2D-Raman THz Spectroscopy of Ionic Liquids

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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.

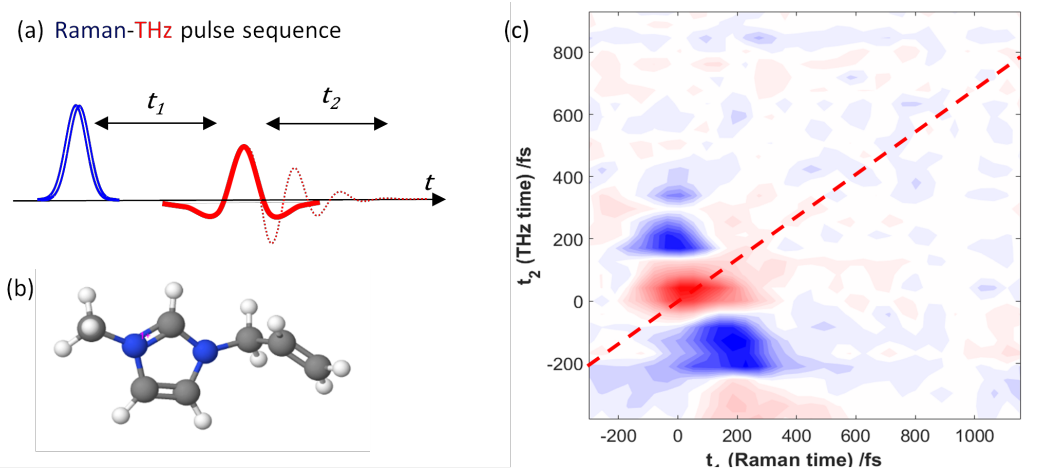

Fig.1 (a) Raman-THz pulse sequence (b) AMI molecular structure (c) 2D Raman-THz response of 1-Allyl-3-methylimidazolium dicyanamide

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)