# **Supporting Material**



Figure S-1. TEM image of Poly-L-Lysine treated graphene



Figure S-2. AFM images of the small rectangle origami. The scale bar is 50nm.



**Figure S-3.** (A) Additional TEM images of the small rectangle origami (B-C) and DNA tetrahedron. Scal bar is 50 nm in all images.



Figure S-4. Histogram of the side lengths of the DNA structures. A) Small Origami, B) tetrahedron

#### S-5. Tomography of tetrahedron

The paper of He et al. [1] not only introduced the DNA tetrahedra, but also demonstrated the use of TEM tomography for analyzing cryo-TEM images of DNA structures. For our work standard tomography is not suitable because the tetrahedra tend to sit with one face nearly flush with the graphene and only rarely are other orientations seen (see Fig. S4-B). Nevertheless, by taking the measuremenets of the observed triangles and assuming these to be projections of regular tetrahedra, one can perform a 2D version of TEM tomography. A histogram showing a distribution of the tetrahedron side lengths so determined is plotted in Fig. S5. The mean is 17.4 nm (with a standard deviation of 1.5 nm), which is in reasonable agreement with the expected hydrated size of about 16.2 nm.



**Fig. S5**. Histogram of the lengths of the sides of the tetrahedra as inferred from the TEM images in Fig. S4-B using a 2D version of TEM tomography.

## S-6. AFM imaging

For AFM sample preparation, freshly cleaved mica was treated with 40  $\mu$ L of 2 mM NiCl solution for 5 minutes. The mica was rinsed in water and allowed to dry. The origami rectangle was imaged under aqueous conditions, with DNA(~10  $\mu$ L of 10nM) in a TAE Mg+ containing buffer placed on the sample and allowed to assemble. Buffer(~30  $\mu$ L) was then added and the sample was imaged in tapping mode. AFM measurements were performed on a Veeco Multimode with the Nanoscope IIa controller (Veeco Instruments). Imaging was done using ultra sharp diamond-like carbon tips (MikroMasch) and the images were analyzed using WSxM software.[2]

## S-7. DNA sequences

Small Rectangle, replicated portion from M13:

5′-

CGAAACTCAGTGTTACGGTACATGGGTTCCTATTGGGCTTGCTATCCCTGAAAATGAGGGTGGTGGTGGCTCTGAGGGTG GCGGTTCTGAGGGTGGCGGTTCTGAGGGTGGCGGTACTAAACCTCCTGAGTACGGTGATACACCTATTCCGGGCTAT ACTTATATCAACCCTCTCGACGGCACTTATCCGCCTGGTACTGAGCAAAACCCCGGCTA-3'

- 1: CACCGTACTCAGGAGGCCCGGAAT
- 2: AGGTGTATGGATAAGT
- 3: GCCGTCGATTGCTCAGTACCAGGC
- 4: TCAATAAGTATAGTTTAGTAC
- 5: TCGTAGCGGGGTTGAGGGTTG
- 6: CGCCACCCTCAGACACCACCC
- 7: ATTTTTCAGGGATACTGAGTT
- 8: CTCAGAGCACCGCCACCCTCAGAA
- 9: CAATAGGACCGCCACC
- 10: ACCCATGTACCGTAACAGCAAGCC

#### References

[1] Y. He, T. Ye, M. Su, C. Zhang, A. E. Ribbe, W. Jiang and C. D. Mao, *Nature*, 2008, 452, 198-U141.

[2] I. Horcas, R. Fernandez, J. M. Gomez-Rodriguez, J. Colchero, J. Gomez-Herrero and A. M. Baro, *Review of Scientific Instruments*, 2007, **78**.