

Project Summary

The Hylleraas Centre for Quantum Molecular Sciences will make a decisive, timely contribution to chemistry, biology, astrophysics, geology, and materials science by developing multiscale methods for complex systems interacting with external fields. The emergence of new experimental facilities such as the MAX IV Laboratory and the European XFEL offers new opportunities but also poses new challenges for theory and computation. With the advent of more powerful computers and the development of advanced computational methods, the goal of the Hylleraas Centre is to treat systems containing thousands and even millions of atoms and to explore processes extending over microseconds, subject to short laser pulses and strong magnetic fields.

The Hylleraas Centre gathers world-leading expertise in the domain of electronic-structure theory, multiscale modelling at the mesoscale, computational spectroscopy, and the use of computation to unravel complex chemical and biological systems. The centre is uniquely positioned to take the lead in the modelling and understanding of the new scientific frontiers that will be opened by the new experimental facilities.

Through an extensive incoming sabbatical program, a generous visitors program, focus bienniums, international workshops, conferences, outreach activities, and seminar series, the Hylleraas Centre will create an internationally visible and attractive centre for the computational modelling and understanding of new chemistry at the frontiers of a wide range of scientific disciplines. Through the training of PhD students and postdocs, the establishment of a Norwegian graduate school and international summer schools, the Hylleraas Centre will educate a new generation of computational scientists skilled to address the new research frontiers. The centre will continue longstanding efforts to improve gender balance in theoretical and computational chemistry.

Primary and Secondary Objectives

The primary objective of the Hylleraas Centre is to develop and apply computational methods to understand, interpret, and predict new chemistry, physics, and biology of molecules in complex and extreme environments.

Secondary objectives are to (1) develop accurate, reliable, and scalable electronic-structure methods for complex systems, (2) develop multiscale methods with quasi-atomistic/mesoscale resolution, (3) understand electron–nuclear dynamics processes in strong fields and spectroscopy at interfaces, (4) predict magnetic bonding in white dwarfs and crystal impurities, (5) model processes for renewable energy sources and eco-friendly production of chemicals, (6) model van der Waals-dominated systems and interface dynamics, (7) train a new generation of theoretical and computational chemists, (8) establish a Norwegian graduate school, and (9) provide innovative, new software to the international community at no cost.

Hylleraas Centre for Quantum Molecular Sciences

new tools, new opportunities, new discoveries

The Hylleraas Centre will develop and apply computational methods to understand, interpret, and predict new chemistry, physics, and biology of molecules in complex and extreme environments.

With the advent of powerful computers and the development of advanced computational methods, **first-principles calculations** of molecular electronic systems have become commonplace. Using advanced software such as our program DALTON [1], quantum-mechanical calculations are being routinely performed on systems containing hundreds of atoms by chemists in academia and industry, complementing or supplementing experimental work, with an accuracy that often rivals that of experimental measurements [2]. However, over the next decade, **new experimental facilities** such as the European Spallation Source (ESS), the MAX IV Laboratory, and European XFEL will revolutionise the way we study matter, by enabling the use of intense laser fields in a wide frequency range and allowing ultrashort pulses to be generated, targeting increasingly complex chemical and biological systems. This development offers new opportunities but also poses **new challenges for theory and computation**: To exploit fully the potential of the new infrastructures, computation will be needed not only to simulate the **complex systems** themselves to complement experiment but also to interpret the new experimental data, bearing in mind that these have been generated by little-understood **matter-field interactions**.

Simulations of complex systems require the simultaneous treatment of phenomena that occur over widely different time and length scales. For a balanced description of different time and length scales, **multiscale modelling** is performed in a hierarchical manner, with a core treated quantum mechanically and its surroundings treated classically, at various levels of detail. The **Hylleraas Centre for Quantum Molecular Sciences**¹ will make a *decisive, timely contribution to chemistry by developing multiscale methods for complex systems, emphasising matter-field interactions*, see Fig. 1. Our goal is to treat systems containing thousands and even millions of atoms and processes extending over microseconds, including their interactions with short laser pulses and ultrastrong fields. With our new tools, we aim to study and uncover new phenomena, often in collaboration with experimentalists.

Illustrations: Two examples from our own recent work illustrate how new tools for computational studies of matter-field interactions may provide crucial insight into natural phenomena under vastly different conditions, see Fig. 2. Using our program package LONDON [3], Helgaker and co-workers published in *Science* in 2012 the discovery of a new bonding mechanism, **paramagnetic bonding**, generated by the motion of the electrons in ultra-strong magnetic fields [4]. As a result, chemistry

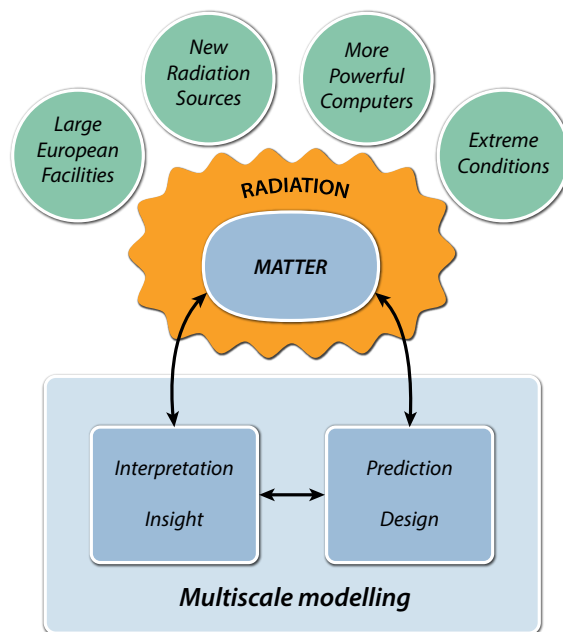


Figure 1: The Hylleraas Centre will make a decisive, timely contribution to chemistry by developing multiscale methods for complex systems, emphasising matter-field interactions

¹The 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 quantum-mechanical calculation of the lattice constant and cohesive energy of an ionic crystal, LiH (1930).

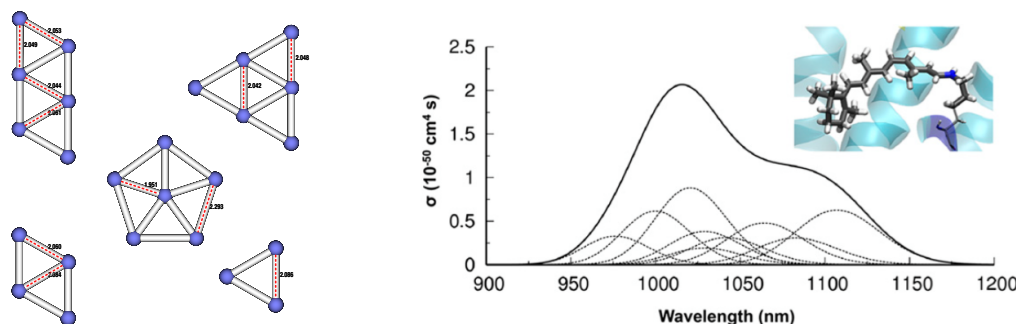


Figure 2: *New tools, new opportunities, new discoveries.* Left: Magnetically bound helium clusters in a field of 470 kT calculated with LONDON [6]. Rare-gas atoms bind in planar clusters constructed from triangular units, adopting a perpendicular orientation to the field. Right: Rhodopsin two-photon absorption cross section (full line) constructed from snapshot curves (dashed lines) obtained from DALTON calculations on the quantum region (inset) during multiscale dynamics [7]. The simultaneous absorption of two photons of wavelength 1020 nm (infrared, maximum of total curve) corresponds to absorption of one visible photon of wavelength 510 nm.

is strongly affected, giving rise to molecules that cannot exist on Earth but are predicted to exist on magnetic white dwarfs—indeed, a collaboration with astrophysicists aims at detecting these unusual molecules. Intriguingly, the effects of magnetic fields can be strongly enhanced by certain crystal impurities and experimental work has begun to detect paramagnetic bonding in such impurities [5].

In 2014, the long-standing puzzle of **human infrared vision** was explained in a publication in *Proceedings of the National Academy of Sciences* by Cascella, in collaboration with experimental groups [7]. Combining classical simulations of proteins with quantum-mechanical calculations of two-photon absorption in rhodopsin performed using DALTON, the authors demonstrated that infrared vision arises from the simultaneous absorption of two infrared photons, whose total energy and absorption cross section are equal to those of a single visible photon and hence detectable by the eye.

These examples illustrate how molecular calculations can provide key insight into chemical, biological, and physical phenomena, replacing or complementing experimental investigations under vastly different conditions: while magnetic bonding occurs in the fierce environment of white-dwarf atmospheres, two-photon activation of rhodopsin occurs in the delicate environment of the human eye. The examples also show how new tools (first developed in our laboratory) pave the way for new research and discoveries, in synergy with experimental work in multiple disciplines.

Research Themes: Our goals require breakthroughs in three areas: *simulation methods* for complex systems; *matter-field modelling* to study such systems perturbed by electromagnetic fields; and *interpretation and prediction of new chemistry, physics, and biology* using advanced tools. In each area, work is organised in two **Research Themes (RTs)** as summarised in Table 1 and described below.

The work on simulation methods for complex systems in RT1 and RT2 constitute the core methodological development at the centre, underlying all other activities. In **RT1 (Electronic Structure)**, we address the need for a more accurate description and understanding of electron correlation, dispersion and relativity, important for complex systems governed by non-electrostatic interactions and core-hole excited states. In **RT2 (Multiscale Modelling)**, we develop accurate, reliable modelling tools for surfaces, interfaces, and biological membranes. Methods to be developed include quantum-based multiscale modelling and the coupling of quantum and atomistic models to coarse-grained and mesoscale models. The challenge of widely different time scales of electronic and molecular motion requires coupling of different time-propagation schemes to be developed.

Matter-field interactions take centre stage in RT3 and RT4. In **RT3 (Spectroscopic Processes)**, the methods of RT1 and RT2 are linked to emerging experimental spectroscopic techniques, including multiphoton processes, high-harmonic generation, and time-resolved spectroscopies, allowing us to unravel processes observed in ultrafast pump-probe experiments and to link multidimensional vibrational spectroscopies to biomolecular structures. In **RT4 (Extreme Environments)**, the exotic chemistry of molecules in the ultrastrong magnetic fields of magnetic white dwarfs and neutron stars will be studied in parallel with field-enhancement effects in complex systems, using a multiscale extension of LONDON. Important goals are the discovery of molecules in magnetic white dwarfs and para-

RT1: Electronic Structure TBP & LF • Projects: correlation in finite and extended systems; relativity; bivariational principle; multiwavelets • Challenges: reliability and affordability; scalability; static correlation; dispersion; time propagation	RT2: Multiscale Modelling MC & BOB • Projects: embedding; atomistic and coarse-grained models; mesoscale statistical-field methods • Challenges: quasi-atomistic/mesoscale resolution; amorphous materials, organelles
RT3: Spectroscopic Processes KR & TBP • Projects: nonlinear and multi-dimensional spectroscopies; electron and nuclear dynamics • Challenges: novel nonlinear spectroscopies; surface/interface spectroscopies	RT4: Extreme Environments TH & KR • Projects: chemistry and bonding in ultrastrong magnetic fields and ultrahigh pressure • Challenges: accurate spectra for detecting magnetic bonding in white dwarfs and crystal impurities
RT5: Chemical Transformations OE & KH • Projects: complex reactions in solution; catalytic reactions, catalysts and protocols design • Challenges: processes for renewable energy sources and eco-friendly production of chemicals	RT6: Multiphase Systems BOB & MC • Projects: macromolecular complexes, protein–membrane interactions, adsorption • Challenges: van-der-Waals dominated systems; Interface dynamics; molecular crowding

Table 1: **Research Themes**; for initials of theme leaders, see Table 2

magnetic bonding under laboratory conditions. Like ultrastrong fields, ultrahigh pressure confines matter, yielding materials with dramatically different properties. Exploiting this link, high-pressure chemistry with relevance to planetary physics, geochemistry, and life sciences will be explored.

Chemical transformations may involve short-lived highly reactive species that are difficult to detect and characterise—in particular, for complex systems such as biological macromolecules or solution reactions with many components. The expected experimental advances and new radiation sources will provide data critical for understanding such processes. **RT5 (Chemical Transformations)** will address challenging chemical transformations, with emphasis on processes that can lead to a more sustainable use of resources such as processes associated with renewable energies (e.g., artificial photosynthesis) and more eco-friendly production of chemical feedstocks and high-value chemicals. Key goals of RT5 will be to predict alterations for improved efficiency and selectivity of these processes.

Advances in imaging techniques are expected to provide high-resolution structural data of large macromolecular complexes in their natural environment, producing a wealth of data on macromolecular localisation at functionalised interfaces. In **RT6 (Multiphase Systems)**, we study the dynamics of macromolecules at interfaces, focusing on molecular transport and on the mechanism of self-assembly and diffusion of macromolecular complexes. With the advances in RT1 and RT2, RT5 and RT6 will be well positioned to make the best possible use of new experimental information and to guide new experiments. Key examples include the role of nanoparticles in water oxidation, related to artificial photosynthesis (RT5), and the activity of antimicrobial compounds targeting specific cell walls (RT6).

Ambitions, Impact, and Feasibility: Complex systems and multiscale modelling are important current areas of research in chemistry. Our contribution will be unique, with the inclusion of advanced methods for describing matter–field interactions. Additionally, our quantum description extends to all (core and valence) electrons, with an explicit treatment of electron correlation and the option for a full four-component relativistic treatment. Timeliness relative to emerging experimental facilities ensures an impact extending far beyond the centre itself, in research and industry. This impact is strengthened by the dissemination of the methods developed to the community through **software distribution**.

The Hylleraas team possesses world-class expertise in the quantum chemistry of molecular properties and spectroscopic processes (Frediani, Helgaker, Pedersen, Ruud); recently, the expertise has been significantly extended to multiscale modelling and complex systems by strategic group additions (Brandsdal, Cascella, Eisenstein, Hopmann). The team includes **all ERC chemistry grant winners in Norway** (Helgaker, Ruud, Kvaal), **all but one of the four RCN Young Research Talent (YRT) grant winners in chemistry** (Fliegl, Hopmann, Tellgren), and **one Marie Curie Career Integration**

name	position	PI	aff	RT1	RT2	RT3	RT4	RT5	RT6	external funding	nationality
Trygve Helgaker (TH)	Prof	director	UiO	✓	✓	✓	✓			ERC ADG	Norwegian
Kenneth Ruud (KR)	Prof	✓	UiT	✓	✓	✓	✓	✓		ERC STG	Norwegian
Bjørn-Olav Brandsdal (BOB)	Prof	✓	UiT		✓			✓	✓	RCN FRIPRO	Norwegian
Odile Eisenstein (OE)	Prof (50%)	✓	UiO				✓	✓	✓		French
Michele Cascella (MC)	Assoc Prof	✓	UiO		✓			✓	✓		Italian
Thomas Bondo Pedersen (TBP)	Assoc Prof	✓	UiO	✓	✓	✓	✓				Danish
Luca Frediani (LF)	Assoc Prof		UiT	✓	✓	✓	✓		✓		Italian
Kathrin Hopmann (KH)	Researcher	✓	UiT			✓			✓	RCN YRT	German
David Balcells (DB)	Researcher		UiO		✓			✓		MC CIG	Spanish
Heike Fliegl (HF)	Researcher		UiO	✓			✓	✓		RCN YRT	German
Bin Gao (BG)	Researcher		UiT	✓	✓	✓					Chinese
Simen Kvaal (SK)	Researcher		UiO	✓	✓	✓	✓			ERC STG	Norwegian
Erik Tellgren (ET)	Researcher		UiO	✓	✓	✓	✓			RCN YRT	Swedish
Simen Reine (SR)	Sr Engr		UiO	✓	✓	✓	✓	✓	✓	n/a	Norwegian
Michal Repisky (MR)	Sr Engr		UiT	✓	✓	✓	✓	✓	✓	n/a	Slovak

Table 2: *Leadership, research and technical staff at the Hylleraas Centre*

Grant (MC-CIG) winner (Balcells). The team is young (all but two members younger than 50 years of age) and international (ten non-Norwegian members of eight different nationalities), see Table 2.

The Hylleraas Centre builds on the success of the Centre of Theoretical and Computational Chemistry (CTCC), a Centre of Excellence established by the RCN in 2007 with emphasis on integrating computation into participating experimental groups. The emerging experimental techniques and facilities (ESS, MAX IV, XFEL) mandate a concerted theoretical effort to model complex system in extreme fields and environments. The Hylleraas Centre will build on the core theoretical competence of the CTCC and recent group additions to achieve its goals.

Organisation of Hylleraas Centre

The centre is organised as a project at the Department of Chemistry, University of Oslo (UiO), and operated as a Centre-of-Excellence (CoE) consortium with two nodes, located at the UiO and UiT The Arctic University of Norway (UiT). In recognition of their theory groups as world leading, both institutions have recently made significant investments by increasing the permanent staff by two positions in each group. Additional in-kind support is provided as detailed in the budget.

Leadership and Management: The **Director**, appointed in an 80%–20% position at the two universities, has responsibility for all centre activities—in particular, the fulfilment of its scientific ambitions. In the first five-year period, the centre is directed by Prof. Trygve Helgaker (UiO, 296 publications, 16500 not-self citations, H-index 66); in the second period, by Prof. Kenneth Ruud (UiT, 274 publications, 7100 not-self citations, H-index 50). Both are **ERC grant winners**. The **Management Team**, consisting of the director and the principal investigators, oversees all research, training, outreach, and dissemination, with responsibility to allocate resources among the research themes, identify new projects, and promote synergies. Each research theme is headed by a theme leader and a co-leader, one from each node, with responsibility for research, proposals, and hiring in that theme.

For an efficient day-to-day management, one **Head of Office** is recruited at each node. The centre is governed by a **Board of Directors** with representatives of each university, industry, and the Norwegian Chemical Society. The board is responsible for strategy in matters of research and training, economy, and budget, exercising a monitoring function in these matters. A **Scientific Advisory Committee** of five prominent international scientists including experimentalists, to be consulted annually and meet biennially, will give independent advice regarding research, management, and funding.

Funding: Internally funded projects will be complemented by **externally funded** projects that support the goals and vision of the centre. All projects, funded internally or externally, will be designed and evaluated with respect to their potential for cross-links and synergies. Centre members are expected to apply for funding; the ambition is to have at least one ERC grant and three FRIPRO grants running at any given time. The centre will also seek to participate in relevant H2020 projects involving data from the new infrastructures and to coordinate an MSCA-ITN network.

Research Environment: To bring freshness, inspiration, and visibility to the centre, each non-methodological theme (RT3–RT6) will have a **Focus Biennium** dedicated to its activities, with an international **Summer School** for PhD students in the first biennium year and a **Hylleraas Conference** in the second year. A **Specialist Workshop** will be organised in tandem with each school and conference. The team members have long experience with the organisation of such schools and meetings.

To attract the best international expertise, the centre will run a broad **Sabbatical Programme**, coordinated with but not exclusive to the biennium activities. Potential sabbatical visitors will be identified and invited to spend three months or more at the centre, with at least one full year offered to a prominent female scientist in each biennium. An extensive **Visitors Programme** will enable members to initiate new collaborations and reinforce established ones. Seminars, including a **weekly seminar series**, will be broadcast to both nodes by video conferencing. Two centre meetings will be organised yearly, one in cooperation with the Norwegian Chemical Society.

Outreach and Dissemination: Scientific results and other centre information will be disseminated through the following **standard channels**: peer-reviewed articles (600 or more in the 10-year period), software distribution, conference presentations, annual reports, web pages, blogs, and popular science articles. **Software** will be maintained, developed, and distributed under the leadership of Sr. Engrs. Dr. Reine and Dr. Repisky, ensuring an extensive impact on the academic and industrial community at large. **Social media** (Facebook, Twitter, YouTube) will be used to promote activities at the centre—for example, software tutorials, new results, popular talks, or didactic videos. Each year the centre will host an open **Hylleraas Day** for science dissemination to non-experts, targeting young people. Yearly, a prominent scientist will be invited to deliver the **Almlöf–Gropen Lecture**, initiated by the CTCC in 2008, intended to be broadly accessible and promote the field to the scientific community.

Training and Mentoring: The centre will establish **national PhD courses** in theoretical and computational chemistry, to be given biennially to students from all Norwegian universities. A **mentorship program** will be created to prepare young researchers for a career as scientific leaders. With the opportunities offered by external funding, the centre is expected to train more than 35 postdocs and 30 PhD students during its 10-year lifespan. All centre members will be engaged in the training of master, PhD, and postdoc students and give PhD courses; all members apart from Helgaker, Ruud, and Eisenstein (50% position) will perform normal, undergraduate teaching duties at their institutions.

Gender Balance: The centre will work towards reducing gender imbalance in theoretical chemistry by making all efforts to ensure female applicants to all positions, by having at least 30% female members including students, by focusing on career hurdles for female researchers, and by organising seminars with focus on gender imbalance and implicit bias. Additionally, the centre will support the conference series **Female excellence in theoretical and computational chemistry (FemEx)**, established by the CTCC and now adopted as a regular international conference [8].

References

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