PhD Position in AI accelerated simulations of chemical reactivity at ChimieParisTech – PSL University

Title: Learning to Choose: Adaptive Electronic-Structure Methods for Excited States in Photocatalysts

Application Deadline: May 22, 2026, at 13h00 (CET)

Start Date: Between September 1, 2026, and November 30, 2026

Supervisor: Junior Prof. Thijs Stuyver, fellow of PR[AI]RIE-PSAI institute

Host Institution: ChimieParisTech – PSL University

Funding: PR[AI]RIE-PSAI cluster

Research Topic

Over 90% of the chemical processes worldwide utilize some form of catalysis.[1] The utilization of excited state reactivity of transition-metal catalysts, where energy is supplied by light, has emerged as an exciting opportunity to drive novel, efficient and selective reactions. Nickel (Ni) photocatalysts in particular have proved especially useful due to their rich spectrum of excited states, relative abundance, and low cost.[2] These catalysts already have remarkable value in processes such as converting carbon dioxide to fuel, generating hydrogen gas, or converting atmospheric nitrogen to ammonia, but predictive modelling of these processes remains largely out of reach, limiting our understanding and ability to rationally design improved systems.[3] Advancing the design of novel Ni photocatalysts is thus crucial for a sustainable future.

In general terms, the photochemical reactivity of transition-metal complexes is governed by excited states with fundamentally different electronic character, including metal-centered (d–d), charge-transfer, and strongly correlated states, which can lead to distinct reaction pathways.

Computational chemistry provides an efficient alternative to experiments for exploring such systems, and most studies rely on time-dependent density functional theory (TDDFT). However, TDDFT employs a fixed exchange–correlation functional and often fails to describe diverse excited states with consistent accuracy. In first-row transition-metal complexes, small geometric distortions can induce rapid changes in electronic structure, leading to regimes where different states require qualitatively different theoretical treatments.[4,5] In practice, this may require not only tuning the parameters of the functional but also switching between methodological variants, such as spin-flip approaches.[6]

Higher-level wavefunction-based methods, such as Restricted Active Space Spin-Flip (RAS-SF), provide more reliable descriptions of excited states in such systems.[7] However, their higher computational cost and the need for system-specific active space selection hinder their routine application across large datasets.

These challenges highlight a fundamental limitation of current approaches: the lack of a state-sensitive, adaptive framework in which the electronic-structure treatment

can be adjusted to individual excited states while balancing accuracy and computational cost. Addressing this limitation requires learning from limited, high-quality reference data. In this context, recent work by Prof. Stuyver and co-workers has shown that learned representations (latent spaces) can capture complex structure–property relationships beyond predefined descriptors.[8] Building on this idea, the present project will leverage such representations to enable data-efficient, AI-driven adaptive modelling of excited states.

Objectives & Expected Research Outcomes

The goal of this project is to develop state-sensitive, machine learning-assisted electronic-structure models for excited states of transition-metal complexes, with a particular focus on Ni complexes.

The project is structured around three main objectives:

Aim 1: Automated generation of high-quality reference data

Develop an automated workflow for active space selection in RAS-SF calculations, enabling reliable descriptions of electron correlation. In practice, descriptors of orbital importance will be designed, and an agentic decision-making framework will be developed to automate the setup and evaluation of these types of calculations.

Aim 2: Learning representations of excited states

Train graph neural networks on the results of ensembles of electronic-structure calculations, in particular TDDFT with diverse exchange–correlation functionals, to learn information-rich latent representations of excited states. By combining multiple approximate descriptions, these representations will encode both state character (e.g., metal-centered vs charge-transfer) and sensitivity to the underlying method.

Aim 3: State-sensitive adaptation of electronic-structure methods

Using a limited set of high-level RAS-SF reference data generated by our agent (see aim 1), develop machine-learning models that relate learned excited-state representations (see aim 2) to the performance of different TDDFT functionals and spin-flip variants. The goal is to detect when standard approaches become unreliable and to identify the most appropriate electronic-structure treatment for each state.

In summary, this project aims to introduce a shift from fixed electronic-structure approximations to adaptive, state-aware methodologies guided by machine learning. By combining multiple approximate descriptions to learn robust, bias-reduced representations of excited states, the approach enables reliable and scalable prediction of photochemical reactivity in transition-metal complexes. The framework is expected to generalize to a broader class of strongly correlated systems, advancing the integration of AI with first-principles simulations for chemical discovery.

References

[1] K. Christensen, in *Chemical Product Design: Toward a Perspective through Case Studies*, **2007**.

[2] F. Julia, *ACS Catal* **2025**, *15*, 4665–4680.

[3] Z. Wang, W. Sun, G. Li, Y. Li, B. Jiang, *ACS Appl Mater Interfaces* **2025**, *17*, 41457–41497.
[4] D. Khan, A. Price, B. Huang, M. Ach, O. A. von Lilienfeld, *Science Advances* **2025**, *11*.
[5] A. G. Garrison, H. J. Kulik, *J. Chem. Theor. Comput.* **2026**, *22*, 2243-2260.
[6] Y. Shao, M. Head-Gordon, A. I. Krylov, *The Journal of Chemical Physics* **2003**, *118*, 4807–4818.
[7] D. Casanova, M. Head-Gordon, *Phys Chem Chem Phys* **2009**, *11*, 9779–9790.
[8] G. Chen, T. Stuyver, *Digital Discovery* **2025**, *4*, 3227–3237.

Eligibility and Selection Criteria
Candidates will be evaluated based on:
- Academic excellence
- Relevance of their background to the research topic

The selection process follows an open, transparent, and merit-based (OTM) recruitment procedure.

Non-discrimination, openness, and transparency:
All PR[AI]RIE-PSAI partners are committed to supporting and promoting equality, diversity, and inclusion within their communities. We encourage applications from diverse backgrounds and ensure a fair selection process.

Application Requirements
Applicants must submit the following documents:

1. Curriculum Vitae (CV)
2. Motivation Letter (max. 1 page) describing:
   - Your interest in the research topic
   - How your background aligns with the project
3. Copy of your most recent diplomas

Application Procedure
- Deadline: May 22, 2026, at 13h00
- Applications should be submitted to: thijs.stuyver@chimieparistech.psl.eu
- The evaluation process consists of two phases:
  1. **Pre-selection** by the supervisor.
  2. **Final selection** by an expert committee, evaluating applications based on excellence and alignment with PR[AI]RIE-PSAI's scientific program.

Final results will be communicated by **June 5, 2026**.

For additional information, please visit https://thijsstuyver.com or contact thijs.stuyver@chimieparistech.psl.eu.