VII-D Theory of Sum Frequency Generation Spectroscopy

Visible-Infrared Sum Frequency Generation (SFG) spectroscopy has been recognized as a powerful experimental technique to optically probe a variety of interface structure. In spite of the recent technical progress in experimental measurement, its analysis remains a challenging issue in general, since experimental spectra are not often amenable to straightforward fitting or interpretation. We have proposed theoretical methods to compute SFG directly *via* molecular modeling and molecular dynamics simulation, without resorting to empirical parameterization. This work aims at developing our methods for practical applications.

VII-D-1 Improved Computation of Sum Frequency Generation Spectrum of Water Surface

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The SFG spectrum of water surface was calculated with significantly improved accuracy *via* the time-dependent formalism we have recently proposed. The revisions include molecular modeling of OH stretching region, sampling statistics, and treatment of boundary conditions. The computation was performed by fully exploiting the massive parallel environment of the NAREGI supercomputers. The calculated spectra show excellent agreement with the recent experiments, allowing us fairly detailed comparison and analysis.

VII-E Theory of Mass Transfer Kinetics at Liquid-Vapor Interfaces

Mass transfer kinetics at liquid-vapor interfaces is of fundamental significance in atmospheric chemistry or fluid engineering, and accordingly it has been studied over a century. Understanding of the mass transfer kinetics ranges from phenomenological macroscopic or thermodynamic description to molecular-scale dynamics, and precise comparison between laboratory experiments and molecular-level simulation poses conflicting, unresolved problems. This study tries to obtain a unified picture of mass transfer kinetics, including bulk transport and surface accommodation.

VII-E-1 Mass Accommodation Coefficient of Water

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The mass accommodation coefficient α of water into liquid water has been a controversial issue for a long time, whether α is essentially unity or smaller than one. We have recently argued that the molecular dynamics study yield an α value very close to one, and the droplet train experiments, which insists a significantly smaller than one, is in fact not inconsistent to the unit α, after accurate analysis of the experimental uptake kinetics. This study strengthens our argument, and showing possible directions to resolve this conflict.