

Separation of Intermolecular Motions in Liquids with Polarization Dependent Stimulated Raman Spectroscopy

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Recently, we demonstrated an experimental scheme to measure stimulated Raman scattering in the THz regime [1]. Here, we demonstrate an extension of our experiment to polarization dependent measurements. For the setup, we use a regenerative amplifier providing linear polarized fs-pulses (800nm, 4W, 50fs) at a repetition rate of 1 kHz. To induce a positive frequency chirp to these pulses, we stretched them to around 4ps (at full width half maximum). Then, the laser beam was sent to an interferometer, where it was split into a pump and a probe beam. At the output of the interferometer, both beams were aligned parallel. A $\lambda/2$ -waveplate in the pump beam allowed us to change the polarization angle between the pump and the probe beam. By focusing the pump and the probe beam onto the sample, we generated stimulated Raman scattering at the frequency corresponding to the optical beat generated with the pump and probe beams. Consequently, we observed an energy transfer between the pump and the probe beam by separating the probe beam with an analyzer and an iris and detecting the signal with a Si-Photodiode. The specific scattering frequency and the direction of the energy transfer depends on the time delay between the pump and probe pulses. By scanning with the delay stage of the interferometer, we are able to investigate stimulated Raman scattering across a frequency range between 100 GHz and 10 THz. Figure 1 depicts the results of the measurements of an aqueous solution of ZnBr_2 at 0° and 90° polarization angle. Such a measurement is

equivalent to the observation of the Raman depolarization ratio [2]. In order to compare both measurements, we removed an offset signal from the 0° measurement, which resulted from insufficient separation between the pump and probe beam, and scaled the signal by a constant factor. As a result, we can separate two different overlapping spectral features: a distinct peak and a broad spectral band. A previous report assigned the peak to a vibration of a ZnBr_2 complex [3]. The disappearance of the peak at 90° implies that the origin of the peak is a highly symmetric vibration, while the broad feature remained at 90° might originated from more non-symmetric intermolecular vibrations.

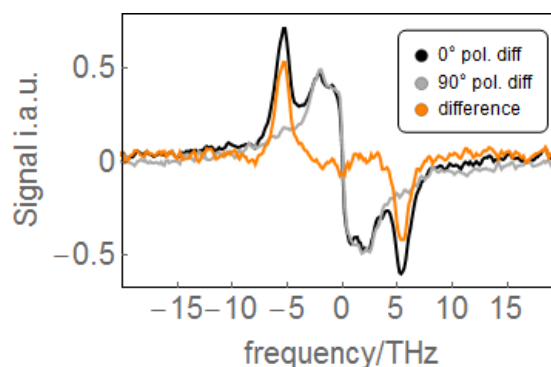


Fig 1. Measurements of aqueous ZnBr_2 (4M).

References:

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