PhD position in Path Integral Molecular Dynamics applied to Geochemistry

Starting date : October 2020.
Directors: Merlin Méheut and Magali Benoît
Location of the position : Géosciences Environnement Toulouse (GET), « Stable Isotopes Geochemistry » group, Observatoire Midi-Pyrénées, Université Paul Sabatier (Toulouse, France)

Scientific context

Isotopic compositions of natural phases constitute a tool of primary importance to assess geological history. Recent progresses in mass spectrometry and analytical chemistry have enabled the detection of isotopic variations of a host of elements (e.g. Fe, Mg, Ca, Li, Zn), opening the path to numerous applications. In particular, Ca isotopes have shown their potential for medical applications (early diagnosis of decalcification), to assess mechanisms occurring at the soil-plant interface, or to assess the Ca cycle in the environment.

To take the full extent of these new measurements, it is necessary to precisely constrain the isotopic effect associated with elementary processes occurring at the atomic level. In this perspective, the fractionation of isotopes reached at thermodynamical equilibrium (called equilibrium fractionation) between two phases (between one mineral and a dissolved species, or between two dissolved species) is of particular interest.

It is possible to estimate isotopic fractionation based on atomistic modeling by estimating the thermo-kinetic properties of atoms in a given bonding environment. For solids, these properties are generally estimated based on the harmonic approximation, starting from the vibrational frequencies of the mineral (e.g. Méheut and Schauble 2014). For liquids, we have developed an approach based on path integral molecular dynamics (PIMD, Dupuis *et al.* 2017). This approach, which was applied to Li isotopes, is computationally very expensive, and that is why it has been limited so far to the use of empirical potentials to describe atomic interactions.

Our goal is to develop a numerical scheme to compute equilibrium fractionation properties dissolved species, based on path integral molecular dynamics methods, but with atomic bonding described by ab initio electronic structure computations (instead of empirical potentials), so as to render its application to virtually any case. This approach will be first tested on the Ca^{2+}-H_2O system for which experimental data exist, and then for systems relevant to the understanding of the Ca cycle in the soil-plant system.

Description of the position:

The PhD student will first model aqueous systems relevant to the Ca natural cycle (complexed species of Ca) by means of classical molecular dynamics (based on empirical or ab initio approach). In the mean time, the set up of the ab initio path integral framework will be performed by M. Méheut and a postdoctoral researcher. Then the PhD student will apply the developed ab initio path integral framework to these systems. The work will be mainly realized with the CP2K and Quantum Espresso codes.

The candidate should have a Master degree in chemistry, chemistry-physics, theoretical chemistry or physics, or equivalent obtained in 2019 or in 2020 and must have a solid training in physical and theoretical chemistry. Experience in ab initio molecular calculations as well as programming skills (Fortran, Python...) are assets. Interest for Geosciences and Environmental Sciences is a plus.

For further information about the project, please contact:
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To apply for this position, please connect to the following link : https://bit.ly/2X5UIbt
CV and motivation letter will be requested.

Some papers in relation to the proposed work::

Dupuis R., Benoît M, Tuckerman M, Méheut M. (2017), Accounts of chemical research 50 (7), 1597-1605

Méheut M. and Schauble E.S. (2014), Geochimica et Cosmochimica Acta 134, 137-154