Dynamics and Nonlinear Spectroscopies of Liquids: Theoretical and Computational Approaches

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Molecules in condensed phases, e.g., solutions and biological systems, show very complicated motions.[1] Knowledge of the molecular motions is indispensable to understand, control, and design chemical reactions in condensed phases. Spectroscopy is a useful method to investigate molecular dynamics. However, it is difficult to get detailed information about molecular dynamics from conventional spectroscopies in which all information is projected onto one variable, time or frequency.

Multidimensional spectroscopy is a powerful way to overcome this difficulty. Various multidimensional spectroscopic methods are intensively exploited to investigate vibrational and electronic dynamics.[2] In the multidimensional spectroscopy, a system interacts with electric fields at several different times and thus it is possible to analyze the complicated dynamics in details. We established *ab initio* theoretical method by using to calculate nonlinear multidimensional spectroscopies. We investigated liquid dynamics by using the two-dimensional Raman spectroscopy and two-dimensional IR spectroscopy of liquids and elucidated the coupling between intermolecular modes and the detailed molecular mechanism of energy relaxation in water which are difficult to reveal by using conventional one-dimensional spectroscopy.[3,4] In the lecture, the theoretical background of spectroscopy will be introduced at the beginning and then our results of nonlinear spectroscopy of liquids will be presented.

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