

Delocalised terahertz phonon-like modes in biomolecules

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Ultrafast optical Kerr-effect experiments have been carried out on proteins, DNA, and other biomolecules demonstrating the presence of coherent delocalised vibrational modes at physiologically relevant conditions as well as dramatically slowed down (20×) solvation-water dynamics.

Underdamped terahertz-frequency delocalised phonon-like modes have long been suggested to play a role in the biological function of proteins and DNA. Such phonon modes involve the collective motion of many atoms and are prerequisite to understanding the molecular nature of macroscopic conformational changes and other related biochemical phenomena. Strong interactions with the surrounding water are expected to cause phonon modes to be heavily damped and localised. We will discuss state-of-the-art femtosecond optical Kerr-effect (OKE) spectroscopy, which is currently the only technique capable of taking low-frequency (GHz to THz) vibrational spectra in solution, applied to aqueous solutions of proteins, DNA, and nucleotides. In a previous publication,¹ we have been able to show the presence of delocalised phonon-like modes in an enzyme and demonstrated their role in ligand binding. We will present our latest results including the demonstration of phonon-like modes associated with the inter-strand hydrogen-bond network in DNA at physiologically relevant conditions. The degree of damping of the delocalised phonon modes in proteins and DNA has likely been optimised by evolution to maximise the efficiency of biologically relevant processes.

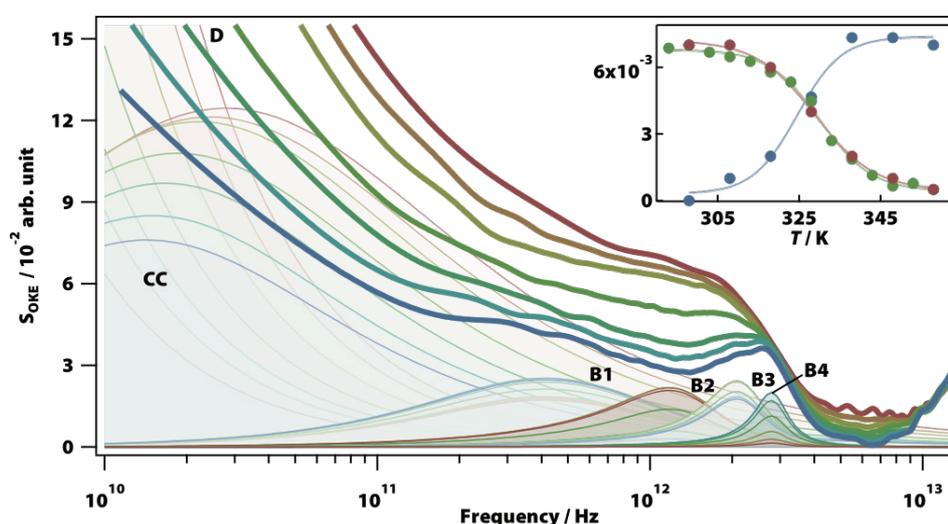


Figure 1. Experimental optical Kerr-effect (OKE) spectra and the component fit-functions used to fit the data for the 20 base-pair AT oligomer. The temperatures run from 298 (blue) to 358 K (red) in steps of 10 K. Each spectrum has been fitted using the combination of a Debye function (D), a Cole-Cole function (CC), and four brownian oscillators (B1 – B4). The features in the figure that change in an unexpected fashion are the bands B2 and B4 (with emphasised colours). The intensity of B2 increases with temperature while the intensity of B4 reduces. The inset shows the intensity of the bands B2 (blue) and B4 (red). They are compared with the scaled results of a circular dichroism experiment (ellipticity at 248 nm, green).

¹ D. A. Turton, H. M. Senn, T. Harwood, A. J. Lapthorn, E. M. Ellis, and K. Wynne, Nat Commun 5, 3999 (2014).