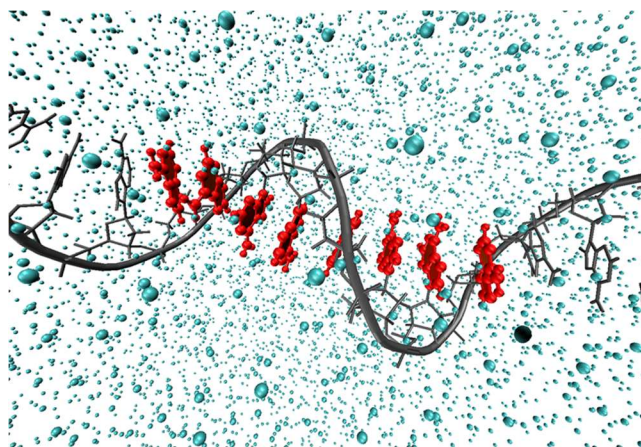


How delocalized is photoexcitation in DNA?

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The question of whether the UV absorption spectrum of DNA is dominated by monomer-like excitations or by collective excitations has intrigued researchers for over 50 years. Here, we report the largest and most accurate multiscale calculations to-date to provide a clear-cut answer to this question. A comprehensive analysis of the electronic properties of the UV spectrum of single- and double-stranded DNA chains, including excitations over a large number of nucleobases, environmental effects, conformational sampling by molecular dynamics and a meaningful interpretation of the electronic structure by quantitative wavefunction analysis is employed to calculate the initial extent of the exciton over the DNA strand. This is a question of fundamental significance, as it decides whether the early electronically excited-state dynamics of DNA is dominated by delocalized excitons undergoing intraband scattering and energy transfer, by dimer excitations promoting excimer formation and dimerization, or by monomer-like processes.



A solvated single stranded polyadenine highlighting 8 nucleobases