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Direct prediction of equilibrium isotope fractionation from molecular dynamics trajectories: Application to metal alloys, silicate melts and hydrothermal fluids.

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First-principles atomistic calculations can be used to determine equilibrium isotope fractionations. By providing these essential thermodynamic parameters and linking these macroscopic isotopic properties to structural properties at the molecular scale, computational studies effectively complement the study of isotopic variability observed in nature and the experimental studies that are very often challenging.

In the case of liquid systems, molecular dynamics (MD) simulations performed at finite temperatures are required. However, the conventional method is to derive fractionation factors from snapshots sampled along the MD trajectory, where the atomic positions are then optimized at zero temperature. Some of the configurational disorder characterising the fluid is thus lost. Here we present an approach based on atomic kinetic energy, which allows the isotopic fractionation factors to be determined directly from the whole MD trajectory.

The advantages and disadvantages of this direct approach and the more conventional "snapshot" approach will be compared and discussed, based on the following applications: Fe isotopes in $Fe_{1-x}S_x$ alloys under conditions relevant to formation of the Earth's core [1], Fe isotopes in various silicate melts [2], and S isotopes of sulphur-bearing species in hydrothermal fluids.

References:

[1] Pinilla, de Moya, Rabin, Morard, Roskosz & Blanchard (2021), Earth and Planetary Science Letters 569, 117059.

[2] Rabin, Blanchard, Pinilla, Poitrasson & Grégoire (2023), Geochimica et Cosmochimica Acta 343, 212-233.