Postdoctoral Researcher Position – 12 months

Sorbonne University, Paris

Complex Phases in Ammonia-Water Mixtures Under Extreme Conditions: Disorder, Plasticity, and Super-Ionicity

Scientific context: The project aims to theoretically model the crystalline phases of ammonia-water mixtures and characterize the associated forms of atomic-scale disorder using molecular dynamics simulations. This understanding is crucial for planetary physics, as these mixtures are found in giant planets like Neptune and Uranus. Despite the recent experimental exploration of the phase diagram of these mixtures [1], simulating the associated phase transitions at the atomic level remains a challenge.

The structure of solid phases of water, ammonia, and their mixtures is governed by hydrogen bonds, resulting in rich polymorphism. Indeed, in mixed crystals, hydrogen bonds vary depending on donor/acceptor molecules and on conditions of temperature and pressure. At high-pressure, proton transfers leads to the appearance of ionic phases that are still poorly understood. Collaboration between theorists and experimentalists is essential : experimental techniques like X-ray diffraction and vibrational spectroscopies evidence various types of disorder (positional, orientational, dynamic/static, concentration-related) but their full interpretation requires comparison to atomic-scale simulations.

Missions: Given the extensive phase diagram of Ammonia-Water mixtures, the recruited person will focus on two main objectives: (i) characterizing the crystal to plastic phase transitions in Ammonia-water mixtures with different stoichiometries and (ii) exploring ionic (and possibly superionic) phases of ammonia hemi-hydrate (AHH).

- **(i)** In particular, we will investigate if the coupling between orientational and translational disorder, that we recently evidenced in AHH [2], can also be at play in other mixtures. For this objective, improving the parametrization of existing interatomic force fields to describe the molecular phases at high pressure is crucial. Parametrization and assessment of the force fields can be done via Bayesian approaches, using DFT calculations as reference.
- **(ii)** Addressing ionic phases will require developing accurate reactive interatomic potentials to describe ionization processes. Combining physical interactions and machine learning techniques [3] (using DFT data for training) should provide an accurate yet computationally affordable description.

The refined force fields will be integrated in highly parallel codes to perform large-scale Molecular Dynamics, to study the atomic dynamics and the stability of the different phases.

[1] L. Andriambariarijaona et al. PRB **108** 174102 (2023). H. Zhang et al. *J. Phys. Chem. Lett.* **14**, 2301 (2023). [2] N. Avallone et al. Orientational disorder drives site disorder in plastic ammonia hemihydrate. *Under review* (2024). [3] T. Plé, L. Lagardère, J.-P Piquemal, Chem. Sci., **14**, 12554 (2023).

Activities:

- Conceive refined force fields, either via Bayesian or machine learning approaches. Prepare and perform molecular dynamics simulations, analyze the resulting data. Propose technical solutions to any difficulties encountered.

- Write reports and scientific papers on the results obtained.

- Participate in meetings concerning the project.

- Communicate the results via seminars or conferences.

- Ensure effective communication between theory and experiment.

Skills:

- The candidate should have a PhD in physics, chemistry or a related field.

- The candidate should be creative, have good analytical skills and be highly motivated by computational work.

- A good knowledge of statistical physics is required. Expertise in at least one of the following topics: parameter optimization via Bayesian statistics ; enhanced sampling techniques in statistical mechanics ; *ab initio* molecular dynamics ; nuclear quantum effects methods ; machine-learning interatomic potentials. Previous experience with several topics in the list will be highly appreciated.

- Good ability to work both independently and in a collaborative working environment.

- Good writing and communication skills are expected. Written and spoken English at B2 level at least is required. Knowledge of French is not required.

Work context :

The project is a collaboration between three groups from Sorbonne University:

- The *low-dimensional oxydes group* at the INSP laboratory (https://w3.insp.upmc.fr/en/institute/) whose theoretical component focuses on atomic scale simulations, with particular interest for nuclear quantum effects simulation methods.

- The PHYSIX team at IMPMC Institute (http://impmc.sorbonne-universite.fr/fr/index.html) whose emphasis is on experimental and theoretical research on materials under extreme conditions of pressure and temperature.

- The multiscale simulations and high performance computing group at the theoretical chemistry laboratory, in which innovative machine learning approaches for the modelization of interatomic interactions were recently developed.

Location : Sorbonne University, Jussieu Campus, Paris.

Constraints and risks :

The work of the candidate will be theoretical and computational and does not involve particular risks. Specific dedicated training will be provided if required.

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