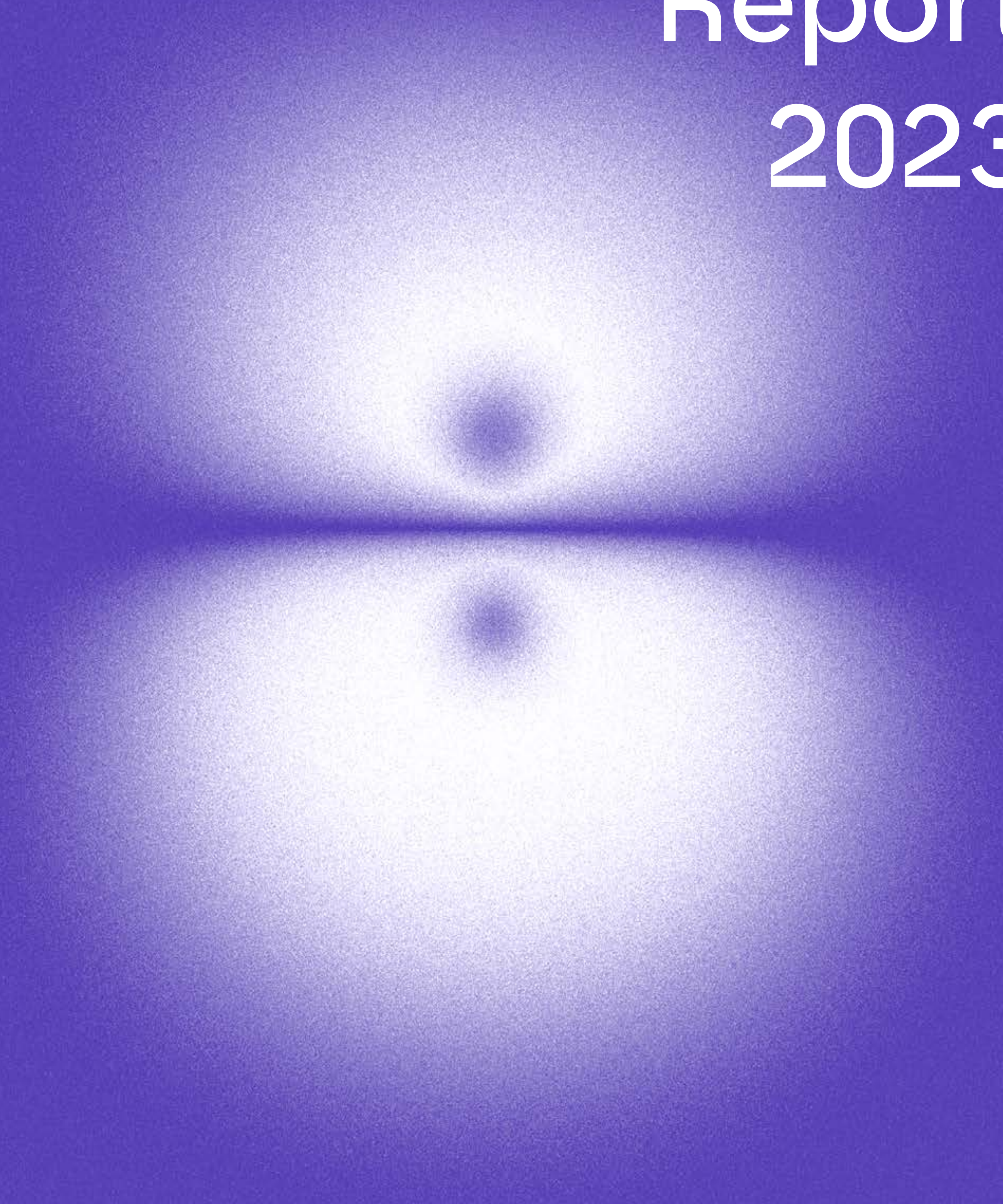
Hylleraas

Annual Report 2023



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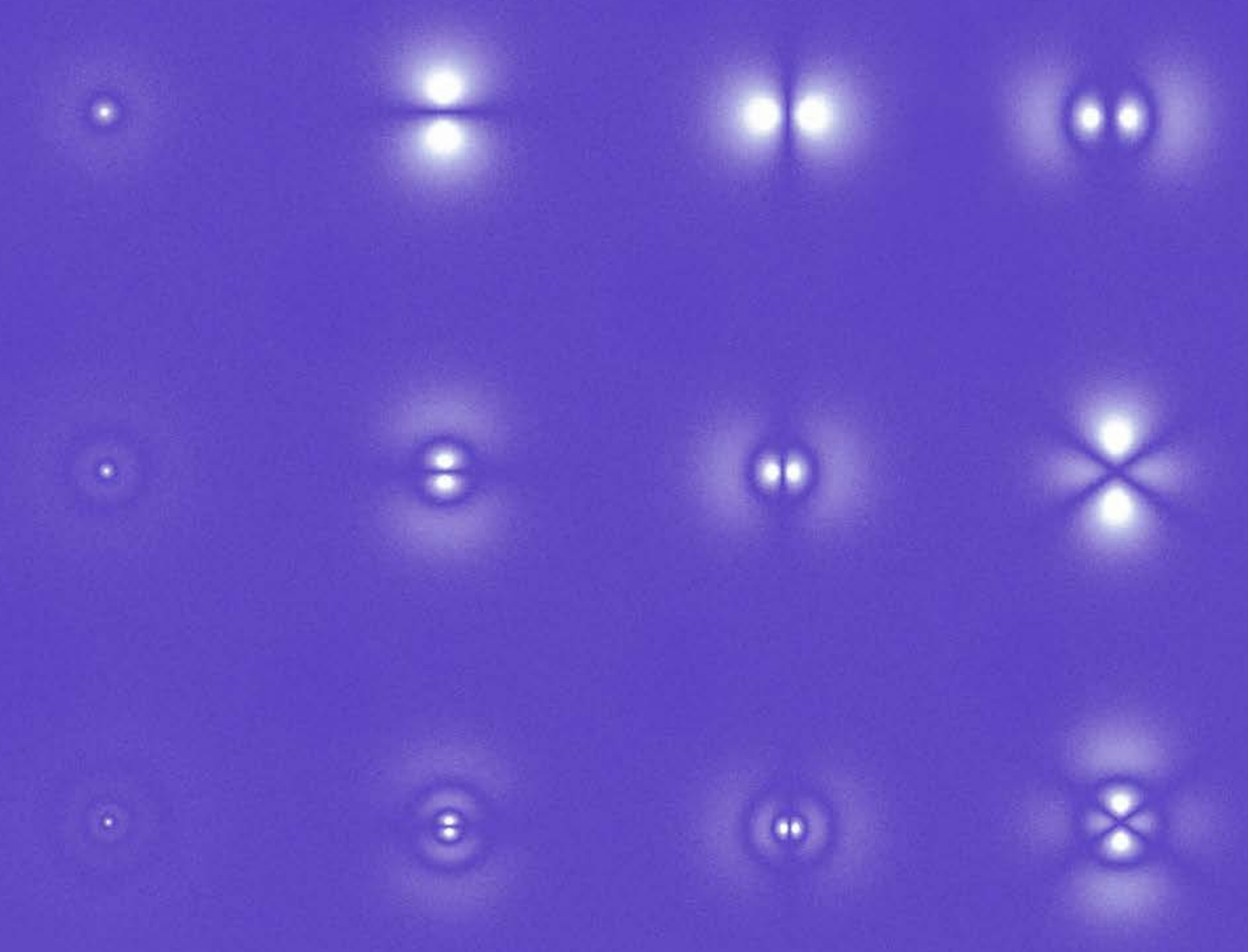

Hylleraas Centre for Quantum Molecular Sciences

The Hylleraas Centre for Quantum Molecular Sciences 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. Established on October 1, 2017, for a period of ten years, the Hylleraas Centre 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.

At the end of 2023, the Hylleraas Centre is one of 19 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.

Contents

From the Directors

Changes characterized the year 2023 at the Hylleraas Centre, as it entered its second period of operations: new leadership, updated research plans implemented in new Research Themes led by a new team of Principal Investigators, new members of the Scientific Advisory Committee, and new members of the Board of Directors. Concomitantly, productivity increased to a record high as a direct consequence of the high success rate in securing research grants from the Research Council of Norway in 2021.

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On July 1, 2023, Prof. Trygve Helgaker stepped down as director of the Hylleraas Centre. We offer our warmest gratitude to Trygve for his dedication over decades to build an internationally leading research environment covering most aspects of modern theoretical chemistry and look forward to continued collaboration with him in his new role as Professor emeritus and affiliate member of the centre. Among other things, Trygve will chair the organization of the 13th Triennial Congress of the World Association of Theoretical and Computational Chemists, which will take place in Oslo in June 2025.

Also on July 1, a younger generation of scientists were appointed Principal Investigators (PIs) at the Hylleraas Centre. We welcome Dr. Abril Castro, Dr. Erik Tellgren, Dr. Simen Kvaal, Dr. Simen Reine, and Dr. Bin Gao to the Management Team. They join the leadership and PIs Michal Repisky, Bjørn-Olav Brandsdal, Ainara Nova, David Balcells, and Michele Cascella in leading the new Research Themes (RTs).

- **RT1:** Molecular Quantum Mechanics (Pedersen, Frediani)
- **RT2/6:** Multiscale Modelling and Complexity (Cascella, Brandsdal)
- **RT3/4:** Matter-Field Interactions (Repisky, Tellgren)
- **RT5:** Chemical Transformations (Nova, Castro)
- **RT7:** Machine Learning and Mathematical Methods (Balcells, Kvaal)
- **RT0:** Hylleraas Software Platform (Reine, Gao)

We are convinced the PIs will excel at the very important tasks of leading the ambitious new research plans and managing the centre with more than 80 members and affiliates.

In 2023, Prof. Anette Bayer stepped down as a member of the Hylleraas Board of Directors. We express our appreciation for her contributions to the Board and extend our sincere thanks. We are pleased to welcome Prof. John Sigurd Mjøen Svendsen, who assumed her role in July 2023. Svendsen is a professor of chemical synthesis and analysis, currently serving as Dean of Research and Development at the Faculty of Science and Technology, UiT – The Arctic University of Norway, and as Chief Scientific Officer at Amicoat AS. His extensive leadership experience will bring important insights to the Hylleraas Board in the second period of operation. At the end of 2023, Prof. Nathalie Reuter announced that she wishes to step down as a member of the Hylleraas Board. She has generously offered to remain in the Board until a replacement has been identified, presumably in early 2024.

Three members of the Scientific Advisory Committee (SAC) stepped down as the centre entered its second period of operation. Chair of the SAC, Prof. Chantal Daniel, and SAC members Prof. Ursula Röthlisberger and Prof. Serena DeBeer were succeeded by Prof. Veronique Van Speybroeck, University of Ghent, and Prof. Carme Rovira Virgili, University of Barcelona. Van Speybroeck is a world-leading researcher in the modelling of nanoporous materials for catalysis, and Rovira Virgili has made important contributions to the understanding of chemical reactions in glycoprocessing enzymes for biomedical and biotechnological applications. They join Prof. Markus Reiher and Prof. Peter Schmelcher as members of the SAC, with Reiher taking over as chair after Daniel. We thank Daniel, Röthlisberger, and DeBeer for their important and exceptionally useful advisory work in the first period of operation and look forward to fruitful collaborations with the new SAC in the coming years.

We offer our warmest gratitude to Trygve [Helgaker] for his dedication over decades to build an internationally leading research environment covering most aspects of modern theoretical chemistry

The Annual Hylleraas Meeting is important to the centre activities and to maintain the close ties between the Oslo and Tromsø sides of the centre. The 2023 edition of the meeting was held at Sommarøy and organized by Tonje Reinholdt Haugen and Gabriel Gerez, the Tromsø representatives of the Young Researcher Parliament (YRP). Our sincere thanks go to them for ensuring the success of the Sommarøy meeting.

Several other impactful events were organized by centre members in 2023:

- **Hylleraas School** (Inga Schmidtke, Tonje Reinholdt Haugen, David Balcells, Simen Reine)
- **Norway-Japan Academic Network Seminar** (Michiko Atsumi, Erik Tellgren, Trygve Helgaker, Simen Reine)
- **Hylleraas Hackathon** (Magnus Ringholm)
- **NordCO2 Annual Meeting** (Ainara Nova)
- **CAS Workshop on Molecular Quantum Dynamics** (Simen Kvaal, Thomas Bondo Pedersen)
- **Numerical Methods in Quantum Chemistry** (Luca Frediani)
- **CECAM Workshop: Computational Molecular Medicine Across Three Decades** (Michele Cascella)
- **TRAINS Conference** (Erik Tellgren, Michele Cascella, Thomas Bondo Pedersen)
- **NordCO2 Winter School** (Ainara Nova)

We extend our gratitude to all organizers for their dedicated efforts in making these events a success.

Hvorfor Det?, a science fair/poster competition for children aged 8-12 from Ila Skole in Oslo, was hosted for the second consecutive year by the Hylleraas Centre and held at the Department of Chemistry in Oslo. We thank the centre members Cathrine Strøm, Morten Ledum, Benedicte Sverdrup Ofstad, and Håkon Emil Kristiansen for their contributions to this event, which was attended by roughly 200 people. We also extend our thanks to Trygve Helgaker and Erik Tellgren who took the lead in several other outreach events throughout 2023.

Despite a one-year suspension of the Groundbreaking Research Programme (FRIPRO) of the Research Council of Norway (RCN) – which is the main source of external funding at the Hylleraas Centre – we are delighted to congratulate Dr. Sigbjørn Løland Bore on winning an RCN Young Research Talent grant, a YoungCAS grant, and a EuroHPC JU grant in 2023. We also congratulate Dr. Marco Bortoli on winning a EuroHPC JU grant and Dr. David Balcells on his participation in a newly granted EU-MSCA Doctoral Training Network. These grants revitalize the centre by bringing in new people and new research activities.

Prof. Thomas Bondo Pedersen
Director

Our congratulations and best wishes are extended to the six young centre members who defended their PhD theses in 2023: Victoria Ariel Bjørnestad, Samiran Sen, Linn Neerbye Berntsen, Manuel Carrer, Mateu Montserrat Canals, and Benedicte Sverdrup Ofstad.

Every year, several social events are organized at the Hylleraas Centre, including the Christmas parties, Easter parties, and Friday beers. In a tradition that goes back to 2009, with a two-year break during the pandemic, an annual summer party has been held in the garden of Trygve Helgaker and his wife Barbara Helgaker, with an ever-increasing attendance. Their last garden party was held in June 2023, with more than 40 guests in glorious weather. We would like to thank Barbara for the efforts she has put into making these parties such a popular and well-attended event at the Hylleraas centre in Oslo.

Finally, we would like to thank all members and affiliates of the Hylleraas Centre for their excellent work in 2023, as detailed in this report.

Prof. Luca Frediani
Deputy Director

From the Board of Directors

The year 2023 marks the transition to the second period of the Hylleraas Centre, which formally began on July 1. As decided in 2022 the new leader of the Hylleraas Centre is Prof. Thomas Bondo Pedersen, who succeeded Prof. Trygve Helgaker. Updated research plans reflecting new scientific directions were implemented in restructured research themes. The new RTs are led by a younger generation of researchers.

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In late 2022, due to extensive budget cuts, the Research Council of Norway (RCN) decided to cancel the mid-term evaluations of the Centres of Excellence established in 2017 and, therefore, the second funding term 2023-2027 was automatically approved. The management team had already laid the groundwork for the mid-term evaluation, which has led to the new structure of the Hylleraas Centre. The board was pleased to see the new structure taking shape: new and exciting themes covering the important aspects of mathematical methods and machine learning (RT7) and recognizing the importance of open-source software development as a unifying aspect and explicit deliverable in theoretical chemistry (RT0). In addition, two of the original RTs have revised goals (RT1 and RT5), and four RTs have been merged into two (RT3/4 and RT2/6) to adapt to the needs of the second term.

The RCN carried out the planned mid-term visit to the Centre in October 2023, and witnessed the strong commitment of the new Centre leader as well as the PIs towards the new strategy.

Compared with the two previous years, 2023 has seen a reduction of additional funding granted to members of the Hylleraas Centre. This is not unexpected, however. First, most PIs already hold one or more grants, including RCN FRIPRO grants, which provide most of the additional funding to the centre. Second, the RCN suspended the entire FRIPRO funding scheme in 2023 except for young researcher talent (YRT) grants. A few notable exceptions for 2023 are the YoungCAS project and the FRIPRO-YRT grant awarded to Sigbjørn L. Bore, and the participation in the MSCA network for machine learning in chemistry and material science of David Balcells, showing how the centre is both a valuable environment to promote young researchers and how the shift towards Machine Learning and artificial intelligence is already paying dividends.

The plentiful availability of computational resources is an essential component for the success of the Hylleraas Centre. The financing model of Sigma2, the Norwegian provider of high-performance computing infrastructure, has recently changed from a fixed price

> The board would like also to thank Prof. Trygve Helgaker for his tenure as Director of the Hylleraas Centre. His dedication and passion for his work have been deeply appreciated. We are glad to see that he will still have an active role in the centre.

agreement (the centre paid 1.6 MNOK/year) to a pay-per-use (1M hours = 50 kNOK) model. Given the extensive use of computational resources by the centre (around 80 Mhours/year or 4 MNOK/year), the new model would heavily affect the budget of the centre. The science faculties at both UiT and UiO realized the severity of the situation and promised to support the centre financially for the extra costs. However welcome, this is a temporary fix and the board is deeply concerned about how the centre and computational research will be affected in the future by the current model: recent development in artificial intelligence and machine learning show that computing will have an ever larger role in cutting edge research and limiting its availability will negatively affect the Hylleraas Centre and theoretical chemistry at large.

The board would like also to thank Prof. Trygve Helgaker for his tenure as Director of the Hylleraas Centre. His dedication and passion for his work have been deeply appreciated. We are glad to see that he will still have an active role in the centre, in preparation for the WATOC 2025 congress in Oslo. We are sure that his mentorship and advice will be extremely valuable for the younger generation of scientists that the centre is nurturing.

We conclude by welcoming Prof. Thomas Bondo Pedersen as the new head of the Hylleraas Centre, wishing him the best of luck for his tenure.

Unni Olsbye (chair)
Professor
Dept. Chemistry
UiO

John Sigurd Mjøen Svendsen
Professor
Dept. Chemistry
UiT

Atle Jensen
Professor
Dept. Mathematics
UiO

Nathalie Reuter
Professor
Dept. Chemistry
UiB

Aslak Tveito
Professor
Simula Research Laboratory

From the Young Researchers

The Hylleraas Centre fosters a supportive environment for young researchers through the Young Researchers Parliament (YRP). All young researchers - Master's students, PhD candidates and postdocs as well as visiting students, actively contribute to shaping the centre's initiatives while supporting their professional growth and development. Two elected representatives from each node bridge the interaction between the YRP and the seniors by being part of the Management Team of the Hylleraas Centre.

Throughout the year, the YRP coordinated events to foster idea exchange and facilitate bidirectional communication between young researchers and the management team. Through monthly pizza meetings at UiO, and social events at UiT, the YRP served as a conduit for sharing discussions from the management level and for bringing forward matters of concern to the management team.

The YRP also oversees the organisation of the annual meeting. This year, the Tromsø node of the YRP took charge of arranging the Hylleraas annual meeting. Held over three days at Sommarøy (September 5-7), the meeting featured 5-minute thesis presentations by the Master's and PhD students, including visiting PhD students, alongside a series of academic seminars. Internal speakers such as Kenneth Ruud and Simen Reine delved into career development in academia and high performance computing, offering insights into effective practices. The lineup also included talks by external speakers from industry and communication, focusing on career differences in and outside of academia, and on strategies for good science communication. The meeting concluded with the YRP annual assembly, facilitating discussions between the YRP nodes to enhance representation, foster internode collaboration, address recent and upcoming changes within the centre and plan future events.

In 2023, the visitor programme of the YRP continued to provide valuable opportunities for young researchers to broaden their academic horizons. By inviting esteemed professors and researchers from external institutions, the programme serves as a pivotal platform for fostering collaborative discussions, hosting seminars, and facilitating knowledge-sharing endeavours. On behalf of the YRP, UiO representatives Marinella de'Giovanetti and Hannes Kneiding invited Prof. Marcel Swart, director of the IQCC institute at the University of Girona (Spain), for a three-day visit to the Hylleraas Centre in Oslo on November 8-10. During his visit, Prof. Swart held two seminars. In the first seminar addressed to the YRP, he shared

In 2023, the visitor programme of the YRP continued to provide valuable opportunities for young researchers to broaden their academic horizons. By inviting esteemed professors and researchers from external institutions, the programme serves as a pivotal platform for fostering collaborative discussions, hosting seminars, and facilitating knowledge sharing endeavours.

his insights into navigating an academic career. In the second seminar addressed to the entire centre, he shared his groundbreaking research on transition metals, spanning applications in chemistry to theoretical developments, thereby fostering interdisciplinary dialogue and knowledge exchange within the scientific community. The YRP visitor programme also includes hosting Hylleraas alumni to give talks at the seminar series. In 2023, former PhD students at UiO Lluís Artús Suàrez and Fabian Faulstich gave Alumni seminars sharing their experience and career development as former members of the centre.

At each node of the Hylleraas centre, two representatives of the YRP are elected to serve as the vital link between early-career researchers and the leadership of the centre. Towards the end of the year, both nodes held an election to replace one of their representatives who were stepping down. At UiO, Hannes Kneiding left his position to Oliver Bohle, while Gabriel Gerez was superseded by Quentin Pitteloud at UiT. The newly elected representatives joined Marinella de'Giovanetti (UiO) and Tonje Reinholdt Haugen (UiT) to represent the young researchers. We would like to thank Hannes and Gabriel for their contributions to the centre.

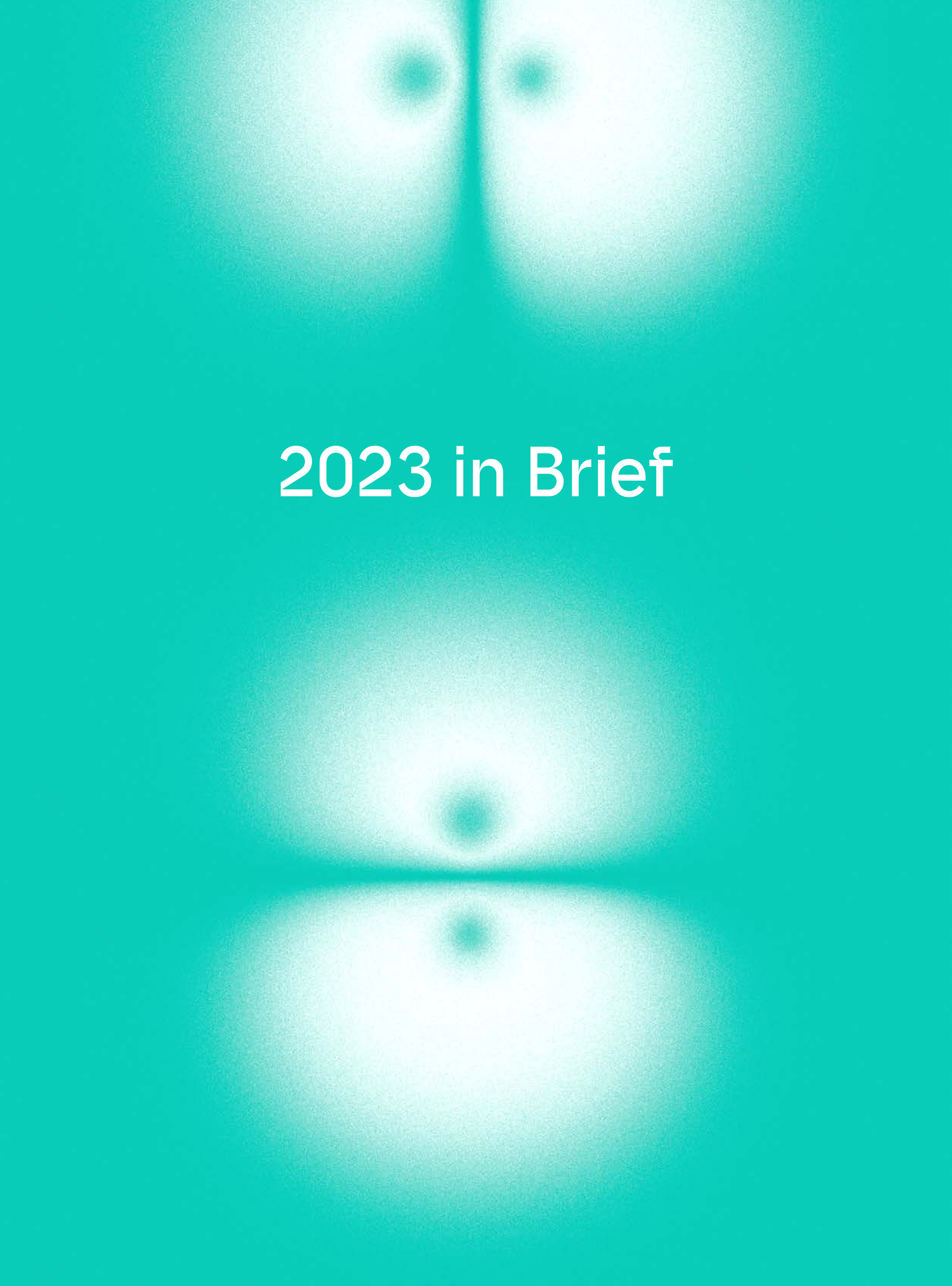
Marinella de'Giovanetti

Quentin Pitteloud

Tonje Reinholdt Haugen

Oliver Bohle

Elected YRP representatives 2023



2023 in Brief

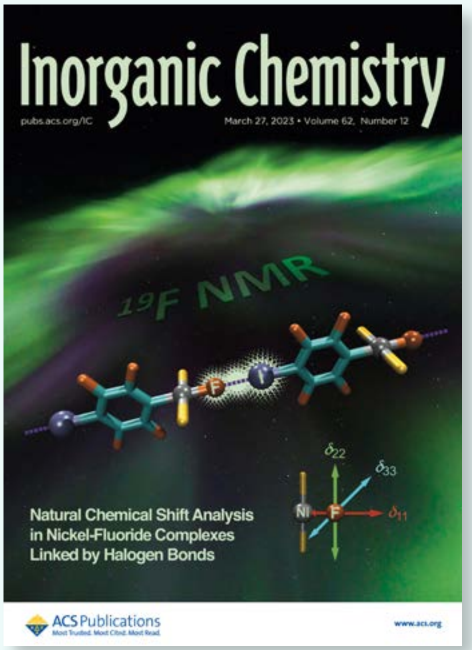


Figure 1: Front cover of volume 62, issue 12 of *Inorganic Chemistry*, promoting the open-access article *"Solid-state 19F NMR chemical shift in square-planar nickel-fluoride complexes linked by halogen bonds"* by A. Castro et al. (Inorg. Chem. 2023, 62, 12, 4835.). Designed by Castro, the cover artwork illustrates a nickel-fluoride complex engaged in halogen bonding and exposed to a magnetic field, symbolized by the Northern Lights. Reproduced with permission from ACS Publications.

Figure 2: Front cover of volume 62, issue 3 of *Biochemistry*, promoting the open-access article *"What Drives Chorismate Mutase to Top Performance? Insights from a Combined In Silico and In Vitro Study"* by H. V. Thorbjørnsrud et al. (Biochemistry 2023, 62, 3,782).

Publications

In terms of scientific publications, 2023 has been the most productive year at the Hylleraas Centre so far. Members of the Hylleraas Centre produced 83 publications in 2023, bringing our total scientific production to 368 publications. Roughly 10% of these—7 in 2023, 36 in total—were published in journals with an impact factor greater than ten. Specifically, in 2023, we published two articles in the Journal of the American Chemical Society, two in WIREs Computational Molecular Science, one in ACS Nano, one in Science Advances, and one in Accounts of Chemical Research. At the end of 2023, the articles published by the Hylleraas Centre since its opening in 2017 had been cited about 4500 times (excluding self-citations), with an H index of 34.

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. International collaborations are very important to the Hylleraas Centre, as documented by the 83 publications in 2023 being co-authored by researchers from more than 100 universities, research centres, and companies across the globe. The broad scope of the research at the Hylleraas Centre is illustrated by four review articles published by Hylleraas members on a wide range of topics from the structure of the density-potential mapping in density-functional theory, over time-dependent coupled-cluster theory of laser-driven electron dynamics, to the chemical reactivity of cyclometalated gold complexes.

Also in 2023, two articles by members of the Hylleraas Centre were featured on the cover of highly regarded international journals. *Inorganic Chemistry* published the cover paper Solid-state *^{19}F NMR chemical shift in square-planar nickel-fluoride complexes linked by halogen bonds* (Figure 1) by Dr. Abril Castro together with Profs. Michele Cascella and Odile Eisenstein from the Hylleraas Centre and international collaborators. *Biochemistry* published the cover paper *What Drives Chorismate Mutase to Top Performance? Insights from a Combined In Silico and In Vitro Study* (figure 2) by Dr. Manuel Carrer and Prof. Michele Cascella from the Hylleraas Centre together with Hylleraas affiliate Prof. Ute Krengel.

Figure 3: Opening of the 2023 Hylleraas Annual Meeting at Sommarøy by the organizers Tonje Reinholdt Haugen and Gabriel Gerez. *Photo: Marco Bortoli.*

Figure 4: Participants at the TRAINS conference honouring the career and achievements of Trygve Helgaker. *Photo: Marco Bortoli.*

Figure 5: Rector Svein Stølen opening the Norway-Japan Academic Network seminar on February 15, 2023. *Photo: Jan Ingar Johnsen.*

Meetings and conferences

Several meetings and conferences were held in 2023. *The Hylleraas Hackathon* was held at Skibotn near Tromsø on April 17–21, organized by Magnus Ringholm, with about 30 participants. The *Annual Hylleraas Meeting* took place at Sommarøy near Tromsø on September 5–7 and was organized by YRP representatives Tonje Reinholdt Haugen and Gabriel Gerez (Figure 3), with 54 participants. On November 27-30, Erik Tellgren, Michele Cascella, and Thomas Bondo Pedersen organized the conference *TRAINS – celebrating Trygve's adventures in science* to honour the career of former Hylleraas Director Trygve Helgaker. The conference, which also served as the annual *National Meeting of the Division of Quantum Chemistry and Modelling* of the Norwegian Chemical Society, was held at Sundvolden and was attended by about 100 participants (Figure 4).

On February 15, the Norway-Japan Academic Network seminar *Knowledge Triangle in Energy Research, Education, and Business* was held at the Hylleraas Centre in Oslo, organized by Michiko Atsumi, Erik Tellgren, Simen Reine, and Trygve Helgaker. More than 80 people participated in-person and via Zoom, and the seminar was opened by Rector of the University of Oslo, Svein Stølen (Figure 5).

Ainara Nova organized the 2023 annual meeting of the NordCO2 consortium in Saariselkä, Finland, on April 24-27, with 40 participants from seven Nordic universities. As part of the Centre for Advanced Study project *Attosecond Quantum Dynamics Beyond the Born-Oppenheimer Approximation*, Simen Kvaal and Thomas Bondo Pedersen organized the workshop *Molecular Quantum Dynamics* on May 23-26 at Hotel Lysebu in Oslo (Figure 6). The workshop gathered 37 scientists from 12 different countries. A couple of weeks later, Luca Frediani and Hylleraas Adjunct Professor Laura Ratcliff organized the workshop *Numerical Methods in Quantum Chemistry*, which was held at UiT – The Arctic University of Norway in Tromsø on June 5-8 with more than 50 international participants. Michele Cascella organized the CECAM workshop *Computational Molecular Medicine Across Three Decades* at the SISSA International School for Advanced Studies in Trieste, Italy, on October 23-25. The workshop was organized in celebration of the 60th birthday of Prof. Paolo Carloni.

Professor Frank Neese from the *Max-Planck-Institut für Kohlenforschung in Mülheim* an der Ruhr, Germany, gave the 2023 Almlöf–Gropen Lecture, *A Perspective on Large Scale Quantum Chemical Codes and Algorithms*, on November 21 in Oslo and November 23 in Tromsø. At both events, the Almlöf–Gropen Young Speakers were Torsha Moitra from Tromsø and Sigbjørn Løland Bore from Oslo.

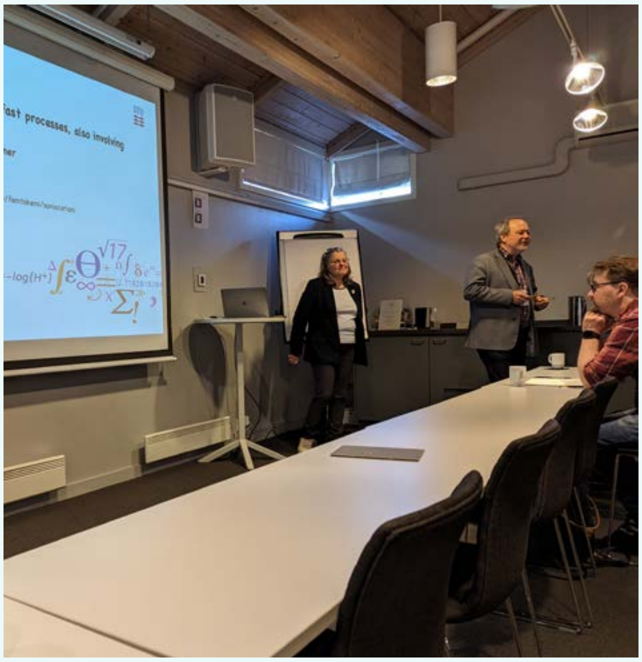


Figure 6: Professor Sonia Coriani about to start her lecture, as Prof. Reinhold Schneider introduces her to the audience at the CAS Workshop on Molecular Quantum Dynamics. *Photo: Thomas Bondo Pedersen*.

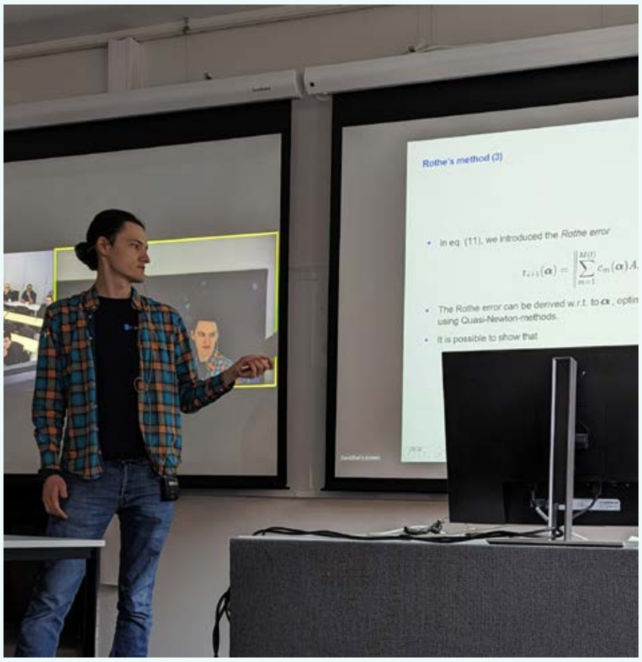


Figure 7: Simon Elias Schrader giving a Friday Seminar on the simulation of high-harmonic generation (HHG) at the Hylleraas Centre in Oslo on September 29, 2023, just days before the Nobel Prize in Physics was awarded to Agostini, Krausz, and L'Huillier for the development of the HHG techniques that are at the heart of attosecond science. *Photo: Thomas Bondo Pedersen.*

Dissemination

The members of the Hylleraas Centre gave a total of 43 oral presentations in 2023. Of these, 18 presentations were given by young members as Friday seminars at the centre, while 25 presentations were given by Hylleraas members at international meetings and conferences. Thomas Bondo Pedersen and Trygve Helgaker gave keynote lectures at the 59th *Symposium on Theoretical Chemistry* (STC 2023) at ETH Zürich, Switzerland, while Michele Cascella gave a keynote lecture at the 9th *International Theoretical Biophysics Symposium* (TheoBio2023) in Cetraro, Italy. David Balcells gave an invited lecture at the 2023 *IMS International Conference on Statistics and Data Science* in Lisbon, Portugal, and Ainara Nova gave a plenary lecture at the *XLIX Congress of the Physical Chemistry Division of the Italian Society of Chemistry* in Turin, Italy.

A total of 28 Friday seminars (Figure 7) were given at the Hylleraas Centre in 2023, of which eight were given by guests to the centre. One of these was given by the YRP-invited guest, Prof. Marcel Swart from the University of Girona, Spain, on November 10. Members of the Division for Quantum Chemistry and Modelling of the Norwegian Chemical Society were invited to follow his lecture on Zoom, marking the beginning of a new seminar series of the Division with two lectures planned per year.

At the 14th *European Conference on Computational and Theoretical Chemistry* held in Thessaloniki, Greece, August 27-31 2023, PhD candidate Aurore Denjean was awarded the best poster prize for her poster *N-Doped Graphene-Based Nickel(II) Single-Atom Catalyst for Hydrogen Borrowing: A DFT Computational Study*. Three young researchers from the Hylleraas Centre won poster awards at the conference *Quantum Systems in Chemistry, Physics, and Biology* (QSCP-XXVI) held in Jaipur, India, October 14-20, 2023. PhD candidate Samiran Sen was awarded for his poster *Life Under Pressure*, while Dr. Lukas Konecny and Dr. Marius Kadek were awarded for their posters *Relativistic Linear Response QEDFT for Cavity Engineering of Excited States and Band Structures of 2D Transition-Metal Dichalcogenides Films from Relativistic Four-Component Dirac-Kohn-Sham Theory using All-Electron Gaussian-Type Orbitals*, respectively.

Figure 8: About 30 posters at the Department of Chemistry, University of Oslo, at *Hvorfor Det?* 2023 while the young presenters and their families were attending educational lectures about Egil Hylleraas, quantum theory, chemistry, and practical aspects of life as an astronaut. *Photo: Thomas Bondo Pedersen.*

Outreach

The Hylleraas Centre was visited by *Talentsenteret i realfag* (part of *Oslo Vitensenter* at the Norwegian Museum of Science and Technology) on January 17, 2023. The twelve visiting students carried out exercises related to quantum chemistry, attended talks by Trygve Helgaker, Hanan Gharayba, and Erik Tellgren, and visited the mass spectrometry laboratory with Mauritz Ryding.

During *Forskningsdagene 2023*, an annual national research festival organized by the Research Council of Norway, Thomas Bondo Pedersen participated in a panel debate about *Energi og en verden i forandring* at Litteraturhuset in Oslo, September 20, and gave a seminar about the role of and need for basic research in solving societal grand challenges in the 21st century. The panel debate was organized by the Centre for Advanced Study in Oslo.

Faglig-pedagogisk dag is an annual event at the University of Oslo aimed at high school teachers. On October 31, 2023, Thomas Bondo Pedersen presented the research themes of the Hylleraas Centre for about 20 chemistry teachers from high schools in and around Oslo.

On Saturday November 18, 2023, the Hylleraas Centre hosted *Hvorfor Det?* (Figure 8), 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 siblings, parents, and grandparents. Hylleraas members Håkon Emil Kristiansen, Morten Ledum, Benedicte Sverdrup Ofstad, and Thomas Bondo Pedersen participated as referees.

Throughout 2023, the Hylleraas Centre ran *Hylleraas Math Help* for students at the Department of Chemistry in Oslo, helping with mathematics and scientific programming at all levels, led by Erik Tellgren.

Management

The Management Team met 11 times in 2023 (all months except July), while the Board of Directors met three times (January 27, May 23, and November 8).

Visitors

In 2023, a total of 20 researchers visited the Hylleraas Centre, with 13 in Oslo and 8 in Tromsø. This count includes the Almlöf–Gropen speaker who visited both places but excludes inter-node visits and visits by affiliate members.

Figure 9: Participants of the Hylleraas School 2023. *Photo: Marco Bortoli.*

Training and career support

Six members of the Hylleraas Centre defended their PhD thesis in 2023: Victoria Ariel Bjørnestad (UiO), Linn Neerbye Berntsen (UiO), Mateu Montserrat Canals (UiO), Manuel Carrer (UiO), Samiran Sen (UiO), and Benedicte Sverdrup Ofstad (UiO). At the end of the year, a total of 23 PhD candidates had completed their degrees at the Hylleraas Centre. In addition, three students completed their master's degree at the Hylleraas Centre in 2023: Mira Mors (UiO), Aleksandar Davidov (UiO), and Jørn Eirik Betten (UiO).

The 2023 edition of the *Hylleraas School* was organized by a committee composed of Inga Schmidtke, Tonje Reinholdt Haugen, David Balcells, and Simen Reine, and held at Hadeland north of Oslo on January 10-13 with 47 participants (Figure 9). Ainara Nova co-organized the Joint NordCO2 and CCU-Net Winter School on carbon capture and conversion in Århus, Denmark. The two-day school was attended by 63 PhD students from Nordic universities.

Young Researcher Parliament

The Young Researcher Parliament (YRP) invited Prof. Marcel Swart, director of the Institute of Computational Chemistry and Catalysis (ICQC) at the University of Girona, Spain, for a three-day visit at the Hylleraas Centre in Oslo, November 8-10. In addition to the Friday seminar mentioned above, Swart gave a seminar for the young researchers on career development in academia.

The Hylleraas Annual Meeting in September 2023 was organized by the YRP representatives Tonje Reinholdt Haugen and Gabriel Gerez at Sommarøy Arctic Hotel.

In 2023, Quentin Pitteloud succeeded Gabriel Gerez as YRP representative in Tromsø, while Oliver Bohle succeeded Hannes Kneiding in Oslo. Pitteloud and Bohle join Marinella de'Giovanetti and Tonje Reinholdt Haugen as YRP representatives.

Figure 10: On July 14, Odile Eisenstein signed the Charter Book of the Royal Society in London, UK. *Photo: Royal Society.*

External projects and funding

With no funding available for Researcher Projects within the Groundbreaking Research (FRIPRO) program of the Research Council of Norway in 2023, the Hylleraas Centre attracted relatively little new funding following the record years 2020 and 2021.

However, Dr. Sigbjørn Løland Bore was awarded a Young Research Talent (YRT) grant from the Research Council of Norway. His YRT project *H2O-MOF: Harnessing Hydrogen by Optimized Ion-Conductive Metal-Organic Frameworks* will run from 2024 to 2028 at the Hylleraas Centre in Oslo.

Bore also received a Young-CAS grant 2024-2025 from the Centre for Advanced Study in Oslo for the project *Can Ice be Described from First Principles?* The grant funds meetings at the Norwegian Academy of Science and Letters in Oslo, with the first meeting planned for January 2024.

Both Bore and Dr. Marco Bortoli received funding for high-performance computing time from the European High-Performance Computing Joint Undertaking (EuroHPC JU). Bore's project *Why is Ice so Slippery* and Bortoli's project *Rational Chemical Synthesis: Empowering Grignard Reagents with Machine Learning* received computing time on the LUMI supercomputer in Finland and the Leonardo supercomputer in Italy. Such grants of computing time are becoming increasingly important due to limited resources at the national level.

Finally, the Hylleraas Centre became part of a new Marie Sklodowska Curie Action (MSCA) Doctoral Network through the participation of David Balcells. The project DEMO: *Discovery of Efficient Enzyme-Like Metal-Organic Frameworks* is a collaboration between six research institutions across Europe and funds one PhD student in Oslo, with two additional PhD students performing extensive parts of their research in Oslo.

Personnel

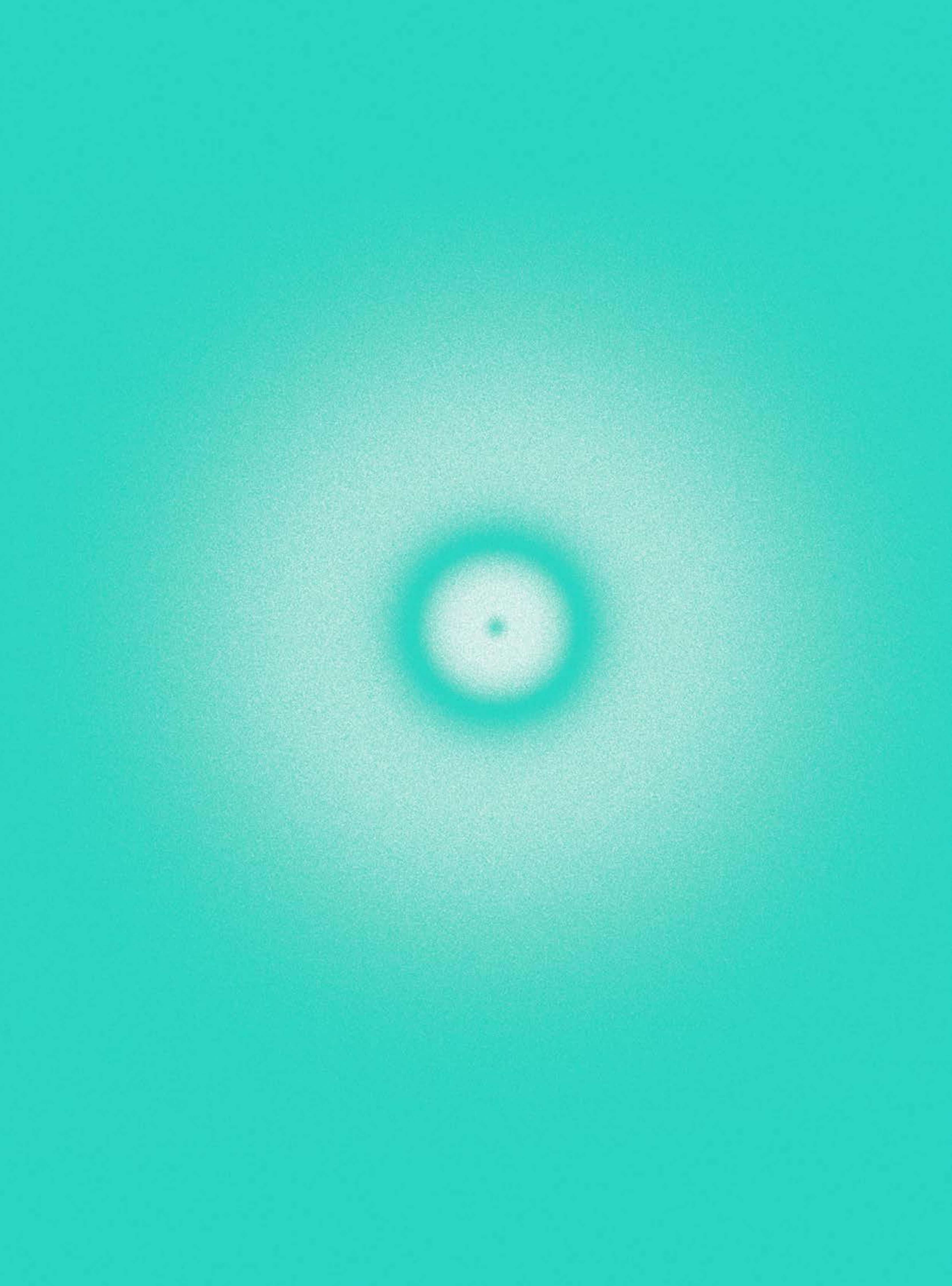
On July 1, 2023, Thomas Bondo Pedersen succeeded Trygve Helgaker as the director of the Hylleraas Centre. At the same time, five new Research Theme (RT) leaders were appointed: Dr. Simen Kvaal, Dr. Erik Tellgren, Dr. Abril Castro, Dr. Simen Reine, and Dr. Bin Gao. Together with the existing RT leaders, they lead our new research themes RT1: Molecular Quantum Mechanics led by Thomas Bondo Pedersen and Luca Frediani, RT2/6: Multiscale Modelling and Complexity led by Michele Cascella and Bjørn Olav Brandsdal, RT3/4: Matter-Field Interactions led by Michal Repisky and Erik Tellgren, RT5: Chemical Transformations led by Ainara Nova and Abril Castro, RT7: Machine Learning and Mathematical Methods led by David Balcells and Simen Kvaal, and RT0: Hylleraas Software Platform led by Simen Reine and Bin Gao.

Odile Eisenstein was elected Foreign Fellow of the Royal Society, signing the Royal Society Charter Book on July 14 (Figure 10).

Richard P. Feynman, Nobel Laureate 1965. In the early 1980s, Feynman proposed the idea of quantum computers, which may revolutionize computational chemistry.
Photo credit: Harvey of Pasadena, courtesy AIP Emilio Segrè Visual Archives

"The physicist could never quite believe that the chemist knew what he was talking about when he described the arrangement of the atoms. For about twenty years it has been possible, in some cases, to look at such molecules [...] by a physical method, and it has been possible to locate every atom, not by looking at colors, but by measuring where they are. And lo and behold!, the chemists are almost always correct."

> Richard P. Feynman – on the concept of molecular structure,
> in *The Feynman Lectures on Physics* (1964)



Research Highlights

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 and data science to address challenging questions in modern science, often in collaboration with experimentalists. For 2023, we present recent research carried out in Research Theme 1, Molecular Quantum Mechanics, and Research Theme 3/4, Matter-Field Interactions.

Unlocking Time’s Secrets: Relativistic Insights in Attosecond Pump-Probe Spectroscopy

Torsha Moitra and
Michal Repisky

Attochemistry, a captivating frontier in modern science, delves into the realm of atoms and molecules at unimaginably swift timescales, measured in attoseconds – billionths of a billionth of a second (1 as = 10^{-18} s). It unravels the intricate choreography of electrons during chemical processes, offering profound insights into fundamental processes underlying chemistry [1]. By harnessing cutting-edge laser technologies, researchers in the past few years have succeeded in molecular transformations at this fleetingly small timescale, paving the way for groundbreaking discoveries with far-reaching implications, from revolutionizing drug design to advancing renewable energy technologies. The importance of this field can be further gauged by the Nobel Prize in Physics awarded to Agostini, Krausz, and L’Huillier in this realm in 2023.

The field lies at the intersection of physics, chemistry, mathematics, and computer sciences, demanding a multifaceted approach. For its understanding and prediction, the need for advanced theoretical models becomes increasingly evident. The Hylleraas Centre therefore emerges as an ideal institution for delving into the intricacies of attochemistry, offering a rich environment for interdisciplinary collaboration and cutting-edge research.

Following electron motion

To study the pure electronic motions, the most straight-forward approach is to solve the time-dependent Schrödinger or Dirac equations by exposing the electron density to an electromagnetic wave or pulse and propagating the electron density in time [2]. The Hylleraas Centre has over the years contributed to this domain, by implementing and extending the real-time propagation schemes to study molecular properties at the relativistic density functional theory [3-6] as well as coupled cluster level [7-9].

The pump-probe setup serves as a conventional means of investigating time-domain processes, wherein a pump pulse initially excites a molecule, generating an out-of-equilibrium coherent superposition of states. Subsequently, this non-stationary wave packet is probed to capture its response. Figure 1 shows a simplified pictorial depiction of the theoretical setup. This type of spectroscopy has additional degrees of freedom in comparison to conventional absorption spectroscopy: namely the pump pulse features (like shape, carrier frequency, duration, etc) and the pump-probe time delay. The time-delay between the pump and probe pulses are of the order of few femto-seconds – that is, on timescales smaller than the onset of nuclear motions.

Attosecond light sources typically operate in the extreme ultraviolet (XUV) and soft X-ray region. In the case of the latter, excitations processes originate from element-specific core level orbitals,

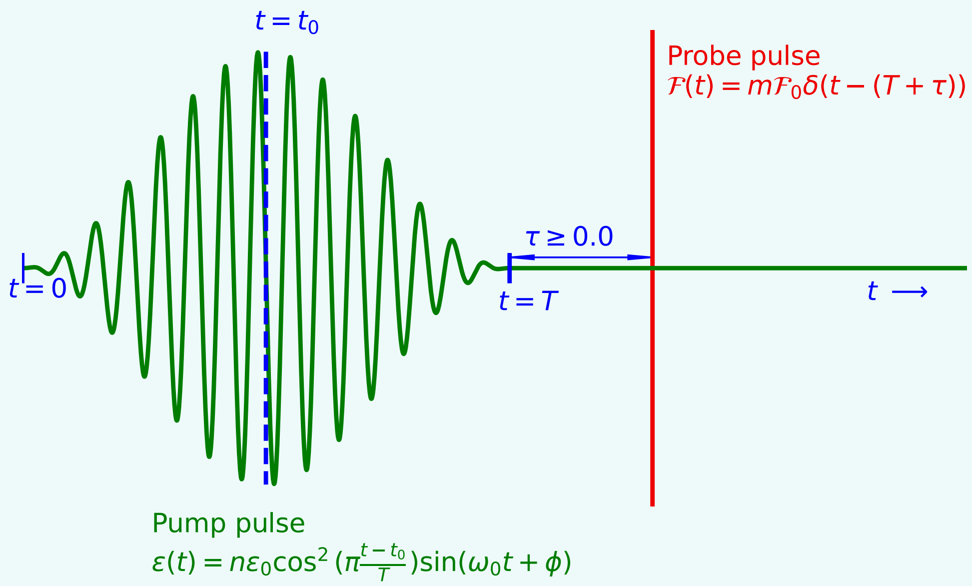


Figure 1: Pictorial representation of computational setup for pump-probe processes, where the pump (ε) generates the non-stationary wave packet, probed after time delay (τ) with field (F). Adapted from J. Phys. Chem. Lett., 2023, **14**, 1714.

profoundly influenced by scalar and spin-orbit relativistic effects. Therefore, requiring the inclusion of relativistic effects into the theoretical model. However, it comes with an additional cost. The gold-standard relativistic calculations using four-component Dirac-Coulomb Hamiltonian are possible with modern super-computers (like the NRIS FRAM or BETZY machines) but they are computationally demanding. Therefore, a quest for accurate yet cheaper two-component frameworks has been a focal interest of many researchers, including those at the Hylleraas Centre. In 2022, Dr. Michal Repisky and co-workers advanced closer to this goal by developing a highly accurate atomic mean-field exact two-component (amfX2C) Hamiltonian, which reproduces the parent four-component results with a high accuracy of 10^{-5} Hartree/atom [10]. Furthermore, in 2023, the amfX2C Hamiltonian model has been extended to study non-equilibrium molecular properties with real-time time-dependent density functional theory (RT-TDDFT) [11] in the ReSpect software package [12].

In this study, several facets of pump-probe spectroscopy were explored. Starting with the effect of the pump pulse strength, showcasing the counter-intuitive analogy that sufficient depopulation of ground state is necessary to generate a transient absorption spectrum from the ground state absorption spectra. Additionally, the appearance of negative intensity spectra peaks with higher pump pulse strength was observed, which are a hallmark of non-equilibrium processes. To interpret this unique spectral feature, the non-equilibrium response function was developed. The response function, in addition to a Lorentzian dependence also consists of a Rayleigh lineshape dependence. The later attributing to the negative intensity feature. Further the authors explore the importance of spin-orbit effects near the sulphur $L_{2,3}$-edge X-ray transient absorption spectra of thiophene as shown Figure 2. The spectra obtained at the non-relativistic one-component (1c) level lacks the spin-orbit splitting between the $2p_{1/2}$ and $2p_{3/2}$ excitation bands, reinstating the need for relativistic treatment. In the relativistic realm, it is important to note that the spectra obtained at the amfX2C (2c) and gold-standard four-component Dirac-Coulomb (4c) level are identical, offering promising prospects.

This theoretical work constitutes a significant leap towards simulating and understanding transient absorption spectra, while being applicable for realistic systems consistently across all elements of the periodic table as well as both core and valence energy regions.

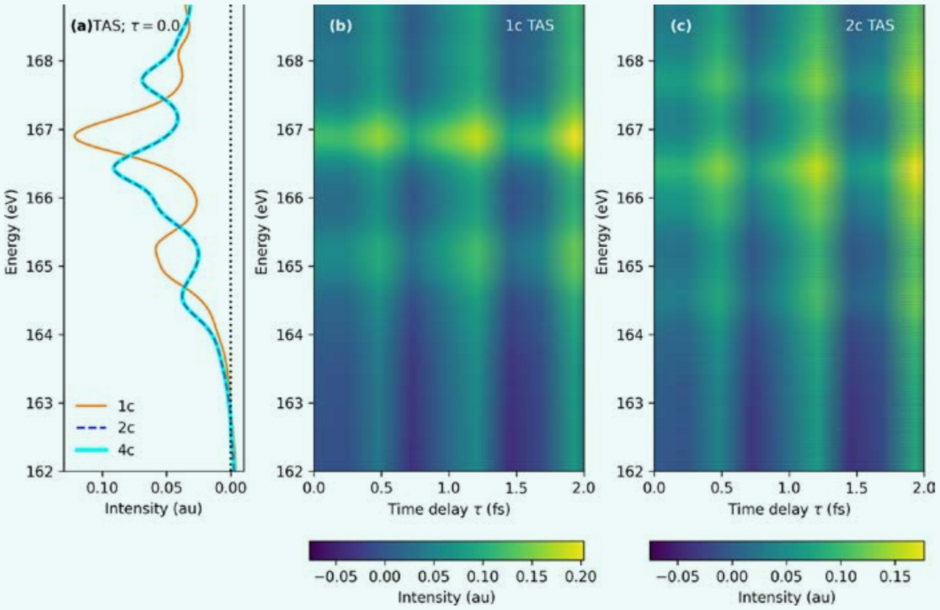


Figure 2: Transient absorption spectra of thiophene (a) at time-delay=0, with one-component (1c) non-relativistic, amfX2C and Dirac-Coulomb Hamiltonian levels shown in orange, blue and cyan, respectively. (b) and (c) are the variation in the spectra with time delay at the one-component non-relativistic (1c) and two-component amfX2C (2c) level, respectively. Adapted from J. Phys. Chem. Lett., 2023, **14**, 1714.

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Molecular Structure in Quantum Mechanics

Thomas Bondo Pedersen
and Henrique Musseli Cezar

The structure of a molecule – that is, the three-dimensional arrangement of the constituent atoms – is a core concept in chemistry. It distinguishes between isomers and thus forms the basis for chemical language, the systematic nomenclature that uniquely identifies a compound. Moreover, molecular structure can be used to rationalize and predict chemical properties, including reactivity [1], often with little or no direct reference to the physical (electromagnetic) forces holding the constituent atoms together.

Indeed, the concept of molecular structure was developed by chemists long before it could be explained by the laws of physics. In the first chapter of his famous Lectures on Physics [2], Nobel Laureate Richard P. Feynman characterizes the pioneering work of 19th-century chemists such as Jacobus Henricus van't Hoff and Joseph Le Bel as *"one of the most fantastic pieces of detective work that has ever been done,"* and continues

"The physicist could never quite believe that the chemist knew what he was talking about when he described the arrangement of the atoms. For about twenty years it has been possible, in some cases, to look at such molecules […] by a physical method, and it has been possible to locate every atom, not by looking at colors, but by measuring where they are. And lo and behold!, the chemists are almost always correct."

Feynman himself, in 1939 [3], made a seminal contribution to the understanding of molecular structure by a quantum-mechanical derivation of the effective forces acting on the atomic nuclei in a molecule. Molecular structure then emerges as an arrangement of the nuclei where the forces acting on them vanish. Feynman's formulation uses an elegant mathematical construction, now known as the Hellmann-Feynman theorem, which is still used today to compute molecular structures – a fundamental task of quantum chemistry. Remarkably, Feynman's paper does not cite a single reference although it explicitly relies on the assumption that the nuclei can be considered classical point particles with charge equal to the atomic number times the elementary charge. This assumption is justified by the observation that atomic nuclei are orders of magnitude heavier than electrons and, therefore, much more sluggish. This is, of course, an approximation, the Born-Oppenheimer approximation, which was first proposed and analyzed by Max Born and Julius Robert Oppenheimer in 1927 [4]. While Oppenheimer went on to make fundamental contributions to, e.g., the theory of black holes and neutron stars as well as leading the Manhattan project during World War II, his most cited scientific work remains the Born-Oppenheimer paper, documenting how crucial it has been to chemistry. The Born-Oppenheimer approximation is almost always used, in one form or other, in theoretical and computational chemistry today, often without even mentioning it or citing the Born-Oppenheimer paper. It thus

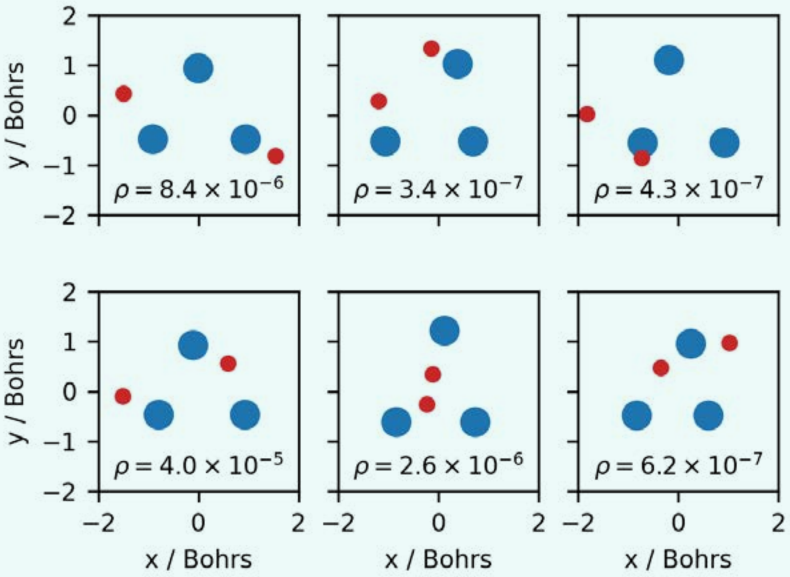

Figure 1: Six aligned snapshots from the random sampling of the probability density for the ground state of D_3^+. Blue dots represent deuteron positions, red dots electron positions projected into the plane defined by the deuterons. Reproduced from [24] under CC-BY 4.0 license.

can be regarded as the fundamental physical framework in which modern chemistry is understood.

The Born-Oppenheimer approximation works exceedingly well in far most cases, which means that it can be successfully applied to understand, interpret, and predict experimental observations. Moreover, the conditions under which it fails are well known from both experience and from theoretical considerations, including rigorous mathematical analyses – see, e.g., [5] for a recent review of the various mathematical treatments. Nevertheless, the Born-Oppenheimer approximation explicitly breaks some of the most fundamental laws of quantum physics, probably our most well-tested theory of matter. First, Heisenberg's uncertainty principle prohibits the exact determination of the position of a quantum particle such as an atomic nucleus. Second, the indistinguishabilty of identical quantum particles prevents us from speaking of "the hydroxyl proton" and "the methyl proton" in, e.g., the ethanol molecules because in doing so, we explicitly distinguish identical fermions, the protons. In other words, the perhaps most fundamental concept in chemistry appears to be in conflict with the fundamental laws of Nature. This conflict has caused continued debates in the scientific literature regarding the concept of molecular structure, see f.ex. [6-19].

It is a postulate of quantum mechanics that the wave function contains all information about the system it describes. Hence, keeping in mind the mathematical vailidity and overwhelming success of the Born-Oppenheimer approximation in chemistry, it should be possible to extract the structure from the *full* molecular wave function for electrons and nuclei interacting via the Coulomb potential. To test this hypothesis, one faces two major challenges:

1. Solve the molecular Schrödinger equation for the Coulomb Hamiltonian without invoking the Born-Oppenheimer approximation at any stage – i.e., treating both electrons and nuclei on the same quantum-mechanical footing.

2. Define a computationally viable procedure that yields the structure from the wave function only.

While no analytical solution of the Coulombic Schrödinger equation has ever been obtained for more than two particles, highly accurate approximate solutions can be computed numerically for few-particle molecules (up to about ten particles in total) by expanding the wave function in a basis of explicitly correlated Gaussians [20,21]. This approach overcomes the first challenge, albeit at a significant computational cost since the proper permutational symmetry – the Pauli principle – must be explicitly imposed. Thus, for each set

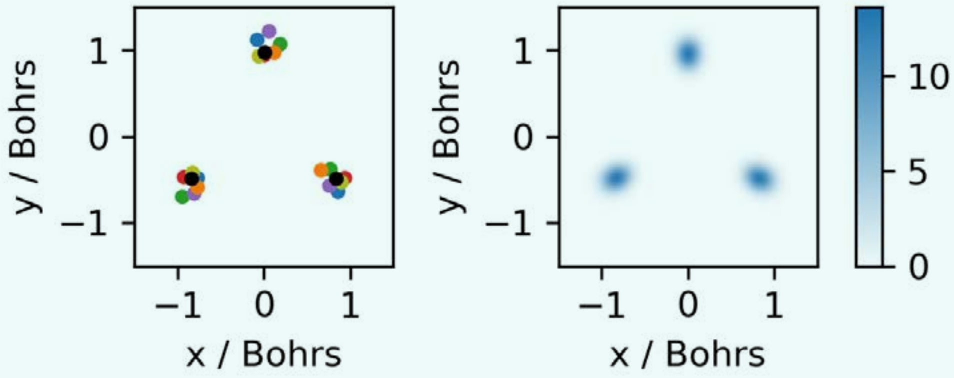

Figure 2: Left panel: The deuteron positions of the six snapshots in Fig. 1 with the medoid structure shown as black dots. Right panel: 2D kernel density estimate (KDE) of the x and y values of the deuteron positions of 30000 selected snapshots after alignment with the medoid configuration. Reproduced from [24] under CC-BY 4.0 license.

of N identical particles within the system (the electrons, the protons, the carbon-12 nuclei, etc.), N! permutations must be explitly computed. The effort is rewarded, however, by extremely good agreement with uninterpreted experimental data such as infrared and microwave absorption frequencies and intensities.

The second challenge is more intricate, since there is no quantum-mechanical operator that represents molecular structure, neither as a whole nor as the different elements of structure such as, e.g., bond distances and angles. This would clearly require distinguishability of identical nuclei ("the methyl proton" etc.). Hence, contrary to what one might expect if molecular structure is to be considered truly observable, the structure cannot be obtained from the wave function as an expectation value of one or more hermitian operators [22]. One is, therefore, left with the fundamental physical interpretation of the quantum-mechanical wave function originally proposed in 1926 by Born [23], who argued that the modulus-square of the wave function yields the probability density for finding the particles at specified points. Hence, molecular structure should manifest itself as a particular arrangement of the nuclei associated with large probability density [10,11].

This idea has been pursued by Hylleraas members Dr. Lucas Lang, Dr. Henrique Musseli Cezar, and Prof. Thomas Bondo Pedersen in collaboration with Prof. Ludwik Adamowicz at the University of Arizona, a world-leading expert in quantum chemistry without the Born-Oppenheimer approximation and a frequent visitor to the Hylleraas Centre in Oslo. In the manuscript, *Quantum Definition of Molecular Structure*, which was accepted for publication in the Journal of the American Chemical Society in late 2023 [24], they hypothesize that *"molecular structure is essentially a manifestation of strong statistical correlation between the positions of the nuclei"*.

Given an approximate yet accurate full non-Born-Oppenheimer wave function computed as a linear combination of explicitly correlated Gaussians for the ground state of three deuterons (heavy hydrogen nuclei composed of a proton and a neutron) and two electrons – i.e., the D_3^+ molecule – they used random sampling of the probability density to locate high-probability regions for the deuteron positions. The resulting positions are randomly oriented in space due to the rotational degrees of freedom, and a rigid alignment procedure originally developed for attitude determination in astronautical science [25] was applied to bring the three deuteron positions for each high-probability region into the same plane. The same procedure can be applied to the electrons, of course, resulting in a large number of structural snapshots, see Fig. 1 for six examples.

To determine if the snapshots represent a single or more structures, the k-medoids

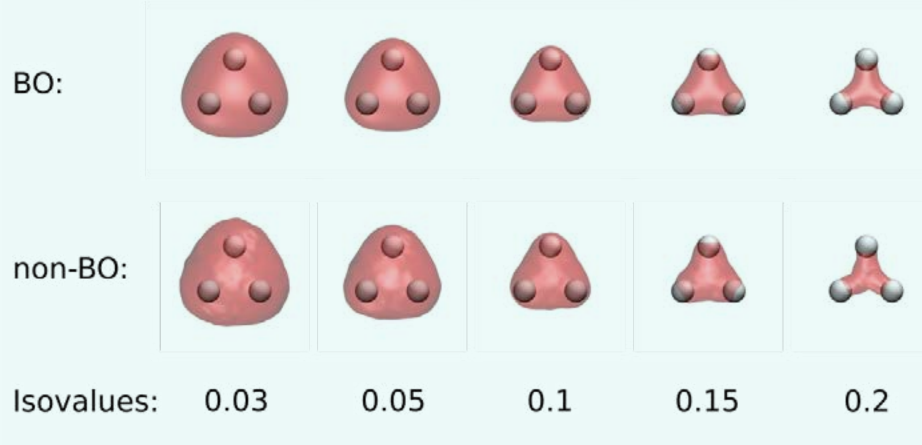

Figure 3: Born-Oppenheimer (BO) structure and electron density computed at the FCI/cc-pVQZ level of theory compared with the results extracted from the full non-Born-Oppenheimer (non-BO) molecular wave function. The electron density is plotted at various isovalues for a more detailed comparison. Reproduced from [24] under CC-BY 4.0 license.

clustering algorithm [26] – an unsupervised machine-learning technique– can be used. This results in a single structure, an equilateral triangle, as shown in Fig. 2.

Repeating the procedure for the electrons, one can use 3D kernel density estimate to obtain the electron density and compare with full configuration-interaction (FCI) results obtained within the Born-Oppenheimer approximation. As seen in Fig. 3, the agreement is striking, with the minor deviations ascribable to fully anharmonic zero-point vibrational effects, which are automatically included in the non-Born-Oppenheimer results but excluded in the Born-Oppenheimer ones.

This is the first-ever demonstration that the Born-Oppenheimer structure of a molecule with more than two nuclei can be extracted from the molecular wave function with full permutational symmetry. Work is in progress aiming at extraction of the structure from excited-state wave functions without the Born-Oppenheimer approximation.

The work is important not only for settling the decades-long debate regarding the concept of molecular structure within quantum mechanics, but also for establishing a computational procedure to extract the structure from a given wave function. This is needed to interpret and visualize the full molecular quantum dynamics induced by intense attosecond and few-femtosecond laser pulses, a current project at the Hylleraas Centre initiated with the 2021–2022 CAS project *Attosecond Quantum Dynamics Beyond the Born-Oppenheimer Approximation* led by Kvaal and Pedersen.

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Almlöf–Gropen Lecture Series

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. Arieh 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 Mülheim, 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

2022 **Prof. Clemence Corminboeuf**
École Polytechnique Fédérale de Lausanne (EPFL), Switzerland

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

Almlöf–Gropen Lecturer 2023

Prof. Frank Neese giving his lecture at the Hylleraas Centre in Oslo on November 21, 2023. *Photo:Jan Ingar Johnsen.*

Professor Frank Neese

Max-Planck-Institut für
Kohlenforschung,
Mülheim an der Ruhr,
Germany
–

A perspective on large scale quantum chemical codes and algorithms
–

Nov. 21, 2023,
University of Oslo
Nov. 23, 2023, UiT The Arctic
University of Norway
–

Frank Neese received both his Diploma (Biology, 1993) and Ph.D (Dr. rer. Nat., 1997) working with Peter Kroneck at the University of Konstanz. He performed Postdoctoral work at Stanford University with Edward Solomon (1997-1999) before returning to Konstanz for his Habilitation (2001). He joined the Max Planck Institute (MPI) for Bioinorganic Chemistry in 2001 as a group leader until accepting the position of Chair of Theoretical Chemistry at the University of Bonn (2006). In 2011, Neese was appointed Director of the MPI for Chemical Energy Conversion. In 2018, Neese relocated his activities to the MPI für Kohlenforschung as the successor of the late Walter Thiel. In 2010, Neese received the highest award of the German Science foundation, the Gottfried Wilhelm Leibniz Award followed by numerous national and international awards, including the Schrödinger medal of the World Association of Theoretically Oriented Chemists (2022). Neese is member of various national and international academies including the German National Academy of Sciences (Leopoldina, 2013) and the Academia Europea (2018). Neese has authored >700 scientific publications and has been listed among the top 1% world-wide most highly cited chemists since 2015. His work focuses on the theory of magnetic spectroscopies (electron paramagnetic resonance, magnetic circular dichroism) and their experimental and theoretical application, local pair natural orbital correlation theories, multi-reference methods, electronic and geometric structure and reactivity of transition metal complexes and metalloenzymes. Neese is the lead author of the ORCA program, as well as co-founder of the company FAccTs (2017) that distributes ORCA commercially.

In his Almlöf–Gropen lecture, Neese honored the legacy of Almlöf and Gropen, two pioneers of electronic structure theory. They paved the way for future generations of quantum chemists who would follow in their footsteps with the aim to make quantum chemistry ever more applicable to chemistry in ever more efficient and accurate ways. In fact, in recent years, quantum chemistry has been an indispensable component of many if not most chemical studies. While this success is endearing, the field also faces some rather severe challenges, many of which center around the sustainability of large-scale software development. In the lecture, Neese discussed some of these challenges and proposed a few (partial) solutions that he and members of his group have pursued in their recent re-development of the ORCA electronic structure package.

Almlöf– Gropen Young Speakers 2023

Torsha Moitra

Hylleraas Centre,
UiT The Arctic University of Norway
–

Relativistic framework for understanding ultrafast pump-probe processes

Torsha Moitra is a PostDoc at the Hylleraas Centre for Quantum Molecular Sciences at UiT The Arctic University of Norway. Moitra´s research focusses primarily on development and application of quantum chemical methods for elucidating ultrafast processes in the atto- to sub-femtosecond timescales. She obtained her PhD in 2021 from the Technical University of Denmark on Coupled-Cluster methods for X-ray photoabsorption and photoelectron spectroscopies.

Sigbjørn Løland Bore

Hylleraas Centre,
University of Oslo
–

Realistic phase diagram of water from first principles

Sigbjørn Løland Bore is a PostDoc at the Hylleraas Centre for Quantum Molecular Sciences at the University of Oslo. Bore's research combines advanced sampling techniques with machine learning to understand from first principles how water behaves under exotic conditions. He is a Young-CAS fellow, leading the project “Can ice be described from first principles?” and an RCN Young Researcher Talent, leading the H2O-MOF project on understanding and developing high-conductive metal-organic frameworks for application as membranes in fuel cells.

Meetings and Events

Hylleraas School

Hotel Hadeland, Gran
January 10-13, 2023
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The 2023 Hylleraas School melded scientific education with social interaction, fostering collaborative innovation at Hotel Hadeland.

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The second edition of the Hylleraas School was held at Hotel Hadeland from January 10-13, 2023. The school series aims to introduce important topics within the Hylleraas Centre to our interdisciplinary student community, which includes chemists, physicists, biologists, mathematicians, and computer scientists. The school also dedicates a generous amount of free time to promote social activities, fostering cross-node research projects, and facilitating interaction between junior and senior members.

The 2023 edition of the School was organized by Inga Schmidtke, Tonje Reinholdt Haugen, David Balcells, and Simen Reine. The first day of the school, started with a lecture on graphical design by Simen Kvaal, followed by a lecture on high-performance computing by Ole Widar Saastad (IT department, University of Oslo). The second day focused on potential-energy surfaces and geometry optimization approaches, with Simen Reine giving background lectures in the morning, and Tilmann Bodenstein, Morten Ledum, Bin Gao, and Simen Reine conducting tutorials in the afternoon. The third day was dedicated to machine learning, featuring lectures by David Balcells and tutorials from Hannes Kneiding, Simen Reine, and Lucas Lang.

Regarding social activities, there was a festive banquet on the first night and an engaging quiz organized by the Young Researcher Parliament on the last night. The quiz involved fun tasks such as matching childhood photos of famous scientists and senior Hylleraas members with their current selves. On the final day, Director Trygve Helgaker provided an overview of the activities of the Centre, followed by "state-of-the-RT" presentations by the PIs.

At the conclusion of the Hylleraas School, a survey was distributed to collect feedback from participants. In general, the students were very satisfied with the contents and organization, providing suggestions for further improvements for the next 2024 edition.

Visit from Talentsenteret on January 17, 2023. Left: Trygve Helgaker introducing quantum chemistry.
Right: Hanan Gharayba explaining Euler's method for simulating classical motion. *Photo: Erik Tellgren.*

Visit from Talentsenteret

Hylleraas Centre, Oslo
January 17, 2023

Talentsenteret, an initiative for talented senior high school students, visited the Hylleraas Centre and got a peak into computational chemistry and experimental chemistry.

Talentsenteret is an initiative for senior high school students by Oslo Vitensenter. Students who are particularly talented and interested in natural science can apply to participate. As part of that initiative, on January 17 twelve students and three accompanying staff visited the Centre. The visit was coordinated by Cathrine Strøm and planned by Erik Tellgren.

The program included an introductory lecture about quantum chemistry by Prof. Trygve Helgaker, smaller lectures and computer demonstrations of classical dynamics, quantum dynamics, and molecular orbital theory by Erik Tellgren and Hanan Gharayba. The students were introduced to computational chemistry using both simple, minimalistic programs for model systems as well as through visualization of molecular orbitals, geometry, and vibrations using advanced software. The latter part benefitted from the Hylleraas Software Platform. In addition, the students visited Mauritz Rydberg for a demonstration of the mass spectrometry lab.

Participants at the Norway-Japan Academic Network Seminar on February 15, 2023, at the Hylleraas Centre in Oslo. Photo: Michiko Atsumi.

Norway-Japan Academic Network Seminar

Hylleraas Centre, Oslo
February 15, 2023
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The Norway-Japan Academic Network Seminar Knowledge Triangle in Energy – Research, Education, Business was held at Hylleraas Centre in Oslo as part of the 20th anniversary of the Norway-Japan national research contract.

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At the seminar, we were fortunate to have a visit from the rector at the University of Oslo, Svein Stølen, who made the opening remarks. The theme of the seminar was post-lithium-ion battery for research and business as well as furthering the Norway and Japanese network for research and education. The main scientific talk was a lecture titled "Grand design of post Li-ion battery" by Prof. Shigeto Okada from Kyushu University. We were also fortunate to have other speakers covering themes ranging from the battery industry and electric car market to educational perspectives on active learning and joint Japan-Norway educational workshops on plasma physics, and institutional collaborations. There were also announcements and information from funding agencies as well as from the Japanese embassy in Norway.

The seminar was held as a hybrid event with in-person attendance at the Hylleraas Centre and virtual attendance via Zoom. The seminar was organized by Hylleraas members Trygve Helgaker, Erik Tellgren, Simen Reine and Michiko Atsumi, and included the attendance of more than 80 participants from Norway and Japan.

The Hylleraas centre, Research Council of Norway, Innovation Norway Tokyo, Norwegian Directorate for Higher Education and Skills, the Norwegian embassy in Tokyo, the Japan Society for the Promotion of Science (JSPS) Stockholm office, JSPS Norway-Japan Academic Network, and Kyoto University ESICB supported and were represented at the symposium.

Participants at the Skibotn-Hackaton 2023.

Skibotn Hackathon

Hylleraas Centre, Oslo
February 15, 2023

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Organized by Magnus Ringholm, the 2023 Skibotn Hackathon took place between April 17 and 21 at the Skibotn field station of UiT The Arctic University of Norway.

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Roughly 30 researchers spent the week April 17-21, 2023, working on their own projects or in collaboration with others at the Skibotn field station. Most participants tend to come from the Hylleraas Centre itself, but participation is also open to external collaborators of Hylleraas members. The field station is situated in relative seclusion in natural surroundings, providing a valuable opportunity to evade the distractions of everyday life. The facilities include a fireplace, a sauna and a barbecue hut (which was used one evening), so between these and the enjoyable company, the Skibotn hackathon is also a great opportunity for socializing with co-workers. Many of the participants enjoyed some nice evenings with board games and other kinds of fun.

In the later part of the week, meltwater started flooding the basement of the main building and this was the source of some concern, with several participants recruiting themselves to help keep the water at bay until proper drainage was put in place. Fortunately, there does not appear to have been any serious damage to the field station such that Tuula and Elisabeth will still be there to welcome us in 2024.

The Hylleraas members Mohamed, Mahika, Dat, Inga, and Raquel at the conference dinner in a “Kota” in Saariselkä on April 26, 2023. *Photo: Marie-Josée Haglund.*

NordCO2 Annual Meeting

Santa’s Hotel Tunturi,
Saariselkä, Finland
April 24-27, 2023

The Annual Meeting of the Nordic Consortium for CO_2 Conversion (NordCO2), led by Dr. Ainara Nova from the Hylleraas Centre, took place in Finland with 40 participants from seven Nordic universities.

Among the 40 participants at the 2023 NordCO2 Annual Meeting were five PhD students from the Hylleraas Centre: Mahika Luthra, Mohamed Eid Safy, Inga Schmidtke, Sahil Gahlawat, and Vu Duc Ha Phan. The meeting was organized by Ainara Nova (NordCO2 leader) and Prof. Timo Repo (NordCO2 PI, University of Helsinki) with the help of Dr. Marie-Josée Haglund Halsør (NordCO2 administrator, UiT The Arctic University of Norway). The scientific sessions included 30-minute student talks on the following topics: CO_2 conversion to chemicals, CO_2 conversion to fuels, and incorporation of catalysts into solid supports. The meeting also included a poster session and a NordCO2 Quiz. The conference dinner took place in a “Kota”, a traditional tent of the Sámi from the Finish Lapland.

Participants at the CAS workshop Molecular Quantum Dynamics at Lysebu Hotel in Oslo on May 23-26, 2023. *Photo: Cathrine Strøm.*

CAS Workshop: Molecular Quantum Dynamics

Lysebu Hotel, Oslo
May 23-26, 2023

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The interdisciplinary workshop *Molecular Quantum Dynamics* was organized as part of the Centre for Advanced Study project *Attosecond Quantum Dynamics Beyond the Born–Oppenheimer Approximation*, led by Hylleraas members Simen Kvaal and Thomas Bondo Pedersen, with 37 participating researchers in mathematics, physics, and chemistry.

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The Centre for Advanced Study project AttoDyn (*Attosecond Quantum Dynamics Beyond the Born-Oppenheimer approximation)* [1] ran from fall 2021 to spring 2023, with a pandemic extension to 2022/23. During May 23-26 an international workshop was organized by project leaders Thomas Bondo Pedersen and Simen Kvaal at Lysebu Hotel, gathering 37 participants from a dozen countries [2]. The title of the workshop was "Molecular Quantum Dynamics", and explored a variety of themes, including attochemistry, chemistry without the Born-Oppenheimer approximation, variational principles, and time-dependent many-body models.

1. https://cas-nor.no/project/attosecond-quantum-dynamics-beyond-born-oppenheimer-approximation
2. https://www.mn.uio.no/hylleraas/english/news-and-events/events/events-in-oslo/2023/CAS_MQD.html

Participants of the NMQC workshop (left) held in Tromsø (right).
Photo: Stig Eide (left) and Yngve Olsen Sæbbe (right).

Numerical Methods in Quantum Chemistry

UiT The Arctic University of Norway, Tromsø
June 5–8, 2023

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Held at the Department of Chemistry, UiT – The Arctic University of Tromsø, the workshop *Numerical Methods in Quantum Chemistry* gathered over 50 scientists from around the world to discuss the development of novel computational methods in quantum chemistry.

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The overarching theme of the conference was the use of unconventional basis sets, including wavelets, multiwavelets, numerical orbitals, finite field and finite difference methods. The workshop was part of a series dedicated to numerical methods, which started in Tromsø in 2015. The main organizer was Luca Frediani, supported by Prof. Robert J. Harrison (Stony Brook University, USA). The scientific committee also included Florian Bischoff, Jakob Kottmann, Laura Ratcliff, Hideo Sekino, Dage Sundholm, and Edward Valeev. The local organizing committee included the whole MRChem group and Stig Eide, the head of office at Hylleraas in Tromsø.

At this edition, in addition to the foucs on numerical methods, the conference dedicated special attention to relativity and correlated methods: the former in connection to the ReMRChem FRIPRO project which financed the conference and the latter because of the general interest in the topic at the Hylleraas centre.

Despite the progress made in the last two decades, since 2004, when wavelets and multiwavelets were first presented as possible alternative for quantum chemistry methods, they remain not so well known in the community at large. We have therefore decided to flank the more traditional part of the conference with a set of tutorials which covered the fundamental aspects of wavelet theory and their use in quantum chemistry applications.

The venue of the conference was the UiT campus which has provided us with excellent support, both in terms of facilities (conference auditorium, rooms for the tutorials, student lodging) and catering from Samskipnaden (coffee breaks and lunches).

Hylleraas Annual Meeting

Participants at the Hylleraas Annual Meeting at Sommarøy. *Photo:Marco Bortoli.*

Sommarøy Arctic Hotel Tromsø
September 5-7, 2023

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The Hylleraas Annual Meeting was held at beautiful Sommarøy and continues to help foster a good scientific and social connection between the Tromsø and Oslo nodes of the Hylleraas Centre.

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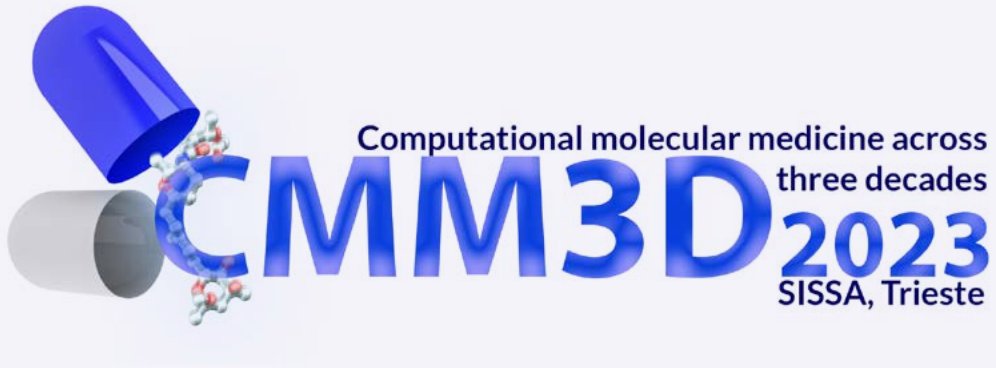
On September 5-7 2023, 54 Hylleraas members gathered at Sommarøy, which is located about 1 hour drive east from Tromsø. It was organized by Young Researcher Parliament representatives from Tromsø, Gabriel Gerez and Tonje Reinholdt Haugen with support from Bjørn Olav Brandsdal and Stig Eide. The topics for the meeting were a mixture of career development and transferable skills. Kenneth Ruud gave a presentation on how to prepare for a lifelong career in academia, while Jan Heier Johansen from Microsoft Tromsø talked about how it is working outside of academia. For transferable skills, Sigbjørn Løland Bore and Simen Reine talked about workflows in high-performance computing, with a practical example using active learning from Sigbjørn. A talk on scientific communication was held by Jørn Weines and Nathalie Isabell Blomstereng from Result at UiT. Master and PhD students participated in a Five-Minute Thesis competition, which was won by visiting PhD student Jonas Vester from the Technical University of Denmark.

In addition to the scientific program, there was a focus on the social aspect in the afternoons and time was set aside to do a hike up Hillesøytoppen as well as to explore the island itself in beautiful weather. The last day focused more on the future of Hylleraas, with the YRP having a joint node meeting about future events such as the Hylleraas School and seeing new and interesting ways to collaborate and help each other between the different projects and RTs.



Logo of the CECAM workshop *Computational Molecular Medicine Across Three Decades* co-organized by Michele Cascella in honor of Prof. Paolo Carloni. *Credit: Alessandra Magistrato.*

CECAM Workshop: Computational Molecular Medicine Across Three Decades: Celebrating the 60th Birthday of Paolo Carloni

International School for Advanced Studies, Trieste, Italy
October 23–25, 2023

To celebrate the 60th birthday of Prof. Paolo Carloni, Forschungszentrum Jülich, Germany, the workshop Computational Molecular Medicine Across Three Decades was organized under the auspices of the CECAM network.

Co-organized by Michele Cascella, the three-day event hosted by the International School for Advanced Studies (SISSA) in Trieste, Italy, was attended by roughly 80 scientists. The workshop featured 26 lectures by international speakers, and 18 flash presentations by alumni of Prof. Carloni. The scientific program of the workshop was dedicated to the state of the art in computational modelling of molecular biological phenomena, with special focus on medical and pharmaceutical research. This also included new algorithms exploiting data driven approaches and new hardware, from exascale architectures to quantum computing.

Prof. Marcel Swart giving his lecture *"A new twist to old stories"* as a Friday Seminar and as the first shared seminar of the NKS Division for Quantum Chemistry and Modelling at the Hylleraas Centre in Oslo on November 10, 2023. *Photo: Marinella de'Giovanetti.*

YRP Invited Scientist: Marcel Swart

Department of Chemistry,
University of Oslo
November 8-10, 2023
–

In November Prof. Marcel Swart, director of the IQCC Institute at the University of Girona, visited the Oslo node of the Hylleraas Centre on invitation of the YRP.

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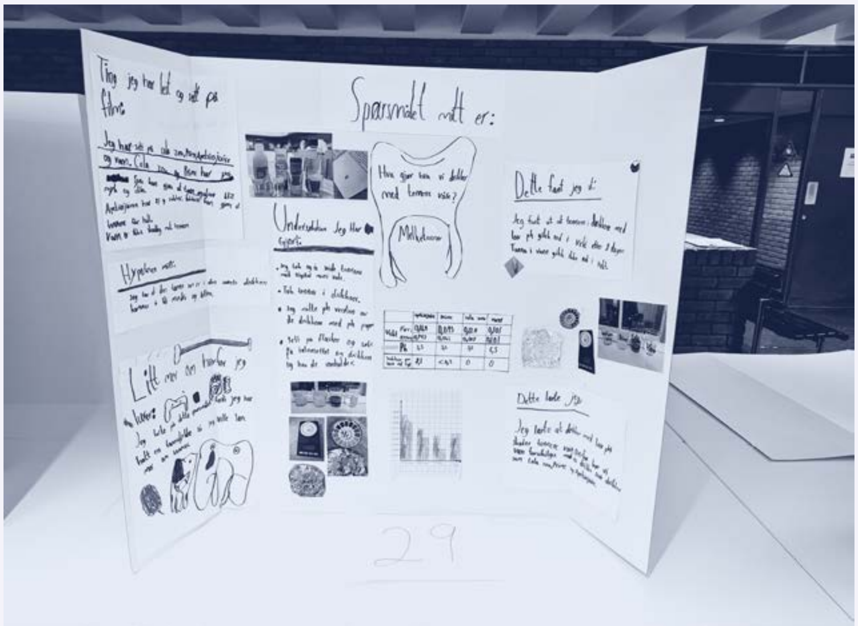
His visit was marked by enriching interactions with the young researchers, with a focus on early career development. In that spirit, on November 9, Prof. Swart delivered a compelling talk entitled *"My career so far"*, offering a glimpse into his own professional journey and navigating career paths.

On the following day, Prof. Swart held a joint Hylleraas seminar entitled *"A new twist to old stories"*, discussing his recent studies in homogeneous catalysis with transition metal complexes, as well as his latest theoretical work in the development of DFT functionals. His lecture was also the first event of a new digital seminar series on theoretical chemistry, organized by the NKS Division for Quantum Chemistry and Modelling.

Prof. Swart's visit also offered Hylleraas members an invaluable opportunity to foster scientific discussions and academic connections. His visit featured personal talks with young researchers as well as senior members of the Centre. Additionally, Prof. Swart met the Oslo PIs and YRP representative Marinella de'Giovanetti over dinner on the first day.



Hva gjør hva vi drikker med tennene våre? ("What do beverages do to our teeth?") – one of the award-winning posters showcased at the poster competition during *Hvorfor Det?* on November 18, 2023. *Photo: Thomas Bondo Pedersen.*

Hvorfor Det?

Department of Chemistry, Universitetet i Oslo November 18, 2023

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Poster presentations and awards of curiosity-driven research by school children from Ila Primary School in Oslo, hosted by the Hylleraas Centre.

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On Saturday, November 18, 2023, 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 more than 30 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 Morten Ledum, Benedicte Sverdrup Ofstad, Håkon Emil Kristiansen, and Thomas Bondo Pedersen from the Hylleraas Centre. The "Audience Award" was determined by votes from about 200 participants. While the referees were discussing, Trygve Helgaker addressed an engaged audience on quantum mechanics and chemistry.

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

Sign made by Amalie Bondo Pedersen marking the entry to the Café at *Hvorfor Det? Photo: Thomas Bondo Pedersen.*

Trygve Helgaker speaking about electron clouds and their many shapes at Hvorfor Det? P*hoto: Thomas Bondo Pedersen.*

Professor Poul Jørgensen (left) and Rector Svein Stølen (right) speaking at the banquet dinner at the TRAINS conference on November 29, 2023. Trygve Helgaker with his back to the camera, and his wife, Barbara Reay Helgaker, in the background. *Photo: Thomas Bondo Pedersen.*

TRAINS Conference

Sundvolden Hotel, Krokkleiva November 27–30, 2023

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The 2023 annual meeting of the Division for Quantum Chemistry and Modelling of the Norwegian Chemical Society was turned into an international conference to mark the 70th birthday of Trygve Helgaker and to celebrate Trygve's Adventures IN Science.

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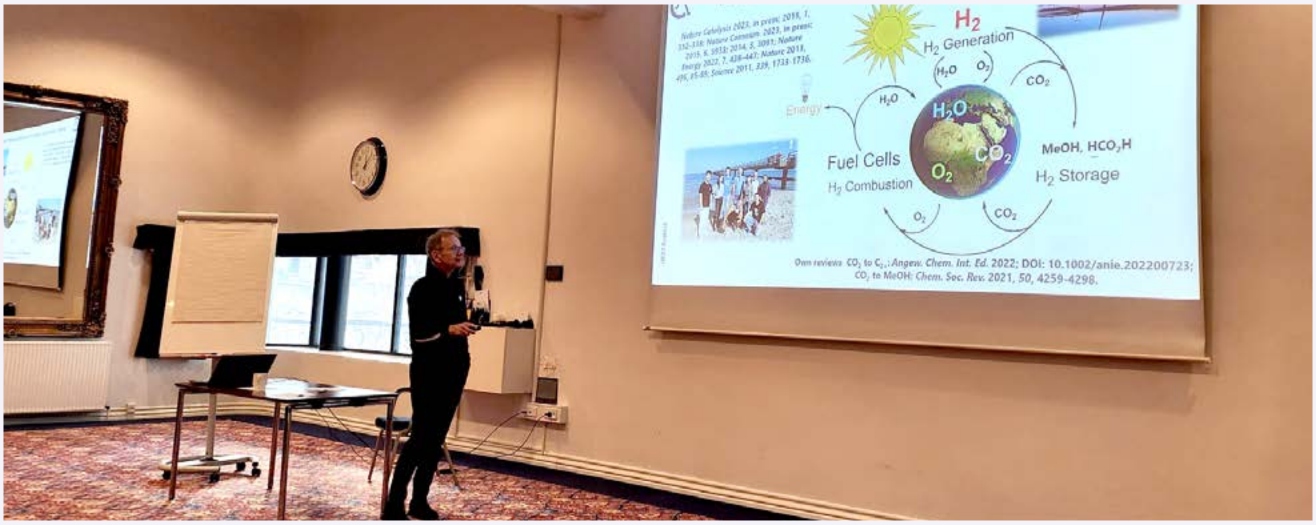
The TRAINS Conference was organized by Erik Tellgren, Michele Cascella, and Thomas Bondo Pedersen with assistance from Jan Ingar Johnsen at Sundvolden Hotel on November 27-30, 2023. The conference featured 29 invited lectures, mainly by former and current collaborators of Helgaker: 6 speakers from Norway, 6 from Denmark, 5 from Germany, 3 from the UK, 3 from the US, 3 from France, 1 from Italy, 1 from Sweden, and 1 from China. The conference also featured a poster session and the annual General Assembly of the Division for Quantum Chemistry and Modelling of the Norwegian Chemical Society. There were roughly 100 participants from Norway and abroad, and the event was sponsored by the Faculty of Mathematics and Science at the University of Oslo, the Hylleraas Centre, the Norwegian Chemical Society, Taylor&Francis, and Aker Scholarships.

Rector Svein Stølen at the University of Oslo attended the banquet dinner in the evening of November 29 and gave a celebratory speech. Professor Poul Jørgensen, Aarhus University, Denmark – a close collaborator through most of Helgaker's career– also spoke at the banquet dinner.



Prof. Matthias Beller giving his lecture at the NordCO2 and CCU-Net Winter School in Århus, on November 29, 2023. *Photo: Petra Agota Szilagyi*

Joint NordCO2 and CCU-Net Winter School

Helnan Marselis Hotel, Århus, Denmark
November 29-30, 2023

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The Nordic Consortia NordCO2 and CCU-Net organized a joint winter school in carbon capture and conversion in Århus, Denmark.

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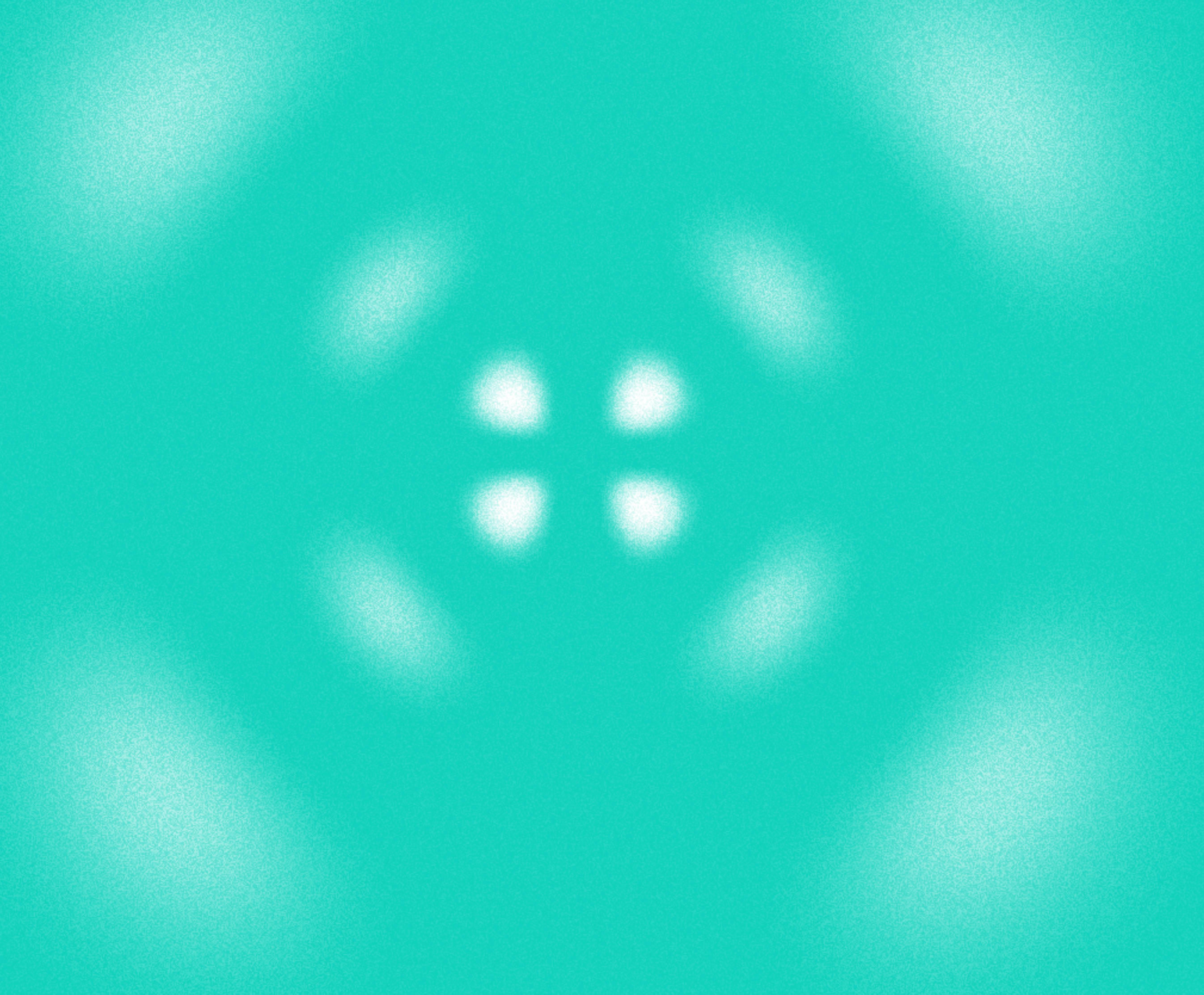
Within the framework of the NordCO2 consortium, Dr. Ainara Nova from the Hylleraas Centre and NordCO2 leader, together with Prof. Andreas Kaiser (Technical University of Denmark) and Dr. Karoline Thorum (Aarhus University), organized a winter School at the Helnan Marselis Hotel, in Århus, Denmark, on November 29-30, 2023. The topic of the school was carbon capture and conversion, and it had 63 participants. The school took place after the annual meeting of the Carbon Dioxide Activation Center (CADIAC), a Danish National Research Center led by Prof. Troels Skrydstrup, also a member of NordCO2. The connection of the school with the CADIAC annual meeting allowed the attendance of CADIAC members and NordCO2 and CCU-Net members at the meeting. Nova contributed to the meeting with an oral presentation, and Prof. Matthias Beller, a member of CADIAC, gave a lecture at the Winter School. Other lecturers from the University of Oslo were Prof. Unni Olsbye, PI at CCU-Net, and Assoc. Prof. Petra Agota Szilagyi.



Alumni Talks

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 seminar about their career choices and the position they hold today, whether in academia or in industry. In June 2023, two Hylleraas alumni presented themselves: Dr. Lluís Artús Suárez and Dr. Fabian M. Faulstich.

Dr. Lluís Artús Suàrez

Postdoc, KTH Royal Institute of Technology, Stockholm, Sweden
Life during and after a PhD

Online seminar from the Hylleraas Centre, University of Oslo, June 9, 2023
–

Dr. Fabian M. Faulstich

Assistant Professor and Eliza Ricketts Foundation Career Development Chair, Rensselaer Polytechnic Institute, Troy, New York, USA
An Academic Career Path

Online seminar from the Hylleraas Centre, University of Oslo, June 15, 2023
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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. Two seminars in June 2023 were dedicated to alumni talks by Dr. Lluís Artús Suàrez and by Dr. Fabian M. Faulstich.

At the Hylleraas Friday Seminar on June 9, Hylleraas alumnus Dr. Lluís Artús Suàrez presented himself and his career path after his PhD studies at the Hylleraas Centre in Oslo. Suàrez carried out his PhD work under the supervision of Ainara Nova and David Balcells together with affiliate Prof. Mats Tilset, and defended his thesis *Computational study on the deaminative hydrogenation of amides catalyzed by base metal complexes* in 2021.

On June 15, Dr. Fabian M. Faulstich presented his views and advice on a career in academia. Faulstich did his PhD research under the supervision of Simen Kvaal, Andre Laestadius, and Prof. Trygve Helgaker at the Hylleraas Centre in Oslo, and defended his thesis *Mathematical Aspects of Coupled-Cluster Theory in Chemistry* in 2020.

PhD Defences 2023

Candidate: Victoria Ariel Bjørnestad

University: University of Oslo
Date: April 12, 2023
Thesis: *Surfactant Micelles and Structural Pathways*
for Solubilization of Lipid Membranes
Trial Lecture: *Applying the principles of lipid self-assembly for encapsulating drugs,*
proteins, and nucleic acids
Supervisors: Prof. Reidar Lund, University of Oslo
Prof. Michele Cascella, University of Oslo
Committee: Prof. Uri Raviv, Hebrew University of Jerusalem, Israel
Assistant Prof. Constanza Montis, University of Florence, Italy
Assoc. Prof. Petra Agota Szilagyi, University of Oslo
Chair: Prof. Harald Walderhaug, University of Oslo
Format: physical

Candidate: Samiran Sen

University: University of Oslo
Date: May 26, 2023
Thesis: *Advances in Hamiltonian Hybrid Particle–Field Theory:*
Improving the description of interfacial systems
Trial Lecture: *Non-equilibrium molecular dynamics simulations and derivation of kinetics*
Supervisors: Prof. Michele Cascella, University of Oslo
Dr. Irep Gözen, University of Oslo
Committee: Prof. Stefano Vanni, University of Fribourg, Switzerland
Assoc. Prof. Zoe Cournia, Academy of Athens, Greece
Dr. Erik Tellgren, University of Oslo
Chair: Prof. Harald Walderhaug, University of Oslo
Format: physical

Candidate:

Linn Neerbye Berntsen

University: University of Oslo
Date: June 9, 2023
Thesis: *Development of New Synthetic Methods for the Preparation of N-functionalized Hydantoins*
Trial Lecture: *Overview of metal-free and metal-catalysed arylation methods*
Supervisors: Dr. Ainara Nova, University of Oslo
Assoc. Prof. Tore Bonge-Hansen, University of Oslo
Committee: Prof. Berit Olofsson, Stockholm University, Sweden
Assoc. Prof. Jørn Hansen, UiT The Arctic University of Norway
Assoc. Prof. Petra Agota Szilagyi, University of Oslo
Chair: Prof. Harald Walderaug, University of Oslo
Format: physical

Candidate:

Manuel Carrer

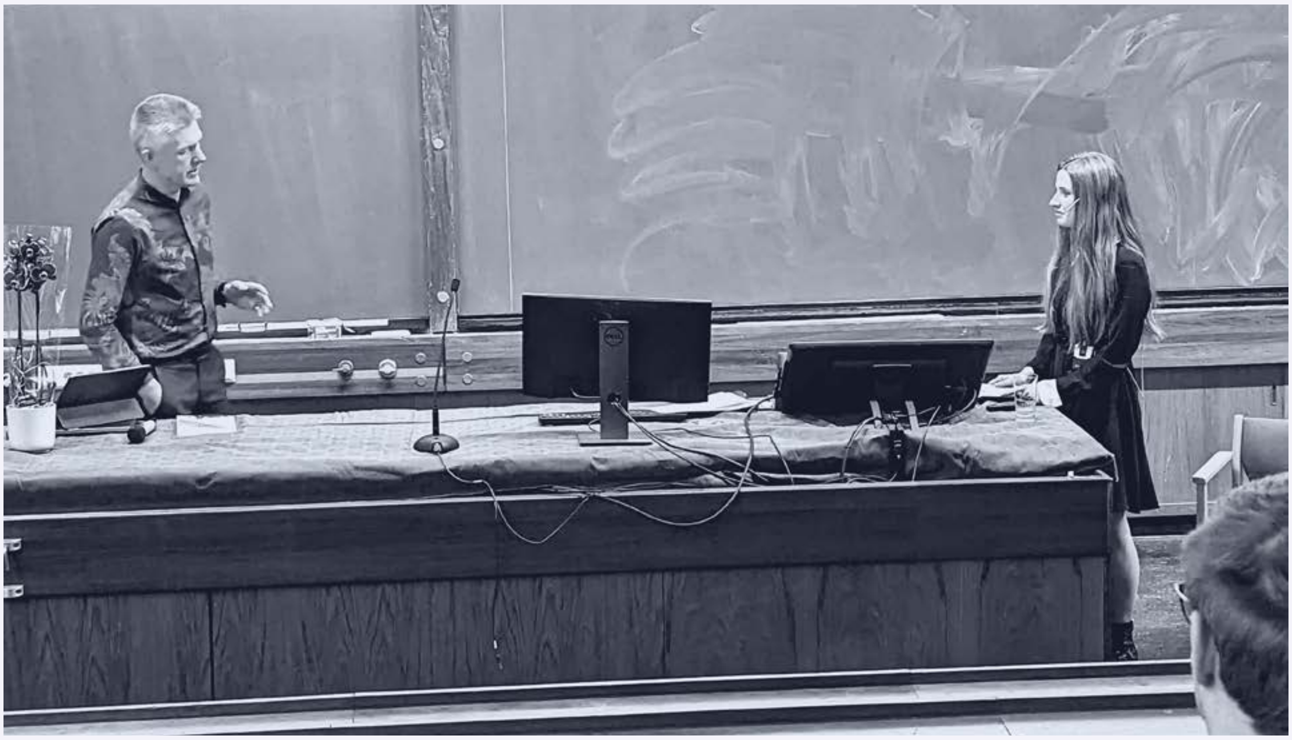
University: University of Oslo
Date: September 19, 2023
Thesis: *From All-Atom to Mesoscale: Bridging the Gap With Differentiable Molecular Dynamics*
Trial Lecture: *Mechanical properties of cell membranes: Helfrich theory*
Supervisors: Prof. Michele Cascella, University of Oslo
Prof. Reidar Lund University of Oslo
Committee: Prof. Tristan Bereau, Heidelberg University, Germany
Prof. Nathalie Reuter, University of Bergen
Prof. Steven R. H. Wilson, University of Oslo
Chair: Prof. Stian Svelle, University of Oslo
Format: physical

Candidate:

Mateu Montserrat Canals

University: University of Oslo
Date: September 25, 2023
Thesis: *Molecular warfare: A structural biology view on pathogen weapons – GbpA from Vibrio cholerae – and host defences – Vg from the honey bee*
Trial Lecture: *The fight against SARS-CoV-2 – a structural biology perspective*
Supervisors: Prof. Ute Krengel, University of Oslo
Prof. Hartmut Luecke, UiT The Arctic University of Norway
Prof. Michele Cascella, University of Oslo
Prof. Reidar Lund, University of Oslo
Committee: Prof. Thilo Stehle, University of Tübingen, Germany
Prof. Inari Kursula, University of Bergen
Senior Lecturer Hans-Petter Hersleth, University of Oslo
Chair: Prof. Harald Walderhaug, University of Oslo
Format: physical

Candidate:	Benedicte Sverdrup Ofstad
University:	University of Oslo
Date:	December 1, 2023
Thesis:	*Time-domain quantum dynamics: Optical properties from time-dependent electronic-structure theory*
Trial Lecture:	*Formulations of time-dependent variational principles for molecular systems*
Supervisors:	Dr. Simen Kvaal, University of Oslo Prof. Thomas Bondo Pedersen, University of Oslo
Committee:	Prof. Ove Christiansen, Aarhus University, Denmark Associate Prof. Ida-Marie Høyvik, Norwegian University of Science and Technology Prof. Reidar Lund, University of Oslo
Chair:	Prof. Steven R. H. Wilson, University of Oslo
Format:	physical

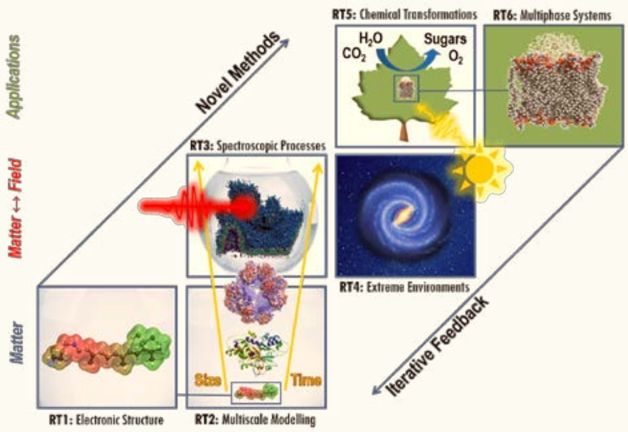


Benedicte Sverdrup Ofstad discussing with Prof. Ove Christiansen at her PhD defence on December 1, 2023.
Photo: Thomas Bondo Pedersen.

Avslør naturens hemmeligheter med **laptopen** som ditt **laboratorium**

Hylleraas-senteret for teoretisk kjemi bringer deg til forskningsfronten

Ved Hylleraas-senteret utvikler og anvender vi simuleringsmetoder 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 molekylær 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 eksperimentalister.
Kontakt: **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**

Sammen med **Ainara Nova** vil du ta i bruk simuleringsmetoder for å utforske struktur og reaktivitet for katalysatorer nødvendig for gjenvinning verdifulle kjemikalier fra CO_2. Forskningen omfatter mange ulike systemer, blant annet homogene og single-site heterogene katalysatorer.
Kontakt: **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**

Sammen med **Simen Kvaal** kan du blant annet utvikle nye metoder for å løse den tidsavhengige Schrödingerlikningen for molekyler på datamaskinen, utforske kvantekaos og grenselandet mellom kvante- og klassisk mekanikk, og benytte maskinlæring for å gjøre kvantekjemiske beregninger kjappere.
Kontakt: **simen.kvaal@kjemi.uio.no**

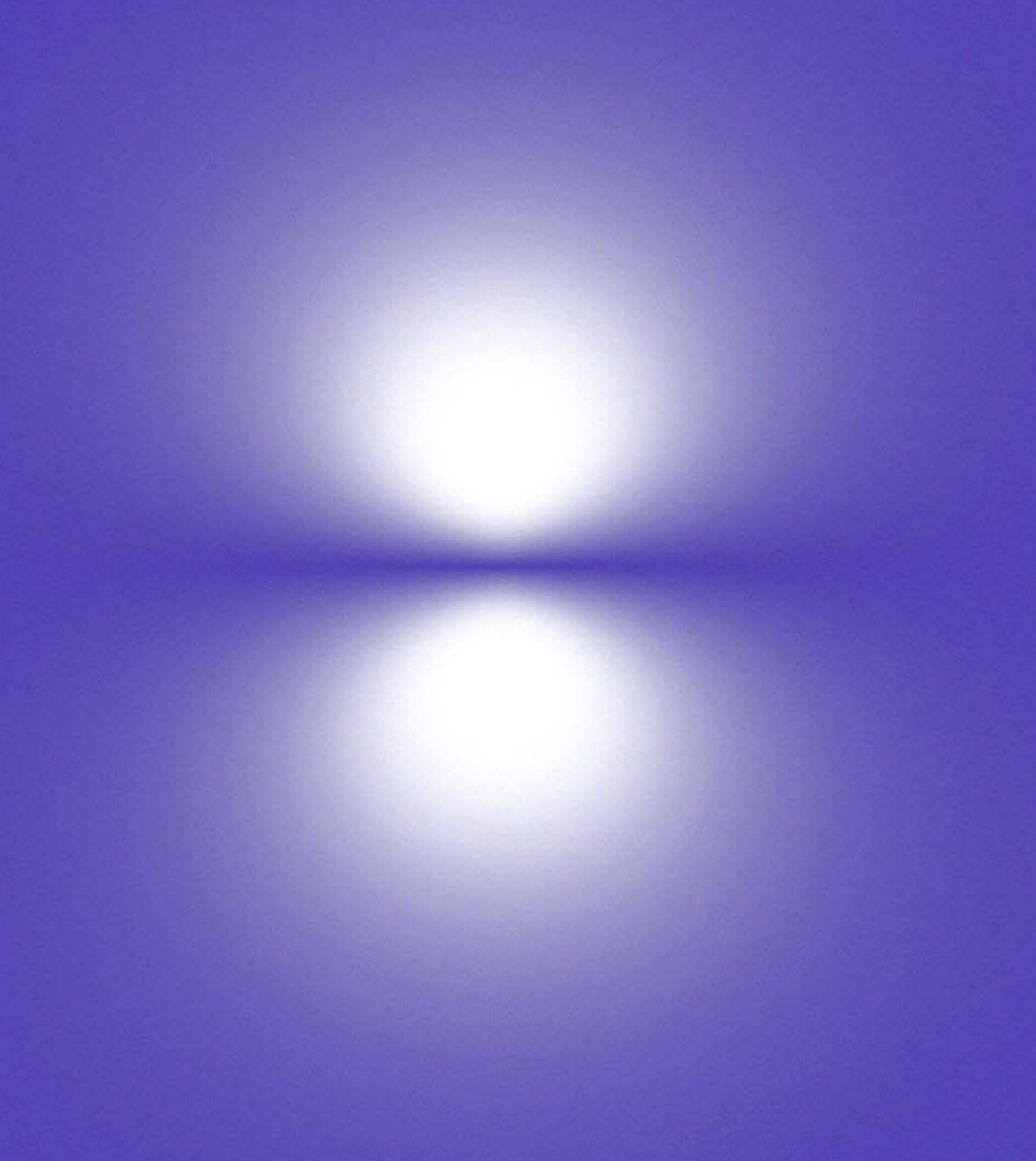
Med **Erik Tellgren** som veileder kan du utforske eksotisk kjemi for molekyler i sterke magnetfelter, hvor nye kjemiske bindinger og fysiske effekter gjør seg gjeldende. Prosjekter kan fokuseres mot anvendelse, programmering av beregningsmetoder, eller matematiske/teoretiske aspekter.
Kontakt: **erik.tellgren@kjemi.uio.no**

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.
Kontakt: **abril.castro@kjemi.uio.no**

← Besøk vår nettside

Hylleraas

Visits and Mobility



There were in total 21 visits and 20 unique visitors to the Hylleraas Centre in 2023.

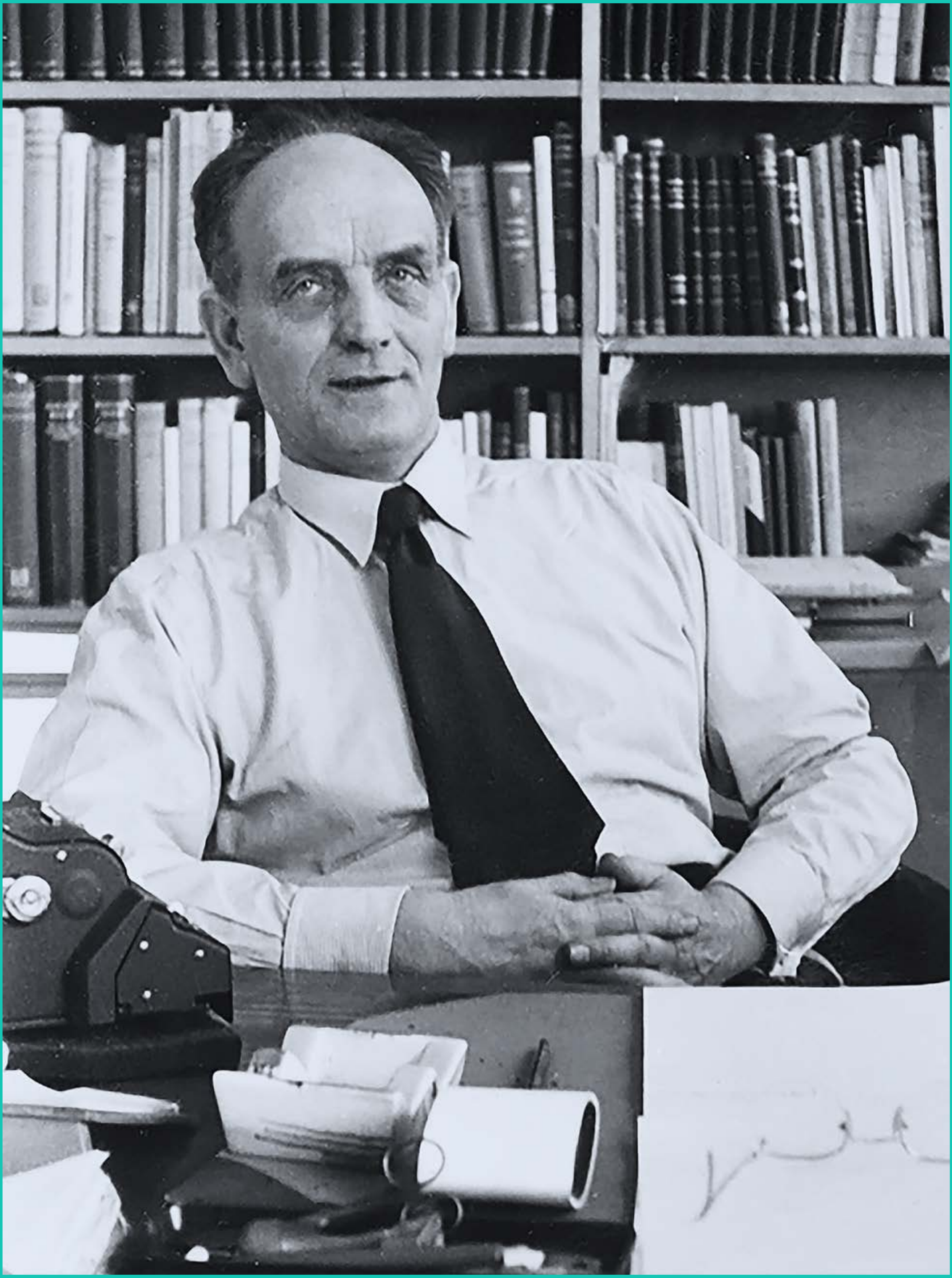
Incoming Visits

Visitors to Oslo

Prof Ludwik Adamowicz	University of Arizona, USA	Jan 5 – Jun 30
MSc Jiali Liu	Evonik Industries AG, Germany	Jan 16 – Jun 23
MSc Pedro Sodré	Federal University of ABC, Brazil	Feb 1 – Jul 31
Prof Caroline Lasser	TU München, Germany	Feb 20 – Mar 31
BSc Inmaculada Ri	University of Ciudad Real, Spain	Mar 1 – May 20
Dr Sylwia Kozdra	Lukasiewicz-Institute of	Mar 1 – Jun 30
	Microelectronics and Photonics, Poland	
MSc Gabriel M. Batista	Aarhus University, Denmark	Apr 17 – Jun 23
MSc Mads Greisen Højlund	Aarhus University, Denmark	Sep 1 – Nov 30
MSc Magnus Stransgaard	University of Copenhagen, Denmark	Sep 1 – Nov 30
MSc Jordan Rio	University of Lyon, France	Sep 18 – Dec 20
MSc Julia Kolodoazhnaia	Aarhus University, Denmark	Sep 25 – Dec 23
Prof Marcel Swart	University of Girone, Spain	Nov 8 – Nov 10
Prof Frank Neese	MPI Mülheim, Germany	Nov 20 – Nov 22

Visitors to Tromsø

Prof Johan Åqvist	Uppsala University, Sweden	Feb 8 – Mar 17
MSc Moritz Gubler	University of Bern, Switzerland	Apr 17 – Apr 21
Prof Robert Harrison	Stony Brooke University, USA	May 30 – Jun 1
Dr Monika Stas	University of Opole, Poland	Jul 27 – Aug 2
MSc Jonas Vester	Technical University of Denmark	Aug 6 – Oct 28
MSc Raunak Farhaz	Humboldt University of Berlin, Germany	Sep 1 – Oct 20
BSc Hugo Åström	Finland	Oct 1 – Oct 15
Prof Frank Neese	MPI Mülheim, Germany	Nov 22 – Nov 24



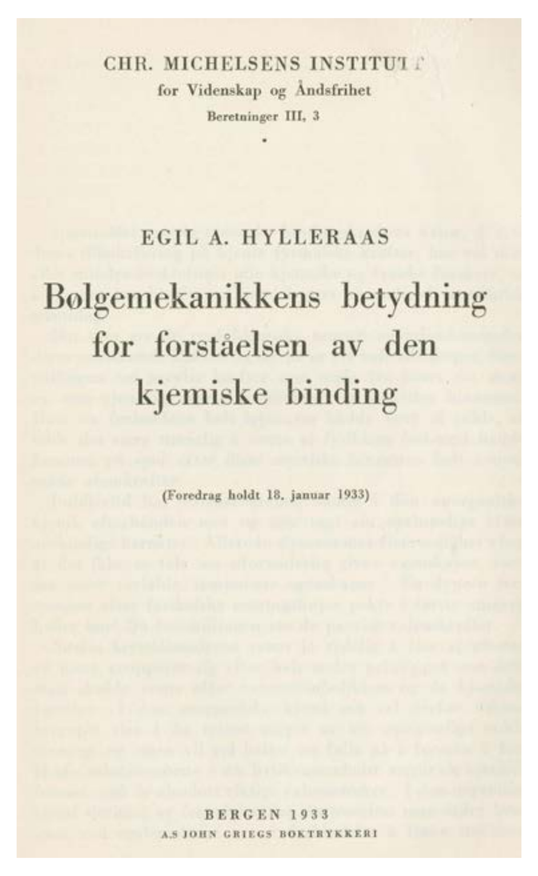
Egil A. Hylleraas

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)



CHR. MICHELSENS INSTITUTT
for Videnskap og Åndsfrihet
Beretninger III, 3

EGIL A. HYLLERAAS

Bølgemekanikkens betydning
for forståelsen av den
kjemiske binding

(Foredrag holdt 18. januar 1933)

BERGEN 1933
A.S JOHN GRIEGS BOKTRYKKERI

Visiting Researcher:

Caroline Lasser

Dr. Caroline Lasser is a professor at Zentrum Mathematik, Technische Universität München, Germany. She visited the Hylleraas Centre in Oslo from February to March 2023, working with Thomas Bondo Pedersen and Simen Kvaal on laser-driven molecular quantum dynamics.

Caroline Lasser obtained her PhD degree in mathematics at Technische Universität München in 2004. After a postdoctoral fellowship at Freie Universität Berlin from 2004 to 2005, she was first appointed Junior Research Group Leader there in 2005, and then professor in 2008. In 2010, she returned to Technische Universität München as professor for numerics of partial differential equations. Her research is focused on numerical analysis of the time-evolution of physical systems, such as the time-dependent Schrödinger equation.

Why did you choose to visit the Hylleraas Centre and how did you learn about the Hylleraas Centre and its visitor program?

I first visited the Hylleraas Centre as a project member of the Centre for Advanced Study project Attosecond Quantum Dynamics Beyond the Born-Oppenheimer Approximation. Even before the joint project, I was aware of the excellent international reputation of the theoretical chemists working at the Hylleraas Centre. I learned about the visitor program from my hosts Thomas Bondo Pedersen and Simen Kvaal.

In what way was the stay at the centre important for your scientific activities?

My stay helped for an improved understanding of the difficulties that have to be overcome for the numerical simulation of quantum dynamical systems using nonlinear model reduction. The visit has been one (of presumably many) building blocks for continuing the joint research with my project partners Thomas Bondo Pedersen and Simen Kvaal. As a trained numerical analysist, I tend to approach the design of numerical methods from a more mathematical point of view. Applicability to a "real" molecular simulation, however, requires more, and my research stay at the Hylleraas Centre helped a lot to develop a better understanding of these important aspects.

How did you find Norway in general and Oslo in particular?

Since I was trying to focus on my research work, I have not visited Norway beyond the Oslo premises. I have been charmed by Oslo's easily accessible natural surroundings, its cultural life (in particular the public library and the opera), but mostly by the relaxed friendliness of its citizens in everyday life.

Prof. Caroline Lasser. *Photo: Camilla Kottum Elmar/CAS.*

Visiting Researcher:

Moritz Gubler

Moritz Gubler is a PhD candidate in Physics at the University of Basel, Switzerland, working under Professor Stefan Goedecker. He visited Luca Frediani in Tromsø in April 2023 and attended the Hylleraas Hackathon at Skibotn.

Gubler's research focuses on areas like superconductivity, crystal structure prediction, and density functional theory. He also explores how to integrate machine learning into physics to improve computational models. His academic path and research interests stem from curiosity about material properties and their practical applications.

Why did you choose to visit the Hylleraas Centre and how did you learn about the Hylleraas Centre and its visitor program?

My visit to the Hylleraas Centre was inspired by a discussion with Luca Frediani at the Psi-k conference in Lausanne, where we discussed potential collaboration on geometry optimization of molecules using a multi-resolution wavelet basis at the DFT level. This conversation introduced me to the centre's visitor program, which seemed like an excellent opportunity to advance our project and access specialized expertise.

In what way was the stay at the centre important for your scientific activities?

My stay at the Hylleraas Centre was highly beneficial. Networking and discussions with the group of Luca Frediani, particularly in multi-resolution analysis, enriched my understanding significantly. This collaborative environment enhanced my ongoing projects and opened new research possibilities.

How did you find Norway in general and Tromsø in particular?

While most of my time was spent in Tromsø, my overall impression of Norway was very positive. The community spirit there was particularly notable. During a ski tour to the top of Imagaisi, a mountain in the Lyngen Alps, I lost a ski when it's binding unexpectedly opened, sending it down into a couloir. Unable to retrieve it myself, I mentioned this to locals, and, remarkably, someone kindly retrieved it for me a couple of days later. Additionally, participating in the Hylleraas Hackathon at Skibotn with other scientists was a definite highlight. These experiences, combined with Tromsø's beautiful scenery and vibrant community activities, made my stay enjoyable and memorable.

PhD candidate Moritz Gubler from the University of Basel, Switzerland, skiing in Troms.

Externally Funded Projects

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 2023. Progress in these projects will in the future be reported as part of the activities of the research themes.

H2O-MOF: Harnessing Hydrogen by Optimized Ion-Conductive Metal-Organic Frameworks

Dr. Sigbjørn Løland Bore (UiO)

–

Research Council of Norway
Young Research Talent (RCN-YRT) grant
2024–2028

Dr. Sigbjørn Løland Bore.

Dr. Sigbjørn Løland Bore has received a RCN-YRT grant with the project H_2O-MOF: Harnessing Hydrogen by Optimized Ion-Conductive Metal-Organic Frameworks. H_2O-MOF is a computational project aiming to model, understand, and optimize proton and hydroxide conductivity of metal-organic frameworks for potential use as an ion exchange membrane in fuel cells. With a budget of 8 MNOK, the project will support Bore for the next four years and the hiring of a postdoc for two-and-a-half years.

–

The development of fuel cell technology capable of efficiently converting hydrogen into electricity is considered a critical component in the transition away from CO_2-emitting energy sources. Ion exchange membrane fuel cells are gathering considerable interest due to their low electricity usage. However, this technology's full commercial potential is held back by the performance-critical ion exchange membranes (IEM) not meeting the challenges of simultaneously achieving high ion conductivity, low manufacturing cost, and high robustness.

Metal-organic frameworks (MOFs), a class of porous materials, exhibit channels that facilitate ion conduction, high structural stability, and chemical tunability, making MOFs a promising material for the next generation of IEMs. To exploit the full potential of MOFs, i.e., to rationally design MOFs, there is a knowledge need for understanding the underlying molecular mechanisms behind ion conduction inside MOFs.

To satisfy this knowledge need, Bore, in ***H_2O-MOF***, will develop a computational approach to model, understand, and optimize the ion conductivity in MOFs. To ensure high accuracy, ***H_2O-MOF*** will develop machine-learning-based models from first principles capable of describing, on the molecular scale, the dynamics of ions in MOFs. From the simulated real-time dynamics of the ions, the conductivity of the ion in the MOF will be extracted and validated against experiments. By investigating a varied set of highly conductive MOFs, ***H_2O-MOF*** aims to understand what makes a MOF a good ion conductor. By applying the molecular insight from the real-time dynamics in the MD simulations, ***H_2O-MOF*** aims to discover high-conductivity MOFs with potential as IEMs in fuel cells.

H_2O-MOF will be led by Bore and carried out at the Hylleraas Centre in Oslo, in close collaboration with Prof. Stian Svelle and Assoc. Prof. Petra Agota Szilagyi at the University of Oslo, and Veronique van Speybroeck from Ghent University in Belgium. The grant will fund Bore for four years and PostDoc for two-and-a-half years.

Can Ice be Described from First Principles?

Dr. Sigbjørn Løland Bore (UiO)
–
Centre for Advanced Study YoungCAS Fellowship 2023–2025

Illustration of the YoungCAS project *Can Ice be Described from First Principle? Illustration: Camilla Kottum Elmar/CAS*

Dr. Sigbjørn Løland Bore was awarded a YoungCAS fellowship grant by the Centre for Advanced Study (CAS) in 2023. Through the fellowship of 750 kNOK, Bore will gather researchers worldwide at the CAS in Oslo for four meetings with the ambitious goal of modeling water from first principles with high enough accuracy to capture the relative stability of different ice polymorphs.
–

From a cold glass of water on a sunny day to the uninhabitable Antarctica, ice is a constant companion for us humans on Earth. Of all the freshwater available, ice makes up 69% and covers 10% of the Earth's surface. On the one hand, ice is fundamental for humans, keeping water levels manageable for current human habitation, reflecting sunlight to slow down global warming, preserving food, or facilitating leisure activities, such as skiing in Nordmarka during winter. But, on the other hand, ice can pose an existential threat, for example, making you miss out on Christmas dinner due to long queues while waiting for the deicing of the airplane.

Understanding this crucial substance, ice, at its most fundamental level requires molecular insight. In theory, modeling from first principles provides just this. In practice, the high demand on accuracy needed to characterize accurately the melting point of ice is not met by the standard first principles approaches of reasonable computational cost, such as density functional theory (DFT) [1], leading to shifts in the melting point of ice by about 20 Kelvin in some cases [2], or incorrect description of the topology of water phase diagram [3].

Through his YoungCAS project, Bore gathers researchers worldwide with complementary expertise to bridge this gap between theory and experiment. Specifically, higher-accuracy alternatives to DFT exist, but they are computionally too expensive to characterize the thermodynamic stability of ice, such as the melting point. However, their cost is just within reach to create training data for machine learning-based models, which in turn can be used to efficiently characterize the thermodynamic stability of ice.

In this regard, the first meeting to be held in January 2024 will be dedicated to exploring the ORCA software developed by the Neese Group at the Max-Planck-Institut für Kohlenforschung, which provides highly efficient and accurate coupled-cluster calculations [4]. This initial workshop will be followed by three additional meetings, which aim to steadily incorporate high-level theory calculations with state-of-the-art msachine learning models to accurately capture the thermodynamic stability of ice from first principles.

References

[1] Gillan, M. J.; Alfè, D.; Michaelides, A. J. *Chem. Phys.* 2016, **144**, 130901.

[2] Zhang, L.; Wang, H.; Car, R.; Weinan, E. *Phys. Rev. Lett.* 2021, **126**, 236001.

[3] Reinhardt, A.; Cheng, B. *Nat. Commun.* 2021, **12**, 588.

[4] Neese, F. *WIREs Comput. Mol. Sci.* 2022, **12**, e1606.

Why is Ice so Slippery?

Dr. Sigbjørn Løland Bore (UiO)

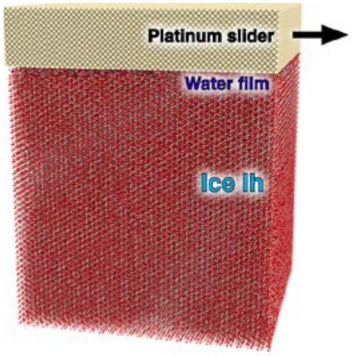
—

The European High-Performance Computing
Joint Undertaking (EuroHPC JU) grant
2024

Dr. Sigbjørn Løland Bore was awarded a EuroHPC JU grant of supercomputer time for the project Why is Ice so Slippery? A total of 134000 GPU node hours was granted on the LUMI supercomputer in Finland.

—

One of the most iconic properties of ice is its slipperiness. While humans exploit this property at a large scale in various sports, such as hockey and speed skating, we have yet to reach a clear scientific consensus on why ice is so slippery. Ultimately, the slipperiness of ice is an emergent phenomenon stemming from the interactions of the atoms at the interface between the sliding body and the ice, but the exact molecular mechanism of the slipperiness has not been determined. Bore, with Henrik Sveinsson at the Department of Physics, University of Oslo, were awarded the computing time needed to simulate the underlying process of metal sliding on ice at the atomic level. To reach the long length- and time scales needed to study this phenomenon under operating conditions (see illustration), they will use a highly scalable state-of-the-art machine learning potential approach [1] at the LUMI supercomputer. To study this phenomenon accurately, the model will be parameterized from first principles calculations at the hybrid DFT level of theory. Through the trajectories of the water molecules, they aim to unravel the molecular mechanism behind the slipperiness of ice.



Full-scale simulation setup for platinum sliding on ice. *Illustration by Sigbjørn Løland Bore*.

References

[1] Zeng, J; Zhang, D.; Lu, D.; Mo, P.; Li, Z.; Chen, Y.; Rynik, M.; Huang, L.; Li, Z.; Shi, S.; Wang, Y.; Ye, H.; Tuo, P.; Yang, J.; Ding, Y.; Li, Y.; Tisi, D.; Zeng, Q.; Bao, H.; Xia, Y.; Huang, J.; Muraoka, K.; Wang, Y.; Chang, J.; Yuan, F.; Bore, S. L.; Cai, C.; Lin, Y.; Wang, B.; Xu, J.; Zhu, J.-X.; Luo, C.; Zhang, Y.; Goodall, R. E. A.; Liang, W.; Singh, A. K.; Yao, S.; Zhang, J.; Wentzcowitch, R.; Han, J.; Liu, J.; Jia, W.; York, D. M.; Weinan, E.; Car, R.; Zhang, L.; Wang, H. J. *Chem. Phys.* 2023, **159**, 054801.

Rational Chemical Synthesis: Empowering Grignard Reagents with Machine Learning

Dr. Marco Bortoli (UiO)

—

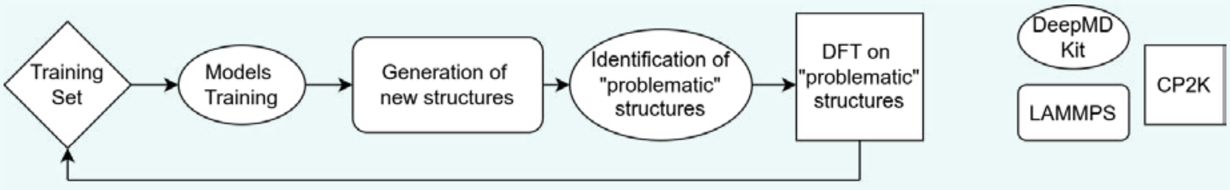
The European High-Performance Computing
Joint Undertaking (EuroHPC JU) grant
2024

Dr. Marco Bortoli was awarded a EuroHPC JU grant of supercomputer time for the project *Rational Chemical Synthesis: Empowering Grignard Reagents with Machine Learning*. A total of 65000 GPU node hours was granted on the LUMI supercomputer in Finland, and 13000 CPU node hours on the Leonardo supercomputer in Italy.

—

The goal of this project is the development of accurate machine-learned force fields to be employed in molecular dynamics simulation of Grignard and turbo-Grignard reagents taking advantage of state-of-the-art GPU accelerated machine learning codes.

Main group organometallic compounds are key components in a broad range of chemical reactions, from drug synthesis to fertilizer manufacturing. Their main application involves the formation of a new C-C bond in the so-called Grignard reaction. In the early 2000s, it was seen that the overall reactivity of these compounds can be improved by the addition of Li salts, creating the so-called turbo-Grignards [1]. However, these solutions are characterized by fast equilibria involving the solvent which makes their experimental characterization extremely challenging. Computational methods represent then the optimal investigation tool, although very accurate dynamical techniques are computationally very expensive and thus greatly limit the possibility to study a broad range of different compounds and environments. Machine learned potentials provide a promising solution to substitute *ab initio* counterparts to decrease the computational demand without any appreciable loss in accuracy. This requires significant computational resources but will allow to achieve close to a 1000x speed-up in production simulations.



Workflow to obtain the machine-learned force field. The different codes used (to the right) are interfaced with in-house written python code. *Illustration by Marco Bortoli.*

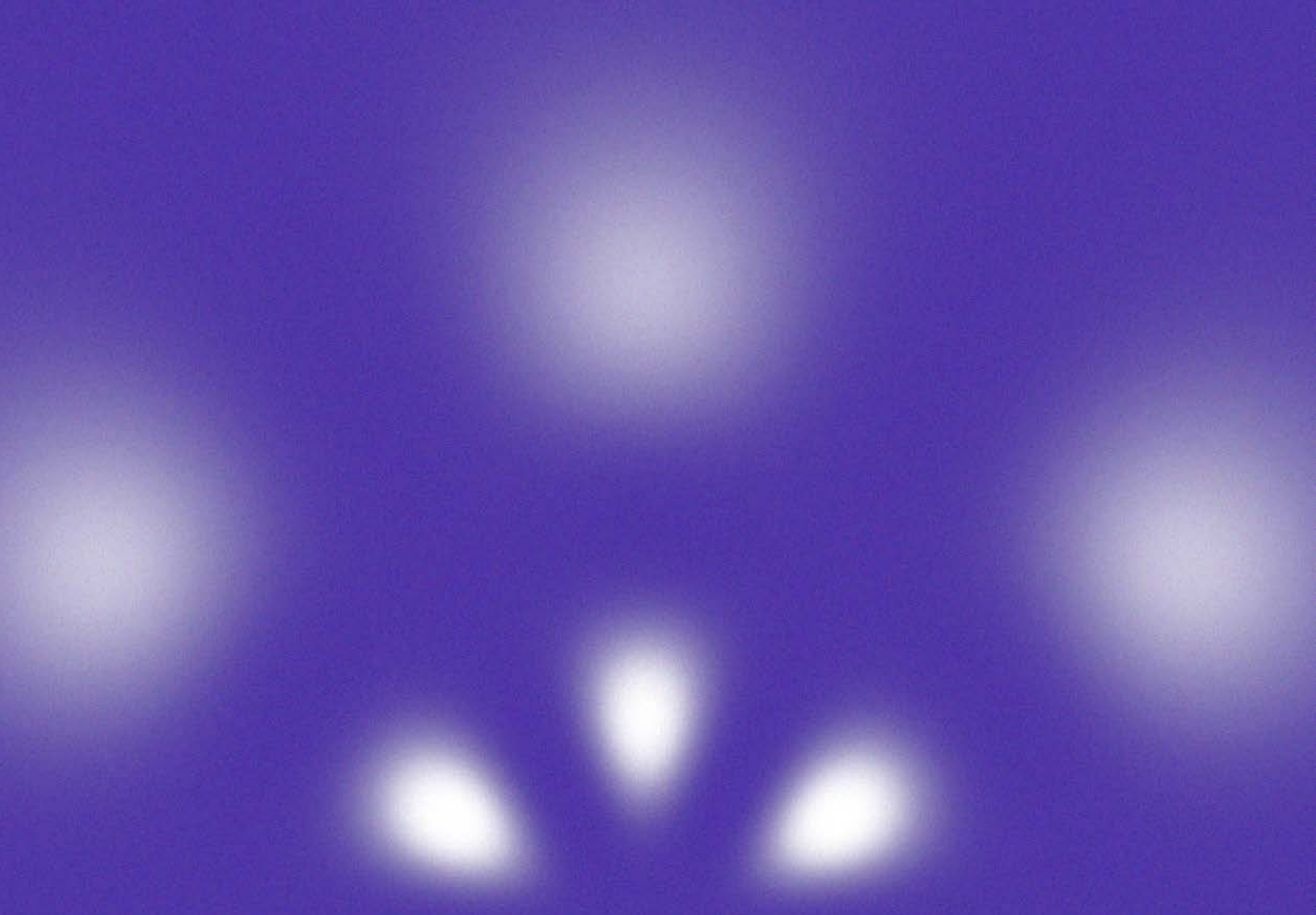
The project is divided in two main phases: model training and production simulations. The first embraces an active learning approach that exploits increasingly more accurate machine learned force fields to efficiently collect the data needed to create the training set used to train the final model (Figure 1), while the second consists in running molecular dynamics simulation using the previously developed machine learned force field on a broad array of Grignard and turbo-Grignard reagents to analyze their different behavior and thus shed light on the intricate mechanisms that govern the reactivity of Grignard and turbo-Grignard compounds.

Dr. Marco Bortoli

References

[1] Krasovskiy, A.; Knochel, P. *Angew. Chem. Int. Ed.* 2004, **43**, 3333.

Research Theme Activity Reports



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. From January to June 2023, our work was organized into six research themes (RTs):

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

On July 1, 2023, six new RTs were introduced, reflecting new research directions and changes in the leadership:

- **RT1** Molecular Quantum Mechanics
- **RT2/6** Multiscale Modelling and Complexity
- **RT3/4** Matter-Field Interactions
- **RT5** Chemical Transformations
- **RT7** Machine Learning and Mathematical Methods
- **RT0** Hylleraas Software Platform

Each RT has its own deliverables, directed towards the common goal of the centre: develop an integrated approach to the investigation of complex molecular systems in electromagnetic fields and in extreme environments, including:

- accurate and efficient modelling of complex systems in space and time
- understanding matter–field interactions and extreme environments

We aim to treat systems containing from a few to thousands and even millions of atoms, to unravel their response to short laser pulses and strong fields, and to explore processes extending from attoseconds to microseconds.

In the following, the 2023 report is provided for each of the new RTs, including activities from the first half of 2023 within the old RTs.

RT1

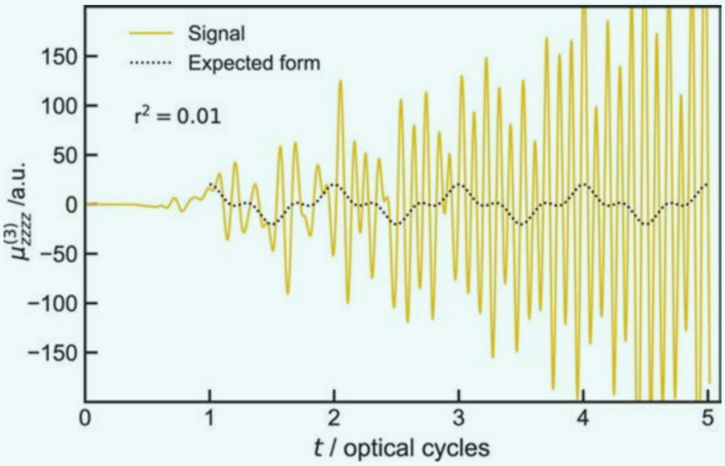
Molecular Quantum Mechanics

Principal investigators:
Thomas Bondo Pedersen
and Luca Frediani
–

Molecular quantum mechanics is the application of the laws of quantum mechanics to study chemical systems. While this is commonly interpreted as electronic structure and dynamics within the Born-Oppenheimer approximation, research in RT1 is based on the wider definition where both electronic and nuclear degrees of freedom are treated quantum mechanically, either with or without the Born-Oppenheimer approximation. A key motivation is to develop accurate computational tools to understand, interpret, and predict molecular properties and reactivity, including laser-induced processes down to the attosecond time scale.

Non-Born-Oppenheimer methods

In the past decades, quantum-mechanical methods and algorithms have emerged that allow us to study the stationary (bound) states of small molecules without assuming the Born-Oppenheimer approximation, i.e., treating the electrons and nuclei on the same quantum-mechanical footing. In this approach, the full molecular wave function is expressed as a linear combination of explicitly correlated Gaussian (ECG) basis functions, with proper treatment of all particle permutation symmetries. In 2023, we have made progress in the generalization of the ECG *ansatz* from the time-independent to the time-dependent domain. First, as described in more detail in the research highlight above, Lucas Lang (UiO) in collaboration with Henrique M. Cezar (UiO), Ludwik Adamowicz (Arizona), and Thomas Bondo Pedersen have developed an algorithm for extracting the molecular structure (or structures, if more than one is present) using only the wave function as input [1]. Second, in collaboration with Simen Kvaal (see also the activity report for RT7), Aleksander Wozniak (Warzaw), and Ludwik Adamowicz, we have demonstrated the ability of fully flexible ECGs – many-body Gaussian wave packets – to capture complicated laser-induced quantum dynamics involving both electronic and rovibrational continua (i.e., ionization and bond dissocia-



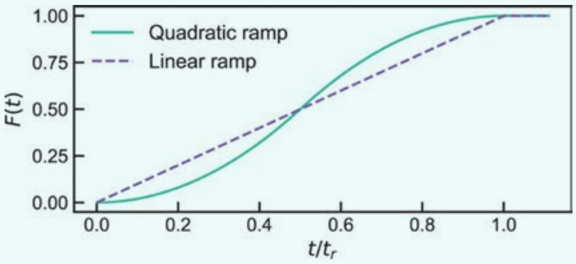


Figure 2: Shape of the linear and quadratic ramps. Both are continuous on the interval 0 to 1, but only the quadratic ramp is smooth at the end points. *Reproduced from [5] under CC-BY 4.0 license.*

Figure 1: Third-order induced dipole moment (yellow) obtained from TDCCSD simulations of the Ne atom with single-cycle linear ramping of the electric-field strength. The black dotted curve shows the form of the signal expected from cubic response theory. Not surprisingly, the second hyperpolarizability component extracted from the simulation does not agree well with that obtained from cubic response theory. *Reproduced from [5] under CC-BY 4.0 license.*

tion processes) [2]. Together with the development of Rothe propagation methods in a direct collaboration between RT1 and RT7 (see the activity report for RT7), these research results form the basis for novel studies of the full molecular quantum dynamics induced by strong attosecond laser pulses generated by means of, e.g., the high-harmonic generation techniques that earned Agostini, Krausz, and L'Hullier the 2023 Nobel Prize in Physics.

Coupled-cluster theory

Laser-driven electron dynamics (with the Born-Oppenheimer approximation) remains a focus area of RT1, and in 2023 we published an invited *Advanced Review* of time-dependent coupled-cluster theory [3] with main emphasis on developments in the past decade on vibrational as well as electron dynamics. The paper describes both bound-state and ionization processes, areas where many efforts in RT1 have been invested during 2023.

In a collaboration with Michal Repisky, Marius Kadek, and Lukas Konecny (RT3/4), former master student Eirill Hauge, together with Håkon Emil Kristiansen and Thomas Bondo Pedersen, published a paper [4] on a dipole-extrapolation method that can accelerate the simulation of linear absorption spectra by more than an order of magnitude while improving the spectral resolution, see the activity report of RT3/4.

Another application area of time-dependent coupled-cluster theory is the calculation of polarizabilities and hyperpolarizabilities in transparent (low-frequency) spectral regions. This is a challenging problem because (hyper-) polarizabilities are only defined in terms of a perturbation expansion of the induced dipole moment in the frequency domain, i.e., in terms of linear (polarizability), quadratic (hyperpolarizability), and cubic (second hyperpolarizability) response theory, while the induced dipole moment obtained from a time-dependent simulation contains all perturbation orders. This implies that one must separate the orders using, e.g., numerical derivatives with respect to the electric-field strength, while carefully ensuring that the field is switched on adiabatically – i.e., very slowly compared with the electronic time scale, in agreement with the fundamental assumptions of time-dependent perturbation theory. In previous studies, including those published earlier by Hylleraas members, the adiabatic switching-on of the electric field was implemented as a linear ramping of the field strength over one or two optical cycles. The linear ramping, however, leads to visible non-adiabatic effects in the higher-order time signals which, in turn, lead to significant disagreement between results obtained from simulations and those from frequency-domain response theory, see Fig. 1. In 2023, PhD student Benedicte Sverdrup Ofstad replaced the linear ramp with a quadratic ramp (Fig. 2) and found that almost per-

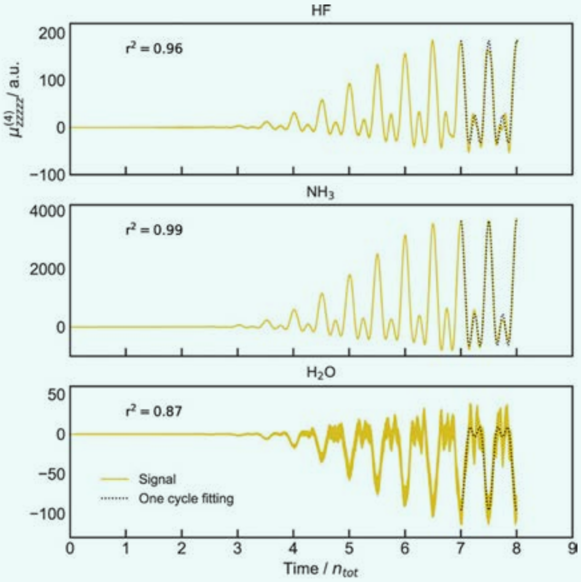


Figure 3: Fourth-order induced dipole moment (yellow) obtained from TDCCSD simulations of the HF, NH_3, and H_2O molecules using a seven-cycles quadratic ramp. The dotted lines show the fitted signals. The agreement between the computed and fitted signals are not as impressive as for the lower orders, but still allows for a reliable estimate of the frequency-dependent third hyperpolarizability. Note that the third hyperpolarizability has never been implemented in a coupled-cluster response code, likely due in part to the significant human effort required. *Reproduced from [5] under CC-BY 4.0 license.*

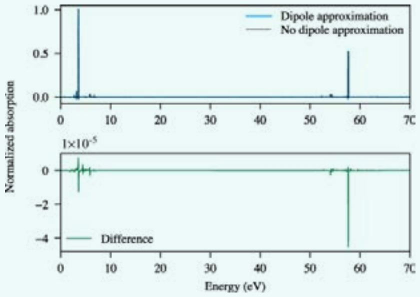


Figure 4: Transient absorption (pump-probe) spectra computed *at the TDCCSD level of theory with and without the electric-dipole approximation for the LiH molecule (top). Beyond-dipole effects are negligible as expected (bottom). *Reproduced from [7] with permission from the American Physical Society.*

fect signals could be achieved, leading to near-perfect agreement with up to cubic response theory at about half the computational cost [5]. Even the third hyperpolarizability, corresponding to quartic response theory, can be quite reliably extracted (Fig. 3).

Subsequently, Ofstad and co-workers used the quadratic-ramp approach to study the limitations of Verdet's law of magnetic optical rotation [6], as described in the RT3/4 activity report.

Simulation of ultrafast transient absorption spectra is an important area where real-time time-dependent electronic-structure theory plays a key role – see the Research Highlight above and the RT3/4 activity report. Within RT1, PhD student Einar Aurbakken has investigated the effects of higher-order multipole components of a laser pulse. Most theoretical studies of transient absorption spectroscopy are based on the electric-dipole approximation where the electric field of the laser pulse is assumed to be spatially uniform while the magnetic field vanishes. This is a very accurate approximation if the wavelength of the radiation field is much greater than the size of the atomic or molecular system. However, since attosecond and few-femtosecond laser pulses have wavelengths that can be similar to molecular dimensions, it becomes necessary to go beyond the electric-dipole approximation. In [7], Aurbakken and co-workers formulate a general, fully gauge-invariant theory of transient absorption that is applicable both within and beyond the electric-dipole approximation.

Using a full plane-wave vector potential, which includes all multipole orders, Aurbakken et al. showed that transient absorption spectra agree well with the electric-dipole approximation for long wavelengths (Fig. 4), whereas beyond-dipole effects, albeit weak, automatically appear for shorter wavelengths in agreement with experiment (Fig. 5).

Our work on time-dependent coupled-cluster theory has been limited to single-reference models, implying that strongly correlated systems cannot be accurately treated. While this is a well-known issue, it is notoriously hard to judge if a system is strongly correlated based on a single-reference coupled-cluster calculation alone. In a collaboration with former Hylleraas member Dr. Fabian Faulstich (Rensselaer Poly-

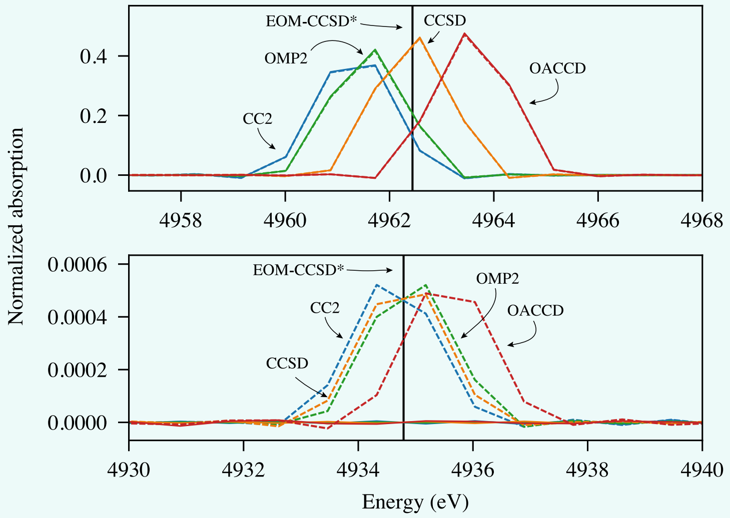


Figure 5: Transient absorption spectra computed with a variety of time-dependent coupled-cluster models with (solid lines) and without (dashed lines) the electric-dipole approximation for the Ti^{4+} ion. **Top:** absorption lines corresponding to the dipole-allowed transition from the core 1s orbital to the 4p orbital. Bottom: absorption lines corresponding to the dipole-forbidden 1s to 3d transition, which is observed experimentally. *Reproduced from [7] with permission from the American Physical Society.*

technic Institute, USA), Simen Kvaal, Andre Laestadius, and Mihaly Csirik (RT7), we have proposed the S-diagnostic to indicate if a single-reference coupled-cluster calculation is reliable or not [8]. The S-diagnostic is based on the mathematical analysis of coupled-cluster methods, which has been a major topic in the first six years of the Hylleraas Centre.

Grid methods

Atom-centered Gaussian-type orbitals are the most widely used basis functions in electronic-structure theory, as they provide a relatively compact and computationally efficient description of the ground and bound excited states. In the context of attosecond chemistry, however, they are significantly less useful because they do not capture the electronic continuum and, hence, cannot be used to describe ionization dynamics. Addressing this issue has been a focus of RT1 in collaboration with RT7 in 2023.

As mentioned above and described in the RT7 activity report, PhD student Simon Elias Schrader has worked on the challenging propagation of Gaussian wave packets. At the same time, postdoc Håkon Emil Kristiansen and PhD student Einar Aurbakken have worked on pseudo-spectral and finite-difference methods that can provide virtually exact results for single-particle problems such as single active electron approaches to strong-field processes like high-harmonic generation and ionization. While these tools provide benchmark results for the work of Schrader, they can also be used to formulate *ab initio* many-body models such as time-dependent Hartree-Fock theory and time-dependent configuration-interaction-singles theory. In a collaboration with Dr. Antoine Camper at the Department of Physics at UiO and member of the AEGIS collaboration at CERN, such many-body methods will be used to simulate the attosecond dynamics of matter-antimatter systems such as positron-chloride, which is planned to be experimentally investigated using attosecond spectroscopies at CERN after the next upgrade.

Multiwavelets

Most quantum chemistry methods make use of atom-centered basis functions. This choice has a clear physical meaning (it regards atoms as the essential building blocks) and numerical advantages (elementary operations with

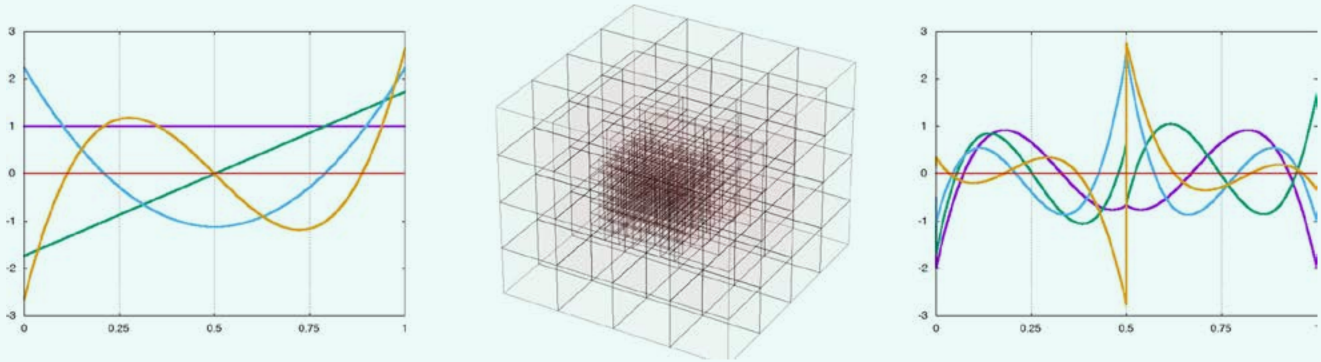


Figure 6: Left panel: scaling functions of order k = 3 defined in the interval [0,1] are simple polynomials. Right panel: the corresponding wavelet functions are piece-wise polynomials with four vanishing moments (orthogonal to polynomials up to the cubic one). Central panel: adaptive grids are constructed on demand to minimize storage and meet precision requirements. *Reproduced from [9] under CC-BY 4.0 license.*

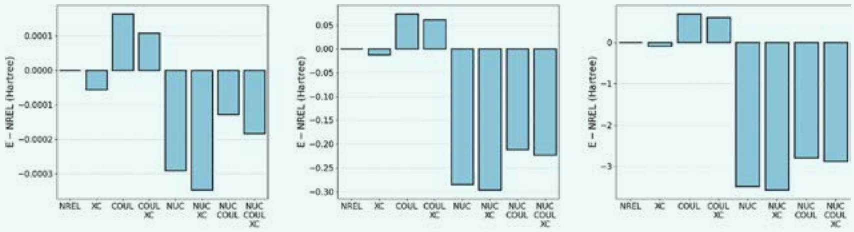
Gaussian-type orbital are efficient and standardized). But obtaining high precision results can be challenging. Grid methods offer an alternative, as mentioned above. Multiwavelets offer an alternative route, providing strict error control and guaranteed precision. A large part of RT1 in Tromsø is focused on developing methods to use Multiwavelets to provide high quality results for energies and properties.

One very active direction we are currently pursuing is the development of relativistic methods using Multiwavelets. The inclusion of relativity can be done at several levels. The most accurate makes use of the Dirac equation and four-component spinors. This is also the most expensive approach. Two-component or scalar methods provide computationally simpler alternatives, at the price of reduced accuracy. Common to all such methods is the fact that describing the electronic structure with atomic orbitals becomes challenging. On the one hand, the choice of basis sets for heavier atoms – where relativistic effects are important – is limited, and on the other hand the relativistic effects are important close to the core – where atomic orbitals struggle to provide a precise representation. Multiwavelets represent functions (orbitals, densities) on an adaptive grid, to guarantee high resolution where necessary.

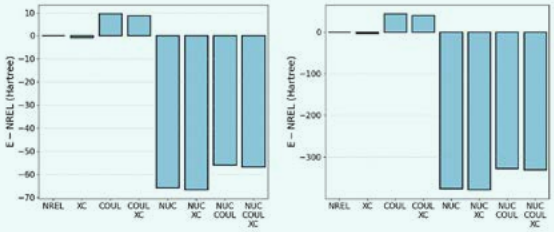
Postdoc Christian Tantardini, adjunct professor Roberto Di Remigio, PhD student Magnar Bjørgve, and researcher Stig Rune Jensen have shown how the MW formalism can be used to compute all terms of the Breit Hamiltonian, which are notoriously challenging to implement using atomic orbitals [9]. Postdoc Christian Tantardini and adjunct professor Roberto Di Remigio and Luca Frediani have implemented an approach based on the square of the Dirac operator to solve the Dirac equation, which allows to use of the same technology employed for the non-relativistic case [10].

Currently PhD student Quentin Pitteloud is also working on the extension to the two-component regime of the ZORA formalism. This follows a previous work published in 2023 which focused on the scalar ZORA formalism [11]. The extension to two components response properties calculations will open the way for a collaboration with Abril Castro (RT5) on the modeling of NMR spectroscopy for heavier elements at the basis set limit.

Another area where using multiwavelets is very beneficial is in providing precise



(a) Helium (b) Neon (c) Argon



(d) Krypton (e) Xenon

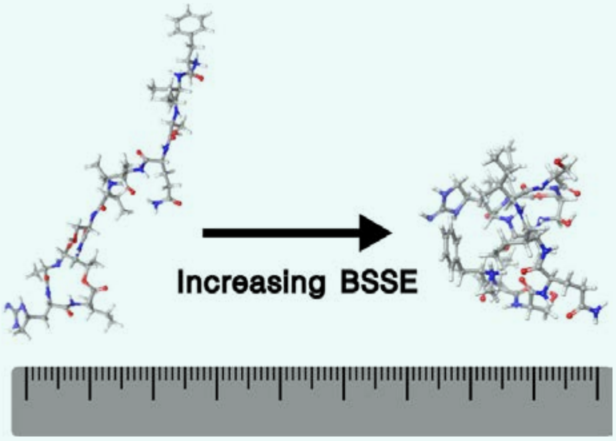
Figure 7: The MW formalism allows an unprecedented flexibility, and the ZORA operator can include any combination of the different potentials (Nuclear, Coulomb, Exchange&Correlation). We could therefore show how the energy changes by including them selectively, compared to the non-relativistic case. The picture shows this effect for all noble gases from He to Xe. Note that the scales on the y-axes are different for each sub-plot. The size of the correction increases with the fourth power of the nuclear charge. *Reproduced from [11] under CC-BY 4.0 license.*

results for larger systems. PhD student Quentin Pitteloud, researchers Peter Wind and Stig Rune Jensen have collaborated with Professor Frank Jensen (Aarhus University, Denmark) have demonstrated how MW methods can be employed to benchmark elusive problems such as the intramolecular Basis Set Superposition Error which arises when different conformations of the same peptide are compared to each other. They have shown that even triple-zeta quality bases can suffer from sizeable errors and quintuple-zeta bases are required to obtain small results [12]. However, making use of such bases for larger molecules is both very expensive and leads to convergence issues. Multiwavelets, which are orthonormal by construction, are both precise and numerically stable.

Another line of development regards the inclusion of solvent effect. We here exploit the strength of the Multiwavelet framework, which allows to solve the generalized Poisson equation in the three-dimensional space rather than transforming it to a boundary-problem. This is both computationally simpler (no need to parametrize a molecular cavity) and physically sounder (there is no real boundary between the solvent and the solute). PhD student Gabriel Gerez, and adjunct professor Roberto Di Remigio implemented a continuum solvation model based on the direct resolution of the Generalized Poisson Equation which includes both surface and volume polarization effects [9]. This work is lately being extended to the calculation of molecular properties (frequency-dependent polarizabilities in particular) and to the inclusion of ionic solutions through the Poisson-Boltzmann equation.

As part of the NFR-funded ReMRChem project we have organized the NMQC conference in Tromsø. It took place at the beginning of June, with more than 50 participants covering, in particular, real-space methods, relativity and correlation.

Dr. Evgueny Dinvay joined the group in Tromsø in 2023 as a postdoc. He is an applied mathematician with experience in methods for time propagation. He has therefore devoted much of his work on the development of a novel time propagation method for Multiwavelets. A manuscript describing the method is currently in preparation.



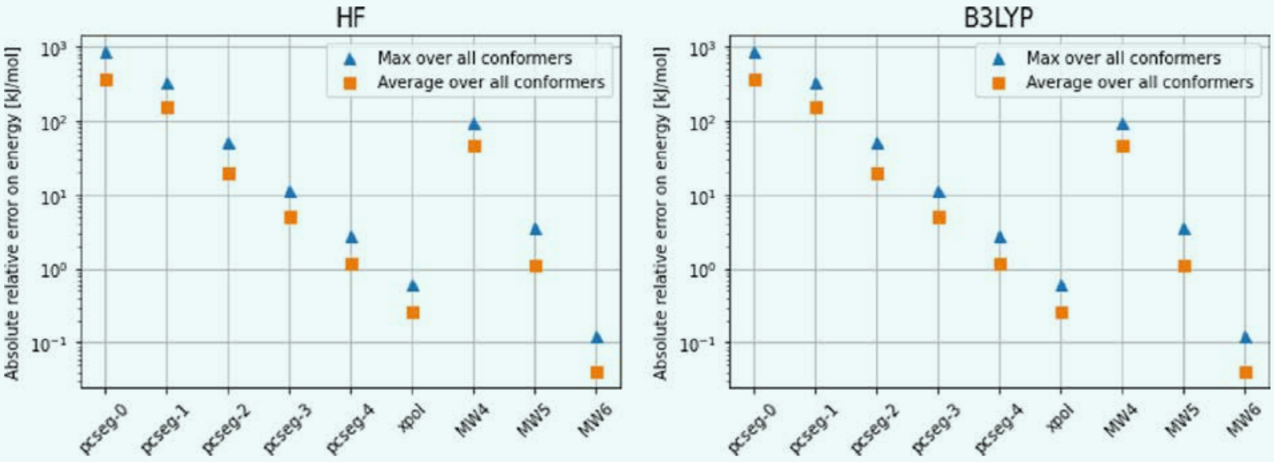


Figure 8: For a sample of conformers of the polypeptide showed above we have used MWs to show that there is a clear correlation between the intramolecular basis set superposition error (BSSE) and the compactness of the conformation: more compact conformations display a stronger BSSE due to the increased overlap between different parts of the molecule. The left and the right panel show the average error obtained for the pcseg-X series of basis sets (X=0,1. 4 and extrapolated) compared to MW calculations. MWY (Y=4,5,6) means a MW calculation with a requested precision of 10^{-Y}. The MW7 calculations have been taken as a reference. MW4 calculations are in between pcseg-1 and pcseg-2. MW5 results are on par with pcseg-4 and MW6 results outperform the extrapolated pcseg results. *Reproduced from [12] under CC-BY 4.0 license.*

wOne of the key advantages of Multiwavelets is a clean formalism which stays true to the original equations. This strength has been exploited by PhD student Magnar Bjørgve, who has developed a Python interface for Multiwavelets, called VAMPyR (Very Accurate Multiresolution Python Routines). He has documented the code in a recent publication, showing how it can be used successfully for a variety of pilot projects and for teaching purposes [13] PhD student Quentin Pitteloud has worked on a pilot response implementation using VAMPyR, which will serve as a basis to interface the Multiwavelet framework to the Open-ended response formalism currently developed by Magnus Ringholm (RT3/4), Vanda Le (RT3/4), and Bin Gao (RT0).

Another area where the strengths of Multiwavelets can be exploited is in the modeling of X-Ray spectroscopy for larger molecules which have interesting applications in material science. Core-properties for large molecules are a particularly hard test for traditional bases, and Multiwavelets are expected to yield precise results combined with stable computational protocols. Adjunct professor Laura Ratcliff has started working on X-Ray spectroscopy simulations by means of DeltaSCF methods using Multiwavelets, in combination with the Maximum Overlap Method.

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Ingoing and outgoing personnel

Benedicte Sverdrup Ofstad defended her PhD thesis on December 1, 2023, and moved on to a position as Scientist at the Institute for Energy Technology in Kjeller, Norway.

Evgueni Dinvay joined the MRChem group at the Hylleraas centre in Tromsø, as a Postdoctoral fellow for three years.

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RT2/6

Multiscale Modelling and Complexity

Principal investigators:
Michele Cascella and Bjørn Olav Brandsdal
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To study large-scale responses to local perturbations, we develop methods, software, and virtual-reality tools bridging electronic structure, condensed matter, and the mesoscale. We apply these methods to bio-soft matter and address mechanisms associated with health threats like bacterial drug resistance.
–

2023 witnessed the completion of several projects developed during the past years, which reached publication status during this year. They spanned methodological and software development, as well as applications in bio-soft matter.

The HylleraasMD software

Motivated by seeding work by Researcher Sigbjørn L. Bore and Michele Cascella [1], a group of scientists at UiO: Eng. Morten Ledum, postdoc Xinmeng Li, former PhD students Manuel Carrer and Samiran Sen, together with Bore and Cascella, developed HylleraasMD (HyMD), a scientific software dedicated to Hamiltonian Hybrid Particle Field (HhPF) Molecular Dynamics simulations. This approach guarantees systematic numerical control over the integration of the equations of motion, guaranteeing correct sampling of the thermodynamic ensembles of interest, and removal of aliases coming from the use of finite meshes. Already anticipated in the Hylleraas 2021 yearly report, HyMD is now fully released as open-source code, and it is available for free download and use under LGPLv3.0 licence [2,3]. This choice is in accord with EU policies for Open Science practice, which promote free access to scientific software, for better data accessibility, reproducibility, and reliability.

The physics behind the hybrid particle field method

Despite having been introduced for more than two decades, the level of approximations and thus, the type of physics described by hybrid particle-field approaches has not been clarified. In particular, past literature has not been ablet to clearly dissect what could be intrinsic methodological boundaries from numerical limitations of specific implementations. Reliably implementing Hamiltonian mechanics equation, HyMD provides a solid base to dissect such conundrum. Starting from [1], Ledum demonstrated the equivalence between HhPF and any two-body potential, solely dependent on the choice of the ana-

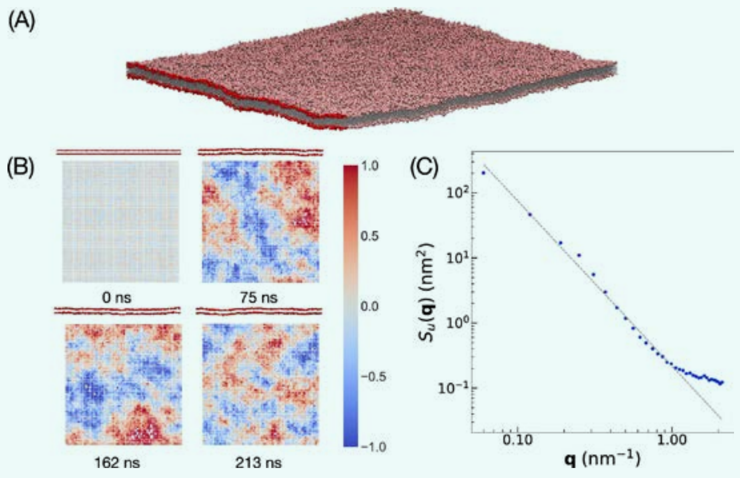

Figure 1: (A) Snapshot of a 100x100 nm2 DPPC bilayer. One edge has been brightened to highlight the curvature; (B) heatmaps of the positions of the lipids with respect to their average height, revealing undulations in the membrane (C) Undulation spectrum Su(q) as a function of wave vector q. Axes are in log-scale. *Reproduced from [5] under CC-BY 4.0 license.*

lytic expression of the filter function [4]. This result evidenced how past research employing hPF methods has worked in a regime of strongly overlapping soft particles, which shows *mean field-like* weak coupling between the bodies included in the system.

Direct consequence of this result is that HhPF simulations must be able to reproduce any property detectable by conventional simulation methods. In particular, they must be able to produce anisotropic pressure tensors in the presence of non-homogeneously distributed moieties like in non-miscible multi-phase systems. The work by Sen, which was showcased in the yearly report 2022, addressed precisely this point in the context of lipid membranes. The study by Sen showed how HhPF is able to reproduce quantitatively well the mechanical properties of lipid bilayers (Figure 1), and reached publication status also in 2023 [5].

Antimicrobial peptide scramble lipid bilayers

Membrane-active peptides constitute an important class of drugs that can help tackling the insurgence of multi-drug resistant bacterial strains. Despite the identification of a wide range of amino acid sequences showing antimicrobial activity, the mechanism of their action is still debated. Small-angle scattering experiments performed by Hylleraas associate Prof. Reidar Lund, using partially deuterated phospholipid bilayers, revealed how these peptides are able to scramble the lipids that compose biological membranes [6]. Particularly interesting it the evidence that such peptides catalyse lipid flip-flip between the two membrane leaflets. This occurs even if peptides bind and reside peripherally on the external leaflet only. Further measurements on the kinetics of the flip-flop reaction also revealed that the catalytic action of the peptide exerts on the entropic component of the activation energy, while it only marginally affects the enthalpic contribution.

To elucidate the molecular means of such action, Carrer performed all-atom simulations of a model lipid bilayer with a 1:3 DMPC:DMPG lipid composition, similar to the one used in Lund's experiment [7]. Furthermore, one indolicidin antimicrobial peptide was bound to one side of the membrane. The equilibrated models were validated against SAXS experiment, thanks to the contribution of Dr. Henrique M. Cezar. Umbrella sampling molecular dynamics simulations forcing one lipid to exchange between the two leaflets revealed that, in the presence of indolicidin, the free energy for the flipping event is reduced by ~16%, in agreement with the experiment.

All atom simulations revealed how binding of indolicidin onto the surface of the membrane perturbs the packing of the lipid heads, facilitating their penetration inside the membrane. The peptide chaperones the flipping lipid by peristaltic fluctuations, until the lipid head reaches the midpoint of the bilayer. The more hydrophilic nature of the peptide also

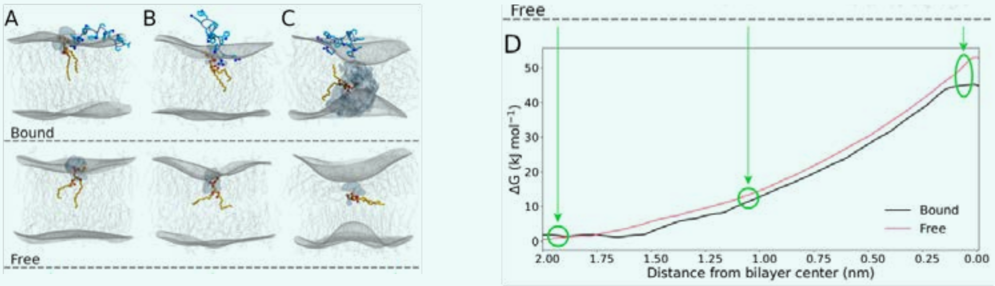

Figure 2: Lipid flipping in a 3:1 DMPG:DMPC bilayer. In A, B, C: selected conformations of the simulated systems depict the lipid flipping across the two leaflets. At the top: membrane containing indolicidin, and at the bottom: in the absence of the peptide. Blue surfaces delineate water penetrating the bilayer with the flipping lipid. Gray surfaces depict the bilayer leaflet surfaces. D reports the Free energy profiles obtained from umbrella sampling for the flip-flop process in the presence (black) and absence (red) of indolicidin. *Adapted from [7] under CC-BY 4.0 license.*

facilitates residual water molecules to accompany the flipping lipid, reducing the overall free energy barrier (Figure 2). Computational models also reveal that, at the transition state, the lipid head detaches from the peptide, which migrates back to its initial position. The fact that the nature of the molecular interactions at the transition state is very similar between the catalysed and uncatalysed events explains why the activation enthalpy is unaffected. On the contrary, binding to the peptide in the ordered leaflet drastically reduces the mobility of the flipping lipid; this produces an entropy loss in the reactant state that results into a lower entropic penalty while reaching the transition state. Understanding of the molecular mechanisms of action of these compounds paves the way for their rationalization and optimization for future medical use.

Machine learning for multiscale modelling

In 2023, activities in RT2 have expanded to incorporate data-driven approaches into multiscaling, in synergy with activities of RT7. As already introduced in the 2022 yearly report, Henrique M. Cezar has been working on the combination of HhPF with Bayesian inference approaches to bias molecular dynamics simulation with respect to predicting small-angle scattering. In this way, the experimental signal is used to drive the computational model to optimal agreement. 2023 saw the appearance of a first publication showcasing the implementation of small-angle neutron scattering (SANS) within a metainference approach 2023 (Figure 3) [8]. Research work as so far focussed on the characterization of the self-assembly of non-conventional surfactants, and lipopeptides, also in

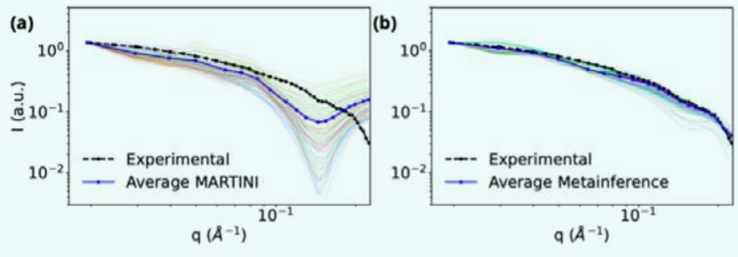

Figure 3: Average SANS intensities computed for MD trajectories using: a) the modified MARTINI 2.0 force field, and b) biasing MD simulations using metainference. Colored thin lines represent the scattering predicted from individual snapshots. *Adapted from [8] under CC-BY 4.0 license.*

collaboration with the group of. Prof. Mauricio Coutinho-Neto (Universidade Federal do ABC, Brazil), for which simplistic core-shell models are not sufficient. Major results from these projects are expected in 2024.

Thanks to the hiring of Sigbjørn L. Bore in late 2022, RT2/6 has started a number of projects dedicated to accurate sampling of molecular systems at the all-atom level with neural network potentials derived from accurate reference ab initio molecular dynamics data. This procedure is now being applied to unravel the nature of organic solution of main group metal salts and related organometallic compounds, in synergy with RT5, as well as the phase diagram of ice, and transport properties of hydrogen in porous materials.

Application studies

Finally, 2023 saw the completion of a series of works in collaboration with experimental colleagues in campus (Hylleraas affiliate Profs. Ute Krengel, Reidar Lund, and Dr. Nikolina Sekulic), dedicated to chorismite mutase [9], photoactive surfactants [10], and Aurora B kinase [11]. These works have been featured in past yearly reports; therefore, they are not described here in detail.

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 etc. A major question is how proteins and enzymes of such organisms have evolved to cope with these environments. The most well studied external factor with respect to protein evolution is temperature, and our focus has 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. The understanding of 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 seems to be a shift in the activation enthalpy-entropy balance, with a reduction in activation enthalpy followed by a more negative entropy. This is also means that the activation free energy is highly similar when comparing cold- and warm-active enzyme orthologs. In order to obtain a deeper understanding of low temperature adaptation we have investigated several different cold- and warm-

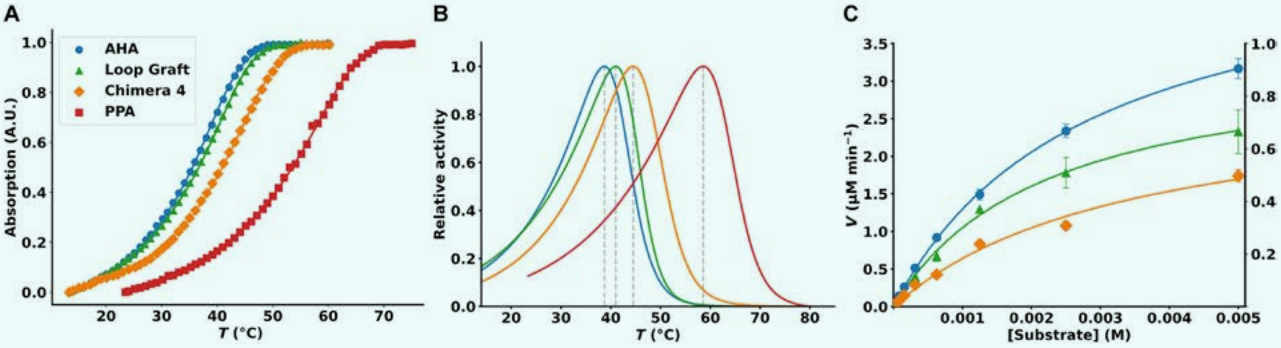

Figure 4: Kinetics of the designed enzyme variants. **(A)** Results from temperature ramping experiments with the 2-chloro-4-nitrophenyl-α-d-maltotrioside substrate for the four enzyme variants AHA (blue), Loop Graft (green), Chimera 4 (orange), and PPA (red). **(B)** Relative k_{cat} for the four variants as a function of temperature obtained from the derivatives of the fitted product formation curves in (A). **(C)** Steady-state kinetics measured at 25°C for AHA (blue, left y axis), the Loop Graft variant (green, left y axis), and Chimera 4 (orange, right y axis). Error bars ± 1σ (n = 3). A.U., arbitrary units. *Reproduced from [14], under CC-BY 4.0 license.*

active enzyme orthologs using a combination of computer simulations and experimental characterizations.

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 [13], allowing us to extract activation enthalpies and entropies from an Arrhenius plot of reaction free energies of the reaction catalyzed by a monofunctional *Bacillus subtilis* chorismate mutase and the promiscuous enzyme isochorismate pyruvate lyase of *Pseudomonas aeruginosa*. Compared to the uncatalyzed reaction, both enzyme-catalyzed reactions exhibit a substantial reduction in activation enthalpy. The effect on activation entropy is relatively minor, demonstrating that enzyme-catalyzed chorismate mutase reactions are enthalpically driven. Furthermore, the monofunctional chorismate mutase from *B. subtilis* catalyzes this reaction more efficient 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 the difference in efficiencies between the two enzymes. This study has been carried out by Ryan Wilkins [13].

In addition to the higher catalytic activity at low temperatures cold-adapted enzymes also have their temperature optimum down-shifted when compared to mesophilic orthologs. Interestingly, the temperature optimum does not coincide with the onset of protein melting, but rather reflects some other type of inactivation. The inactivation for the psychrophilic α-amylase from an Antarctic bacterium is thought to originate from a specific enzyme-substrate interaction that breaks around room temperature. We investigated this through computational redesign of the α-amylase aiming at shifting its temperature optimum upward [14]. Our computer simulations of the catalytic reaction at different temperatures predicted several mutations with the potential to stabilize the enzyme-substrate interaction. These were subsequently verified by kinetic experiments and crystal structures of the redesigned α-amylase, showing that the temperature optimum is indeed markedly shifted upward and that the critical surface loop controlling the temperature dependence approaches the target conformation observed in a mesophilic ortholog (Figure 4).

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RT3/4

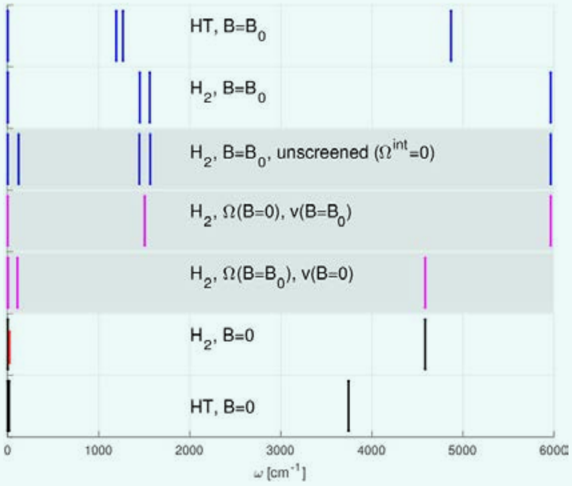
Matter-Field Interactions

Principal investigators:
Michal Repisky and Erik Tellgren

In mid-2023, RT3 and RT4 fused into a single research theme RT3/4. Since then, most of the previous lines of research now continue under the unified heading of *matter-field interactions.*

Molecules in Strong Magnetic Field

The study of the Berry curvature in magnetic fields continued to be a prioritized activity during the year. When viewed from the perspective of the nuclei, the electrons in a molecule give rise to a potential energy surface, described by a non-local scalar potential. In a magnetic field, the electrons also give rise to a non-local quantity that resembles a magnetic vector potential, and its curvature---the Berry curvature---give rise to exotic dynamical effects on the nuclei. The Berry curvature may be thought of as a non-local magnetic field that gives rise to velocity-dependent forces that screen the external Lorentz force. The Lorentz force, due to a magnetic field $\vec{B}$, on a bare nucleus with velocity $\vec{V}$ and charge Q is given by $\vec{F}=Q\vec{V}\times\vec{B}$. Considering that the electrons screen the nuclei from the external magnetic field results in Berry forces that in molecules are generally very complicated. Nonetheless, for neutral atoms, the Berry force exactly cancel out the Lorentz force. From this fact, one may realize that the Berry forces contain information about the partial electric charges, and therefore the electron population, on different nuclei within a molecule. This was exploited in the paper *Berry Population Analysis: Atomic charges from the Berry curvature in a magnetic field* [1], which presented a novel quantum-chemical population analysis. Moreover, both the Lorentz force and the Berry force are velocity-dependent forces, which is untypical in ab initio dynamics and the dominant computational methods employ simplifications that implicit assume the absence of such forces. For this reason, we have developed methods for computing nuclear trajectories in the presence of velocity-dependent forces and the paper *Propagators for molecular dynamics in a magnetic field* [2] contains the latest refinement of these methods. Another area that implicitly assumes the absence of magnetic or velocity-dependent forces is the calculation of molecular rotational and vibrational spectra. Hence, to enable the calculation such spectra in magnetic fields, we have, in the work *Molecular vibrations in the presence of velocity-dependent forces* [3], generalized the standard theory of small vibrations. What is normally a straightforward eigenvalue problem involving the geometric Hessian matrix then becomes what in mathematics is known as a Quadratic Eigenvalue Problem and the magnetic forces result in rotational, vibrational, and even translational motion becoming coupled (Figure 1). This coupling is a very exotic



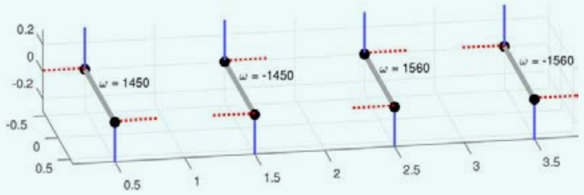


Figure 1: Left, calculated vibrational frequencies for a hydrogen molecule in a magnetic field of B_0=235 kT for various cases illustrating the effect of magnetic forces as well as isotope effects. Above, the two real and imaginary parts of the modes around 1500 cm^{-1} visualized. In the absence of velocity-dependent forces, these would be librations changing the angle to the magnetic field, and in the presence, they are split into clockwise and counter-clockwise precessions. *Reproduced from [3] with permission from the American Institute of Physics*.

phenomenon that is seen clearly in calculated dynamical trajectories, although it turned out to be surprisingly small in vibrations of small molecules.

While nuclei are almost always treated as classical particles they are in principle quantum mechanical. A compromise between accuracy and computational tractability is the nuclear-electronic orbital method (NEO), which treats hydrogen nuclei quantum mechanically and other nuclei as classically. In the work *Time-dependent nuclear-electronic orbital Hartree–Fock in a strong uniform magnetic field* [4], the NEO method was extended to strong magnetic fields. This enabled us to study the magnetic field dependence of quantum mechanical effects. Moreover, it also enabled comparison of semiclassical calculations of molecular (ro-)vibrations with the quantum treatment. In general, the agreement is good and we could verify numerically that the NEO method correctly captures the screening effect that at the semiclassical level is described in terms of the Berry curvature.

An accurate treatment of strong magnetic field effects is important in astrophysics, where spectra from magnetic white dwarf stars are modified by magnetic fields. This effect enables astronomers to identify the composition of the white dwarf atmospheres as well as the strength of the magnetic field they generate. In *A DZ white dwarf with a 30 MG magnetic field* [5], Hylleraas affiliate Stopkowicz and coworkers employed finite magnetic field calculations at the equation-of-motion coupled-cluster level to calculate atomic spectra of atoms in a magnetic field. Comparing observed lines from a white dwarf designated SDSS J114333.48+661531.83 to calculated lines for Na, Mg, and Ca, it could be estimated that this white dwarf has a magnetic field of around 30 MG.

Another important research line is density-functional theory in the presence of magnetic fields. In the two-part article *The Structure of the Density-Potential Mapping* [6,7], the status of the Hohenberg–Kohn theorem was reviewed from a mathematical perspective for various flavors of density-functional theory (DFT). The Hohenberg–Kohn theorem historically served as the mathematical foundation of DFT and remains an important aspect today. The first part was devoted to standard DFT, which underlies most of all applied computational studies. The second part was devoted to flavors that include magnetic field effects and summarizes some lesser-known results.

Laser-driven Many-electron Dynamics

The work *Magnetic optical rotation from real-time simulations in finite magnetic fields* [8] combined several research lines. By directly explicitly simulating magnetic field effects on spectra using real-time propagation, the study avoid-

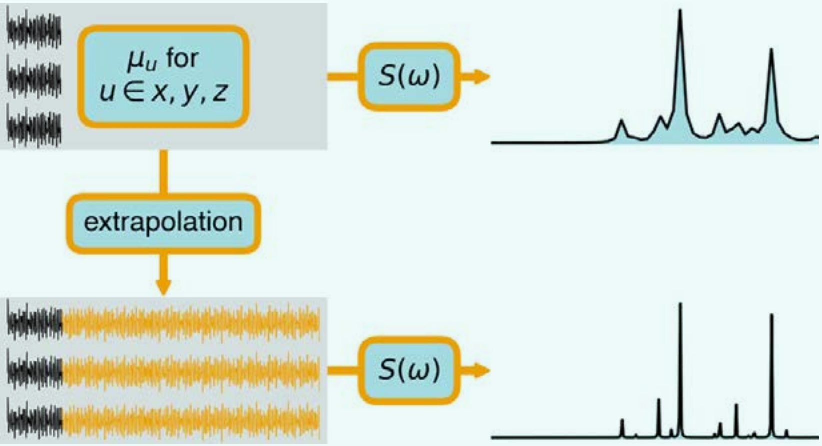


Figure 2: Electronic absorption spectra (left) obtained by the Fourier transformation of induced electric-dipole moments (left) recorded in the course of real-time simulations. A developed fitting method results in arbitrary spectral resolution through extrapolation from shorter dipole moment trajectories. *Reproduced from [9] under CC-BY 4.0 license.*

ed the standard implicit treatment using perturbation theory. Optical rotation occurs when the polarization direction of light is rotated as it passes through a medium. When induced by a magnetic field, this is known as magnetic optical rotation or Faraday effect. A time-dependent coupled cluster-level approach was developed and used to verify what is normally just assumed, namely that the optical rotation to a good approximation is linear in experimentally accessible magnetic field strengths. Moreover, a pragmatic approach to time-dependent current-density-functional theory, which incorporates magnetic-field effects via the kinetic energy density, could also be tested against the more reliable coupled-cluster results.

Obtaining electronic absorption spectra from real-time simulations involves subjecting the molecular systems to a short electric-field pulse, and Fourtier transforming the induced electric-dipole moments recorded in the course of simulations. A significant computational challenge of the method lies in the long simulation times required to achieve the necessary resolution in absorption spectra. Eirill Hauge, in collaboration with Pedersen, Repisky, Dr. Håkon Emil Kristiansen, Dr. Marius Kadek, and Dr. Lukas Konecny, have addressed this challenge by introducing a novel function fitting method based on shorter trajectories of the dipole moment, achieving arbitrary spectral resolution through extrapolation (see Fig. 2) [9]. Numerical testing demonstrates convergence with as few as 100 atomic units of dipole trajectories for most studied systems. To ensure the reliability of the approximated spectrum, the authors have also introduced an error estimate for the fit.

Real-time electron dynamic simulations are pivotal in the field of attosecond pump–probe transient absorption spectroscopy (TAS), wherein an ultrashort laser pulse is divided into two portions; a stronger beam (pump) is used to excite the sample, generating a non-equilibrium state, and a weaker beam (probe) is used to monitor the pump-induced changes in the optical properties of the sample (Fig. 3). Theoretical modeling of TAS for heavy elements poses a challenge due to significant influences from scalar and spin-orbit relativistic effects. To tackle this challenge, in 2023, post-doc Dr. Torsha Moitra, in collaboration with Repisky, Dr. Lukas Konecny, Dr. Marius Kadek, and Prof. Angel Rubio (MPI Hamburg), propose and integrate into the ReSpect program a methodology for simulating TAS within the relativistic real-time time-dependent density functional theory (RT-TDDFT) framework, covering both

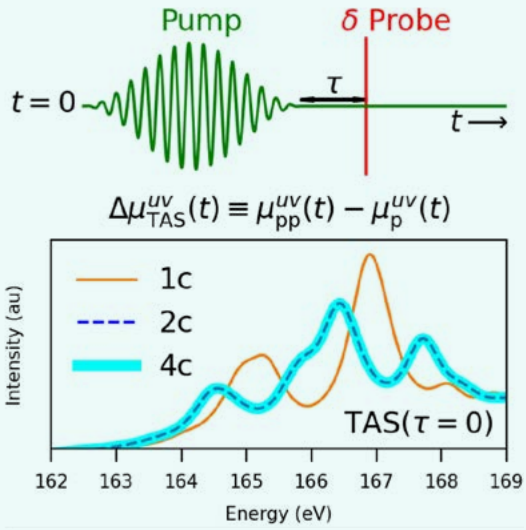


Figure 3: Pump–probe transient absorption spectra of thiophene near the sulphur L2,3 absorption edge (bottom) as obtained with various relativistic RT-TDDFT Hamiltonian models. A sequence of applied ultrashort laser pulses consists of two portions (top); a stronger beam (pump) is used to excite the sample, generating a non-equilibrium state, and a weaker beam (probe) is used to monitor the pump-induced changes in the optical properties of the sample. *Reproduced from [10] under CC-BY 4.0 license.*

valence and core energy regimes [10]. In addition to the 4c approach, the team introduced the atomic mean-field exact two-component (amfX2C) Hamiltonian for TAS, considering one- and two-electron picture-change corrections within RT-TDDFT. An illustrative example in Fig. 3 depicts the TAS spectra of thiophene near the sulphur $L_{2,3}$ absorption edge, emphasizing the necessity of incorporating relativistic effects for core spectroscopies and showcasing the notable agreement between amfX2C and 4c spectra. Furthermore, non-equilibrium response theory was extended to the relativistic regime, providing theoretical insights into the distinctive features of pump-probe TAS.

Spectroscopic Processes in Molecules and Solids

X-ray absorption spectroscopy (XAS) has become increasingly popular due to its ability to investigate matter with high spatial and elemental sensitivity. However, the theoretical modeling of XAS poses challenges, particularly near L and M absorption edges, where spectra exhibit fine structure due to spin–orbit (SO) relativistic effects. To address this challenge, post-doc Dr. Lukas Konecny collaborated with Repisky, Dr. Stanislav Komorovsky, Dr. Jan Vicha, and Prof. Kenneth Ruud on the extension of the 2c Hamiltonian models based on the (extended) atomic mean-field exact two-component approach, (e) amfX2C, for XAS calculations using linear-response and damped-response time-dependent density functional theory [11]. Unlike the commonly used one-electron X2C (1eX2C) Hamiltonian, both amfX2C and eamfX2C account for the two-electron and exchange–correlation picture-change (PC) effects arising from the X2C transformation. This work also showed that the absence of PC corrections in the 1eX2C approximation leads to a significant overestimation of SO splittings. In contrast, (e) amfX2C Hamiltonians provided results in excellent agreement with the 4c references, encompassing spectral features such as shape, position, and fine structure splitting, as illustrated in Fig. 4.

Two-dimensional (2D) materials exhibit a wide range of interesting properties, many of which find their origin in the effects of relativistic spin-orbit coupling (SOC). For a comprehensive understanding and accurate prediction of the properties of 2D materials with heavy elements, such as transition-metal dichalcogenides (TMDs), it is imperative to consider relativistic effects in first-principles calculations. In 2023, post-doc Dr. Marius Kadek and PhD Marc Joosten, in collaboration with Repisky, Prof. Kenneth Ruud, and the group of Prof. Arun Bansil from Northeastern University in Boston), introduced an all-electron method based on

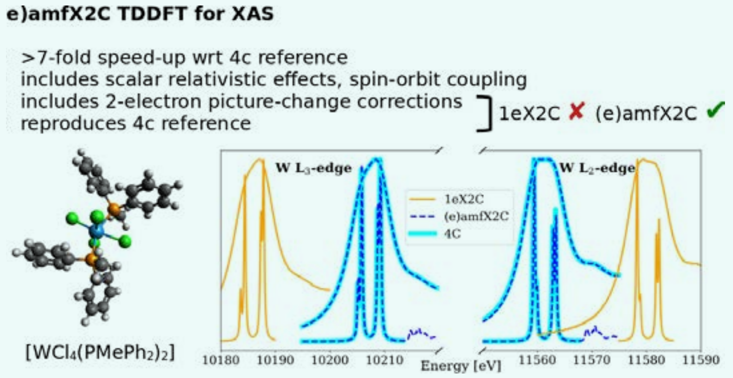


Figure 4: X-ray absorption spectra of [WCl4(PMePh2)2] near the tungsten L2,3 absorption edge as obtained with various relativistic RT-TDDFT Hamiltonian models. *Reproduced from [11] under CC-BY 4.0 license.*

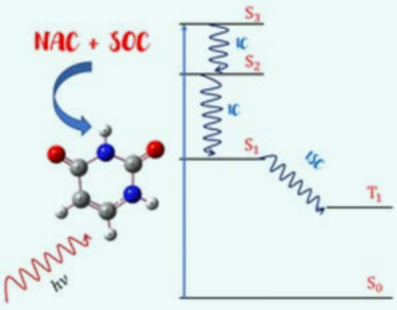


Figure 5: Schematic non-radiative deactivation processes (internal conversion (IC) and intersystem crossing (ISC)) for azulene molecule. *Reproduced from [13] with permission from Royal Society of Chemistry.*

the four-component Dirac Hamiltonian and Gaussian-type orbitals (GTOs) that overcomes complications arising from linear dependencies and ill-conditioned matrices associated with diffuse functions in the GTO basis [12]. As a demonstration, the authors study the relativistic band structure of the 2D TMDs, along with a discussion of their SOC-driven properties such as Rashba splitting and Z2 topological invariants. Since the presented methodology neglects the pseudopotentials altogether, it provides access to all electronic states within uniform framework and paves the way for direct investigations of material properties driven by inner shells where relativistic effects are most pronounced.

Another molecular properties difficult to evaluate accurately are the rates of non-radiative deactivation processes, namely internal conversion (IC) and intersystem crossing (ISC). In 2023, post-doc Dr. Torsha Moitra and Prof. Kenneth Ruud, in colloboration with the group of Prof. Swapan Chakrabarti from Calcutta, presented a composite framework for calculating the rates of non-radiative deactivation processes, accounting explicitly for the non-adiabatic coupling (NAC) and spin–orbit coupling (SOC) constants, respectively [13]. The applicability of the framework was validated on the IC rate of azulene (Fig. 5), obtaining comparable rates to experimental and previous theoretical results. In addition, the authors investigated the photophysics associated with the complex photodynamics of the uracil molecule and obtained rates that corroborate with experimental observations. Detailed analyses using Duschinsky rotation matrices, displacement vectors and NAC matrix elements were presented to interpret their findings. The presented stationary-state approach uses a time-dependent generating function based on Fermi's golden rule, suitability of which was explained in terms of single-mode potential energy surfaces.

A category of DNA folds/structures collectively referred to as G-quadruplexes (G4) frequently emerges in regions of genomes rich in guanine. G4-DNA is believed to play a crucial functional role in regulating gene transcription, making it a target for drug development. However, the intricate molecular interactions responsible for the stacking of guanine-tetrads remain poorly understood, hindering a systematic approach to targeting G4 sequences with drugs. In 2023, researcher Dr. Magnus Ringholm collaborated with the goup of Prof. David R. Klug from Imperial College London on the exploration of the G4-DNA interactions as formed in the Myc2345 nucleotide sequence, utilizing electron-vibration-vibration two-dimensional infrared (EVV 2DIR) spectroscopy [14]. In addition, the authors monitored structural changes associated with G4-folding in response to varying K^+-ion concentrations. Combining experimental 2DIR data with DFT calculations suggests that 102 coupling peaks may exist throughout the folding process, and the guanine-quartets may exist before the addition of K^+-ions, but they remain unstacked until the introduction of K^+-ions.

Another class of biologically important compounds are cyclic peptides. They exhibit a broad spectrum of biological activities, notably serving as antibacterial agents. However, due to their substantial size and flexibility along with multiple chiral centers, the determination of the stereochemistry of cyclic peptides remains challenging task. In 2023, PhD Karolina Di Remigio Eikås and post-doc Dr. Monika Krupová, in collaboration with Dr. Maarten T. P. Beerepoot, Tone Kristoffersen and Prof. Kenneth Ruud, investigated the extent to which the absolute configuration (AC) of cyclic peptides can be definitively determined using vibrational circular dichroism (VCD) [15]. The publication presents both experimental and computed VCD spectra for a series of eight tetrapeptides and hexapeptides, featuring two, three, and four chiral centers. The developed computational protocol for a conformational search based on meta-dynamics reveals that the AC of investigated peptides with two chiral centers can indeed be unambiguously assigned with VCD. However, this proves challenging for cyclic peptides with three chiral centers and impossible for those with four chiral centers. In these cases, at best, one can limit the number of possible stereoisomers. The work also shows the importance of computing the spectra of all stereoisomers before attempting to match them to an experimental spectrum, to prevent inadvertent and erroneous matches.

Personnel and Funding

Research in RT3/4 in 2023 has been supported by the RCN (FRIPRO) project *Magnetic Chemistry* led by Prof. Helgaker; RCN (FRIPRO) project *EPR and paramagnetic NMR of solids from relativistic two- and four-component density-functional theory* led by Kenneth Ruud and Michal Repisky; RCN (Mobility) grant *First-principles modeling of magnetic topological materials from relativistic hybrid density functional theory* led by Marius Kadek; RCN (Mobility) grant *Linear and Non-linear Response Properties and Spectroscopy of Solids from Relativistic TDDFT* led by Lukas Konecny. Torsha Moitra was further supported by an ancillary travel grant for post-doctoral fellows in conjunction with the RCN (FRIPRO).

In the Oslo node, postdocs Tanner Culpitt and Laurens Peters left the Oslo node in June 2023 and December 2023, respectively, while Prof. Trygve Helgaker retired. Currently E. Tellgren (researcher) is primarily active in RT3/4 and hiring of postdoc is planned, while Hylleraas members Pedersen (professor) and Laestadius (researcher) contribute partially to RT3/4.

Tromsø RT3/4 research team in 2023 consists of Ruud (professor), Repisky (researcher), Ringholm (researcher), Kadek (postdoc), Konecny (postdoc), Krupova (postdoc), Joosten (PhD), Le (PhD), Moitra (postdoc) and Rasmus Vikhamar-Sandberg (PhD).

In addition, Hylleraas affiliates Assoc. Prof. Andy Teale, University of Nottingham, and Assoc. Prof. Stella Stopkowicz, Sarland University, have contributed to research in RT3/4.

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RT5

Chemical Transformations

Principal investigators:
Ainara Nova and Abril Castro

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RT5 applies computational methods to understand chemical reactions and develop more sustainable processes in collaboration with experimental groups. Our approach combines the simulation of spectra to support the experimental characterization of reactive species and free energy calculations to construct microkinetic models, allowing us to compare computed and experimental data directly. In 2023, additional effort has been put in the characterization of organometallic compounds involving main-group elements.

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CO_2 reduction

Understanding the reaction pathways associated with the hydrogenation of CO_2 to methanol is one of the main objectives of the FRIPRO CO2pCat project led by Ainara Nova and co-funded by NordForsk via the NordCO2 consortium. One of the most challenging systems approached in this project involves a platinum nanoparticle encapsulated in a UiO-67 metal-organic framework (PtNP@MOF), which catalyzes the hydrogenation of CO_2 to methanol, with CO and methane as by-products. This work was done in collaboration with Prof. Unni Olsbye from the catalysis section at the UiO and Prof. Egill Skúlason from the University of Iceland. The two initial studies published in 2020 provided insight into the initial and final phases of the reaction, the formation of the Pt-NP [1] and the competitive absorption of water and methanol products [2]. The most extensive work on the reaction mechanism for hydrogenating CO_2 to methanol, CO and methane was published in 2023 [3].

The first step in this study was building the models to represent the active sites. Five different models were constructed: (a) a defective zirconium cluster isolated, (b) a 55-atom cuboctahedral nanoparticle of platinum to simulate the edges of the nanoparticle, (c) a platinum (111) surface to simulate the flat surfaces of large nanoparticles, and (d) a defective zirconium cluster on top of a platinum (111) surface and (c) on top of an edge of a platinum surface to simulate two different node-Pt interfaces. With all of them, free energy profiles leading to CH_3OH, CH_4, and CO were computed using PBE functional with the DZVP-MOLOPT-SR-GTH basis set in CP2K. Only combining the results using the interface models and the Pt(111) surface was possible to explain the experimental observations. The formation of methanol was preferred on the interface between the node and the Pt-edge (Figure 1a). The rate-limiting step for this reaction is the hydrogenation of formate, which is consistent with the experimental detection of formate by IR and an inverse KIE. Instead, the formation of CO and methane was preferred on the interface between the node and the Pt surface (Figure 1b and

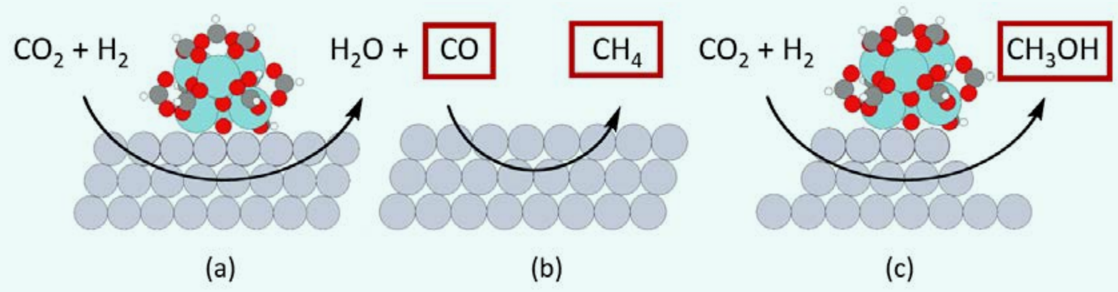

Figure 1: Computational models used on the study of the reaction mechanism for the CO2 hydrogenation to methanol with PtNPs@UiO MOF: (a) a defective zirconium cluster on top of an edge of a platinum surface, (b) a defective zirconium cluster on top of a platinum (111) surface and (c) a platinum (111) surface. *Illustration: Harsha Pulumati and Ainara Nova*

c). This pathway initially forms CO, which migrates from the interface to the Pt surface for its final hydrogenation to methane or desorption at high CO coverage. To support these results, the hydrogenation of CO_2 was tested experimentally on calcinated MOFs containing small and large NPs. The results showed that only the system with large NPs yields methane, consistent with the need for a flat surface to generate methane. This work was performed by Dr. Harsha Pulumatti, who defended his PhD in early 2024. Pulumati was hired by the NordCO2 project and his thesis co-supervised by Nova.

In 2023, NordForsk invited all project leaders to write a Policy article with recommendations on accomplishing a green, competitive, and socially sustainable Nordic Region by 2030. The article submitted by Nova and other NordCO2 members was selected for publication in DIVA, the common publication platform of the Nordic Council of Ministers [4]. In this work, entitled "Recommendations for future CO_2 management in the Nordic Countries", it was suggested to use waste and biomass as materials for capture and to increase the number of intermediate species directly formed from CO_2. This article promoted an interview with Nova in the NordForsk online news [5].

Another project running on RT5 in the field of CO_2 conversion is the EU-ITN CO2PERATE network, which involves student exchanges between European Universities and Industries. The PhD student Mahika Luthra visited Haldor-Topsøe (Denmark) in 2020 to investigate the structure of Sn-Beta Zeotypes [6]. Periodic density functional theory calculations using GPAW software were used to examine the geometry changes of the unit cell of Polymorph A upon mono substitution of tin into the zeolite framework. By substituting silicon with a tin atom at each T-site, nine models containing one tin atom per unit cell in a closed configuration (Si-O-Si bridge) were obtained. In addition, open structures with Sn-OH and Si-OH moieties were also considered. The optimized structures indicate that the unit cell contracts along the *a* axes and expands along the *c* axes, resulting in an overall increase in the *c*/*a* ratio for all T-sites. Interestingly, this change is more pronounced for the open tin site than the closed one. These results are consistent with the experimental parameters obtained for the hydrothermal samples, which show an increasing trend in the *c*/*a* ratio upon tin insertion and contain a mixture of closed and open sites.

Hydride migration in alkene isomerization reactions

A reaction of industrial interest is the selective isomerization of double bonds. In particular, the formation of Z-alkenes is challenged by unfavourable thermo-

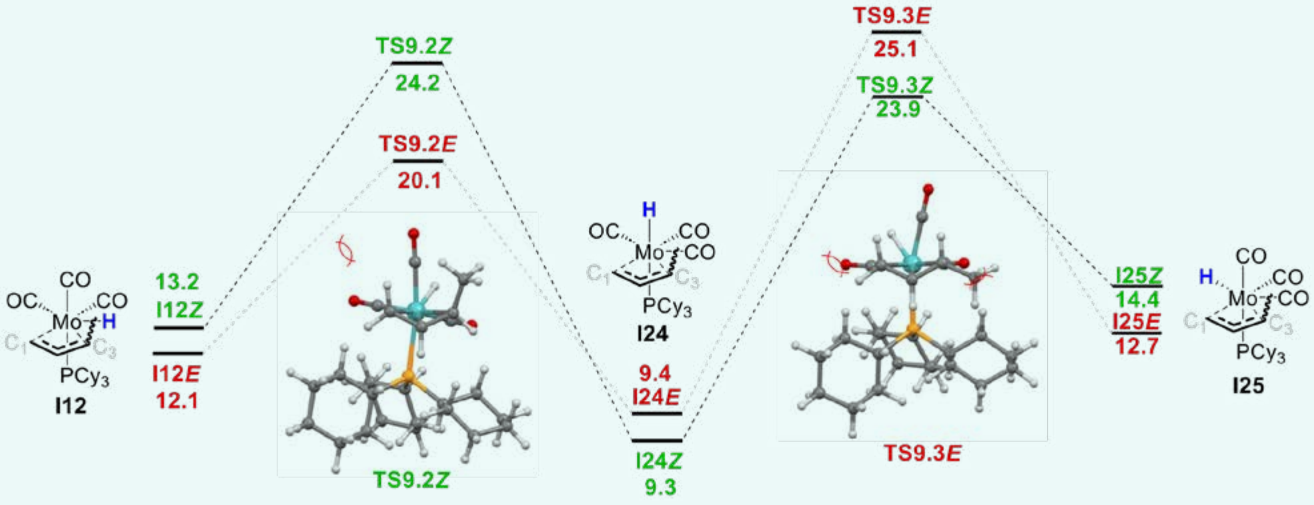

Figure 2: Transition states along the E and Z-leading pathways from intermediates I12E and I12Z. Highest energy transition state geometries are shown with steric interactions noted. *Illustration: Sarah E. Jenny and Ainara Nova.*

dynamics. In a collaboration with the group of Prof. Graham E. Dobereiner, from Temple University, in Philadelphia (US), the mechanism for the preferred formation of Z-2-alkenes by a *cis*-[Mo-$(CO)_4(PCy_3)$(piperidine)] was studied [7]. The PhD Sarah E. Jenny at Temple University performed the experimental and computational work. The latter was supervised by Nova. In a first stage, experiments and DFT calculated thermodynamics were used to determine the nature of the active catalyst and exclude an alkyl isomerization mechanism. Next, the energy profiles for forming the E and Z 2-butene considering different isomers of $[Mo(CO)_3PCy_3$(1-butene)] were computed. Even though the reaction follows the well-established allyl mechanism, including a C-H bond oxidative addition to Mo(0) followed by a reductive elimination, the mechanism for the hydride rearrangement required to obtain the final product was unknown. After considering different mechanisms such as an allyl-rotation or an η_3 to η1 allyl slippage, a hydride migration assisted by the CO ligands was the most energetically preferred. NBO analysis on intermediate **I12Z** (Figure 2) showed a H-C Wiberg index between the hydride and a CO ligand of 0.35 suggesting an attractive interaction. In addition, the acceptor CO showed the expected slight lengthening, with a C-O Wiberg index of 1.95, relative to the 2.06 and 2.08 for the other COs. This CO assisted hydride rearrangement involved the highest energy transition states of the overall reactions and accounts for the product selectivity. The experimental excess of Z isomer could be explained by a preference for **TS9.2*Z*** compared to **TS9.3*E*** by 0.9 kcal/mol. The *fac* CO orientation forces the allyl to be cis to PCy_3. Steric pressure between phosphine and allyl substituents thus plays a role in favoring the *Z* conformation.

Noble metal catalysis

After a long-term collaboration of Nova with Prof. Mats Tilset, the results of several studies on the reactivity of cyclometalated (N,C)Au(III) complexes were reviewed in 2023.[8] This work highlights the impact of trans effects in guiding the synthesis, structure and reactivity in Au(III) complexes. In these studies, computational methods were used to analyze the electronic structure of Au(III)-complexes and provide information on reaction thermodynamics and kinetics. Most of the experimental work was performed by M. S. M. Holmsen during her PhD in the group of Tilset and co-supervised by Nova. After being two years in France, with a postdoctoral position funded by the Research Council of Norway, Holmsen joined the Hylleraas Centre in September to use computational methods to study new Au(III) reactions.

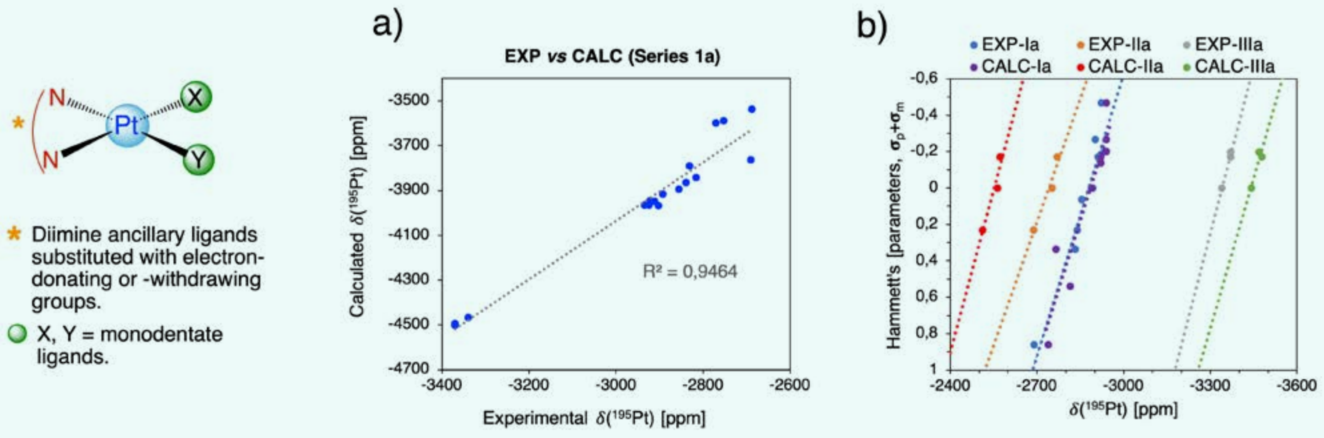

Figure 3: Selected platinum(II) complexes with diamine ancillary ligands. a) Comparison between calculated and experimental 195Pt NMR chemical shifts for a series of selected complexes. b) Calculated 195Pt NMR chemical shifts vs empirical Hammett's parameters. *Illustration: Alberto Guerrero and Abril Castro.*

The study of a Pd(0)- and a Cu(I)-catalyzed N-arylation reactions by using DFT methods were published in 2023 [9,10]. The work involving copper was performed by Linn Neerbye Berntsen, who defended her thesis in June under the supervision of Nova. The N-arylation of hydantoin (2,4-imidazolidinedione) is a reaction of interest as it gives direct access to N-aryl hydantoins, considered valuable scaffolds in medicinal chemistry. The Cu(I)-catalyzed N3-arylation of unsubstituted hydantoin with unsymmetrical aryl(TMP)iodonium salts (TMP=2,4,6-trimethoxyphenyl) was developed by Berntsen under the supervision of Alexander Sandtorv, Assoc. Prof at the Dept. of Chemistry until 2021 [11]. In the computational study of this reaction [10], two different mechanisms were explored by DFT methods. In pathway I, the imide NH-group is deprotonated by a Cu(I)-species before the oxidative addition of the diaryliodonium salt. In pathway II, the oxidative addition of the diaryliodonium salt is the first step followed by the NH-deprotonation. In both cases, the final product was formed by reductive elimination of a Cu(III)-species. The computed energies showed that Pathway I was the most preferred one, in contrast to the proposed mechanism for C-H arylation reactions with diaryliodonium salts. The deprotonation could take place by triethylamine or the Cu-catalyst. In both cases, the reaction leads to a Cu-amido intermediate, which reacts with the diaryliodonium salt. The oxidative step and reductive elimination of the final product involves four transition states of similar energies. In addition to the C-I bond cleavage and decoordination of TMP, a ligand rearrangement is required before the reductive elimination takes places. The calculation of these energy barriers with aryl groups with different substituents showed that the highest transition step depends on the substituent. This result was consistent with the yields obtained experimentally for these reactions.

Interpretation of NMR parameters

The RCN-YRT project SpecTraM *(Accurate Prediction and Interpretation of NMR Spectra in Transition-Metal complexes)*, led by Abril Castro, has in 2023 focused on using the power of NMR enhanced by DFT calculations to understand the electronic properties and reactivity of transition-metal complexes with relevance to catalysis.

Because of its high sensitivity to the local environment of nuclei, Nuclear Magnetic Resonance (NMR) spectroscopy provides valuable insights into the electronic structure of molecules. The chemical shift is, in fact, directly linked to the frontier molecular orbitals, frequently implicated in reactivity. Consequently, quantum chemical calculations can be conveniently employed to interpret NMR signatures by consid-

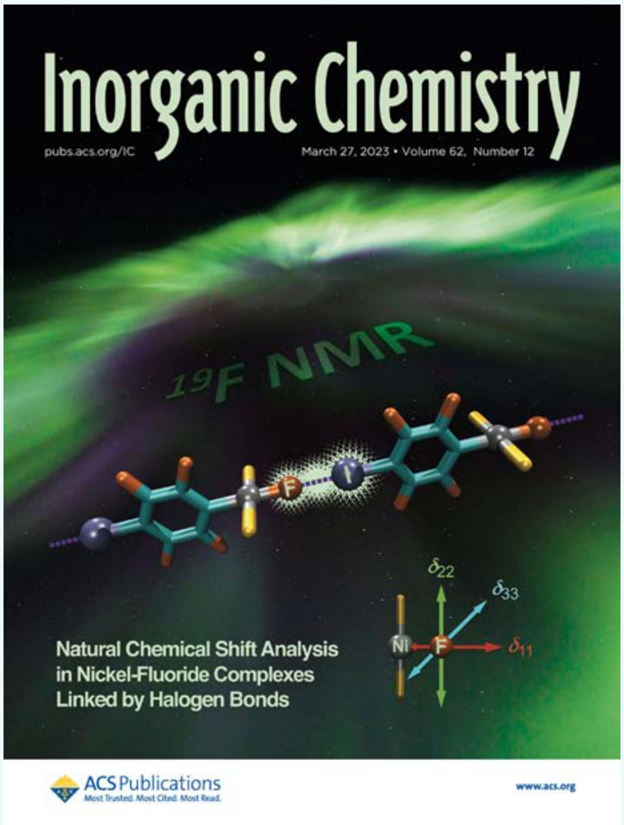

Figure 4: Cover picture in the Inorganic Chemistry Journal, highlighting the open-access article "Solid-state 19F NMR chemical shift in square-planar nickel-fluoride complexes linked by halogen bonds" by Abril Castro et al. (Inorg. Chem. 2023, 62, 12, 4835.). Designed by Castro, the cover artwork illustrates a nickel-fluoride complex engaged in halogen bonding and exposed to a magnetic field, symbolized by the Northern Lights. *Reproduced with permission from American Chemical Society.*

ering localized molecular orbitals and their response to magnetic-field perturbations. In the SpecTraM project, we are interested in using this approach to investigate and understand electronic properties of transition-metal complexes considering both ligand's and metal's perspectives.

Castro, in collaboration with the experimental group of Prof. Mats Tilset (UiO) and her PhD student Alberto Guerrero, has investigated square-planar platinum(II) complexes containing ancillary ligands (Figure 3), which are promising candidates for C-H bonding activation under direct oxidative conditions, leading to the production of valuable hydrocarbons. Varying the substituents of the ancillary ligand directly influences the local environment of the Pt nucleus, which is evident from the broad range of ^{195}Pt NMR chemical shifts observed. Hence, this work aimed to use ^{195}Pt NMR chemical shift calculations to rationalize the observed NMR signatures and establish correlations with descriptors such as the empirical Hammett's parameters.

The study revealed that subtle changes in the geometries can significantly influence the isotropic chemical shift, demonstrating the suitability of ^{195}Pt NMR to decipher small differences in local electronic structure. At the same time, the accurate modelling of NMR chemical shifts at the platinum nucleus was challenging because of the necessity of a relativistic approach. Compared with the experimental data, differences of approximately 1000 ppm were obtained using a two-component ZORA method. Nonetheless, the chemical shift trend is correctly reproduced and attributed without ambiguity for all the series of complexes (Figure 3a). A linear correlation between the Hammett's parameters and the ^{195}Pt NMR chemical shift values was also confirmed (Figure 3b). Furthermore, we used a natural chemical shift (NCS) analysis in the paramagnetic terms of the ^{195}Pt chemical shifts. The paramagnetic terms come from the coupling via the angular momentum operator of occupied and vacant orbitals. In these square-planar complexes, the analysis has shown a large deshielding along the component oriented in the plane of the ancillary ligand, which can be attributed to the overlap of the Pt lone pairs and the antibonding σ^* orbitals. Moreover, the deshielding effect is specific to each ancillary ligand series, rationalizing correlations with the electron-donating or -withdrawing groups present on the ligands [12].

Another project in RT5 using a NCS analysis of calculated NMR chemical shifts, has focused on rationalizing the halogen-bond effect in the ^{19}F NMR signature of nickel-fluoride complexes. The analysis revealed that the deshielding of fluoride in the halogen-bonded systems is attributed to an increase in the energy gap between the occupied and vacant orbitals that are mostly responsible for the paramagnetic terms, notably along the most shielded direction [13]. This work has been highlighted on a journal cover designed by Castro (Figure 4). Furthermore, we are extending the modelling of ^{19}F NMR chemical shift to group 4 metallocene difluorides and other square-planar d^8 metal complexes. The goal is to analyze the role of the metal in the NMR signature and obtain a general interpretation of metal-fluoride complexes. The PhD candidate Alberto Guerrero is currently performing these studies under the supervision of Castro and in collaboration with the experimental group of Prof. Torsten Beweries from LIKAT and the University of Rostock in Germany.

Renewable phosphine ligands for homogeneous catalysis

Together with the experimental groups of Dr. Ashot Gevorgyan from UiT - The Arctic University of Norway and Prof. Nazari from Yale University, Castro has investigated renewable phosphines ligands for homogeneous catalysis.[14] The study was focused on the synthesis of new renewable analogues of Beller's ligand, which is among the most electron-rich phosphines commercially available. The stereoelectronic properties of the phosphine ligands were analyzed by measuring the phosphorus-selenium coupling constants and estimating the steric hindrance through computational methods. Since phosphines are flexible ligands with rich structural dynamics, DFT methods were used to perform a conformational search. Moreover, the steric effects were quantified using topographic steric maps and corresponding percent buried volumes (Figure 5). It was found that the reduced catalytic efficiency of phosphinites derived from renewable phenols can be primarily attributed to a distinctive interplay between enhanced steric hindrance and diminished σ-donor abilities.

Chemistry of main-group organometallic compounds

Part of the research has been focusing on ethereal solutions of lithium halides salts (LiX) as well as methyl lithium (MeLi), and on the interaction of LiCl with Grignard compounds. This latter study, most prominently the merit of PhD student Marinella de Giovanetti, is now completed and published in the Journal of the American Chemical Society [a]. This study showed how LiCl interacts preferentially with $MgCl_2$, which, in turn solubilizes the alkaline salt. On the other hand, this study also revealed the severe limits of ab initio molecular dynamics (AIMD) in sampling the vast combinatorial chemical space accessible to these compounds. Because the final goal of the project is to characterise the so-called turbo-Grignard (highly concentrated 1:1 mixture of LiCl and Grignard reagent), the past year de Giovanetti, together with postdocs Marco Bortoli, Sigbjørn L. Bore, and Sondre H. Hopen Eliasson initiated the calibration of Neural-Network interaction potentials trained on AIMD data for LiX, MeLi, and Grignard compounds. The goal is to obtain empirical potentials with accuracy similar to DFT but with superior size-scalability and computational performance orders of magnitude faster than conventional DFT-based methods, thus making the exploration of the complex chemistry of turbo Grignard accessible.

Synergy with alkaline salts is not the only way to improve the Grignard reaction. In fact, an experimental setup involving liquid-liquid phase co-existence between an ether and a deep eutectic solvent was shown to be effective in making the reaction cleaner, and more controllable. Despite striking experimental evidence, chemical intuition does not help understanding how such combination of non-mixing solvents may help the reactivity. In 2023, Bortoli established a multi-scale model mimicking the experimental conditions. The two liquids are represented at the empirical molecular-mechanics level of theory. Enhanced simulations using umbrella sampling indicate that the substrate prefers to partition into the ethereal phase. On the contrary, the Grignard reagent may localise at the interface between the liquids. Currently, enhanced-sampled QM/MM simulations are being used to compare the Grignard reaction at the interface with that in the bulk ether, aiming to

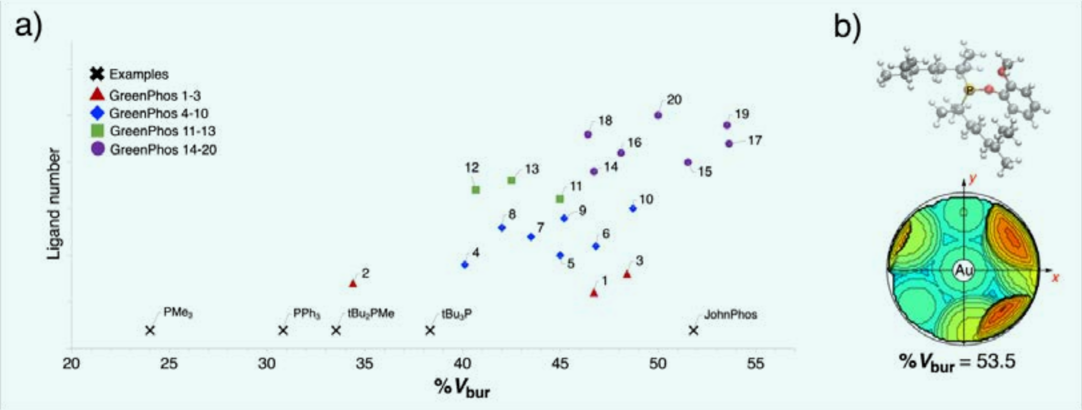

Figure 4: a) Computed percentage of buried volume for linear [R3PAuCl] complexes. b) Example of steric map computed for a phosphinite derived from a renewable phenol ligand. *Illustration: Abril Castro.*

identify how the interface can improve the process. This part of the project is in collaboration with the experimental groups of Prof. Karen Adler (Lund University), and Prof. Eva Hevia (University of Bern).

Personnel and Funding

Three PhD students of the CO2perate network visited the Hylleraas Centre from two to three months in 2023: Jiali Liu from Evonik, in Germany; Gabriel M. Batista, from the University of Aarhus in Denmark, and Sahil Gahlawat, from UiT. Li, in collaboration with Raquel J. Rama, PhD at UiO, used DFT methods and microkinetic models to study the formation of syngas from methanol using bifunctional catalysts. Batista used DFT methods to study the mechanism for the deconstruction of epoxy polymers with a base and a Ru catalyst. Gahlawat performed ab-initio molecular dynamics (AIMD) simulations for advancing ^{19}F NMR prediction. The projects of Li and Batista were supervised by Nova, and the project of Gahlawat by Castro. These projects were finalized by the end of 2023, and their publication is expected in 2024.

Two members of the NordCO2 consortium also visited the Hylleraas to work with Nova. Juan Miranda Pizarro, from the University of Helsinki in Finland, was introduced to AIMD simulations to study a Rh-hydride complex in collaboration with PhD Inga Schmidtke and co-supervised by Castro. Luliia Kolodiazhnaia, from Aarhus University in Denmark, worked on electrochemical CO$_2$ reduction with Mn-complexes involving organo-boranes. This work is connected with Luthra's PhD project on the use of Mn-complexes containing pendant amines [15].

The BSc student Inmaculada Ri, from the University of Ciudad Real in Spain, visited the Hylleraas Centre for three months to perform her MSc project under the supervision of Nova. Ri used DFT methods to study the mechanism of a hydrogen-borrowing reaction using bi-functional catalysts. This work connects with the PhD project of Aurore Denjean, PhD student at the Hylleraas center, who co-supervised Ri during her stay. Another visitor during 2023 was Dr. Sylwia Kozdra, from Łukasiewicz - Institute of Microelectronics and Photonics in Poland. Kozdra came with her own project to work with Nova in the modelling of polyvinylidene and its interaction with Li salts by using cluster models using the Gaussian software. During her stage, she also collaborated with Dr. Tamal Das, a postdoc at the Hylleraas Centre, on the use of DFT methods to study the structure of MoS2/MoO3 heterostructure [16].

In addition to these incoming visitors, there were two outgoing visits from Hylleraas PhD students. Ning Cao visited the group of Prof. Serena Debeer at the Max Planck Institute in Germany, to use computational methods for the simulation of EXAFs spectra of Cu attached to a Zr-nodes on a UiO MOF in the context of the Synergy grant CUBE. Inga Schmidtke, funded by NordCO2 visited the group of Prof. Timo Repo at the University of Helsinki to participate in the experimental work on one reaction she is studying in her thesis, the hydrogenation of ethylene by a Rh-hydride complex. This visit was funded by NordCO2.

Several PhD students participated in international conferences during 2023. In August, Cao, Luthra, Guerrero, and Denjean attended the European Conference on Computational and Theoretical Chemistry that took place in Thessaloniki, Greece. The poster of Denjean entitled "N-Doped Graphene-Based Nickel(II) Single-Atom Catalyst for Hydrogen Borrowing: A DFT Computational Study" won a prize for the best poster presentation. Denjean and Schmidtke participated in the XIV International school of organometallic chemistry in September in Camerino, Italy. Prof. Odile Eisenstein, adjunct professor at Hylleraas, was one of the lecturers in the school. In addition, Schmidtke and Mohammed E. Safi contributed to the NordCO2 annual meeting in Helsinki with oral presentations. Guerrero and Denjean presented a poster at the international TRAINS conference, organized in recognition of Helgaker's 70th birthday and held in November at Sundvolden.

Nova was invited to attend the Nobel Symposium NS193: “Metal-organic frameworks - Fundamental science enabling transformative materials”, which took place in September in Karlskoga, Sweden. Attendance at this symposium, organized by the Nobel Foundation and the Royal Swedish Academy of Sciences, was only possible by invitation. In the same month, she participated at the XLIX Congress of the Physical Chemistry Division of the Italian Society of Chemistry in Torino, Italy, as invited speaker, where she presented the work done on metal-organic frameworks [1-3]. Nova also organized a Virtual Workshop on Single Atom Catalysis (SACs) together with Profs. Bert Maes (University of Antwerp, Belgium), Matthias Beller (LIKAT, Germany), and Radek Zboril (Palacký University Olomouc, Czech Republic), in August.

The Grignard project is funded by the RCN (FRIPRO: MetalSynergy, Profs. Odile Eisenstein and Michele Cascella) and the EU (MSCA-IF: Ratio, Bortoli). In 2023 Marco Bortoli has obtained additional support for his project, by the EU in the form of high-performance computational resources, through the Euro-HPC JU initiative. In its methodological aspects, this project has strong connections to RT2/6, RT7, and RT0.

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RT7

Machine Learning and Mathematical Methods

Principal investigators:
David Balcells and Simen Kvaal

RT7 focuses on the development of machine learning (ML) and mathematical methods (MM) and their application to a broad range of problems in chemistry, physics, biology, and computer science. Regarding ML, the main activities include the computation of large datasets, the development of novel graph representations, generative methods, active learning approaches, and machine learned interatomic potentials. Regarding MM, the main activities include analysis of electronic-structure methods and development of novel time-integration schemes. A long-term goal is to integrate the ML and MM lines of research, focusing on topics like the mathematical foundations of ML.

ML. Natural Quantum Graphs.
Graphs are among the most expressive representations available for molecules and, when used to input, structure, and optimize (graph) neural networks, the resulting ML models are among the most accurate in the prediction of molecular properties. For transition metal complexes (TMCs), building graphs from xyz geometries or string representations like SMILES is challenged by the complex, often multi-center, nature of the metal-ligand bonds. We thus developed a method for deriving graph representations based on natural bond orbital theory (Figure 1). The fundamental idea was to use natural orbitals to define and inform the topology of TMC graphs, which were thus called natural quantum graphs (NatQG). These data were leveraged in a graph neural network based on a message-passing architecture to predict quantum properties like the HOMO-LUMO gap. We showed that the gain in accuracy originating from electronic structure information was significantly larger than that originating from geometric information. The code developed for this work, HyDGL, allows for building both undirected and directed graphs, where the latter express donor-to-acceptor orbital interactions. Further, we compiled all NatQG graphs in a 60k graph dataset, tmQMg, which, like the code, is openly available on the Internet.

The main author of this work was Hannes Kneiding, a PhD student in the Oslo node of the Center. The work also resulted from a fruitful collaboration between several partners, including Ruslan Lukin, Lucas Lang, Simen Reine, Thomas Bondo Pedersen, and Riccardo De Bin, under the supervision of David Balcells, who also designed the project. Ruslan, a PhD student from the University of Kazan (Russia), visited the Hylleraas Center in 2022. Lucas was an MSCA postdoctoral fellow also contributing to other ML projects, who is currently a Liebig Junior Research Group Leader at TU Berlin, Germany. From other RTs, both Simen (RT0) and Thomas (RT1) contributed, and, from other faculties, Riccardo, Assoc. Prof. at the UiO Department of Mathematics, also contributed.

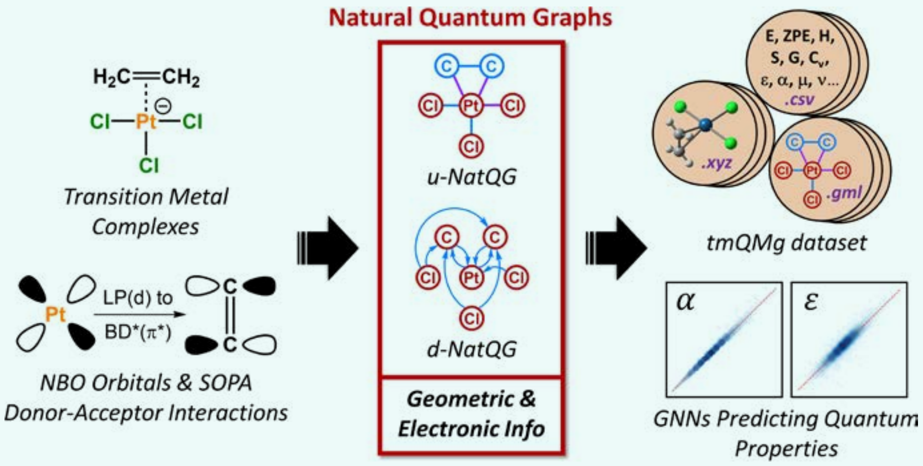

Figure 1: TMC graph representations from natural bond orbitals, including the tmQMg dataset and the prediction of quantum properties with graph neural networks. *Illustration: David Balcells.*

This work was first published as a preprint in the ChemRxiv, where it already attracted interest (>2000 views, >1000 downloads). Shortly after, it was peer-reviewed and published in the Digital Discovery journal of the RSC [1]. Further, it was highlighted in *Nature Computational Science* [K. McCardle, Learning properties of metal complexes, *Nat Comput Sci*, 2023, 3, 281–281] and presented by David Balcells in several invited contributions abroad, including two lectures, at Trinity College (Ireland) and the University of Geneva (Switzerland), and three keynote talks, at the CANBIC (Parry Sound, Canada), IMS-ICSDS (Lisbon, Portugal), and TSRC (Telluride, USA) conferences. The ML work summarized in the following three sections was also presented in these contributions. Furter, Hannes gave a talk about this project at the EuChemS CompChem conference (Thessaloniki, Greece) and, in relation to this work, he visited the group of Prof. Kulik at the MIT (Cambridge, USA) for three months, with the aim of developing active learning approaches to the exploration of vast chemical spaces derived from the tmQMg dataset. Further, to expand his training on ML methods based on graph theory, Hannes published a review on this topic in *Nordic Machine Intelligence* [2].

ML. Extended Autocorrelations.

Several ML models take a vector representation as input, which acts as a fingerprint encoding molecular similarity. These vectors are often derived from the associated graphs and a popular approach to such transformation is the Moreau-Broto autocorrelation function (ACF), recently adapted to TMCs. This function loops over a graph from a given origin, collecting properties and computing arithmetic operations with them – normally a summation of multiplication terms that is truncated at an arbitrary depth, consisting in the number of bonds "walked" from the origin. The ACF returns fixed-length vectors from variable size graphs, operating on the atomic properties of the nodes. We extended the ACF concept by adding the bond properties of the edges, yielding what we called the AABBA autocorrelation vector, which contains both pure, atom-atom (AA) and bond-bond (BB) terms, as well as crossed, bond-atom (BA) terms (Figure 2). Given the high dimensionality of the resulting vectors, we used gradient boosting machine models to identify the most relevant components of the AABBA vectors, which contained contributions from the three terms. These components were used to predict oxidative addition barriers with Gaussian processes and graph neural networks. The results showed that the addition of the BB and BA terms increased the accuracy of the predictions relative to the pure AA baseline. The AABBA code is openly available in the Internet.

This work was published as a preprint in the ChemRxiv [3]. The main author was Lucía Morán-González, who developed and implemented the code during her 3-month visit to the Center in 2022. Jørn Eirik Betten contributed with the ML models and this work was part of his MSc thesis, which he defended before moving to a PhD candidate position at the Simula Research Laboratory in Oslo. Hannes Kneiding also contributed with the NatQG graphs used to derive the AABBA vectors and, together with Nishamol Kuriakose, he recently started to implement this representation for metal-organic frameworks. Nishamol computed a 10k MOF dataset containing data for hydrogen adsorption capacity, in relation to the EU HYSTRAM project. She presented part of this work at the EUROMOF conference (Granada, Spain). The whole AABBA project was supervised and designed by David Balcells.

ML. Generative methods.

In recent years there has been a growing interest on generative ML models, which use data to produce more data of added value which can be, for example, texts, as in ChatGPT, or images, as in DALL-E. In chemistry and materials science, generative ML enables inverse design tasks in which a model is prompted with a desired property to generate molecules or materials that have it. One of the most powerful approaches to generative ML is evolutionary learning with genetic algorithms. In this approach, the system is expressed as a chromosome combining a collection of genes that yields a combinatorial explosion defining an implicit chemical space. Generations are evolved with genetic operations, including mutation and crossover, between elite individuals above a fitness threshold that relates to the property optimized. We developed a novel algorithm: the Pareto lighthouse multiobjective genetic algorithm (PL-MOGA), which allows to optimize multiple properties over the Pareto front in a directional manner including fine control over both the aim and scope of the optimization (Figure 3), without requiring prior knowledge on the properties optimized. In order to augment the spaces explored by the PL-MOGA, we also developed the tmQMg-L ligand library, which was derived from the tmQMg graph dataset. This library contains 30k diverse and synthesizable ligands. The method was benchmarked in chemical spaces containing millions of TMCs and thereafter applied to the exploration of larger spaces containing billions of TMCs.

This work was published as a preprint in the ChemRxiv [4] The main author was Hannes Kneiding, who developed the tmQMg-L dataset and implemented the PL-MOGA algorithm. Together with David Balcells, Ainara Nova, from RT5, contributed with a method for determining the ligand charges based on natural bond orbital theory. The project was supervised and designed by David Balcells. Hannes presented this work in two posters – one at the ICQC conference (Bratislava, Slovakia) and another at the Ellis Unit Summer School of the University of Cambridge (United Kingdom). Both the tmQMg-L dataset and the PL-MOGA code are openly available on the Internet.

More recent work on generative models has focused on the junction tree variational autoencoder (JT-VAE) method. In particular, Magnus Strandgaard, a PhD student from the University of Copenhagen (Denmark), visited the Center for a period of three months with the goal of implementing a JT-VAE model for TMCs. This work is being extended by Trond Linjordet who was recently hired in a postdoctoral position in the catLEGOS RCN FRIPRO project led by David Balcells. Further, Hannes is exploring ways of connecting the JT-VAE model with the PL-MOGA optimizer and there is one manuscript already in preparation.

ML. Catalyst high-throughput screening.

Catalyst discovery and optimization is a hard problem for ML since it involves multiple properties, in a context in which the training data needed to optimize the models is expensive and difficult to acquire. It is thus important to have a complete and deep understanding of the problem tackled before adopting any ML approach. In this regard, we used the tmQMg-L ligand library to define and explore a chemical space of 1500 palladium TMCs with potential activity in the Suzuki-Miyaura cross-coupling (SMCC), a reaction widely used in the pharmaceutical industry. This space was systematically screened by computing these three different properties with a DFT method: the oxidative addition barrier, the reductive elimination barrier, and the dissociation energy of acetonitrile. The screening showed that these properties are mutually exclusive to a large extent, that is: both barriers can be lowered though at the cost of maximizing the binding of acetonitrile, which is a metric for catalyst deactivation. Interestingly, several of the few systems identified as optimal in the screening have not been reported in the literature yet as active catalysts for the SMCC reaction.

The main contributor to this work was Arron Burnage, who computed the screening and performed the statistical analysis of the resulting data. Arron was recently hired in a postdoctoral position in the catLEGOS RCN FRIPRO project led by David Balcells. Hannes Kneiding contributed with the tmQMg-L library whereas Ainara Nova contributed to the definition of the chemical space and the properties screened. The project was supervised and designed by David Balcells. There is one manu-

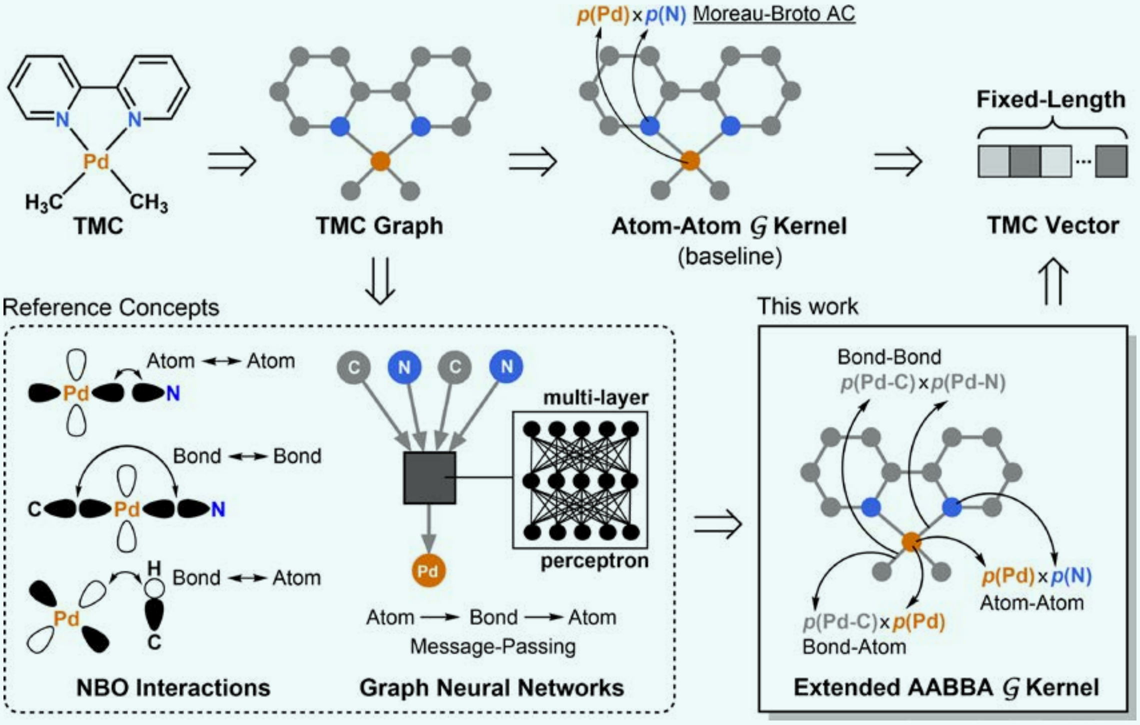

Figure 2: Extended AABBA graph autocorrelations for TMCs. *Illustration: David Balcells.*

script in preparation and Arron already presented the results in a poster at the TRAINS conference held in Sundvolden in honor of Trygve Helgaker.

ML. Interatomic potentials for metal alloys.

Christian Tantardini, postdoc at the UiT node, introduced a machine learning interatomic potential for multi-component magnetic materials uniquely incorporating magnetic moments as essential degrees of freedom alongside atomic positions, types, and lattice vectors [5]. The training set, meticulously constructed through constrained DFT, encompassed a broad configuration space, enabling the accurate prediction of properties in excited states, particularly those with non-equilibrium magnetic moments. Validation on the Fe-Al system underscored the potency of the developed machine-learning model, revealing its impressive reliability by demonstrating excellent agreement with DFT results for key properties such as formation energies, equilibrium lattice parameters, and total magnetic moments. This successful application highlighted the robustness and validity of machine-learning interatomic potentials in capturing complex magnetic behaviors, showcasing their potential as efficient tools for advancing material science research.

CCerror

The RCN project CCerror combines mathematics, physics and chemistry and has been carried out at the Hylleraas Centre since December 2019. It directly involves PI Andre Laestadius and researcher Mihaly Csirik (both in adjunct positions). During the year the project had four publications [6,7,8,9] that addressed coupled-cluster theory using homotopy and topological degree theory and a diagnostic based on strong monotonicity.

Moreover, A. Laestadius was invited to the 62nd Sanibel Symposium to give the talk "Some Recent Mathematical

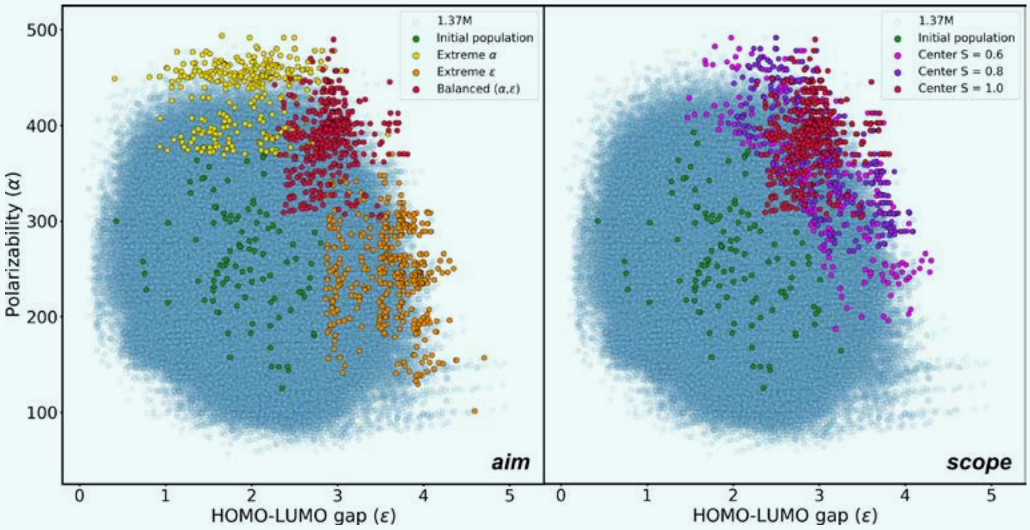

Figure 3: Aim and scope control over the optimization of Pareto solutions for the HOMO-LUMO gap and polarizability in a chemical space containing 1.37 million TMCs. *Illustration: David Balcells.*

Advances in Coupled-Cluster Theory" that disseminated results of the CCerror project. M. Csirik also attended the SIAM Conference on Computational Science and Engineering, Recent Advances in Numerical Methods for Electronic Structure Calculations, and presented the talk "Topological Index and Homotopy in Coupled-Cluster Theory" (based on the publications [6,7]). At the moment the researchers of CCerror are collaborating with former Hylleraas member F. Faulstich.

The QOMBINE Project: New Mathematical Foundation for CC Theory

The QOMBINE project is an RCN Fellesløft IV large-scale interdisciplinary researcher project funded jointly by RCN and the Faculty of Mathematics and Natural Sciences. The project consortium comprises physicists, chemists (Kvaal), and mathematicians, is led by Prof. Nadia S. Larsen (Department of Mathematics), and has a total budget of 25 MNOK, with project time frame 2022–2027. QOMBINE is interdisciplinary and seeks to develop new mathematical methods for central problems of quantum technology. Among the topics to be developed is a new mathematical foundation for coupled-cluster theory, the most popular quantum-chemical computational method besides DFT. Simen Kvaal, Sergiy Neshveyev (Mathematics), and Nadia S. Larsen are together with PhD students Håkon R. Fredheim (Hylleraas) and Snorre Bergan (Deartment of Mathematics) use C*-algebra theory and differential geometric notions to unify theory for electrons, protons, and other elementary particles, possibly also more exotic particle-like phenomena such as anyons and Majorana fermions. The latter are important building blocks in the development of putative topological quantum computers. On the other hand, a version of coupled-cluster theory called unitary coupled-cluster theory is classically intractable, but very fast on a quantum computer. Thus, the chemistry arm of QOMBINE aims not only at a better understanding of coupled-cluster calculations to be run on quantum computers, but also better understanding of how to build future quantum technologies. The QOMBINE project introduced a new, exciting research direction and interdisciplinary collaboration for the Hylleraas centre, involving several departments, including the industry sector.

Density-Functional Theory

An important research topic at the Hylleraas Centre is the mathematical development of density-functional theory, in its various forms. Hylleraas members have pioneered the Moreau–Yosida regularization technique in formal DFT (MY-DFT), and this continues to be a fruitful research topic. In 2023, a book chapter on the mathematics of the early developments of MY-DFT was published by Springer [10].

The ERC Starting Grant project *REGAL* of Prof. II Andre Laestadius (OsloMet, 20 % at Hylleraas) aims to make MY-DFT a practical tool. Laestadius and Mihaly A. Csirik have published articles on density-functional theory together with Hylleraas member Erik Tellgren. This was done as a two-series invited review article covering the structure of the density-potential mapping [11,12]. The second part, addressing the theory including magnetic fields, was selected as "Editors' choice".

Rothe's Method for Time Propagation

Electronic and molecular wavefunctions are highly complicated and their representation and time propagation presents enormous challenges. Since the AttoDyn CAS project (2021-23), we have been working on an adaptive scheme for propagating wavefunctions represented as linear combinations of *explicitly correlated gaussians*. These wavefunction representations yield an enormous compression ratio compared to conventional grid or fixed-basis representations, as is conventional in quantum chemistry. In 2023, the focus has been on a formulation for the three-dimensional hydrogen atom subject to intense laser pulses, and PhD candidate Simon E. Schrader has demonstrated that extremely accurate high-harmonic spectra can be reproduced with very few basis functions. Together with PhD candidate Aleksander Wozniak (Warsaw) and Ludwik Adamowicz (Arizona), Simen Kvaal and Thomas Bondo Pedersen have been working on proof of concept simulations on 2d model systems, yielding highly encouraging results.

Differential Programming

MSc student Aleksandar Davidov studied a model using *Differential Programming*, a machine learning paradigm that embeds computational methods artificial neural networks to enhance their capabilities. This represents a new research direction that combines machine learning with more formal mathematical tools. In particular, the embedding necessitates a differentiation of the computational method. Davidov and Kvaal studied the parameterized semi-empirical method PM3 embed-

Figure 4: The QOMBINE project anno 2023. Back row, second from right: PhD candidate Håkon Fredheim. Front row, from right to left: PhD candidate Snorre Bergan, Simen Kvaal, Nadia S. Larsen, and Sergiy Neshveyev. *Photo: Elisabeth Seland.*

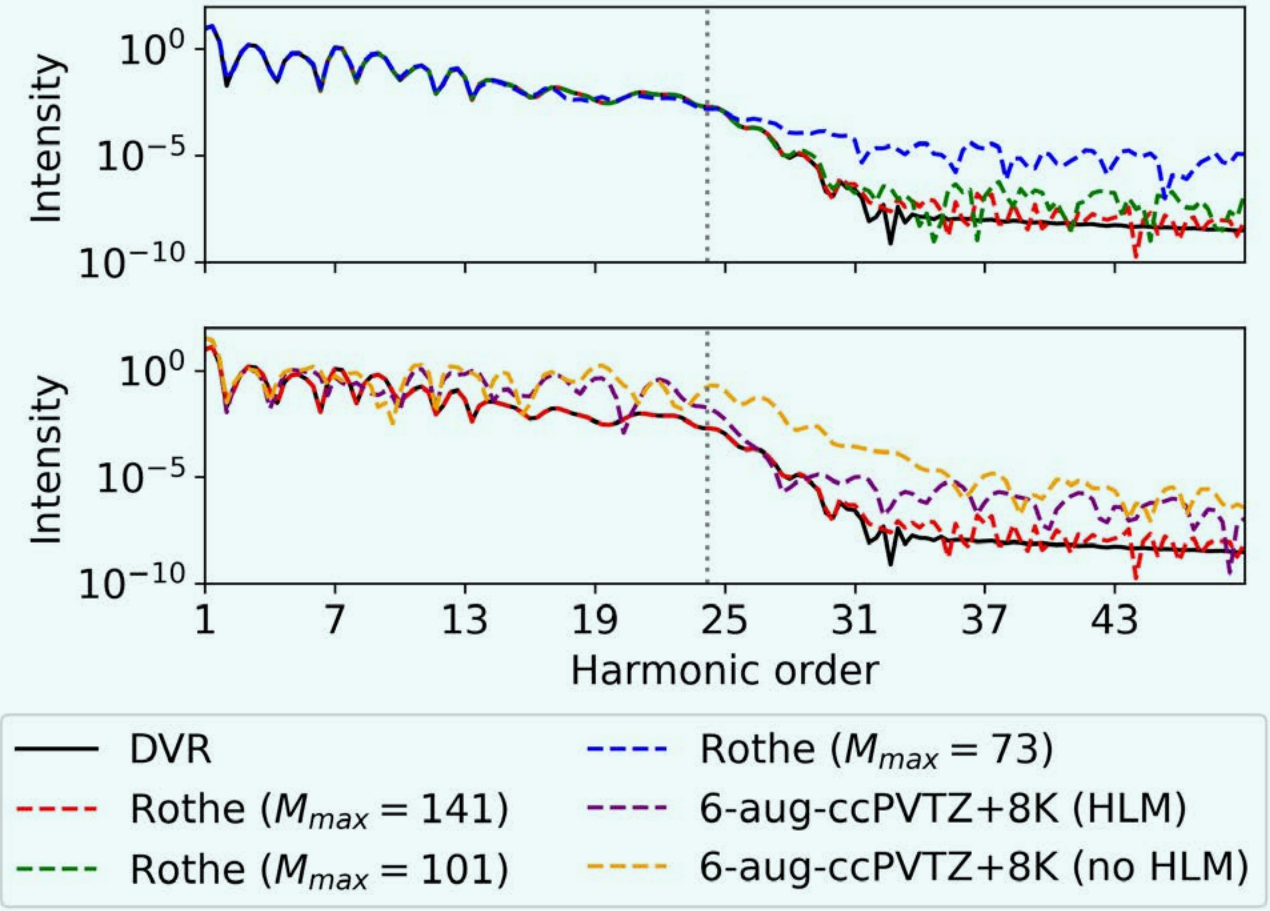

Figure 5: High-harmonic generation spectrum of a hydrogen atom subject to an intense infrared laser pulse of attosecond duration, computed using a variety of methods. The DVR reference is a highly accurate grid representation. The Rothe simulations demonstrate that with only a few basis functions, results that rival the DVR calculations in quality are possible, vastly outperforming conventional gaussian basis sets that are universally used in quantum chemistry, but that are not designed for quantum dynamics. *Illustration: Simon E. Schrader.*

ded in a graph neural network. The parameters of the PM3 model were optimized on a per molecule basis using this approach. The initial results yielded a mean absolute error in energies one order of magnitude smaller than plain PM3, with negligible overhead cost.

External Funding

catLEGOS FRIPRO, RCN, 2021-2025, 12 MNOK.
DEMO Doctoral Network, EU, 2023-2027, 3.5 MEUR.
HYSTRAM, EU, 2022-2025, 6.0 MEUR.
QOMBINE, Fellesløft IV, RCN, 2022-2025, 3 MNOK share (25 MNOK total)

Ingoing and outgoing personnel

Trond Linjordet started as a postdoc in the catLEGOS project.

Jørn Eirik Betten finished his MSc thesis and moved on to a PhD position at the Simula Research Laboratory in Oslo.

Aleksandar Davidov finished his MSc thesis and started a position in industry.

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RTO

Hylleraas Software Platform

Principal investigators:
Simen Reine and Bin Gao

–

2023 marked a pivotal year to advance the Hylleraas Software Platform into a powerful platform for computational research at the Centre.

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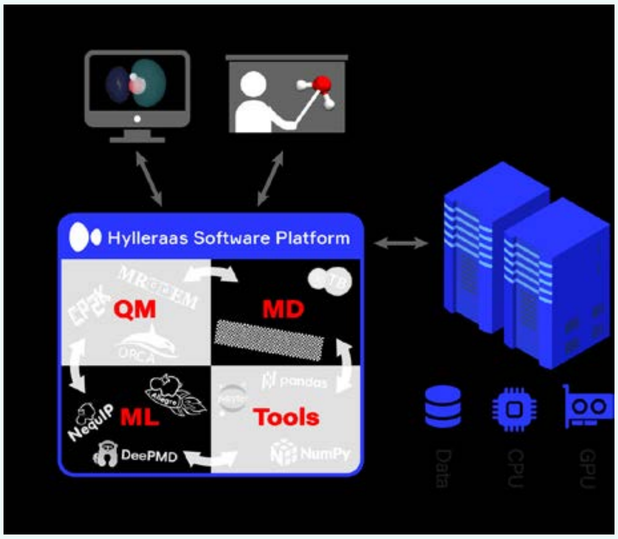
This year, the Hylleraas Software Platform (HSP) team has focused on enhancing the core functionalities of the platform to support the Hylleraas Centre's computational research needs. The team is composed of key personnel dedicated to the project to various extents. Bin Gao from UiT and Simen Reine from UiO serve as principal investigators, contributing 33% and 50% of their time, respectively, to the development of the platform. Tilmann Bodenstein, a researcher at UiO, is fully committed to the platform, working on it 100% of his time. Morten Ledum, a senior engineer at UiO, divides his efforts evenly between platform development and serving in an advanced user-liaison role at the IT department, a position partially supported by Sigma2, dedicating 50% of his time to each role.

An important achievement this year is a unified framework that allows running computational codes interchangeably through the HSP across various platforms. These include Anaconda environments, Docker containers, and external Slurm compute clusters such as the Norwegian Sigma2 machines Betzy and Saga, as well as the EuroHPC supercomputer LUMI. This significant development required a comprehensive overhaul of the computational settings and code interfaces within the HSP, ensuring a more efficient and flexible platform.

A notable highlight of 2023 was the development of functionality for the handling of datasets, designed for the automatic processing of inputs and outputs, and with smooth integration with machine learning engines to facilitate streamlined workflows for computational experiments and data analysis. Alongside this development, the platform has seen the addition of several new features. These include automated unit handling to simplify the use of different units across computations, a module for performing atomic-orbital transformations to facilitate interoperability between various software package representations, and support for asynchronous input/output that enables simulations to be run concurrently.

A flagship project in 2023 was the development of a fully automated active-learning framework to train machine-learning potentials for atomistic simulations. In collaboration with Sigbjørn Løland Bore, the project aims to study complex scientific phenomena such as ion transfer processes, phase transitions, and chemical reactions. It not only serves as a practical application of HSP's capabilities but also provides a rigorous testbed for refining the platform's core functionalities. It



Outline of the architecture of the Hylleraas Software Platform.
Illustration: Simen Reine and Bin Gao.

involves complex workflows that utilize multiple software tools running in parallel, with integrated job submissions across different computing environments and automated data handling.

Furthermore, the team has engaged in enhancing the platform's capabilities through a series of collaborations and developments aimed at supporting the Hylleraas Centre's broader research objectives. Among these are efforts to implement fully relativistic natural chemical shielding (NCS) analysis of chemical shieldings in collaboration with RT5. The team also works on integrating the CREST interface and workflow setup to streamline chemical reaction studies, also in collaboration with RT5. Additionally, to support RT1 developments, a general visualization tool for studying attosecond processes has been developed to simplify the visualization of complex quantum phenomena.

The platform now interfaces with a broad spectrum of software, expanding its utility across quantum mechanics codes such as xTB, Orca, Dalton Project, ReSpect, LONDON, MRChem, CP2K, and VASP; molecular mechanics codes like LAMMPS and HyMD; and machine learning engines including DeepMD and Nequip. These interfaces significantly broaden HSP's applicability in computational research, ensuring it meets the diverse needs of the Hylleraas community.

In summary, while 2023 has seen a focus on enhancing the platform's fundamental capabilities, the progress made significantly contributes to the Centre's computational efficiency and research flexibility. The developments in code interfacing, environment adaptability, and data processing set a strong foundation for future expansions and research initiatives within the Hylleraas community.

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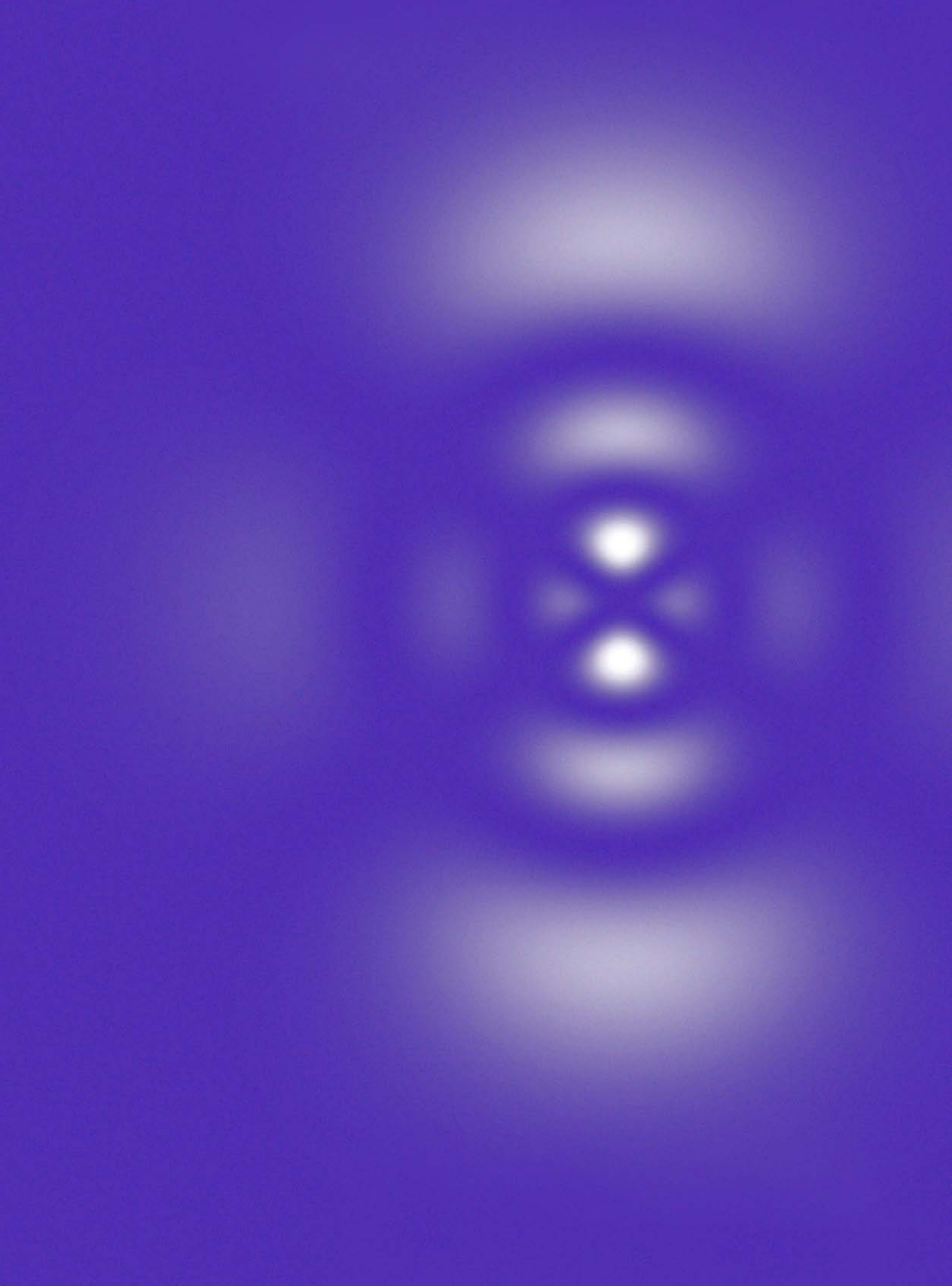

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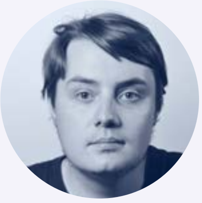

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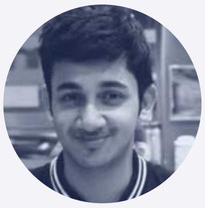

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Funding and Staff Report

Funding 2023

(in NOK 1000)

Basic funding as agreed in the Centre contract	
Funding from Research Council (CoE grant)	18 436
Funding from home institutions	11 825
Total basic funding	**30 261**
Additional funding from external projects	
Research Council funding	26 227
International funding	2 647
Public funding	0
Private funding	0
Total additional funding	**28 874**
Total funding	**59 135**

Staff Report 2023

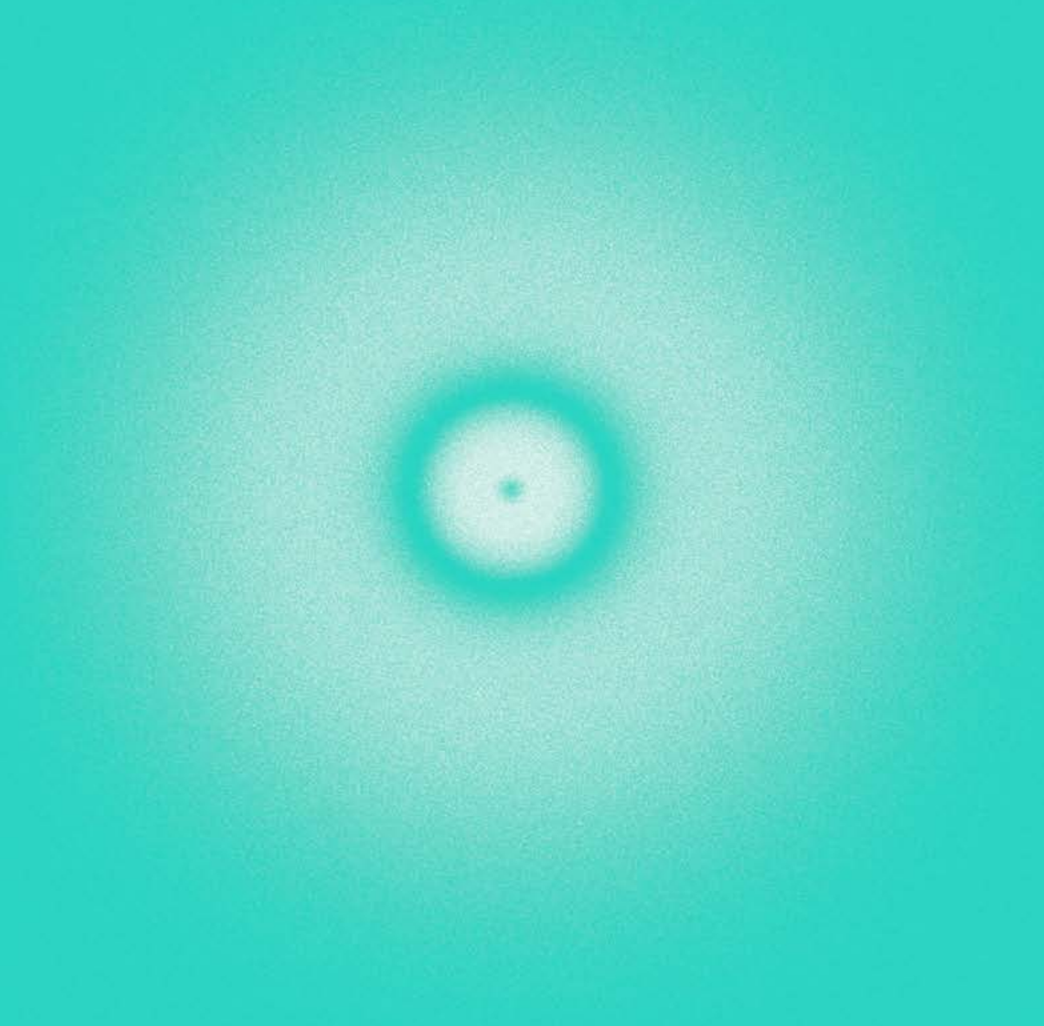
(in man-years)

Professors and researchers	15.7
PhD Candidates	22.4
Postdoctoral Fellows	14.5
Visiting Researchers	4.1
Administrative and technical staff	3.7
All staff	60.4

> “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.

Design: Anagram Design



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