

2D-Raman THz Spectroscopy of Ionic Liquids

S. Shukla¹, A. Shalit¹, P. Hamm^{1*}

¹Dept. of Chemistry, University of Zurich

2D Raman-THz spectroscopy is a novel spectroscopic method that measures the 2D response of intra and intermolecular degrees of freedom of liquid samples in the low frequency range (below 300cm⁻¹) Fig. 1(a). It has been successfully used to study the dynamical heterogeneity in liquid water and various aqueous salt solutions [1,2]. The current work aims to extend the application of this technique to study ionic liquids, such as 1-Allyl-3-methylimidazolium dicyanamide (AMI) Fig. 1(b).

Room temperature ionic liquids (RTILs) are an interesting class of salts that are liquids at or below room temperature, and their modularity allows for a wide range of desirable properties, making them a promising candidate for various industries such as energy storage, chemical synthesis, CO₂ capture, and green chemistry. The presence of charges in the chemical structure of ionic liquids might facilitate heterogeneity in the structural dynamics, and 2D Raman-THz spectroscopy is expected to provide new insights into the dynamics of these materials. Fig. 1(c) shows a response of AMI liquid obtained by means of our recently developed fast acquisition single shot 2D Raman-THz spectrometer [3] where extent of the signal beyond the strong instantaneous response along $t_1 = t_2$ diagonal (dashed line) encodes inhomogeneity of the liquid.

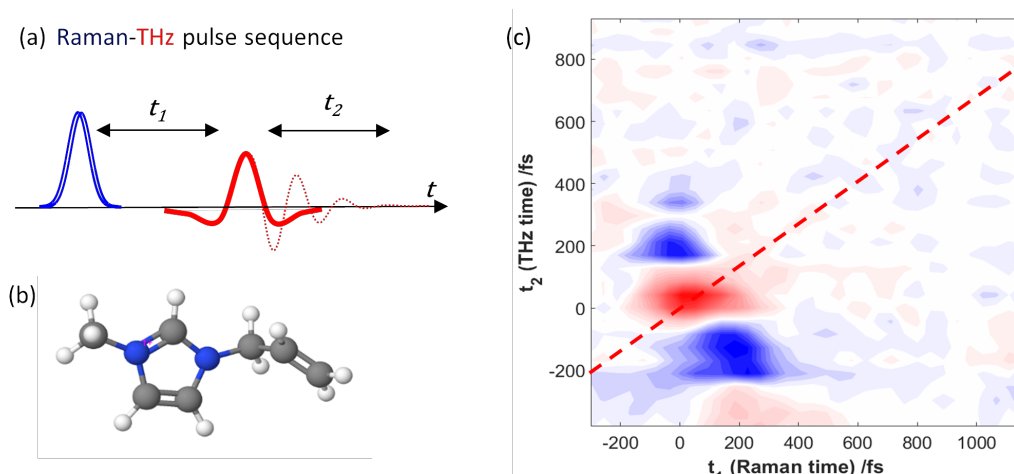


Fig.1 (a) Raman-THz pulse sequence (b) AMI molecular structure (c) 2D Raman-THz response of 1-Allyl-3-methylimidazolium dicyanamide

We are currently analyzing the data with respect to the possibility of seeing an echo along this diagonal.

[1] Janne Savolainen, Saima Ahmed, Peter Hamm, Proc. Natl. Acad. Sci. U.S.A. **110** (51), 20402–20407 (2013).

[2] Andrey Shalit, Saima Ahmed, Janne Savolainen & Peter Hamm, Nature Chem **9**, 273–278 (2017).

[3] Marta Duchi, Saurabh Shukla, Andrey Shalit, Peter Hamm, J. Chem. Phys. **155**, 174201 (2021)