

Supporting information for

**IR Emitting Quantum Dots: DNA conjugation and DNA origami
directed Self-assembly**

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Experimental Methods

Chemicals:

Cadmium nitrate tetrahydrate ($\text{Cd}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$, 99.8%), Lead nitrate ($\text{Pb}(\text{NO}_3)_2$, $\geq 99\%$) Tellurium (Te, powder, -200 mesh, $\geq 99\%$, powder), Sodium borohydride (NaBH_4 , powder, $\geq 99\%$), 3-Mercaptopropionic acid (MPA, $\text{HSCH}_2\text{CH}_2\text{CO}_2\text{H}$, $\geq 99\%$), Glutathione (GSH, $\geq 99\%$), were purchased from Sigma-Aldrich and used without further purification. M13mp18 single stranded DNA was purchased from New England Biolabs and was used without further treatment. All unmodified helper strands were purchased from Integrated DNA Technologies, Inc. (IDT, www.idtdna.com) in 96-well plate format, suspended in nanopure water (H_2O , with resistivity up to $18.2 \text{ M}\Omega \cdot \text{cm}$) and used without further purification. All modified helper strands as QD capturing strands were purchased from IDT and purified by denaturing PAGE gel electrophoresis. Phosphorothioate backbone modified ps-po-chimeric ssDNA strands for capping QDs were purchased from IDT and used after denaturing PAGE purification.

DNA conjugation during the synthesis of the CdPbTe alloyed QDs:

First, a NaHTe solution was freshly prepared by dissolving Te powder (1 mmol) with NaBH₄ (4 mmol) in 2 mL degassed water in a thick walled glass tube. A needle was inserted into the capped tube to release the pressure of the evolved gas, and the solution was stirred for a few hours at 4 °C. Meanwhile the precursor solution was prepared by mixing Cd(NO₃)₂·4H₂O (1.75 mM), Pb(NO₃)₂ (0.75 mM) and GSH or MPA (4 mM). Solid DNA (G*G*G*G*G*T₂₀, * represents the phosphothioate linkage) was added to this mixture to get a final concentration of the DNA 0.1 mM. The pH was adjusted to 9 by adding 1 M NaOH drop-wises. Calculated amount of freshly prepared NaHTe solution was micro injected into N₂ saturated precursor solution so that the concentration of NaHTe in the reaction mixture was 0.5 mM. The color of the solution immediately changed from colorless to brownish after the injection. The reaction mixture was incubated at 90°C for 30 minutes and followed by quenching the reaction through quickly cooling down to 0°C in an ice-bath. The resulted QDs particles were purified to remove the excess DNA and the unreacted small ions and molecules by washing with nanopure water 3 times using an Amicon centrifugal filter (30kD MWCO).

Preparation of Triangular DNA Origami:

Triangular DNA Origami was synthesized following the typical procedure described by Rothmund in 2006 (Nature, 2006). The long single stranded M13 scaffold and each of the short staple strands without purification were mixed at molar ratio of 1:5. The binding sites on the origami for each of the QD were generated by modifying 3 adjacent staple strands (arranged in a triangle) at selective positions on the origami by adding 20A nucleotide at the 5' ends, which act as the capturing strands. The ratio between the M13 DNA and the modified staple strands (purified) was 1:20 in the mixture. The assembly was done in 1×TAE-Mg²⁺ buffer (Tris base 40 mM; Acetic Acid 20 mM; EDTA 2 mM; Magnesium Acetate 12.5 mM; pH 8) by cooling down slowly from 90°C to 4°C. In order to get rid of the excess staple strands and the capture strands,

the assembled origami was washed 3 times with $1\times$ TAE-Mg²⁺ buffer in an Amicon filter (100kD MWCO). The purified origami was then mixed with the purified QDs and the mixture was cooled down from 37°C to room temperature over 24 hours.

Structure and Optical Characterization of the QDs:

Steady state fluorescence spectra were collected with a Horiba Nanolog spectrophotometer (Horiba Jovin Yvon) equipped with 450W Xenon lamp and Liquid N₂ cooled DSS-IGA020L InGaAs detector. High-resolution electron microscopy (HRTEM) and energy dispersive X-ray spectroscopy (EDS) were done in a JEOL JEM 2010F electron microscope operating at 200 kV. Carbon coated copper grid (400 mesh, Ted Pella) was used to deposit the sample, which was then washed, stained and air dried before imaging. Powder X-ray diffraction measurements were performed in a PANalytical X'pert Pro Materials research X-ray diffractometer with Cu K α radiation ($\lambda= 1.5418$ Å). Atomic force microscopy (AFM) was performed using a Veeco 8 AFM in tapping in air mode. ICP-MS was performed on a Thermo X-series with Q-ICP-MS with CCT (Collision Cell Technology) instrument.

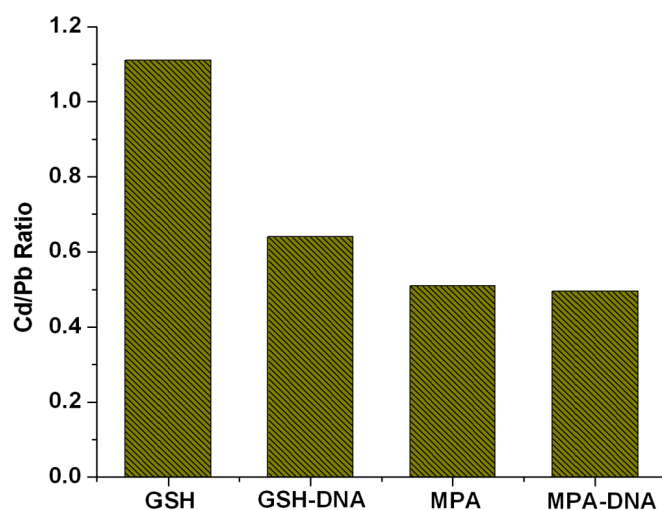


Figure S1. The atomic ratio of Cd/Pb in the QDs synthesized using different capping ligands measured by ICP-MS.

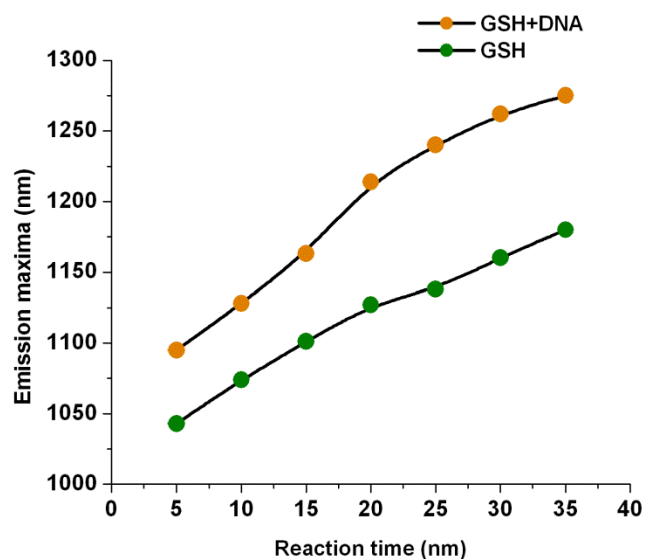


Figure S2. Comparison of the red shift of the QD photoluminescence peak positions monitored during the formation process of the GSH capped QDs in the presence (orange) and absence of DNA (green). The continuous red shift of the QDs obtained may reflect the growing of the size of the nanocrystals. In the presence of both GSH and DNA, the QDs formed at short time already showed a red shift compared to that in the presence of GSH alone. This may reflect a higher incorporation of Pb ions in the nanocrystals.

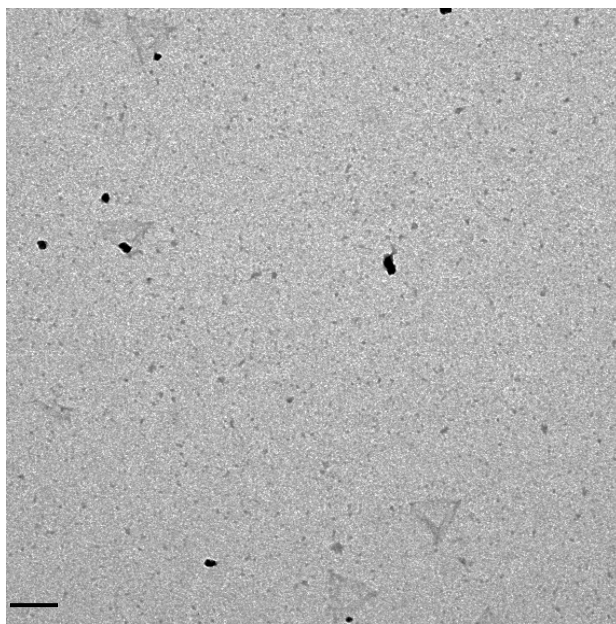


Figure S3. Additional zoom out TEM images of CdPbTe capped with GSH/DNA on DNA origami. (Scale bar 100nm) The DNA origami was quite diluted in this sample.

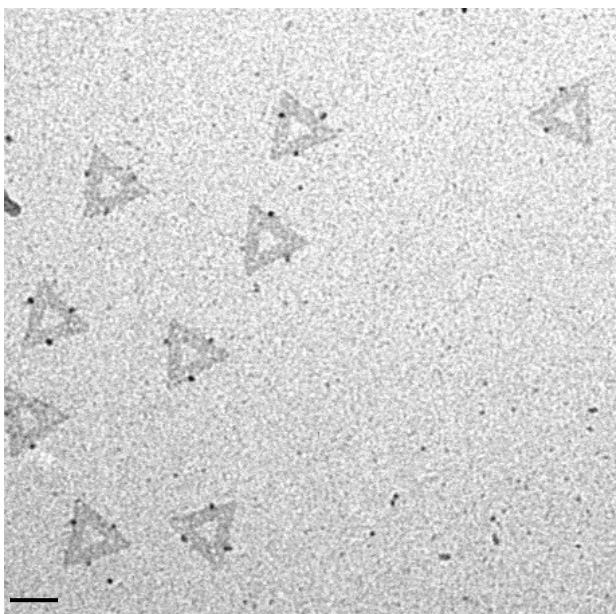


Figure S 4: Additional zoom out TEM images of CdPbTe capped with MPA/DNA on DNA origami. (Scale bar 100nm)

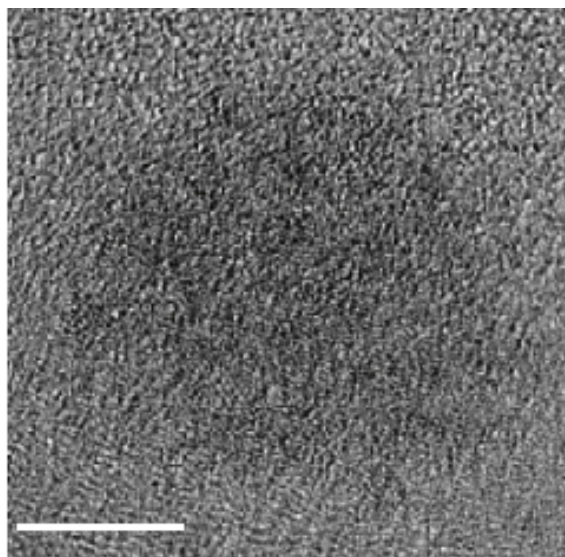


Figure S 5: Zoom in TEM image of $\text{Cd}_x\text{Pb}_{1-x}\text{Te}$ QDs encapsulated with GSH and ps-DNA. Scale bar 5 nm. Multiple 1-2 nm crystal domains can be identified inside the ~ 10 nm diameter particle.

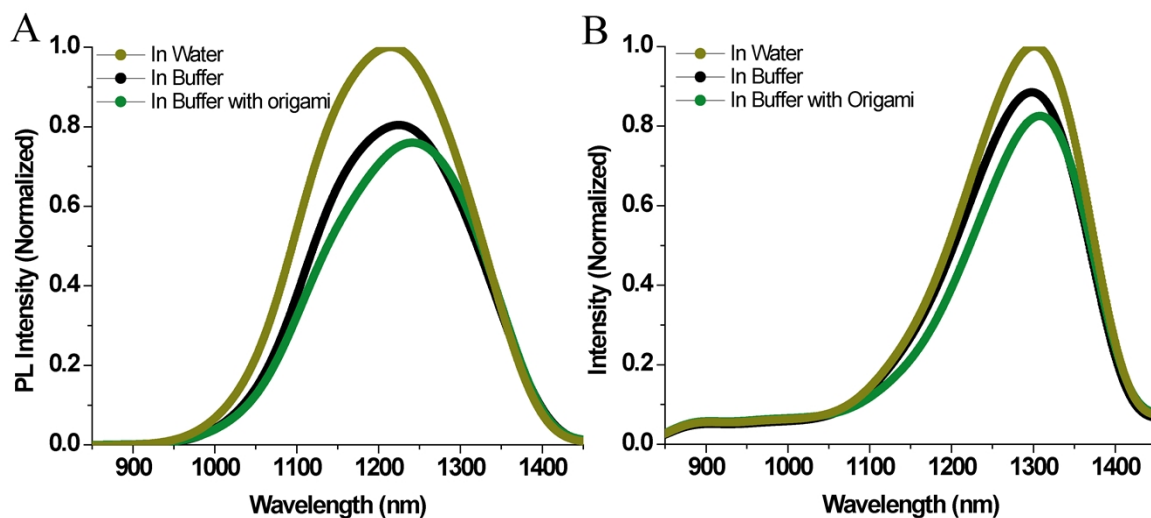


Figure S 6: PL emission spectra of $\text{Cd}_x\text{Pb}_{1-x}\text{Te}$ QDs capped with GSH (A) and MPA (B) in water, incubated in $1\times\text{TAE-Mg}^{2+}$ buffer (containing 12.5 mM MgCl_2) or attached to the DNA origami in the same buffer. In both cases quenching of fluorescence is observed along with slight red shift in the emission maxima.

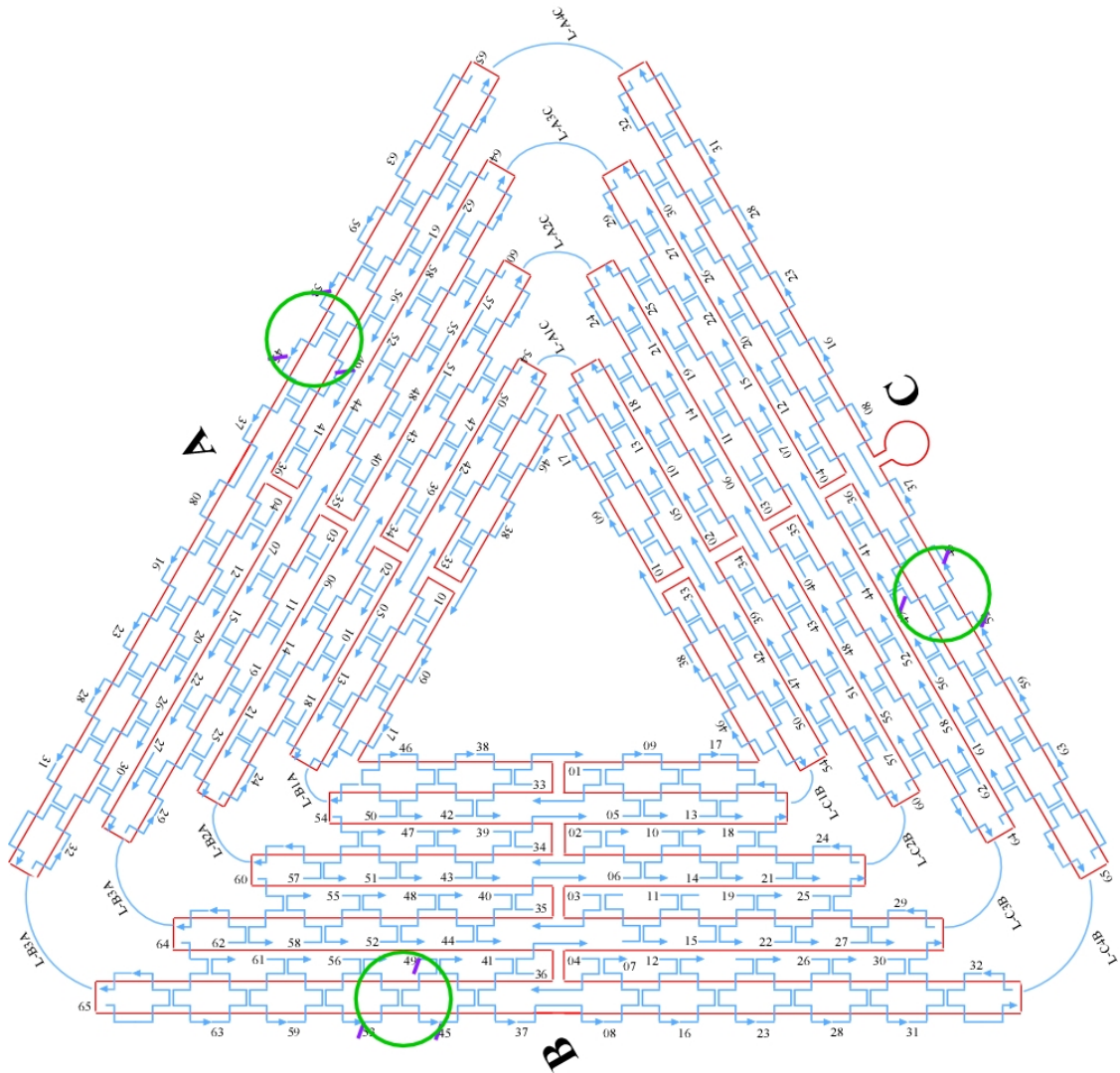


Figure S 7: Schematic design of the triangular DNA origami. The green circles represent the binding sites for the QDs. The short purple lines mark the extension of the A20.

Sequences of the capture strands:

B49-capture:

AAAAAAAAAAAAAAAAAAAAA**TTT****TTATCATCGTTGAAAGAGGACAGATGAAGAAAA**
ATCTACG

B45-capture:

AAAAAAAAAAAAAAAAAAAAA**TTT****TTAATAAAACGAACTAACCGAACTGACCAACTCC**
TGATAA

B53-capture:

AAAAAAAAAAAAAAAAAAAAAATTTTACCAGTCAGGACGTTGGAACGGTGTACAGACCG
AAACAAA

A49-capture:

AAAAAAAAAAAAAAAAAAAAAATTTTAGCATGTATTTTCATCGTAGGAATCAAACGATTTT
TTGTTT

A45-capture:

AAAAAAAAAAAAAAAAAAAAAATTTTAACGTCAAAAATGAAAAGCAAGCCGTTTTTATGA
AACCAA

A53-capture:

AAAAAAAAAAAAAAAAAAAAAATTTTCCCAATCCAAATAAGATTACCGCGCCCAATAAA
TAATAT

C49-capture:

AAAAAAAAAAAAAAAAAAAAAATTTTGTTGCGTCACGCTGGTTTGCCCCAAGGGAGCC
CCCGATT

C45-capture:

AAAAAAAAAAAAAAAAAAAAAATTTTAGAGCTTGACGGGGAGTTGCAGCAAGCGGTCA
TTGGGCG

C53-capture:

AAAAAAAAAAAAAAAAAAAAAATTTCTAAATCGGAACCCTAAGCAGGCGAAAATCCTT
CGGCCAA

Sequences of the rest of helper strands:

A01, CGGGGTTTCCTCAAGAGAAGGATTTTGAATTA,
A02, AGCGTCATGTCTCTGAATTTACCGACTACCTT,
A03, TTCATAATCCCCTTATTAGCGTTTTTCTTACC,
A04, ATGGTTTATGTCACAATCAATAGATATTAAAC,
A05, TTTGATGATTAAGAGGCTGAGACTTGCTCAGTACCAGGCG,
A06, CCGGAACCCAGAATGGAAAGCGCAACATGGCT,
A07, AAAGACAACATTTTCGGTCATAGCCAAAATCA,
A08, GACGGGAGAATTAACCTCGGAATAAGTTTATTTCCAGCGCC,
A09, GATAAGTGCCGTCGAGCTGAAACATGAAAGTATACAGGAG,
A10, TGTACTGGAAATCCTCATTAAGCAGAGCCAC,
A11, CACCGGAAAGCGCGTTTTTCATCGGAAGGGCGA,
A12, CATTCAACAAACGCAAAGACACCAGAACACCCTGAACAAA,
A13, TTTAACGGTTCGGAACCTATTATTAGGGTTGATATAAGTA,
A14, CTCAGAGCATATTCACAAACAAATTAATAAGT,
A15, GGAGGGAATTTAGCGTCAGACTGTCCGCCTCC,
A16, GTCAGAGGGTAATTGATGGCAACATATAAAAGCGATTGAG,
A17, TAGCCCGGAATAGGTGAATGCCCCCTGCCTATGGTCAGTG,
A18, CCTTGAGTCAGACGATTGGCCTTGCGCCACCC,
A19, TCAGAACCCAGAATCAAGTTTGCCGGTAAATA,
A20, TTGACGGAAATACATACATAAAGGGCGCTAATATCAGAGA,
A21, CAGAGCCAGGAGGTTGAGGCAGGTAACAGTGCCCG,
A22, ATTAAGGCCGTAATCAGTAGCGAGCCACCCT,
A23, GATAACCCACAAGAATGTTAGCAAACGTAGAAAATTATTC,
A24, GCCGCCAGCATTGACACCACCCTC,
A25, AGAGCCGCACCATCGATAGCAGCATGAATTAT,
A26, CACCGTCACCTTATTACGCAGTATTGAGTTAAGCCCAATA,
A27, AGCCATTTAAACGTCACCAATGAACACCAGAACCA,
A28, ATAAGAGCAAGAAACATGGCATGATTAAGACTCCGACTTG,
A29, CCATTAGCAAGGCCGGGGGAATTA,
A30, GAGCCAGCGAATACCCAAAAGAACATGAAATAGCAATAGC,

A31, TATCTTACCGAAGCCCAAACGCAATAATAACGAAAATCACCAG,
A32, CAGAAGGAAACCGAGGTTTTTAAGAAAAGTAAGCAGATAGCCG,
A33, CCTTTTTTCATTTAACAATTTTCATAGGATTAG,
A34, TTTAACCTATCATAGGTCTGAGAGTTCCAGTA,
A35, AGTATAAAATATGCGTTATACAAAGCCATCTT,
A36, CAAGTACCTCATTCCAAGAACGGGAAATTCAT,
A37, AGAGAATAACATAAAAAACAGGGAAGCGCATT,
A38, AAAACAAAATTAATTAATGGAACAGTACATTAGTGAAT,
A39, TTATCAAACCGGCTTAGGTTGGGTAAGCCTGT,
A40, TTAGTATCGCCAACGCTCAACAGTCGGCTGTC,
A41, TTTCTTAGCACTCATCGAGAACAATAGCAGCCTTTACAG,
A42, AGAGTCAAAAATCAATATATGTGATGAAACAAACATCAAG,
A43, ACTAGAAATATATAACTATATGTACGCTGAGA,
A44, TCAATAATAGGGCTTAATTGAGAATCATAATT,
A45, AACGTCAAAAATGAAAAGCAAGCCGTTTTTATGAAACCAA,
A46, GAGCAAAAGAAGATGAGTGAATAACCTTGCTTATAGCTTA,
A47, GATTAAGAAATGCTGATGCAAATCAGAATAAA,
A48, CACCGGAATCGCCATATTTAACAAAATTTACG,
A49, AGCATGTATTTTCATCGTAGGAATCAAACGATTTTTTGTGT,
A50, ACATAGCGCTGTAAATCGTCGCTATTCATTTCAATTACCT,
A51, GTTAAATACAATCGCAAGACAAAGCCTTGAAA,
A52, CCCATCCTCGCCAACATGTAATTTAATAAGGC,
A53, TCCCAATCCAAATAAGATTACCGCGCCCAATAAATAATAT,
A54, TCCCTTAGAATAACGCGAGAAAACCTTTTACCGACC,
A55, GTGTGATAAGGCAGAGGCATTTTCAGTCCTGA,
A56, ACAAGAAAGCAAGCAAATCAGATAACAGCCATATTATTTA,
A57, GTTTGAAATTCAAATATATTTTAG,
A58, AATAGATAGAGCCAGTAATAAGAGATTTAATG,
A59, GCCAGTTACAAAATAATAGAAGGCTTATCCGGTTATCAAC,
A60, TTCTGACCTAAAATATAAAGTACCGACTGCAGAAC,
A61, GCGCCTGTTATTCTAAGAACGCGATTCCAGAGCCTAATTT,
A62, TCAGCTAAAAAAGGTAAAGTAATT,
A63, ACGCTAACGAGCGTCTGGCGTTTTAGCGAACCCAACATGT,
A64, ACGACAATAAATCCCGACTTGCGGGAGATCCTGAATCTTACCA,

A65, TGCTATTTTGCACCCAGCTACAATTTTGTTTTGAAGCCTTAAA,
B01, TCATATGTGTAATCGTAAACTAGTCATTTTC,
B02, GTGAGAAAATGTGTAGGTAAAGATACAACCTT,
B03, GGCATCAAATTTGGGGCGCGAGCTAGTTAAAG,
B04, TTCGAGCTAAGACTTCAAATATCGGGAACGAG,
B05, ACAGTCAAAGAGAATCGATGAACGACCCCGGTTGATAATC,
B06, ATAGTAGTATGCAATGCCTGAGTAGGCCGGAG,
B07, AACCAGACGTTTAGCTATATTTTCTTCTACTA,
B08, GAATACCACATTCAACTTAAGAGGAAGCCCGATCAAAGCG,
B09, AGAAAAGCCCCAAAAAGAGTCTGGAGCAAACAATCACCAT,
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B11, CATCCAATAAATGGTCAATAACCTCGGAAGCA,
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B14, CAGGCAAGATAAAAATTTTTAGAATATTCAAC,
B15, GATTAGAGATTAGATACATTTGCAAATCATA,
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B17, GCAAATATTTAAATTGAGATCTACAAAGGCTACTGATAAA,
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B19, TTAGCAAATAGATTTAGTTTGACCAGTACCTT,
B20, TAATTGCTTTACCCTGACTATTATGAGGCATAGTAAGAGC,
B21, ATAAAGCCTTTGCGGGAGAAGCCTGGAGAGGGTAG,
B22, TAAGAGGTCAATTCTGCGAACGAGATTAAGCA,
B23, AACACTATCATAACCCATCAAAAATCAGGTCTCCTTTTGA,
B24, ATGACCCTGTAATACTTCAGAGCA,
B25, TAAAGCTATATAACAGTTGATTCCCATTTTTG,
B26, CGGATGGCACGAGAATGACCATAATCGTTTACCAGACGAC,
B27, TAATTGCTTGGAAGTTTCATTCCAAATCGGTTGTA,
B28, GATAAAAACCAAAATATTAACAGTTCAGAAATTAGAGCT,
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B30, TGCTGTAGATCCCCCTCAAATGCTGCGAGAGGCTTTTGCA,
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B33, AGGGATAGCTCAGAGCCACCACCCCATGTCAA,

B34, CAACAGTTTATGGGATTTTGCTAATCAAAAGG,
B35, GCCGCTTTGCTGAGGCTTGCAGGGGAAAAGGT,
B36, GCGCAGACTCCATGTTACTTAGCCCGTTTTAA,
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B52, GTACAACGAGCAACGGCTACAGAGGATACCGA,
B53, ACCAGTCAGGACGTTGGAACGGTGTACAGACCGAAACAAA,
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B55, AACAGCTTGCTTTGAGGACTAAAGCGATTATA,
B56, CCAAGCGCAGGCGCATAGGCTGGCAGAACTGGCTCATTAT,
B57, CGAGGTGAGGCTCCAAAAGGAGCC,
B58, ACCCCCAGACTTTTTTCATGAGGAACTTGCTTT,
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C56, TTAATGAAGTTTGATGGTGGTTCCGAGGTGCCGTAAAGCA,
C57, TGGCGAAATGTTGGGAAGGGCGAT,
C58, TGTCGTGCACACAACATACGAGCCACGCCAGC,
C59, CAAGTTTTTTGGGGTCGAAATCGGCAAAATCCGGGAAACC,
C60, TCTTCGCTATTGGAAGCATAAAGTGTATGCCCGCT,
C61, TTCCAGTCCTTATAAATCAAAGAGAACCATCACCCAAAT,
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C63, CGATGGCCCACTACGTATAGCCCGAGATAGGGATTGCGTT,
C64, AACTCACATTATTGAGTGTTGTTCCAGAAACCGTCTATCAGGG,
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Link-A1C, TTAATTAATTTTTTACCATATCAAA,
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Link-A3C, CTGTCCAGACGTATACCGAACGA,
Link-A4C, TCAAGATTAGTGTAGCAATACT,
Link-B1A, TGTAGCATTCCTTTTATAAACAGTT,

Link-B2A, TTTAATTGTATTTCCACCAGAGCC,
Link-B3A, ACTACGAAGGCTTAGCACCATTA,
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Link-C1B, GTGGGAACAAATTTCTATTTTGTAG,
Link-C2B, CGGTGCGGGCCTTCCAAAAACATT,
Link-C3B, ATGAGTGAGCTTTTAAATATGCA,
Link-C4B, ACTATTAAAGAGGATAGCGTCC,
Loop, GCGCTTAATGCGCCGCTACAGGGC.