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Ab initio theory for computing sum frequency generation spectra at aqueous interfaces

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Understanding aqueous interfaces at the molecular level is not only fundamentally important, but also highly relevant for a variety of disciplines. For instance, electrode–water interfaces are relevant for electrochemistry, as mineral–water interfaces for geochemistry and air–water interfaces for environmental chemistry; lipid–water interfaces constitute the boundaries of the cell membrane, and are thus relevant for biochemistry. One of the major challenges in these fields is to link macroscopic properties such as interfacial reactivity, solubility, and permeability as well as macroscopic thermodynamic and spectroscopic observables to the structure, structural changes, and dynamics of molecules at these interfaces. Simulations, by themselves, or in conjunction with appropriate experiments, can provide such molecular-level insights into aqueous interfaces. We study aqueous interfaces, by assessing computations of the sum-frequency generation (SFG) spectra, which selectively detect the interfacial molecules, at aqueous interfaces. To avoid bias in the computational results and interpretation originating from the choices of the details of FF models, applying a parameter-free *ab initio* molecular dynamics (AIMD) simulation technique to the SFG calculation seems to be a promising route. However, the huge computational cost required for AIMD simulation has prohibited the widespread use of AIMD simulations for computing the SFG spectra. We have recently presented an efficient calculation algorithm for computing the SFG spectra of the water O–H stretch mode based on the surface-specific velocity–velocity correlation function, by separating degrees of freedom of the nuclei from solvation effects such as the induced dipole and polarizability. This methodology has been applied to the fundamental water-air, water-lipid, and aqueous solution-air interfaces. We are going to extend our method to solid-liquid interfaces.

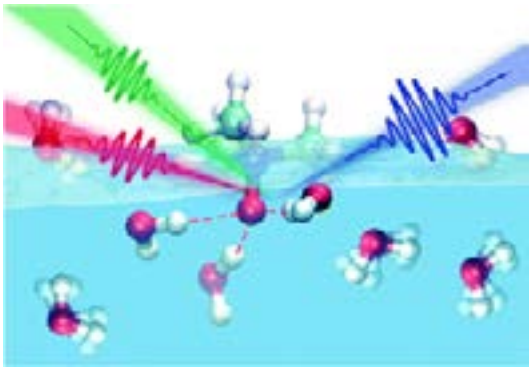

Figure: Trimethylamine-N-oxide solution-air interface

Biography

Tatsuhiko Ohto is an Assistant Professor at Osaka University, Osaka, Japan. He received his PhD degree from University of Tokyo in 2013. During his PhD course, he spent five months as a visiting student at Max Planck Institute for Polymer Research, Mainz, Germany. After Post-doctoral research at Advanced Institute of Science and Technology, Tsukuba, Japan, he joined Osaka University. His research interest is in theoretical modeling, primarily based on first-principles calculations of the structure and dynamics of molecules at interfaces and the electron transport of metal–molecule–metal systems.

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