Exploring Relaxation Processes in Components of DNA with UV Nonlinear Spectroscopy

ANDREW MORAN, BRANTLEY WEST, JORDAN WOMICK, University of North Carolina — Underlying photoinduced relaxation in DNA is a complex world of solute-solvent interactions and fluctuations in the geometries of macromolecules. Electronic excitations are rapidly deactivated by nuclear motions through conical intersections, thereby suppressing the formation of lesions (e.g., thymine dimers) known to inhibit cellular function. At the instant following internal conversion, the bases are left in “hot” quantum states, wherein a subset of vibrational modes possess a highly non-equilibrium distribution of excitation quanta. The transfer of this energy to the surrounding also involves intriguing fundamental physics. We examine these processes in small components of DNA by conducting femtosecond laser spectroscopies at cryogenic temperatures. Our experiments utilize several recent advances in nonlinear optics. Parametric processes in argon gas are used to generate 25fs pulse durations at 265nm. These short pulses are employed in a variety of measurements (e.g., transient grating, 2D photon echo, fluorescence down-conversion) with the goal of understanding relaxation mechanisms. Our data suggest that excited state deactivation in DNA is quite sensitive to the exchange of vibrational energy between the bases and segments of the backbone.

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