

DNA-Mediated Charge Transport Requires Conformational Motion of the DNA Bases: Elimination of Charge Transport in Rigid Glasses at 77 K

Melanie A. O'Neill, and Jacqueline K. Barton

*Division of Chemistry and Chemical Engineering, California Institute of Technology,
Pasadena, California 91125*

Supporting Information

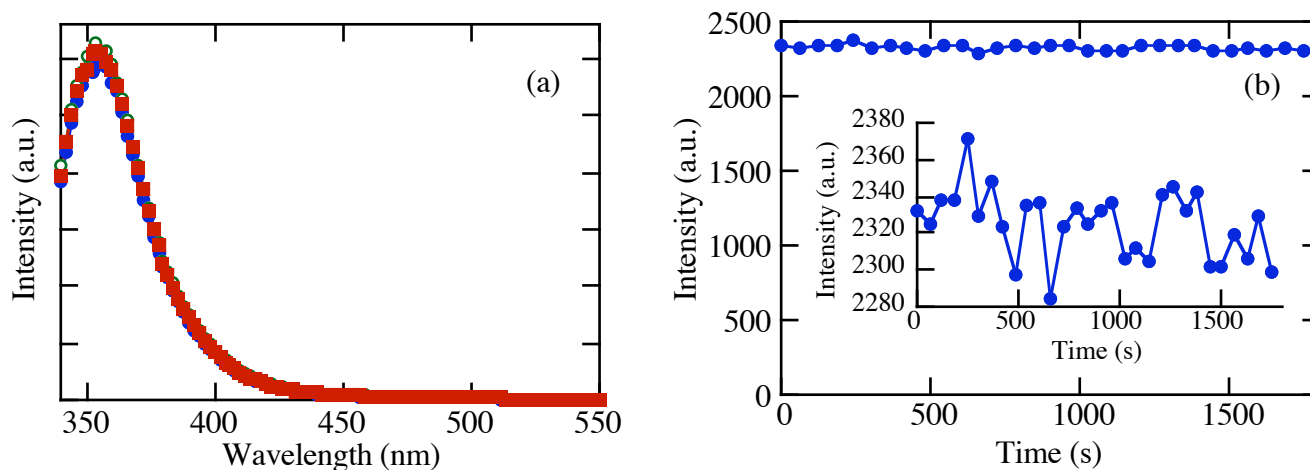


Figure S-1. Reproducibility (a) and stability (b) of 10 M aqueous LiCl glasses, as monitored by the fluorescence emission of photoexcited 2-aminopurine (Ap^* , $25 \mu\text{M}$, $\lambda_{\text{ex}} = 325 \text{ nm}$). Shown in (a) are the emission spectra of obtained in three independently prepared glassy matrices at 77 K. These emission spectra are very stable over periods of at least several hours. Shown in (b) is the variation in emission intensity Ap^* (monitored at $\lambda_{\text{max}} = 360 \text{ nm}$) in a LiCl glass at 77 K as a function of time (total time of 30 minutes). The expanded plot (inset) reveals intensity fluctuation of $< 5 \%$. Similar results were observed for Ap -DNA samples.

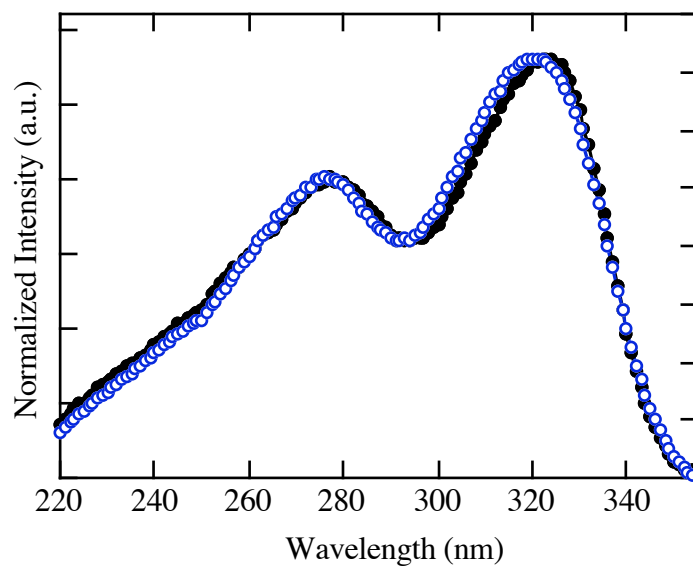


Figure S-2. Fluorescence excitation spectra ($\lambda_{em} = 370$ nm) of 2.5 μ M ApA4I in 100 mM sodium phosphate pH 7 (closed circles) and 10 M LiCl pH 7 (open circles) at ambient temperature (~ 298 K). The lower duplex concentration is necessary due to the strong absorption of the natural DNA bases at $\lambda < 300$ nm.

Table S-1. A comparison of the photophysical properties of Ap-DNA assemblies, and yield of charge transport between Ap* and G in 10 M LiCl pH 7 versus 100 mM sodium phosphate pH 7 (NaP) at ambient temperature (~ 298 K).

Sample ^a	$\Phi_{\text{rel}} Y=I^b$		Anisotropy $Y=I^c$		Fq (298 K) ^d	
	NaP	LiCl	NaP	LiCl	NaP	LiCl
ApY	0.1	0.1	0.25	0.26	0.87	0.68
ApAY	0.06	0.1	0.23	0.25	0.47	0.47
ApA2Y	0.04	0.06	0.25	0.24	0.20	0.23
ApA3Y	0.03	0.05	0.26	0.23	0.24	0.18
ApA4Y	0.04	0.07	0.23	0.23	0.14	0.14

^a25 μM Ap in 35-mer DNA duplexes, 5'-GAT TAT AGA CAT ATT IAp(A)_n YIT ATT AAG TAC ATT AC-3', Y=I,G, in 10 M LiCl, pH 7 or 100 mM sodium phosphate pH 7 at ~ 298 K.

^bFluorescence yields relative to Ap in respective solution ($\lambda_{\text{ex}} = 325 \text{ nm}$).

^cMonitored at 370 nm.

^dEvaluated as $Fq = 1 - \Phi_G / \Phi_I$, relative uncertainties $\pm 10 \%$.