On the nature of DNA Self-Assembled Monolayers on Au: measuring surface heterogeneity with electrochemical in situ fluorescence microscopy

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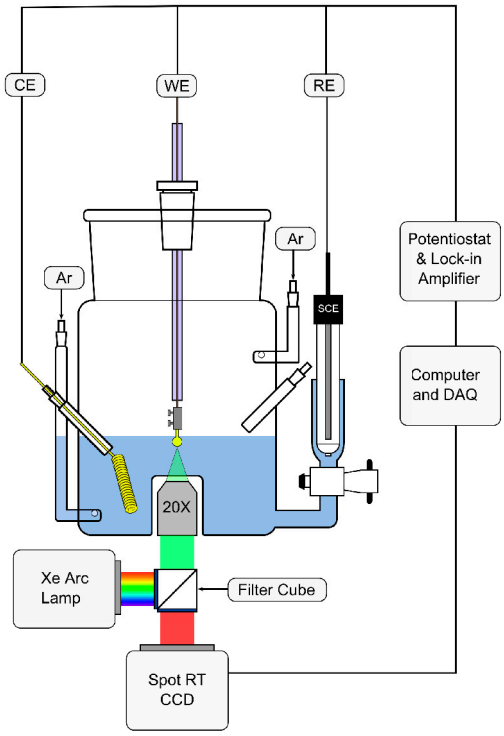
Supporting Information

S1 Schematic of the Experimental Setup

The fluorescence - electrochemistry measurements were performed in a specially designed spectro-electrochemical cell that was created in-house and used a cover-glass window for improved image quality.



The spectro-electrochemical cell is depicted with key parts of the epi-fluorescence microscope (20× objective, filter cube, xenon arc lamp, and Spot RT CCD) and control components (potentiostat with lock-in amplifier and computer with data acquisition board). The cell itself contains a sample as working electrode (WE) - a thiolate-DNA-modified gold bead - suspended above a 0.17mm thick coverglass window through which the objective can be focused. The counter electrode (CE) is a coil of gold wire immersed in the electrolyte solution. A saturated calomel electrode (SCE) functions as the reference electrode (RE), which is connected to the bulk solution via a Teflon stopcock salt bridge. The cell contains two ports for argon gas; one for bubbling argon (Ar) to deaerate the solution and another to maintain a blanket of argon during experimentation.

S2 Fluorescence images of the reductive desorption process

The changes in fluorescence observed during potential excursions are most easily viewed through creation of movies that start at E_{base} (0 mV) and sequentially step to negative potentials (in 25 mV increments). Only the images recorded at the E_{step} potentials are presented. Four movies representing the measurements taken from the ssDNA/MCH (Figure 2 in manuscript), dsDNA/MCH (Figure 3 in manuscript) , MCH/ssDNA (Figure 6 in manuscript), and MCH/dsDNA (Figure 8 in manuscript) surfaces are provided:

ssDNA/MCH (Figure 2 in manuscript): ssDNA_MCH_Figure2_movie.avi

dsDNA/MCH (Figure 3 in manuscript): dsDNA_MCH_Figure3_movie.avi

MCH/ssDNA (Figure 6 in manuscript): MCH_ssDNA_Figure6_movie.avi

MCH/dsDNA (Figure 8 in manuscript): MCH_dsDNA_Figure8_movie.avi

S3 Fluorescence images of particle movement across the surface during the reductive desorption process

The observation of the movement of small particles across the surface during negative potential steps (Figure 7 in manuscript) is presented as a movie: MCH_ssDNA_Figure7_movie.avi Four colored tracks are shown superimposed onto a series of fluorescence images that have been filtered to enhance the spots. The movie starts at -650mV and is complete at -875mV. All E_{step} and E_{base} images are included in the movie which results in a step like increase-decrease in the background.

S4 Fluorescence intensity modulation for various regions of MCH/ssDNA and MCH/dsDNA surface

The modulation of fluorescence during potential steps from E_{base} of 0mV to a maximum E_{step} of -400mV is shown for the both the MCH/ssDNA and MCH/dsDNA modified surfaces for various regions on the surface supplementing Figure 5 in the manuscript.

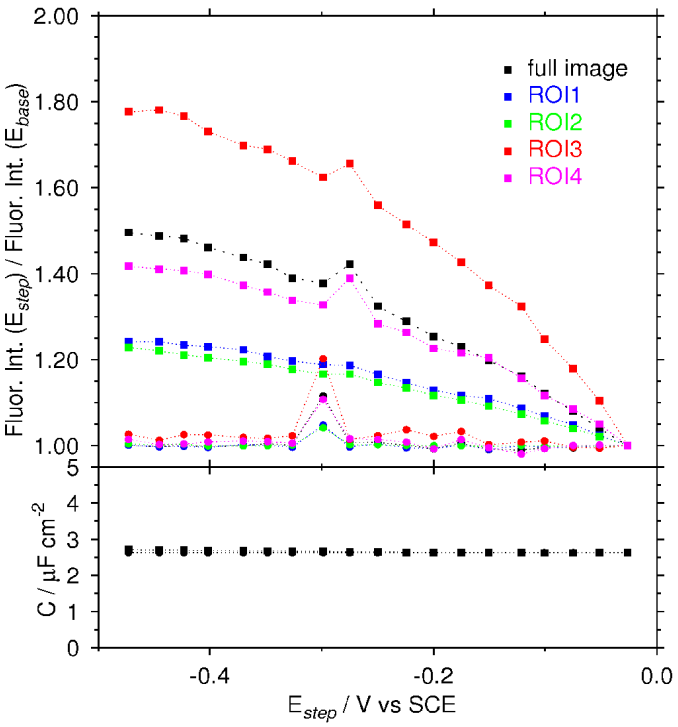


Figure S1: Relative changes in fluorescence for ssDNA/MCH coated gold surface measured for the ROIs shown in Figure 6 in the manuscript

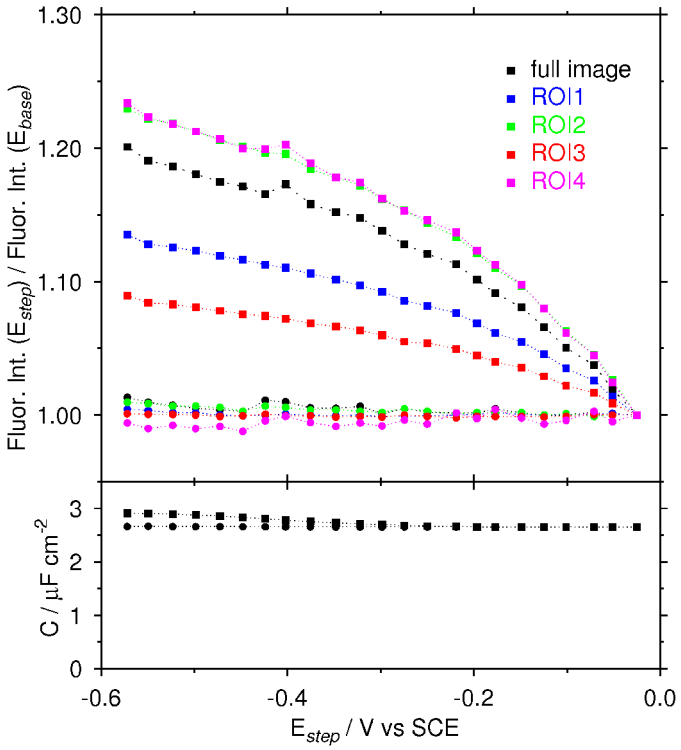


Figure S2: Relative changes in fluorescence for dsDNA/MCH coated gold surface measured for the ROIs shown in Figure 8 in the manuscript