

“Cold, Large Molecules: Chirality, Trace Detection and Sticking”

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【講演要旨】

We are studying polyatomic molecules in an experimental apparatus that can provide large collision rates under constant, low energy controlled conditions. This has been made possible through the advent of a novel cooling method that allows the introduction of hot polyatomic molecules into a cryogenically cooled, low density inert gas.

As part of these studies we have developed methods for trace detection of molecules in mixtures, in both the optical and microwave regime. The cooling of the molecules leads to dramatic increase in the inverse of the internal molecular ro-vibrational partition function. For example, cooling of anthracene ($C_{14}H_{10}$) from 300K to 4K would lead to more than a ten order of magnitude increase in the per quantum state phase space density of the molecular ensemble.

In addition to species identification, we have also demonstrated a method to measure the absolute chirality of the molecular species, and enantiomeric excess. Chirality plays a fundamental role in the activity of many biological molecules and in broad classes of chemical reactions. Spectroscopic methods for determining enantiomeric excess include optical circular birefringence (CB), circular dichroism (CD), vibrational circular dichroism (VCD), and Raman optical activity (ROA). All of these chiral analysis methods yield zero signal in the electric-dipole approximation. In contrast, the electric-dipole signal from sum-frequency generation can be non-zero in a bulk chiral environment. Sum-frequency generation (SFG) in the infrared and visible has been observed in samples of chiral molecules in solution in previous experiments. Doubly resonant SFG in both the infrared and microwave regime has been proposed but not observed. We recently demonstrated enantiomer-sensitive spectroscopy by combining a resonant microwave field with a strong adiabatically switched orthogonal non-resonant (DC) electric field.

Following on that previous work, and using our novel cooling method, we demonstrated true sum-frequency generation, a type of three-wave mixing, on a chiral sample in the microwave regime. We use two orthogonally polarized resonant applied electric fields to induce a third mutually orthogonal field at their sum frequency. The phase of this induced field changes sign with enantiomer, and its amplitude provides a sensitive, quantitative measure of enantiomeric excess. The narrow rotational resonances used make this measure of enantiomeric excess fundamentally mixture compatible: in contrast to other spectroscopic methods, the measurement of E.E. of a given species will not be confused by other chiral or non-chiral mixture components in the sample.

I will discuss this new chiral detection method, our efforts on trace detection, and if time permits, our progress on answering the question of sticking of single atoms to large molecules at low temperatures.