Supporting information

Materials:

All chemical synthesized DNA strands were purchased from Integrated DNA Technologies, Inc. (www. Idtdna.com). The unmodified helper strands ordered in a 96-well plate format, suspended in ultrapure water without purification. All modified strands were ordered from IDT with HPLC purification. Colloidal solution of 5 nm AuNPs was purchased from Ted Pella Inc. 100kDa MWCO centrifuge filters purchased from Pall, Inc.

Experimental Methods:

Self-Assembly of rectangle DNA origami template

DNA origami template was formed according to Rohtermund (reference S1). M13 viral DNA and all the staple strands were mixed together at a 1:10 ratio, in a 1 x TAE buffer solution containing 40 mM Tris-HCl, 20 mM acetic acid, 2 mM EDTA and 12.5 mM magnesium acetate. The DNA origami solution was slow cooled from 90 °C to 16 °C with PCR over 1.5h. The final concentration of M13mp18 DNA genome in the solution was 10 nM. DNA origami was then purified to remove the excess DNA helper strands using 100kDa MWCO centrifuge filters. The formation of dimer was annealed from 45 degree to room temperature in a 2-liter water bath stored in a styrofoam box over the course of 18 hr.

Preparation of DNA coated 5 nm AuNPs

The detailed procedure can be found in reference S2 with slight modification. Briefly, the 5 nm AuNPs were stabilized with adsorption of Bis(p-sulfonatophenyl)phenylphosphine dihydrate dipotassium (BSPP), then sodium chloride was added slowly to this mixture while stirring until the color changed from deep burgundy to light purple. The resulting mixture was centrifuged and supernatant was carefully removed. The concentrated AuNPs were resuspended in BSPP solution and concentration was estimated at ~520 nm.

The thiol functionalized single stranded oligonucleotides (5'-TTTTTTTTTTTTTTT-S) were first reduced by TCEP in water, followed by purification using G25 column (GE Healthcare) to remove the small molecules. Then thiol modified oligonucleotides mixed with phosphinatd AuNPs at 100:1 ratio in 0.5 x TBE buffer containing 50 mM NaCl for two days at room temperature. AuNP-DNA conjugates were washed with 0.5 x TBE buffer with 100kDa MWCO centrifuge filters to get rid of the extra oligonucleotides. The concentration of conjugates was estimated from the optical absorbance at ~520 nm.

Characterization of AuNP -DNA origami structure by atomic force microscopy

5 µl of sample solution were spotted onto freshly cleaved muscovite mica (Ted Pella inc) and absorbed for ~3 min. To remove buffer salts, 20-30 µl of doubly distilled H_2O was placed on the mica, the drop was wicked off and the sample was dried with compressed air. Atomic force imaging was done utilizing Nanoscope IV (Digital Instruments) tapping in air, with ultrasharp 14 series (NSC 14) tips purchased from MikroMasch (www. SPMTIPS.com).

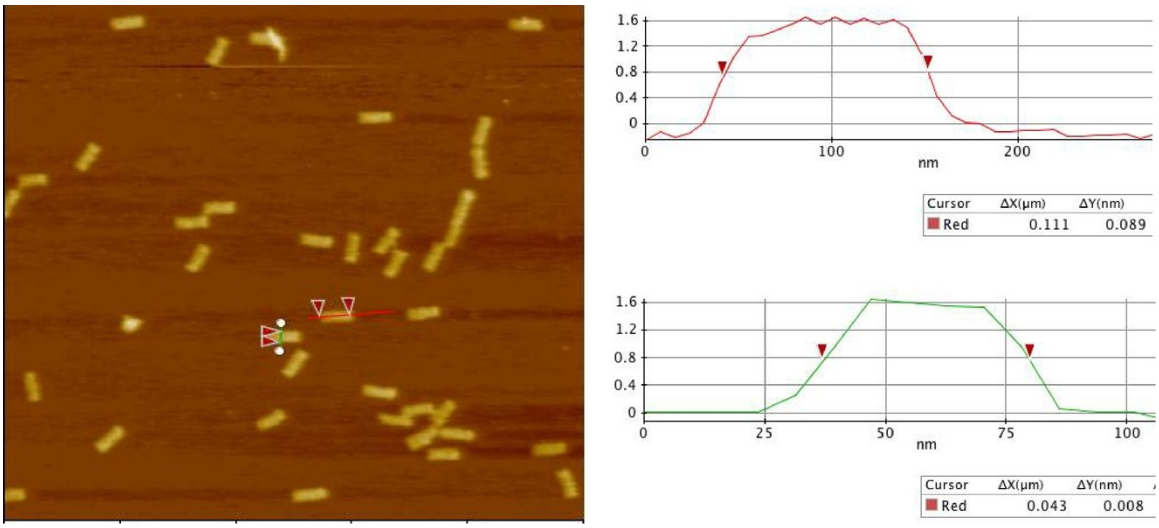


Figure S1. AFM image of DNA origami rectangle monomer and its size analysis. The sample was deposited on the mica surface and scanned in ambient conditions.

Calculation of the yield of DNA origami dimer by atomic force microscopy

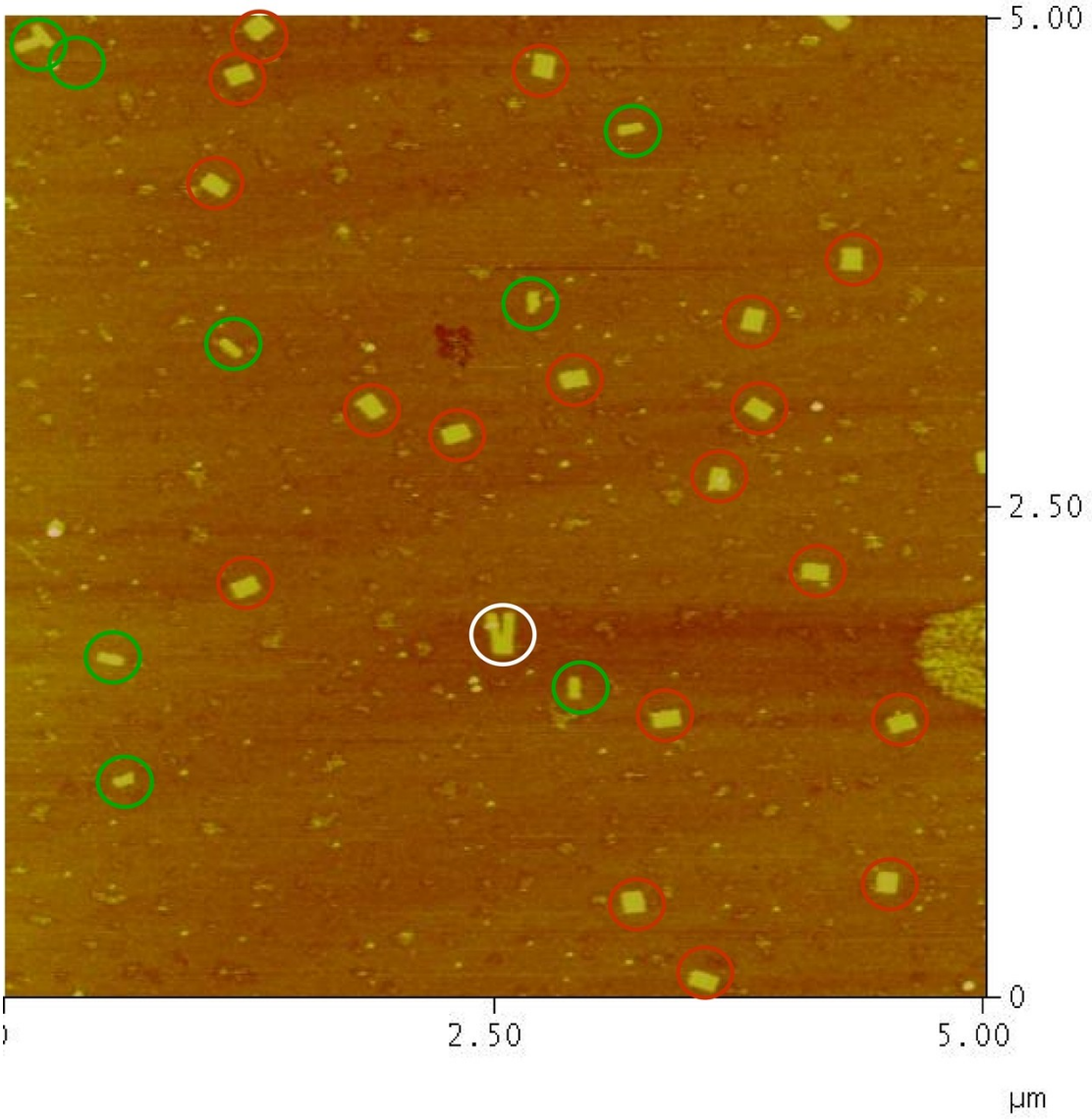


Figure S2. An example of AFM image used to calculate the yield of DNA origami dimer formed with 1X TAE buffer containing 30.0 mM Mg^{2+}. The red circles highlight fully formed dimers, the green circles highlight monomers, and the white circle represents non-counted DNA origami. The final yield is the average yield of each image.

Equation used to calculate the yield of dimer:

$$\%\text{dimer} = \frac{number\ \ of\ \ \text{dim}er\ \times\ 2}{number\ \ of\ \ \text{dim}er\ \times\ 2 +\ \ number\ \ \ of\ \ monomer} \times 100$$

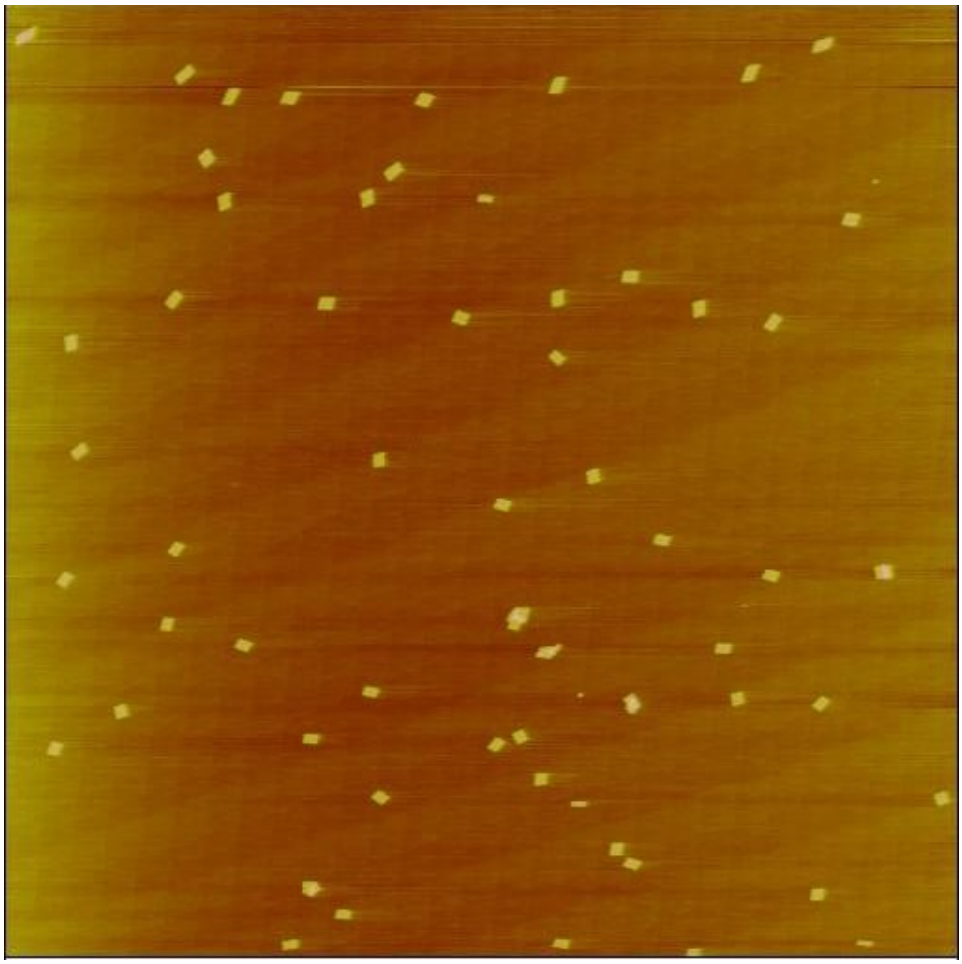


Figure S3. AFM image of DNA origami rectangle dimer formed with 1X TAE buffer containing 60.0 mM Mg^{2+}. The size of image is 8 μm.

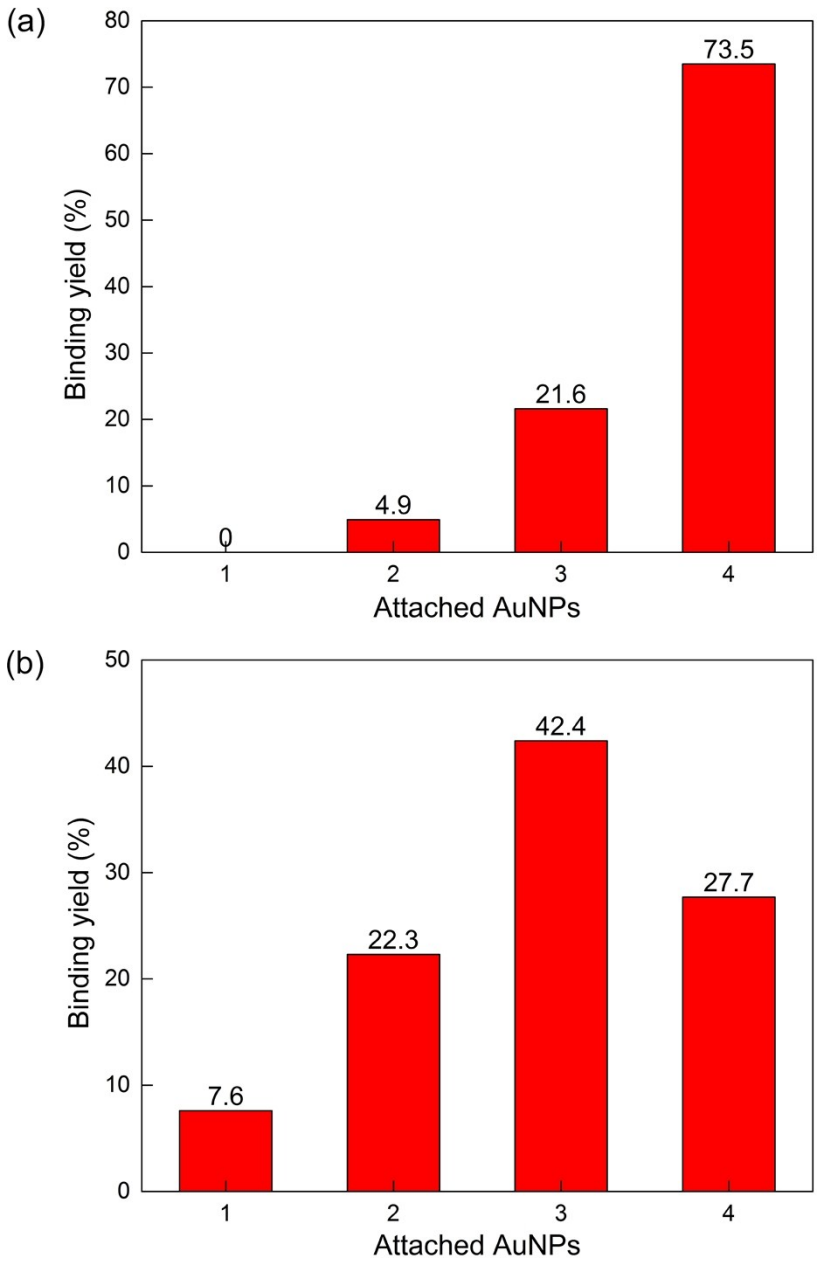


Figure S4. Histograms and calculated binding efficiency of AuNPs on the single-side DNA origami dimer template. (a) The attachment of AuNPs on the four center sites. The percent of 1 particles is 0%, 2 particles is 4.9%, 3 particles is 21.6%, 4 particles is 73.5%, (N = 102). (b) The attachment of AuNPs on the four corner sites on another side. The percent of 1 particle is 7.6%, 2 particles is 22.3%, 3 particles is 42.4%, 4 particles is 27.7%, (N = 184).

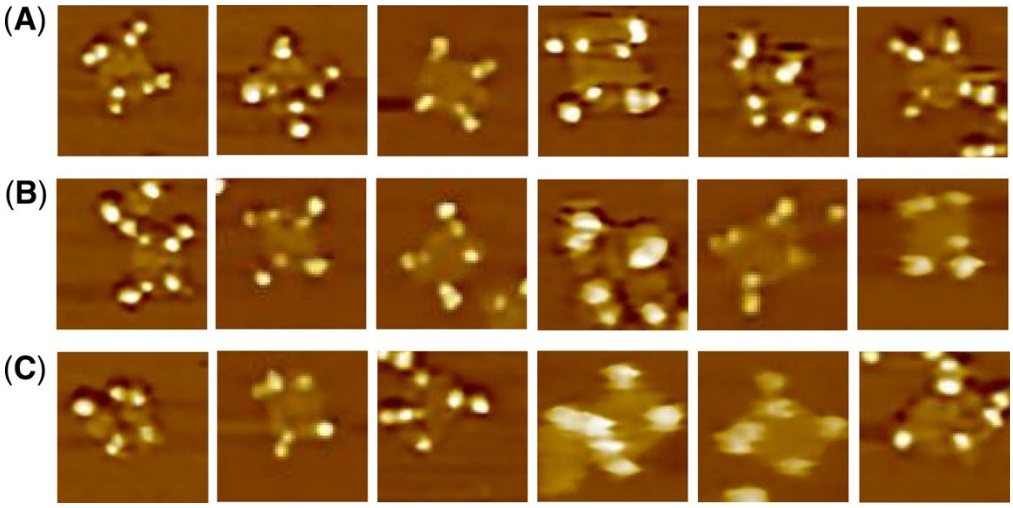
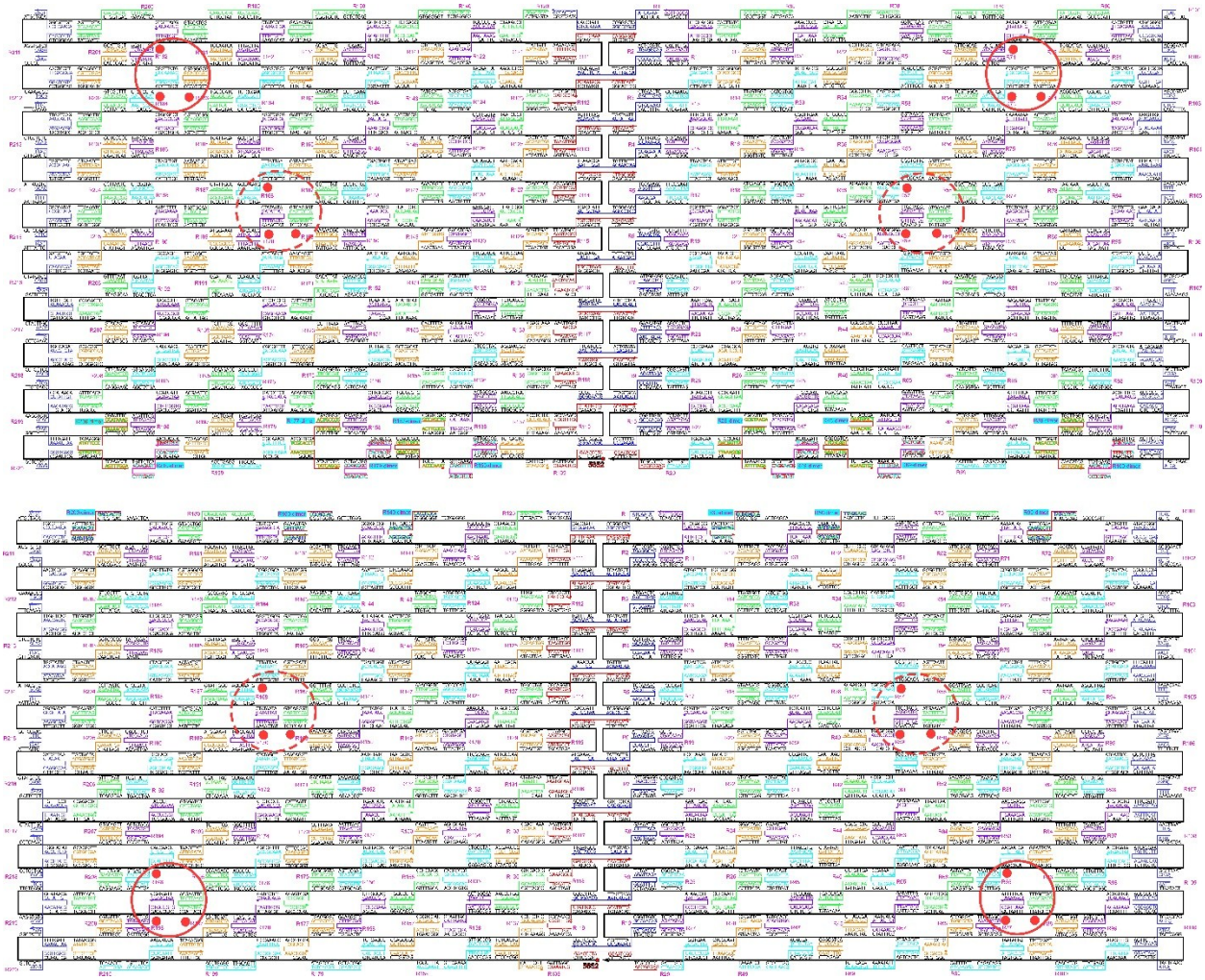


Figure S5. Zoomed AFM images of DNA origami dimer attached with varied AuNPs. A) eight AuNPs B) seven AuNPs C) six AuNPs.

The design and sequences of DNA origami:



The design of DNA rectangular origami and the unmodified staple strands are same as reference S2. The modified sequences starting from 5' are listed as below:

D209-dimer: GCATAAAGGGCAAAATCCCTTATAGTTGTTCCAGTTTGGA
D210-dimer: TGAGTTTCACAAGAGTCCACTATTTGGTGGTT
D177-dimer: ACTGCCCGGCAAGCGGTCCACGCTAAACCGTCTATCAGGG
D179-dimer: CTCAGAGCCGATGGCCCACTACGTCCCTGAGA
D157-dimer: GCCAGCTGGCCCTTCACCGCCTGGGAACCATCACCCAAAT
D159-dimer: CCCTCAGACAAGTTTTTTGGGGTCCGGGCAAT
D28-dimer: GCGTAAGATTATTTACATTGGCAGTTAAAGGGATTTTAGA
D49-dimer: TGCTCAGTCAGGAACGGTACGCCAAATCGTCT
D48-dimer: GAATGGCTCTACATTTTGACGCTCGAATCTTGAGAAGTGT
D69-dimer: CCTCAAGATTTTATAATCAGTGAGACGCTCAT
D88-dimer: ACGAACCACTGGTAATATCCAGAAAATTAACCGTTGTAGC

D100-dimer: AATGCCCCAATACTTCTTTGATTACAAACTAT
D200-dimer: GTCACCAGTACAAACTATAGTTAG
D160-dimer: CACCACCCTCATTTTCTGGGATTT
D140-dimer: ACCGCCACCCTCAGAAAGCGGAGT
D30-dimer: ACCAGGCGGATAAGTGTGATATTC
D50-dimer: GAAGGATTAGGATTAGGCCAGAAT
D90-dimer: CTGCCTATTTCGGAACGATACAGG

D57-center:
AAAAAAAAAAAAAAAAAAAAAAAAAGTTGCTTGTTTATCACGTAGGAATCATTACCACGCTAAC
D59-center:
AAAAAAAAAAAAAAAAAAAAAAAAAGTTGCTTAACTTTTTTAAAGCCAACGCTCAAACGCGCCT
D60-center:
AAAAAAAAAAAAAAAAAAAAAAAAAGTTGCTTATTTTCATACAATAGATAAGTCCTCAAATTCT
D168-center:
AAAAAAAAAAAAAAAAAAAAAAAAAGTTGCTTTTTTGATATATTATAGTCAGAAGCTCGTTTAC
D169-center:
AAAAAAAAAAAAAAAAAAAAAAAAAGTTGCTTACCCTGACAGAGGTCATTTTTGCGGGTCAATA
D170-center:
AAAAAAAAAAAAAAAAAAAAAAAAAGTTGCTTCAAAAACAAGCTATATTTTCATTTATTGCTCC

D71-corner:
AAAAAAAAAAAAAAAGTTGCTTAACCAGAGATACATGGCTTTTGATCTATTATT
D73-corner:
AAAAAAAAAAAAAAAGTTGCTTTCACAATCAATCAGTAGCGACAGAATCACCGG
D73-corner:
AAAAAAAAAAAAAAAGTTGCTT TCACAATCAATCAGTAGCGACAGAATCACCGG
D182-corner:
AAAAAAAAAAAAAAAGTTGCTTATATATTCTCTAAAGTTTTGTCGTCGTAACAC
D183-corner:
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D184-corner:
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D85-corner:
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D87-corner:
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D88-corner:
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D196-corner:
AAAAAAAAAAAAAAAGTTGCTTTCATAGCTCACTCCAGCCAGCTTTCGTTGGTG
D197-corner:
AAAAAAAAAAAAAAAGTTGCTT GTGAGCTACGAAAATCCTGTTTGAAAAGAACG
D198-corner:
AAAAAAAAAAAAAAAGTTGCTTCCGAAATCTGTAAAGCCTGGGGTGAATCATGG