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WITHDRAWN - 2-Dimensional Vibrational Sum Frequency Generation Spectroscopy of Organic Monolayers

Monday 3 June 2019 14:00 (15 minutes)

Vibrational sum frequency generation (SFG) spectroscopy is a non-destructive interface-specific non-linear optical spectroscopy that delivers information about the structure and composition of interface environments. Coupling and energy flow through vibrational modes at surfaces are important and of interest in areas ranging from membrane biophysics and biochemistry, to materials science, electrochemistry, and heterogeneous catalysis. SFG spectroscopy leverages the non-centrosymmetric environment of the interface to mix two incident electric fields and provide a coherent scattered field at their sum frequency. For fields resonant with the vibrational modes of interface molecules, the sum frequency signal provides valuable information about local interface structure and energetics. 2-Dimensional SFG (2D-SFG) spectroscopy extends these studies to provide direct information about molecular interactions and dynamics. Here, we describe the construction of a novel collinear 2D-SFG spectrometer and its application to the vibrational spectroscopy of highly ordered organic monolayers at the CaF2/Air interface. A three pulse IR sequence generated from a dual output Ti:sapphire-based optical parametric amplifier (OPA) and birefringent wedge pair create a highly phase stable train of IR pulses with controllable time delays. This 100 fs IR pulse sequence is combined with a narrow bandwidth picosecond up-converting 800 nm pulse to yield a broadband SFG spectrum. Spectra captured as a function of IR pulse sequence delays are then processed to yield 2D-SFG spectra, providing information about the molecular interactions, vibrational couplings and vibrational dynamics of the organic monolayer. Here we report the vibrational couplings and dynamics of the alkyl chain vibrational modes of supported ordered stearate chains in the CH spectral region to reveal their unusual dynamics and strong mode couplings.

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