

## Supporting Information:

# Direct Imaging and chemical analysis of Unstained DNA Origami performed with a Transmission Electron Microscope

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## Experimental

**DNA origami preparation** - The synthesis of the DNA origami was previously described in details. A molar ratio of 1:10 between viral, ssDNA M13 and short unmodified staple strands was used. Origami triangles were assembled in 1x TAE-Mg<sup>2+</sup> buffer (40 mM Tris, 20 mM acetic acid, 2 mM EDTA, 12.5 mM Mg(OAc)<sub>2</sub>, pH 8.0) by cooling slowly from 90°C to room temperature.

**TEM experiments** - 3 nm thick amorphous Carbon films and 5 nm thick non-porous amorphous silicon membranes were used as a substrate for TEM observations. These ultra-thin films are commercial TEM support from Ted Pella Inc. and SiMPore Inc. respectively. Several microscopes were used for these TEM investigations: EELS spectra were acquired with a VG 501 STEM operating at 100kV and equipped together with a cold field emission gun and an aberration corrector. Phosphorous maps obtained on the aggregated DNA origami were acquired with TEAM 0.5 microscope operating at 80kV and equipped together with a monochromator and a Gatan GIF Tridiem. P-maps were extracted with the three window technique (peak edge at 132 eV and 10 eV slit width). Triangular-shaped DNA origami on amorphous silicon were analyzed (bright field, EFTEM imaging) with a monochromated Zeiss Libra 200FE TEM equipped with an in-column omega filter. We used 10 eV slit to acquire EFTEM images. EFTEM images at the carbon plasmon peak present an optimal contrast at 24eV. We note that the broad shape of the plasmon carbon peak allows a good observation of DNA origami in an energy range from 20 to 100 eV.

**AFM experiments** - Droplets of DNA origami solution were deposited on freshly cleaved mica and HOPG substrate for 5 minutes. After wash once with distilled water,

samples were dried in air. AFM measurement was carried out with multimode scanner (Veeco V) in tapping mode in air. The AFM tip is NSC19 purchased from MikroMasch with resonant frequency  $\sim 80$  kHz and force constant  $\sim 0.6$  N/m.