Visible Optical Resonances in Electrically Doped DNA

– SUPPORTING INFORMATION –

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S1. CONVERGENCE WITH THE BASIS SET USED FOR INDIVIDUAL NUCLEOBASES

The polariton wave function (PWF) formalism relies on the induced charge distributions of excited states of individual nucleobases (NBs) to compute the optical response of DNA chains (see main text). The accuracy of the method depends on the precision of the level of theory used to obtain those charge distributions. In Figure S1, we compare the optical spectra obtained with three different basis sets of increasing complexity (6-31G, 6-311++G, and 6-311+G(d,p)). In the main text, we select the intermediate level (6-311++G) as a compromise between physical accuracy and computational efficiency.

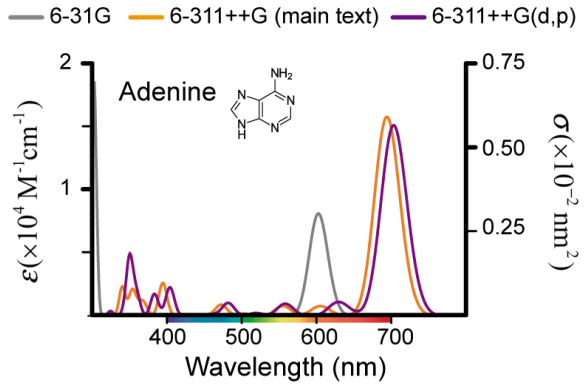

FIG. S1: Spectra for negatively charged adenine calculated from Gaussian 16 at the B3LYP level of theory with three different basis sets (6-31G, 6-311++G, and 6-311+G(d,p)) of increasing complexity and accuracy (see also Figure 1b in the main text, where we use 6-311++G).

S2. COMPUTATIONAL DEMAND OF THE PWF METHOD

The PWF formalism comprises two steps: (1) computation of the excited states of each individual NB and (2) calculation of the coupling coefficients $M_{ljl'j'}$ describing the interaction of the excited states j and j' in the NBs l and l'. In contrast, Quantum Chemistry methods typically deal with the system as a whole, in particular during the calculation of excited states, thus requiring more resources to simulate any physically relevant quantity. Table S2 shows an estimate of CPU time required by the Gaussian 16 suite and the PWF formalism for systems comprising 1-3 NBs, showing that the PWF formalism reduces the computational demand.

	Gaussian			PWF
	6-31G	6-311++G	6-311++G(d,p)	6-311++G
1 NB	10	26	30	26
2 NBs	45	117	135	52
3 NBs	75	195	225	78

TABLE S1: Computation time (minutes) used to evaluate systems consisting of 1-3 NBs using different levels of theory with Gaussian 16 and the PWF formalism.