponding states in a strong magnetic field. The changes in molecular structure in the field were examined by performing excited-state geometry optimizations using the MOM.

It was found that the  $^3\text{E}'$  state of  $\text{H}_3^+$  is significantly stabilized by the field, be-

coming the ground state in a strong field with a preferred orientation perpendicular to the applied field; see Figure 5. Its potential energy surface evolves from repulsive to bound, with an equilateral equilibrium geometry. In contrast, the  $^1\text{A}'_1$  state is destabilized by the field and its structure distorts to an isosceles form with the longest H-H bond parallel to the applied field.

To investigate why these structures are preferred, orbital correlation diagrams were constructed, using the molecular orbitals (MOs) determined by MOM calculations with constrained geometry optimization. The changes in the orbital energies along the lowest energy pathway for bending the molecule revealed that the structure of the lowest  $M_s = -1$  state of  $\text{H}_3^+$  is determined

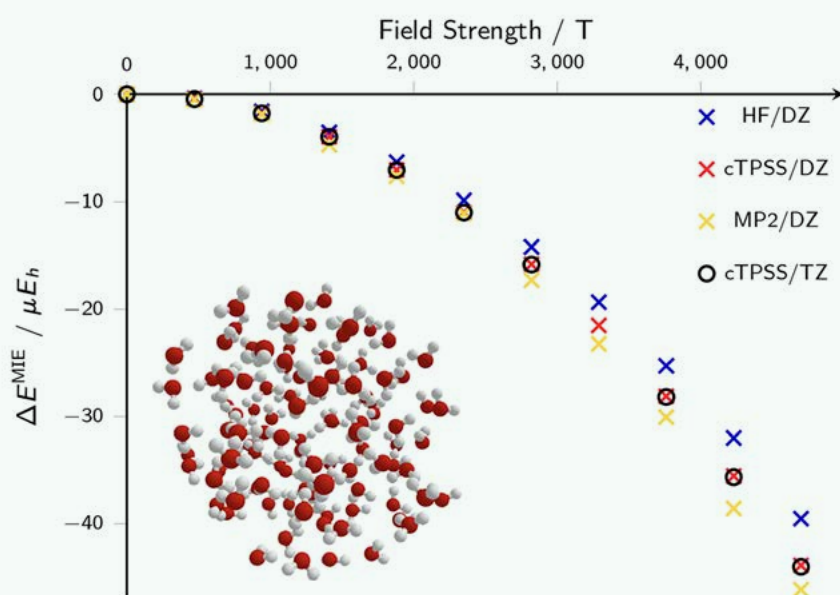


Figure 4: Change in the mean interaction energy for a molecular cluster consisting of 103 water molecules, determined at the Hartree-Fock (blue), CDFT/cTPSS (red), and MP2 (yellow) theory levels in the aug-cc-pVDZ basis and using resolution-of-the-identity theory for two-electron interactions. Equivalent CDFT/cTPSS calculations in the aug-cc-pVTZ basis set are shown as black circles. Reproduced from Ref. [5] under CC-BY 4.0 licence.

by its highest occupied MO (HOMO), which is strongly stabilized at the equilateral geometry. For the neutral  $H_3$  species, this MO, despite not being the HOMO, is still key to determining the molecular structure in a high field, its preference for an equilateral geometry outweighing the preference of the other occupied MOs for a linear or isosceles geometry. In spite of this, the lowest  $M_s = -3/2$  state of  $H_3$  was found to become significantly bound only at  $1.5B_0$ , unlike the lowest  $M_s = -1$  state of  $H_3^+$ , which is strongly bound already at  $0.5B_0$ .

#### Conceptual density-functional theory

In conceptual DFT as opposed to computational DFT, the goal is to extract chemically relevant concepts and principles from DFT. In a collaboration of Andrew Teale with the research groups

of Frank de Proft and Paul Gerlings at the Vrije Universiteit Brussel, Belgium, the first attempts were made at generalizing conceptual DFT to include the influence of an external magnetic field [7,8].

In conceptual DFT in a magnetic field, we are particularly interested in the electronegativity  $\chi(B)$  and chemical hardness  $\eta(B)$  as functions of the field strength  $B$ , which are related to the ionization potential  $I(B)$  and electron affinity  $A(B)$  of the species at the same field strength in the manner:

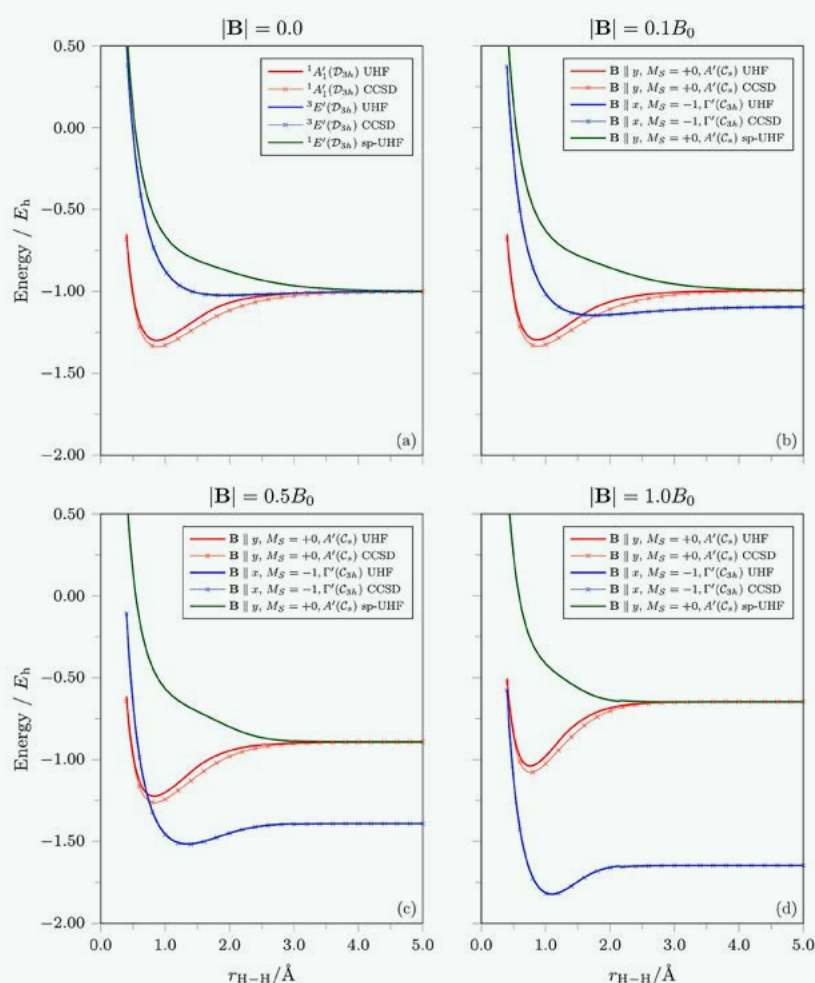
$$\chi(B) = -\frac{1}{2} (I(B) + A(B)),$$

$$\eta(B) = \frac{1}{2} (I(B) - A(B)).$$

In Ref. [7], *Extending conceptual DFT to include external variables: the influence of magnetic fields*, these basic quantities

of conceptual DFT are examined as functions of the field strength. It turns out that these quantities change in a complicated manner with increasing field strength, following the changes in the ground-state electronic configurations with increasing field strength. The electronegativity and hardness for main-group atoms at zero field and at field strength  $0.5B_0$  are illustrated in Figure 6.

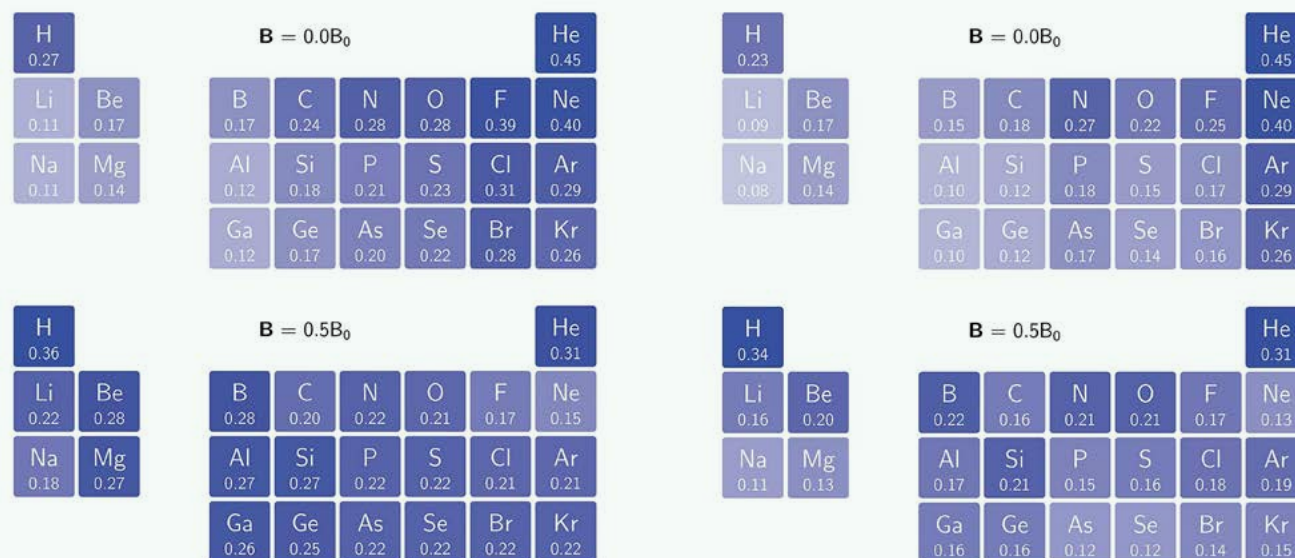
Both for the electronegativity and for the hardness, an overall increase in values is observed on the left side of the periodic table, whereas a decrease is seen on the right side, with a similar behaviour of elements within the same column. The overall picture is a compression of the electronegativity and hardness range across each period, with



important changes to be expected for bond polarity.

The derivatives of the electronegativity  $\chi(B)$  and hardness  $\eta(B)$  with respect to the magnetic field strength at zero field (i.e., response functions in the conventional sense for conceptual DFT) present a simpler picture of the field behaviour since no change in electron configuration occurs as derivatives are taken, making it easier to compare across the periodic table; see Figure 7. In general, all atoms in a given group of the periodic table behave in a similar manner. For the p-block, the general picture is that the increase in the electronegativity from the bottom left to top right of the periodic table at zero field is reduced by the tendency of the electronegativity to decrease by an

⬆ **Figure 5:** Potential-energy curves for equilateral geometries of  $H_3^+$  calculated using unrestricted Hartree–Fock (UHF) theory, spin-purified UHF (sp-UHF) theory and coupled-cluster singles-and-doubles (CCSD) theory. At zero field, the nondegenerate ground-state is bound, while the degenerate excited states are dissociative. In an ultrastrong magnetic field, the  $M_s = -1$  component of the excited state is stabilized by paramagnetic bonding and eventually becomes bound with the magnetic field perpendicular to the (equilateral) molecular plane, while the  $M_s = 0$  component remains dissociative, with the magnetic field along one of the H–H bonds. The MOM calculations are in good agreement with the corresponding CCSD calculations. *Reproduced from Ref. [6] under CC-BY 4.0 licence.*



⬆ **Figure 6:** Schematic representation of the electronegativity (left) and hardness (right) of atoms at zero field and at field strength  $0.5B_0$ . Darker colours indicate higher electronegativity and greater hardness, respectively. Reproduced from Ref. [7] with permission from the Royal Society of Chemistry.

increasing amount from left to right across the periodic table in a magnetic field. For the hardness, the atoms in the group with the highest  $\eta(B)$  in a period at zero field have a decreasing or unchanged hardness in the presence of a magnetic field, whilst all other elements have an increasing hardness.

In Ref. [8], the influence of a strong magnetic field on the charge distribution of hydrogen halides, as manifested by the electric dipole moment, atomic partial charges and charge densities themselves, is considered in two different ways. The first of these is with a conceptual DFT model, making use of the atomic electronegativity and hardness evaluated in Ref. [7] across a range of magnetic field strengths for main-group atoms up to krypton, with which the charge transfer within a bond can be

predicted in the presence of a strong magnetic field.

The second approach is through current DFT calculations on the molecules themselves, with a strong magnetic field applied parallel and perpendicular to the internuclear axis and with the electric dipole moment and Mulliken charges evaluated from the converged solutions. The inversion of polarity predicted by the conceptual DFT model, and the trend expected for the three halogen atoms, is observed in the current DFT calculations – with an inversion in polarity of HF, HCl and HBr with a strong magnetic field applied perpendicular to the internuclear axis, at decreasing magnetic field strength in the series HF  $\rightarrow$  HCl  $\rightarrow$  HBr.

### Personnel and funding

Research in RT4 was in 2022 supported by the RCN FRIPRO project *Magnetic Chemistry* (Grant No. 287950) led by Trygve Helgaker. Andrew Teale is supported by the European Research Council under H2020/ERC Consolidator Grant topDFT (Grant No. 772259).

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⬆ **Figure 7:** Initial responses of the electronegativity (left) and hardness (right) in a magnetic field as a periodic table representation showing both the numerical values and categorizing them with a colour code in which blue indicates a positive derivative, red a negative derivative and yellow a derivative that is zero or close to zero. Reproduced from Ref. [7] with permission from the Royal Society of Chemistry.

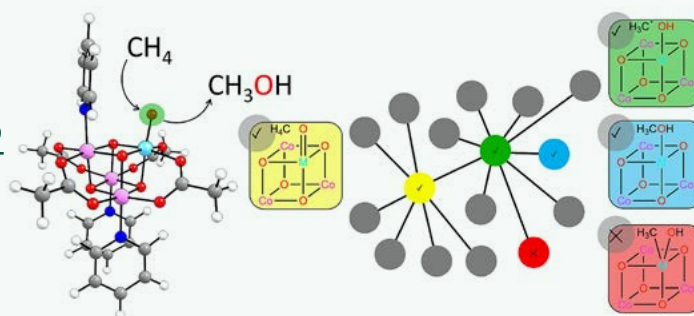
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RT5

## Chemical Transformations

Principal investigators:  
David Balcells and Ainara Nova



↑ **Figure 1:** AFIR derivation of reaction pathways in the oxidation of methane by metal cubanes yielding expected (yellow, green, and blue boxes) and unexpected (red box) intermediates. *Illustration: Bastian Skjelstad and David Balcells.*

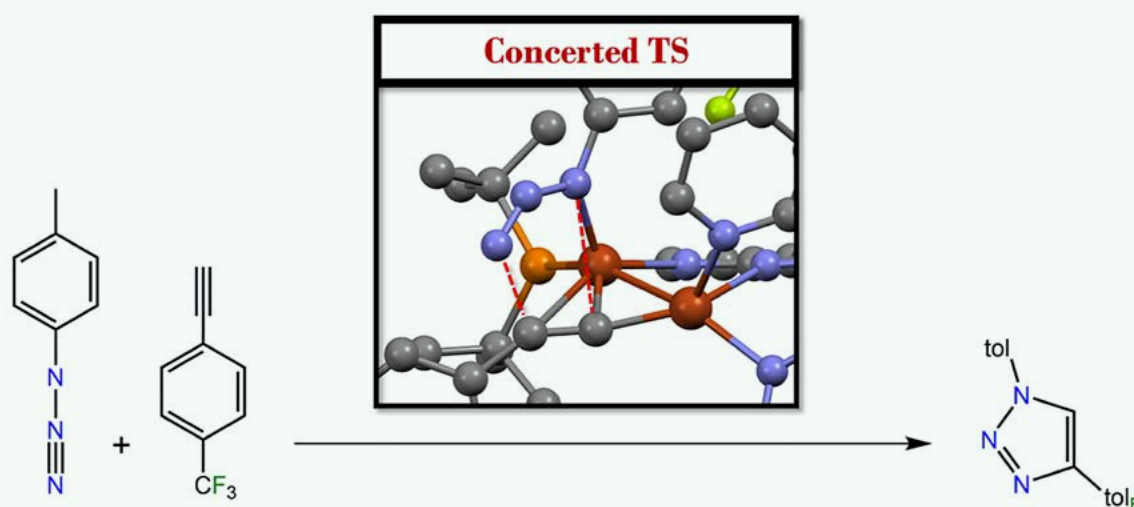
RT5 applies computational methods to understand chemical reactions, often in collaboration with experimental groups. A key goal is to simulate reactivity in complex environments, including reaction networks, spectroscopic processes in solution, and metal-organic frameworks. Machine learning is being introduced with the goal of exploring large chemical spaces in homogeneous catalysis.

### Catalytic oxidations

The reaction pathways associated with the catalytic oxidation of alkanes have been computed by using automation methods. In particular, the artificial force-induced reaction (AFIR) method was used at the level of density-functional theory (DFT) to explore the potential-energy surface of a remarkably complex system known in the literature as a cubane, consisting in a cubic oxide structure with alternating metal and oxo vertices (Figure 1). The study focused on the hydroxylation of methane, which is the alkane with the strongest C-H bond. The calculations showed that the

barrier for methane oxidation can be minimized by doping cobalt cubanes with iron. Further, the AFIR calculations confirmed the rebound mechanism postulated for this reaction and revealed the existence of exotic intermediates (Figure 1). The results were published in *ACS Catalysis* [1], adding to a series of studies on cubanes done in collaboration with the experimental group of Prof. T. Don Tilley at UC Berkeley [2-5]. This work was part of the MSc project of Bastian Skjelstad at the Hylleraas Centre, which was co-supervised by David Balcells in RT5 and Trygve Helgaker in RT4. Bastian is currently doing his PhD at the University of Hokkaido (Japan), in the group of Satoshi Maeda, who also co-supervised part of this work. As part of this collaboration, Balcells gave an invited lecture on the topic of machine learning at the ICRDD Institute of Hokkaido in February 2022, while Skjelstad visited the Hylleraas Centre in Oslo in the last week of September to present this work and his current PhD project.

The selective hydroxylation of methane by mimicking the active site of enzymes in metal-organic frameworks (MOFs) is the goal of the ERC-SyG CUBE project, which includes Ainara Nova as computational partner and chair of the Hylleraas Board of Directors Unni Olsbye as the corresponding PI. One of the challenges of this project is the characterization of Cu-active sites in the MOF cavities and the interpretation of the experimental data generated during the testing of these materials. Computational methods can play a key role in addressing these challenges by providing structural information on the preferred coordination numbers of Cu in different oxidation states and determining potential reaction mechanisms for different Cu species. These studies are carried out by PhD candidate Ning Cao, under the supervision of Nova and Balcells. Cao has used DFT to optimize the geometry of tri- and tetra-dentate Cu complexes in the UiO-67 MOF by using cluster and periodic models, and determined the coordination number preferences for Cu(I) and Cu(II) species.



⬆ **Figure 2:** The concerted cycloaddition transition state of the CuAAC reaction computed at the DFT(PBE0/def2SVP) level of theory. Colour code: C = grey, P = orange, red = Cu, blue = N, green = F.  
Illustration: Julie Héron and David Balcells.

This work was presented in a poster at the virtual International Conference on Theoretical Aspects of Catalysis (ICTAC) in June 2022.

### Base-metal catalysis

The CuAAC reaction, one of the most powerful reactions in the field of click chemistry, has been thoroughly studied by means of DFT. We have focused on the synthesis of triazole rings catalysed by dicopper complexes with nonsymmetric naphthyridine ligands, which, by combining phosphine and pyridine coordination moieties, allow for differentiating the properties of the two metal centres. The calculations revealed a new mechanism in which the cycloaddition reactions take place in a concerted manner (Figure 2), in contrast to the two-step pathway commonly proposed in the literature for similar systems. Furthermore, the study showed that there is a subtle interplay between different phenomena, including ion-pairing as well as the conformation flexibility and partial decoordination of the ligand, which, in combination

yield a large and complex reaction network. The results were published in *ACS Catalysis* [6]. Julie Héron did this work under the supervision of Balcells, becoming one of the main contributions from her thesis and adding to a previous publication on the activation of benzyl-bridged pre-catalysts [7]. After obtaining her PhD degree, Héron left the Hylleraas Centre in 2022 and was hired as a DevOps engineer at the Sopra Steria Company in Toulouse (France).

Besides copper, another base metal of high interest is nickel. Similar to the CuAAC reaction, there is an increasing interest in building molecular systems in which two or more nickel atoms enable cooperative effects that can be leveraged in catalytic reactions. We characterized a new dinuclear nickel complex supported by phenyl-lithium ligands and a benzyne bridging moiety between the two metal centres. Together with X-ray studies, DFT calculations revealed the complex electronic structure of this novel compound and, in particular, the presence of 1) weak s-d

donor-acceptor interactions between the metal atoms, despite the lack of a covalent Ni-Ni bond, 2) strong d-π interactions between the metals and the benzyne bridge, involving both donation and backdonation, and 3) σ-coordination of the phenyl-lithium ligands. The work was published in *Chemical Science* [8], including DFT calculations done by Balcells and Nova in a new collaboration with the experimental groups of Dr. Jesús Campos (University of Sevilla, Spain) and Prof. Eva Hevia (University of Bern, Switzerland).

### Machine learning

Following our previous work in this new line of research at the Hylleraas Centre [9,10], we began in 2022 new activities at the interface between machine learning and computational catalysis, including the automation of transition-state optimizations, deep graph learning of transition-metal complex representations, and Δ-ML models predicting xTB-to-DFT corrections to the extended tight-binding (xTB) model.



In the first project, we collaborated with the group of Dr. Daniel Ess (Brigham Young University, USA) in the development of a new method automating transition-state optimization from a structural core that can be decorated with molecular fragments. We applied this concept to the C–H activation of alkanes by palladium catalysts, in which a Pt–CH<sub>4</sub> core was stabilized by a diverse collection of mono-, bi-, and tri-dentate ligands. The key component of the automation algorithm is a random forest model that predicts the distance of the C–H bond when it is cleaved at the transition state. This work was published in *Topics in Catalysis* [11], which also includes a contribution from Skjelstad.

For deep graph learning applied to organometallic compounds, we explored the transformation of natural bond orbitals into undirected and directed graphs that were fed to message-passing neural networks for the prediction of quantum properties. This work is part of the PhD project of Hannes Kneiding, supervised by Balcells. Kneiding presented his initial results in two posters – one at the Young Researcher's Workshop on Machine Learning for Materials, in Trieste (Italy) in May, and the other in the WATOC 2022 conference, in Vancouver (Canada) in July. With the aim of exploring the use of generative models for inverse design, Kneiding visited the group of Prof. Alán Aspuru-Guzik at the University of Toronto for three months in 2022.

With regard to the  $\Delta$ -ML models, Dr. Lucas Lang worked on the use of

semi-empirical Hamiltonians for the prediction of xTB-to-DFT corrections with equivariant graph neural networks, as part of an inter-RT collaboration between Balcells (RT5) and Thomas Bondo Pedersen (RT1). Lang presented his results in a poster at the Trieste workshop in May and gave an oral presentation at the National Meeting of the Division of Quantum Chemistry and Modelling in Bergen in November 2022. He also visited the Aspuru-Guzik group for two weeks and the group of Prof. Markus Reiher (ETH Zürich, Switzerland) for one week, where he explored the use of automation methods for the systematic exploration of the potential-energy surface of catalytic processes.

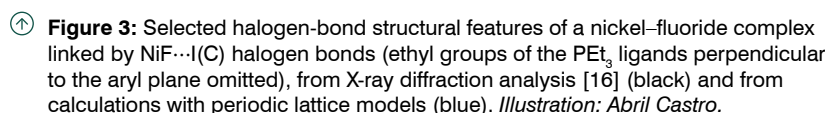
Balcells presented the work of Kneiding and Lang in his invited talk at the WATOC 2022 conference. In the fall, three more hirings were made in machine learning for computational catalysis: Aurore Denjean, in a PhD position, and Dr. Arron Burnage and Dr. Nishamol Kuriakose, in postdoctoral positions. These three new positions were supported by the external funds provided by the RCN catLEGOS and EU HYSTRAM projects led by Balcells.

### NMR simulations

The FRIPRO project SpecTraM, which was awarded to Dr. Abril Castro in 2021, experienced a year of transition in 2022. Following up on the successful modelling of NMR chemical shifts in metal polyhydride complexes [12] and platinum anticancer drugs [13–14], which showed that relativity and dynamics are both crucial for obtaining a good

agreement with experiment, the purpose of the SpecTraM project is to provide a broader modelling of NMR properties in heavy-transition-metal complexes either in solution or in the solid state, for which the environment cannot be neglected. Beyond applications to support the experimental analysis and to help with the interpretation and prediction of spectra, this project is focused on giving a detailed interpretation of the observed NMR chemical shift trends. The interpretation of NMR chemical shifts in transition-metal complexes is at the heart of a deeper understanding of their stability and chemical reactivity. Detailed information about the coordination sphere can be extracted from the nuclear shielding, and thus the computed shielding tensors can be used to identify the origin of the observed chemical shift changes and their relationship to the electronic structure of the system.

Together with Prof. Robin Perutz (University of York) and Dr. Christophe Raynaud (University of Montpellier), and in close collaboration with Odile Eisenstein and Michele Cascella, Abril Castro began analysing a series of nickel-fluoride complexes capable of forming polymers that are linked by NiF...I(C) halogen bonds (Figure 1). The final goal is to understand how the <sup>19</sup>F NMR chemical shift is affected by the halogen bond [15]. Another relevant activity has been the hiring of the PhD candidate Alberto Guerrero, who will work under the supervision of Castro. Guerrero started working on structural and spectroscopic properties of platinum complexes containing ancillary ligands, in collab-



Investigations are currently progressing with studies on alkyllithium and LiBr, LiI by Dr. Sondre Eliason. In addition, the structure of the Grignard reagent in deep eutectic solvent/THF biphasic systems is being investigated by Dr. Marco Bortoli. These studies are done in collaboration with the experimental group of Prof. Eva Hevia at the University of Bern, Switzerland. Finally, to address the complex and dynamical nature of the species involved in these systems, Dr. Sigbjørn L. Bore is exploring the parameterization of empirical models by neural networks (DeePMD)



[20] to replace the excessively costly AIMD simulations based on DFT. This project has strong synergy with RT2, for the AIMD/multiscale component and the forthcoming RT7 dedicated to machine learning.

### Cross-coupling reactions

In 2020, Dr. Raquel Jimenez-Rama joined the group of Ainara Nova to study computationally the mechanism of palladium-catalysed C–N cross-coupling reactions by terphenyl phosphane ligands. As in previous studies of Nova and Balcells [21], the focus was on off-cycle reactions and, in particular, the role of the NH-carbazole generated during pre-catalyst activation in the cross-coupling reaction.

By combining the experimental results obtained by Jimenez-Rama during her PhD work with the computational data generated during her work at the Hylleraas Centre, it became possible to build a microkinetic model that fitted the experimental data [22]. This study concluded that NH-carbazole reduces

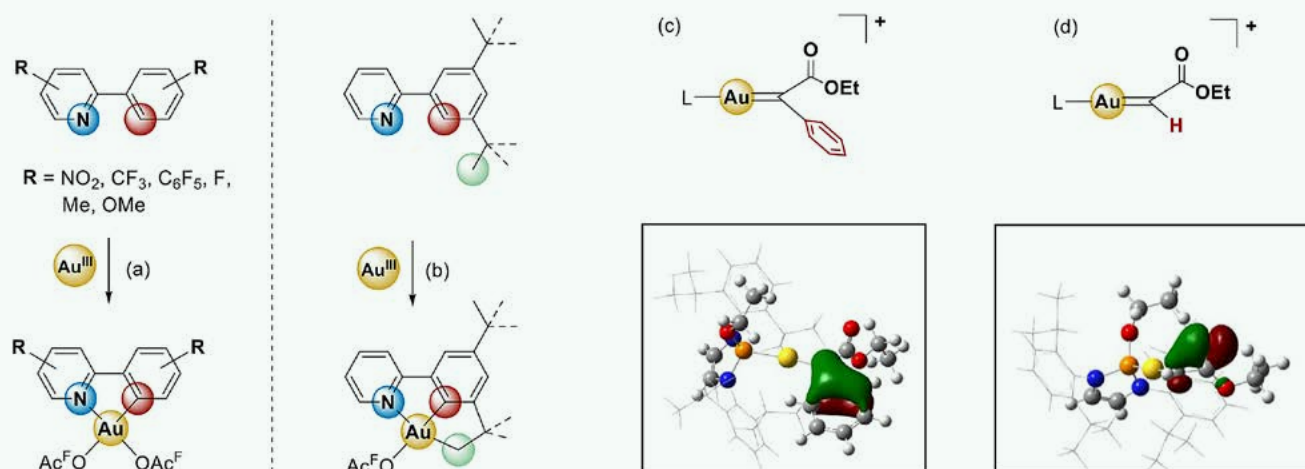
the rate of reactions with alkyl amines by forming an aryl carbazolyl Pd(II) complex. In contrast, aryl amines are easily deprotonated and exchanged by the carbazolyl ligand, avoiding the high activation barrier for the N-arylcarbazole reductive elimination. In addition, formation of this Pd(II)-complex reduces catalyst decomposition, which can be beneficial for coupling substrates that require high temperatures.

In 2022, this work was completed and presented by Jimenez-Rama at the 38th Biennial Meeting of the Spanish Royal Society of Chemistry (RSEQ) in Granada in June 2022 and the Norwegian Catalysis Symposium in Stavanger in December 2022.

### Gold chemistry

Following the collaboration of Ainara Nova with Mats Tilset on the functionalization of olefins with (N,C) and (N,C,C)Au(III) pincer complexes [23,24], PhD candidate Inga Lena Schmidtke used computational methods to gain understanding of the synthesis of these

complexes [25]. The analysis of sterics and electronics of the Au-ligand substituents allowed her to conclude that the C–H activation required for the synthesis of (N,C,C)Au(III) complexes is kinetically preferred by using bulky groups such as tert-butyl, which destabilize the (N,C)Au(III) reactant. On the other hand, the formation of (N,C)Au(I–II) complexes is prevented by highly electron-deficient aryl groups with more than one CF<sub>3</sub> substituent. In the second publication, in collaboration with Prof. Pedro Perez (University of Huelva), Inga accounted for the high selectivity observed in the functionalization of toluene with a Au(I) carbene complex by using DFT and natural-bond-order (NBO) analysis [26]. After seeing the large impact on the C–H bond activation energy by changing the R group of Au=CRCO<sub>2</sub>Et from H to Ph, the NBO analysis revealed a higher stability of the carbene with R=Ph due to the delocalization of its lone pair (Figure 4). The lower reactivity of Au=CPhCO<sub>2</sub>Et promotes its closer contact with toluene in the transition state, differentiating



⬆ **Figure 4:** Synthesis of (N,C)Au(III) (a) and (N,C,C)Au(III) (b) pincer complexes. LAu=CRCO<sub>2</sub>Et complex with R=H (c) and R=Ph (d) with the results of the NBO analysis. Illustration: Inga Lena Schmidtke.

the C–H bonds in the *ortho*, *meta*, and *para* positions.

This work was presented in the CCP5 Summer School on Molecular Simulation Methods in Durham (UK) in July 2022, at the NordCO<sub>2</sub> Annual Meeting near Reykjavik (Iceland) in August 2022, and at the National Meeting of the Division of Quantum Chemistry and Modelling in Bergen in November 2022.

### Conversion of CO<sub>2</sub> to fuels

One of the main goals of the FRIPRO project CO<sub>2</sub>pCat led by Ainara Nova is the design of organometallic complexes able to hydrogenate CO<sub>2</sub> for their incorporation into MOFs. With this goal, PhD candidate Mohamed Eid Safi uses DFT to generate volcano plots to identify the best candidates for this reaction. The first results of Safi were presented at the NKS annual meeting in Bergen in November 2022, at the NordCO<sub>2</sub> annual meeting near Reykjavik, and at the 44th International Conference on Coordination Chemistry (ICCC) in Rimini

in August 2022. This project also motivated a visit of Safi to the group of Prof. Vidar Jensen and Dr. Marco Foscato at the University of Bergen, for de novo design of catalysts using the DENOPTIM software.

Dr. Raquel Jimenez-Rama has already started the synthesis of the organometallic complexes studied by Safi. She was initially hired as a researcher in CO<sub>2</sub>pCat but was awarded an EU MSCA grant in 2022. Her project, TUNEMOF, will allow experimental benchmarking of the computational results of CO<sub>2</sub>pCat and evaluate the scope and industrial implementation of these materials in collaboration with Prof. Matthias Beller at LIKAT (Germany) and Haldor-Topsøe (Denmark). The first results of Jimenez-Rama in the synthesis of linkers were presented in the NordCO<sub>2</sub> annual meeting.

In addition to study the thermal reduction of CO<sub>2</sub> to methanol, we are interested in understanding how this reaction is achieved under electrochemical conditions. In particular, we were

puzzled by the reactivity of manganese bipyridine complexes that lead to the formation of different products (CO, formic acid or H<sub>2</sub>) depending on the incorporation of amines in the ligand.

To understand this reactivity, PhD candidate Mahika Luthra in collaboration with Profs. Kim Daasbjerg and Troels Skrydstrup at Aarhus University (Denmark) used AIMD simulations to assess the proximity of the amine ligands to Mn in the complex shown in Figure 5. In addition, she calculated the energy profiles leading to CO, formic acid, and H<sub>2</sub> to account for the product selectivity. The results of this work [27] were presented by Mahika in a poster at the DFT 2022 conference in Brussels in August–September 2022. This study was supervised by Nova, Castro and Balcells and is part of the EU-ITN CO<sub>2</sub>PERATE network. As a part of her training, Luthra performed two secondments in 2022, in the group of Prof. Belén Martín-Matute at Stockholm University and in the group of Dr. Esben Taarning at Haldor-Topsøe, Denmark.

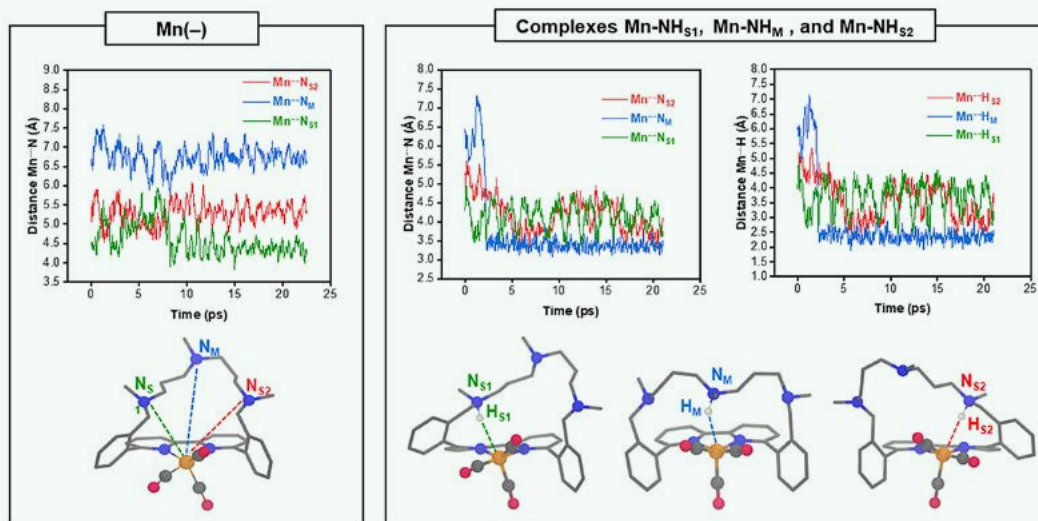


Figure 5: Time-evolution of the Mn...N and Mn...H distances for a reduced Mn-bipyridine complex (Mn(-)) and its protonated forms (Mn-NH). Illustration: Mahika Luthra and David Balcells.

During these visits, Luthra was trained in the synthesis of organometallic complexes and metal-organic frameworks and in the preparation and structural characterization of zeolites.

In a MOF, single metals can be attached to the linkers (as in the work of Cao, Safy, Jimenez-Rama, and Luthra) or to the secondary building units (SBUs), also known as nodes. The incorporation of metals such as Mg, Li and Zn to the Zr-node of UiO-66 and -67 MOFs has been investigated by Dr. Tamal Das under the supervision of Ainara Nova. In this study, which was initiated by Robert Brevik during his MSc thesis, DFT was used to evaluate the stability of different configurations of the MOFs named [4,0], [3,1] and [2,2]. It was found that the relative energy for these configurations depends on the nature of

the metal, its substituent and the length of the linker. Understanding the reason for the highest stability of the [4,0] configuration required the use of periodic and cluster models, AIMD simulations and non-covalent-interaction (NCI) plots.

In 2022, Tamal Das presented these results in a poster (for which he won a poster prize) at the ICTAC meeting in Lyon and in oral presentations at the NordCO<sub>2</sub> meeting in Iceland, at the National Meeting of the Division of Quantum Chemistry and Modelling in Bergen, and at the CCU-Net Symposium in Kuopio.

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## RT6

# Multiphase Systems

Principal investigators:

Bjørn Olav Brandsdal and Michele Cascella

The RT6 team applies computer simulations to important problems in chemistry, biology and physics. Activities in 2022 have focused on antimicrobial resistance, temperature dependence of chemical rates, aggregation of charged surfactants and lipids.

### Antimicrobial resistance

The escalation of multi-resistant bacteria combined with the low success rate of discovery of novel classes of antibiotics in the last decades poses a direct threat to human health globally. The need for new classes of drugs, as well as other strategies, is ever increasing. The bacterial membrane has in this context attracted increased attention as a drug target. Antimicrobials that target the cellular membrane are expected to cause less resistance compared with those that have intracellular targets.

We have in 2022 continued our focus on antimicrobial resistance and mem-

brane active compounds, both barbiturates and peptides consisting of up to six amino acids. Antimicrobial peptides (AMP) are typically larger than and different from the traditional small-molecule druglike compounds, for which the majority of today's drug discovery and design tools have been developed. In order to optimize the properties of the AMPs, it is necessary to understand how the AMPs exert their mode of action. So far, it appears that most AMPs act by causing bilayer leakage through disruption of membrane integrity.

Hydration plays an important role in lipid bilayer and function – for example, by defining the stability of lipid vesicles in solution and controlling the permeability of small molecules across lipid bilayers. Water and other small uncharged molecules passively pass through the membrane to maintain the osmotic equilibrium, while ions and other larger biomolecules are restricted. Interestingly, the rate of diffusion through the hydrophobic core of the

bilayer is limited by space and is suggested to be coupled to the order and motion of the lipid core. Lipid bilayer hydration is also associated with changes in lipid bilayer properties like increased permeability, increased area per lipid and reduced electric membrane potential.

In a recent study [1], we present a new approach that explored the possibilities of measuring permeability across immobilized phospholipid bilayers using a phospholipid vesicle-based permeation assay with NMR combined with all-atoms molecular-dynamics (MD) simulations. The MD simulations showed that, when the AMPs enter into the membrane, the hydrating water molecules follow and thus increase the overall hydration of the lipid core. None of the antimicrobials tested exerted an effect on barrier integrity in the absence of charge lipids in the membrane, but with a small concentration of charged lipids, a destabilizing effect was observed. We also observed that the most potent antimicrobials had a more pro-



nounced effect on the water and ion permeability.

Tonje Reinhold Haugen begun her PhD study in January 2022, working with cyclic hexapeptides. Four different peptides were selected (cLLIWWW, cLWLWLW, cRWRWRW and cRRRWWW) based on their biological activity and potency to kill resistant bacteria and were examined with computer simulations. Simulations were carried out with different concentrations of the peptides in pure water and also in the presence of a model membrane system.

#### **Temperature dependence of catalytic rates**

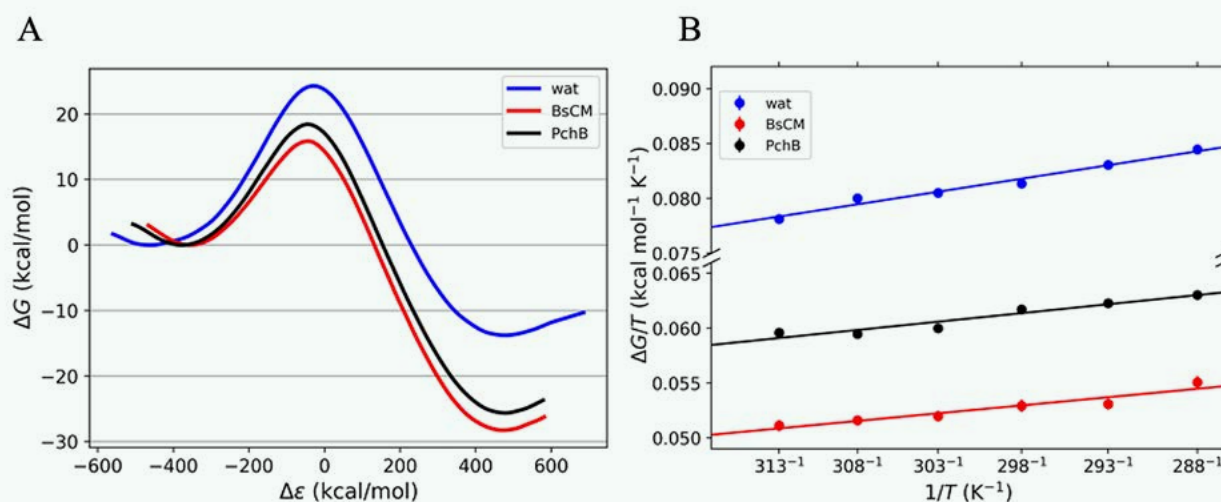
Organisms have through evolution adapted to different extreme environments, such as low or high temperature, high pressure, low or high pH, and so on. A major question is how proteins and enzymes of such organisms have evolved to cope with these environments. The most carefully studied external factor with respect to protein evolution is temperature. Our focus

has therefore been on how enzymes from organisms living around the freezing point of water are able to catalyse their reactions with rates comparable to their orthologs from warm-adapted organisms.

How enzymatic reaction rates are maintained at low temperature is still not fully understood from a structural point of view. However, the universal fingerprint of cold-adapted enzymes characterized experimentally appears to be a shift in the activation enthalpy-entropy balance, with a reduction in activation enthalpy followed by a more negative entropy. This means that the activation free energy is highly similar when comparing cold- and warm-active enzyme orthologs. To obtain a deeper understanding of low-temperature adaptation, we have investigated several cold- and warm-active enzyme orthologs by a combination of computer simulations and experimental characterizations.

From a computational point of view, very few methods are suitable for determination of activation enthalpies and entropies due to the requirements of conformational sampling. Here, we investigate the enthalpy-entropy balance by describing the chemical reaction with the empirical valence-bond (EVB) method. This method allows us to determine the activation free energy, from which the activation enthalpy and entropy can be derived by performing the calculations at five to eight different temperatures and constructing a computational Arrhenius plot. This approach has been successfully used to study several enzymatic reactions as well as solution reactions.

Chorismate mutase (CM) catalyses the pericyclic rearrangement of chorismate to prephenate, which is a key step in the shikimate pathway for biosynthesis of phenylalanine and tyrosine in bacteria, fungi, and higher plants. We utilized EVB/MD free-energy perturbations at a range of temperatures [2], allowing us to extract activation enthalpies and



⬆ **Figure 1:** A) Reaction free-energy profiles obtained from EVB simulations at 298 K and B) the corresponding Arrhenius plots in the temperature range 288 – 313 K for the conversion of chorismate to prephenate in aqueous environment (blue) and in the chorismate mutase enzymes from *B. subtilis* (red) and isochorismate pyruvate lyase from *Pseudomonas aeruginosa* (PchB) (black). Error bars are displayed but are smaller than the data point. Illustration: Ryan Scott Wilkins.

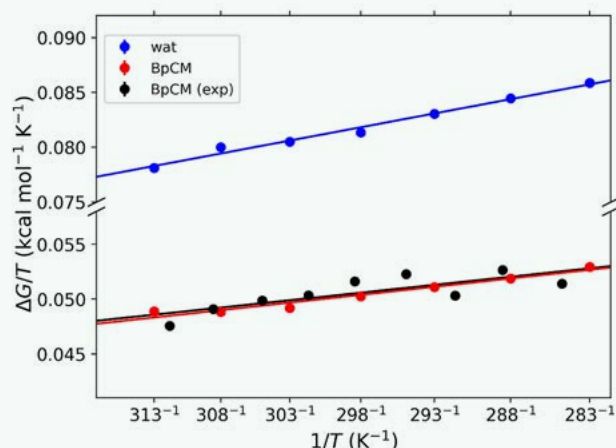
entropies from an Arrhenius plot of reaction free energies of the reaction catalysed by a monofunctional *Bacillus subtilis* chorismate mutase and the promiscuous enzyme isochorismate pyruvate lyase of *Pseudomonas aeruginosa* (Figure 1). In comparison to the uncatalysed reaction, our results show that both enzyme-catalysed reactions exhibit a substantial reduction in activation enthalpy, while the effect on activation entropy is relatively minor, demonstrating that enzyme-catalysed chorismate mutase reactions are enthalpically-driven. Furthermore, we observe that the monofunctional chorismate mutase from *B. subtilis* more efficiently catalyses this reaction than its promiscuous counterpart. This is further supported by a structural analysis of the reaction pathway at the transition state, from which we identified key residues explaining the enthalpically-driven nature of the reactions, and also the

difference in efficiencies between the two enzymes. This study has been carried out by Ryan Wilkins and has been submitted [2].

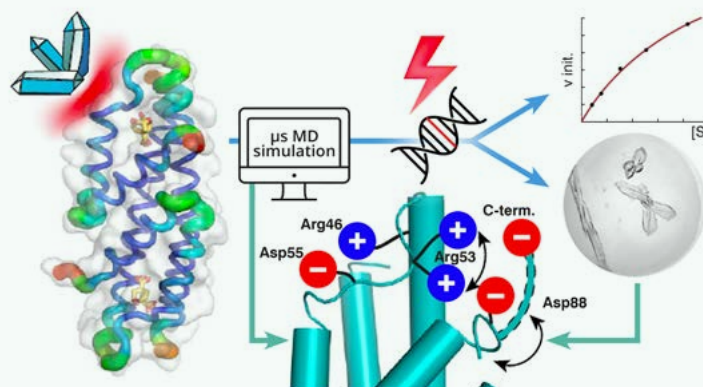
With the successful calculation of the thermodynamic activation parameters in mind, the search for a cold-adapted chorismate mutase was initiated. The psychrophilic bacterium *Bacillus pumilus* has a gene that encodes for a chorismate mutase, which was produced, purified, and characterized. The crystal structure is nearly identical to that from *Bacillus subtilis*, with a root-mean-square deviation of 0.28 Å, a sequence identity of 69% and a sequence similarity of 93%. To our surprise, the thermodynamic characterization and calculations revealed activation parameters virtually identical to the mesophilic chorismate mutase (Figure 2) and it is therefore not a cold-adapted chorismate mutase, as hoped

for. The Arrhenius plot was constructed by fitting  $\Delta G^\ddagger/T$  vs.  $1/T$  using a linear regression, yielding thermodynamic activation parameters of  $\Delta H^\ddagger = 12.9 \pm 0.5$  kcal mol<sup>-1</sup> and  $T\Delta S^\ddagger = -2.1 \pm 0.5$  kcal mol<sup>-1</sup>. An Arrhenius plot and similar regression of experimental results, obtained by fitting  $\ln(k_{\text{cat}})$  versus temperature values obtained from UV/VIS spectroscopy in a chorismic acid assay, gave activation parameters of  $\Delta H^\ddagger = 13.1 \pm 0.4$  kcal mol<sup>-1</sup> and  $T\Delta S^\ddagger = -2.1 \pm 0.5$  kcal mol<sup>-1</sup>. The thermodynamic activation parameters predicted from the Arrhenius-plot approach applied to both simulation and experimental results are in excellent agreement with each other. This study, which was carried out by Ryan Wilkins supported by Bjarte Aarmo Lund, has also been submitted [2].

In addition to chorismate mutase, we are currently investigating the enthalpy-entropy balance using several other



⬆ **Figure 2:** Arrhenius plots from 283 to 313 K comparing spectrophotometry (black) and EVB results (red) for *bacillus pumilus* chorismate mutase (BpCM) to EVB results of the reaction in an aqueous environment (blue).



⬆ **Figure 3:** The structure of MtCM (left); key interactions responsible for the conformation of the catalytic loop, investigated by computational and experimental means (right). Illustration from Ref. [4], under CC 4.0 license.

enzymes, including lipases, esterases, and nucleases. For all of these enzymes, we have developed a pipeline that allows us to produce enzymes and variants for biochemical characterization and computer simulations. The combined theoretical and experimental setup opens up for rational redesign of catalytic activity and thermostability of enzymes based on the thermodynamics and structural insights obtained with EVB simulations.

#### Structure-activity relationship in chorismate mutase

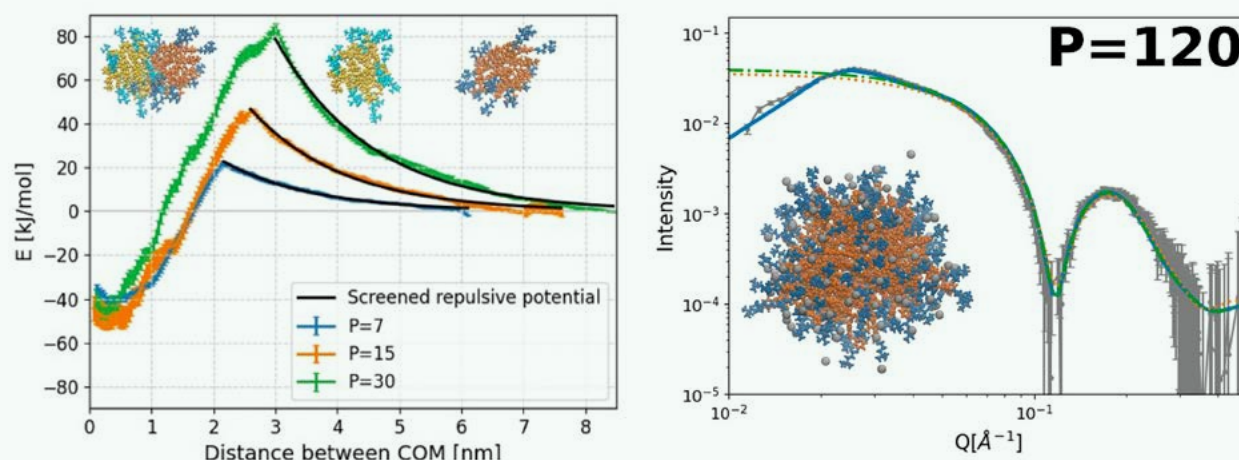
Chorismate mutase is the only known natural enzyme that catalyses a pericyclic reaction: the Claisen rearrangement, transforming chorismate into prephenate in a key step of the shikimate metabolic pathway toward the synthesis of tyrosine and phenylalanine amino acids.

Unlike chorismate mutase, the enzyme from *Mycobacterium tuberculosis* (MtCM) has only low activity on its own. However, its activity is accelerated more than 100-fold by complex formation with partner enzymes. In the past year, PhD candidate Helen V. Thorbjørnsrud in the group of Prof. Ute Krengel (Hylleraas affiliate at UiO) investigated the structural and dynamical properties of this enzyme by means of all-atom classical molecular dynamics. Her studies comprised microsecond dynamics of the wild-type enzyme, together with highly efficient mutants obtained in the lab by directed evolution. Successively, interesting point mutations were investigated by additional simulations run by PhD candidate Manuel Carrer and probed experimentally in collaboration with the group of Prof. Peter Kast at ETH Zürich.

The study revealed how the catalytic power of this enzyme is enhanced by the stabilization of a kinked conformation in the catalytic H2–H3 loop, which guarantees the preorganization of the enzyme, thus reducing the enthalpy penalty upon substrate binding (Figure 3).

#### Micelle kinetics in photo-switchable surfactants

Controlling the process of self-assembly is important for many technological applications. In this regard, photocontrollable micelles based on molecular switches offer promising means to manipulate nano-structures without intervening on the thermodynamic parameters. However, the kinetic pathways for molecular aggregation and relaxation after photoexcitation are not known, hampering rational design for such systems.



⬆ **Figure 4:** Left: Potential of mean force for the fusion of two micelles of size  $P$  as a function of the distance between the two centres of mass. Right: Experimental SAXS spectrum (grey line), fits from theoretical models (dashed lines), and prediction from MD simulations (continuous blue line). Illustration from Ref. [5], under CC 4.0 license.

In this study, PhD candidate Victoria A. Bjørnstad combined small-angle X-ray scattering (SAXS) and all-atom molecular dynamics studies to investigate the kinetics of azo-TMA, a prototypical photo-switchable system characterized by a photo-active diazobenzene hydrophobic core and a hydrophilic positively charged amine group. We found that the quasi-equilibrium aggregation kinetics of *trans*-azo-TMA is controlled by the repulsive electrostatic interactions between the charged micelle surfaces, with a repulsion potential fitting very well standard models for charge screening in electrolyte solutions. The repulsion potential can become unsurmountable, the activation barriers of micelles larger than 30 surfactants becoming higher than 80 kJ mol<sup>-1</sup>. Umbrella sampling simulations quantified how a mechanism of micelle formation based on exchange of monomeric surfactants, where the charge

repulsion increases less with the size of the micelle, is more effective in achieving micellar growth (Figure 4).

Comparison with experimental SAXS data confirmed that the experimental samples are dominated by micelles formed from about 120 surfactants. Complementary coarse-grained simulations using smoothed hybrid-particle-field potentials ran by Dr. Xinmeng Li with the HyMD software showed that this approach can be very effective at achieving aggregates of such size without recourse to computationally expensive enhanced sampling.

Bjørnstad further investigated the nonequilibrium dynamics that occurs upon sudden *trans*-to-*cis* photoisomerization and upon mixing with co-solvent dimethylformamide (DMF). Her combined experimental and computational investigation demonstrated how the

relaxation from an out-of-equilibrium state can follow a fast relaxation via micellar fission, depending on the ratio of photo-isomerized molecules, followed by a slow re-equilibration by individual molecular exchange (Figure 4). This study is carried out under the supervision of Hylleraas affiliate Prof. Reidar Lund [5].



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3. R. Wilkins, B.A. Lund, G.V. Isaksen, J. Åqvist and B.O. Brandsdal, *Submitted*.
4. H. V. Thorbjørnsrud, L. Bressan, T. Khatanbaatar, M. Carrer, K. Würth-Roderer, G. Cordara, P. Kast, M. Cascella, and U. Krengel, *Biochemistry*, 2023, **62**, 782–796.
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# Revised Research Themes

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In preparation for the (later cancelled) midterm evaluation and in response to changes in centre personnel and in the field of theoretical chemistry itself, new research plans were drawn up in 2022. These plans will become effective from July 1, 2023.

Following extensive discussions among PIs and senior researchers in early 2022, it was decided that the new project plan and strategy should not deviate more than necessary from the original plans but should be updated to reflect the current state of the research directions at the centre, some of which were not part of the original strategy, and some of which are related to new developments in the field (such as machine learning). In addition, some changes are forced upon us by the departure of senior members of the centre. At the same time, several talented young researchers have risen to prominence since the establishment of the centre and are ready to take on new responsibilities in the second centre period.

In recent years, the members of the centre have secured support for many external projects that are now either ongoing or starting up – about half of these are owned by younger researchers. Our new strategy has been developed such that these projects would complement our plans and highlight the success of the young members of the centre.

A general trend in our research has been towards more complex systems and greater complexity in the modelling with a stronger focus on time-dependent phenomena and time-dependent modelling. These were important elements of the original strategy but are more strongly emphasized in the new strategy. In the following, the new RTs are briefly described.



## RT1: Molecular quantum mechanics

### Leaders:

Luca Frediani and  
Thomas Bondo Pedersen

*To help control matter at the scale of the electron, reliable yet affordable approaches to strong electron correlation and basis-set completeness will be developed, including methods accounting for the coupling of electronic and nuclear motion in the presence of electromagnetic fields.*

### Comments:

With the increased focus on electronic and nuclear dynamics induced or affected by external fields during the first five years of the Hylleraas Centre, RT1 will be extended beyond traditional electronic-structure theory to include the development of computational tools for solving the full molecular Schrödinger equation, both time-dependent and time-independent, within and without the Born-Oppenheimer approximation. Reflecting these changes, the title of RT1 is changed to *Molecular quantum mechanics*. The leadership of RT1 remains unchanged.



## RT2/6: Multiscale modelling and complexity

### Leaders:

Bjørn-Olav Brandsdal and  
Michele Cascella

*We develop methods, software and virtual-reality tools at multiple resolutions to enable the study of large-scale responses from local perturbations. We apply them to bio-soft materials and address mechanisms associated with health threats like bacterial drug resistance.*

### Comments:

New multiscale modelling tools, including virtual-reality tools and coarse-grained molecular dynamics software, have been developed within RT2 and applied within RT6. These tools have now reached a level of maturity that allows combinations of methods ranging from quantum-mechanical to mesoscale models. These activities are merged into RT2/6, which aims at developing multiresolution schemes tailored to specific classes of problems for investigating phenomena at different scales and how they affect each other. The leadership is that of RT2 and RT6.



### RT3/4: Matter-field interactions

Leaders:  
Michal Repisky and Erik Tellgren

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*We develop high-accuracy quantum dynamics methods to simulate ultrafast laser-driven processes and to describe nontrivial topological phases in periodic magnetic systems. We develop theory and methods for strong-field chemistry, including the simulation of spectra from magnetic white dwarf stars.*

#### Comments:

Advanced molecular spectroscopies and the physics and exotic chemistry of molecules in strong magnetic fields have been subject of RT3 and RT4, respectively. At the same time, studies of laser-induced electronic quantum dynamics in the presence of magnetic fields have been initiated within RT3, while the prediction of spectra from magnetic white dwarf stars is a central goal in RT4. With Ruud and Helgaker stepping down, it becomes natural to merge the RT3 and RT4 into RT3/4, led by Repisky and Tellgren, who have been key personnel of these RTs since the beginning.



### RT5: Chemical transformations

Leaders:  
Abril Castro and Ainara Nova

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*We apply computational methods to understand chemical reactions in collaboration with experimental groups. A key goal is to simulate reactivity in complex environments, including reaction networks, spectroscopic processes in solution, and metal-organic frameworks.*

#### Comments:

In collaboration with experimentalists, understanding and predicting catalysts for sustainable chemical transformations through state-of-the-art *ab initio* simulations and data analysis has been and remains a major goal of RT5. Nova continues as leader of RT5 alongside Castro, who brings in expertise in advanced computational studies of spectroscopic processes in solution. This is a new research line in RT5, which brings the theoretical investigations even closer to experimental research, where advanced molecular spectroscopies are important tools for understanding catalysis.



### RT7:

## Machine learning and mathematical methods for chemistry

### Leaders:

David Balcells and Simen Kvaal

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*To simulate more complex chemical systems and to advance electronic-structure methods, we will use a synergistic approach, combining the development of new machine learning models with the mathematical study of coupled-cluster and density-functional theories.*

### Comments:

In the first centre period, machine learning and mathematical methods have developed into important research areas at the centre. Machine learning is used for data-driven design of catalysts, for parameterization of coarse-grained force fields and density functionals, and for time-series forecasting; mathematical methods are used for analysing and developing electronic-structure theories, and for quantum-mechanical time propagation schemes. Their combination is a unique strong point for the centre, making it natural to collect them in RT7, led by Balcells and Kvaal.



### RT0:

## Hylleraas Software Platform

### Leaders:

Bin Gao and Simen Reine

---

*To support, enhance, and synergize the research among the five other research themes, we will develop a unifying software platform that seamlessly links and runs diverse software in simple, efficient, and novel ways.*

### Comments:

Researchers at the centre have produced and contributed to several open-source software packages, for a large variety of simulation purposes on multiple scales in space and time. At the same time, complex computational protocols and data acquisition and analysis have been developed. To unite these efforts, the Hylleraas Software Platform project (initiated in 2020) will be developed in RT0 under the leadership of Gao and Reine, both experienced software developers. The platform will act as a catalyst for both internal and external collaborations and will further be utilized for teaching purposes.





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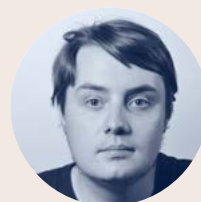
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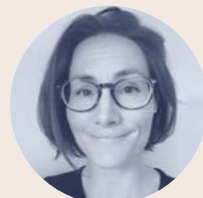
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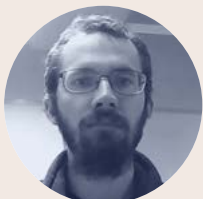
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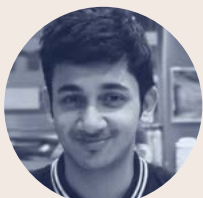
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**Odile Eisenstein**

*Professor  
UiO*



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Haugaard Olsen**

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**Laura Elizabeth  
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**Stella Stopkowicz**

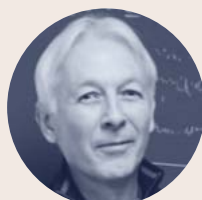
*Professor  
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**Andrew Michael Teale**

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## Affiliates



**Tor Flå**

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**Reidar Lund**

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**Mats Tiset**

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**Cathrine  
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*Communication  
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**Morten  
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**Mauritz  
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**Audun Skau  
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EPF Lausanne,  
Switzerland*



# Publications 2022





# Publications 2022

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# Funding and Staff Report

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# Funding 2022

(in NOK 1000)



<b>Basic funding as agreed in the Centre contract</b>	
Funding from Research Council (CoE grant)	14 855
Funding from home institutions	11 700
<b>Total basic funding</b>	<b>26 555</b>
<b>Additional funding from external projects</b>	
Research Council funding	26 676
International funding	2 643
Public funding	
Private funding	1 359
<b>Total additional funding</b>	<b>29 848</b>
<b>Total funding</b>	<b>56 403</b>



# Staff Report 2022

(in man-years)

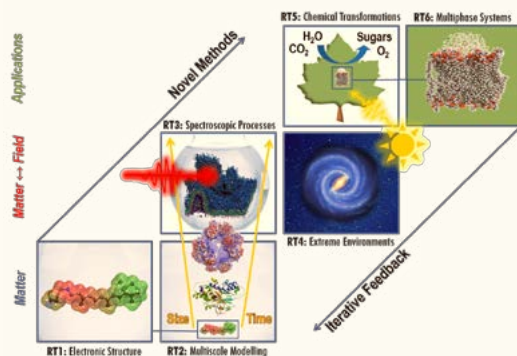


Professors and researchers	16.7
PhD Candidates	22.6
Postdoctoral Fellows	12.9
Visiting Researchers	4.1
Administrative and technical staff	4.1
<b>All staff</b>	<b>60.4</b>

# Avslør naturens hemmeligheter med laptopen som ditt laboratorium

## Hylleraas-senteret for teoretisk kjemi bringer deg til forskningsfronten

Ved Hylleraas-senteret utvikler og anvender vi simuleringsteknikker for å forstå, tolke og forutsi ny kjemi, fysikk og biologi for molekyler i komplekse og ekstreme miljøer.



Våre forskningstema (RT)

Gjennom et bachelor- eller masterprosjekt ved Hylleraas-senteret kan du fordype deg i tema som matematiske modelleringsmetoder, programmering og maskinlæring innen teoretisk kjemi, og du vil få en dyp innsikt i hvordan naturen frembringer molekyler struktur og dynamikk.

I tillegg vil du tilegne deg etterspurte spesialkunnskaper for en fremtidig karriere.

**Finn ditt prosjekt hos oss i kolonnen til høyre.**



Med **Michele Cascella** som veileder vil du krysse ulike størrelsesskalaer, fra kvantemekanikk til statistiske felter, for å utforske kjemiske reaksjoner, selv-aggregering og myke materialer. En tett tilknytning mellom teori og eksperiment gir også muligheter for samarbeid med eksperimentallister.

Kontakt: [michele.cascella@kjemi.uio.no](mailto:michele.cascella@kjemi.uio.no)

Sammen med **Thomas Bondo Pedersen** kan du blant annet utvikle helt nye beregningsmetoder for å simulere kvantedynamikken til molekyler i ultrakorte laserpulser og sterke magnetfelter i tidsskalaer ned til femto- og attosekunder. Du vil utvikle *high-performance computing* algoritmer og få innsikt i avanserte spektroskopiske teknikker.

Kontakt: [t.b.pedersen@kjemi.uio.no](mailto:t.b.pedersen@kjemi.uio.no)



Sammen med **Aina Nova** vil du ta i bruk simuleringsteknikker for å utforske struktur og reaktivitet for katalysatorer nødvendig for gjenvinning verdifulle kjemikalier fra CO<sub>2</sub>. Forskningen omfatter mange ulike systemer, blant annet homogene og single-site heterogene katalysatorer.

Kontakt: [a.n.flores@kjemi.uio.no](mailto:a.n.flores@kjemi.uio.no)

Med **David Balcells** som veileder vil du ta i bruk maskinlæring og high throughput virtuell screening for å utforske katalysatorer for fornybare energiprosesser. Du vil tilegne deg ferdigheter innen dataanalyse, programmering og evaluering av prediktive modeller.

Kontakt: [david.balcells@kjemi.uio.no](mailto:david.balcells@kjemi.uio.no)



Sammen med **Simen Kvaal** kan du blant annet utvikle nye metoder for å løse den tidssavhengige Schrödingerlikningen for molekyler på datamaskinen, utforske kvantekaos og grenselandet mellom kvante- og klassisk mekanikk, og benytte maskinlæring for å gjøre kvantekjemiske beregninger kjappere.

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Med **Erik Tellgren** som veileder kan du utforske eksotisk kjemi for molekyler i sterke magnetfelter, hvor nye kjemiske bindinger og fysiske effekter gjør seg gjeldende. Prosjekter kan fokuseres mot anvendelse, programmering av beregningsmetoder, eller matematiske/teoretiske aspekter.

Kontakt: [erik.tellgren@kjemi.uio.no](mailto:erik.tellgren@kjemi.uio.no)



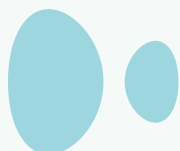
Sammen med **Abril Castro** vil du lære hvordan du simulerer strukturelle, elektroniske og spektroskopiske egenskaper. Mulige emner er modellering av NMR-egenskaper for å forutsi og tolke eksperimenter, analyse av kjemiske bindinger i løsnings- og faststoffsystemer, og studier av molekyler som inneholder tunge atomer.

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 Hylleraas



“The [Hylleraas] Centre has been highly successful in its nationwide goals to train young people and to become a beacon of excellence in chemistry. It is internationally recognized as a world-leading centre for theoretical chemistry in its broadest sense. This accomplishment of the Centre is truly remarkable and cannot be rated high enough. As a result, it puts a burden onto the universities, as it is now their duty to make sure that the investment can be sustained after the end of the Centre in 2027.”

– excerpt from the 2022 Report of the Scientific Advisory Committee of the Hylleraas Centre; see page 47.





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