Atomistics Simulation of Vibrational Signals in Liquid Water and Aqueous Solutions Using Machine Learning Potentials

Axel Gomez, Miguel de la Puente, Rolf David, Damien Laage, Rodolphe Vuilleumier, **Ari Paavo Seitsonen**[1]

[1]Département de Chimie, École Normale Supérieure, 24 rue Lhomond, Paris, France

Ari.P.Seitsonen@iki.fi

The liquid water is a conceptually simple but ultimately a complex liquid rich in details that still has several complicated aspects that are not yet well understood. Most of the peculiarities in water derive from the hydrogen bond network (HBN) that connect the water molecules to each other. The vibrational spectroscopy is a powerful approach to extract information of the HBN.

Recently we have studied the infra-red and Raman spectroscopy signals in liquid water using the density functional theory-based molecular dynamics (MD) simulations to yield the dynamics and the spectroscopic intensities of the liquid water. Furthermore we were able to gain insight into the dependence of the vibrational *fingerprints* of distinct hydrogen bond configurations around a central molecule by restraining the surrounding hydrogen bonds to a given setup.

We are now extending the simulations using machine learning potentials (MLP) to accelerate the molecular dynamics simulations and thus allowing to collect improved statistics and thereby more accurate determination of those fingerprint frequencies. We will further expand the scope of the to more complex cases of solvation of ions and ionic solutions. The extended length of the achievable simulations and the accuracy of the MLP will allow us to tackle also questions related to the (very) low frequency oscillations that have remained elusive until recently.

Figures

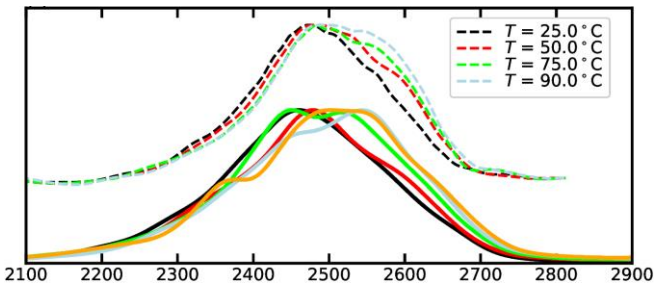

Figure 1. The measured – dashed lines – and simulated – solid lines – temperature dependence of the Raman spectra in the isotropic – left – and anisotropic – right – mode in the O-D stretching frequency range in liquid D_2O [1]

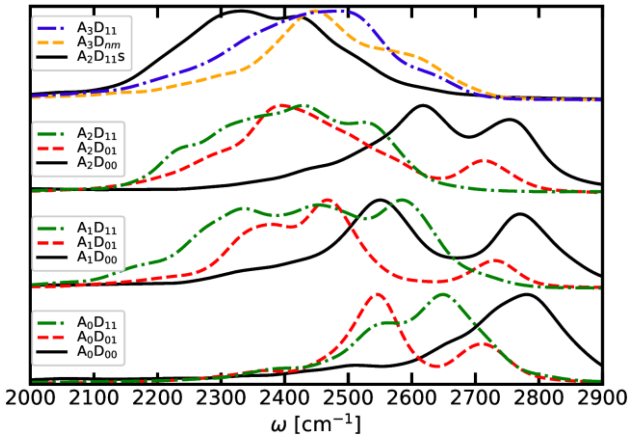

Figure 2. The simulated vibrational infra-red spectrum at distinct local configurations of the hydrogen bond network around a water molecule in the O-D stretching frequency range in liquid D_2O [1]

References

[1] Rodolphe Vuilleumier *&* Ari Paavo Seitsonen, Condensed Matter Physics, 26 (2023) 33301; DOI: 10.5488/CMP.26.33301