Center for Molecular Spectroscopy and Dynamics, IBS-Korea University

IBS Center for Molecular Spectroscopy and Dynamics

COLLOQUIUM

■ SPEAKER

Dr. Yoshitaka Tanimura (Department of Chemistry, Kyoto University)

■ TITLE

Simulating, Modeling, and Analyzing Two-Dimensional THz-Raman and IR-Raman Spectroscopies

■ ABSTRACT

Understanding dynamics in complex environments of molecular liquids and biological systems has been a central topic of investigation in chemistry and biology, because many important chemical processes occur exclusively in such media. Recently, two-dimensional (2D) THz-Raman spectroscopy has been developed to investigate the intermolecular modes of liquid.[1] It is also possible to extend this measurement for infrared region instead of THz to investigate the intramolecular modes. We calculate such 2D spectroscopy signals for water using an equilibrium-non-equilibrium hybrid MD simulation algorithm [2-4] originally developed for 2D Raman spectroscopy.[5] These signals are analyzed in terms of anharmonicity and nonlinear polarizability of vibrational modes using a Brownian oscillator (BO) model with linear-linear (LL) and square-linear (SL) system-bath interactions [6] from the hierarchal equations of motion (HEOM) approach for a non-Markovian noise.[7] All of the characteristic 2D profiles of the signals obtained from MD are reproduced using the LL+SL BO model, indicating that this model captures the essential features of the inter-molecular motion. We analyze the fitted the 2D profiles in terms of anharmonicity, nonlinear polarizability, and dephasing time. The origin of the echo peaks of librational motion and the elongated peaks parallel in the probe direction are elucidated via optical Liouville paths. [3,6] Moreover, we find that this OH-stretching and HB-intermolecular vibrational coupling should be observed as off-diagonal cross peaks in the 2D spectra.[3]

References

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■ DATE AND VENUE

November 21, 2016 (Monday, 5:00-6:00 p.m.)
Seminar room 116, KU R&D Center

■ Language

English