## Supporting Information

## Programming Surface-Enhanced Raman Scattering of DNA Origami-templated Metamolecules

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\# Equal contribution
Materials: Short DNA staple strands were purchased from IDT and used as received without purification. M13 bacteriophage single stranded DNA (p7560) were used as scaffold for origami construction and produced following a published protocol ${ }^{1} .10 \mathrm{~nm}$ AuNPs were purchased from Ted Pella, Inc.

DNA origami hexagon tile design and assembly: DNA origami hexagon tile was designed using caDNAno ${ }^{2,3}$. In a typical annealing protocol, staples were mixed with single stranded DNA scaffold ( $\mathrm{p} 7560,10 \mathrm{nM}$ ) in 5 -fold molar excess in folding buffer ( 5 mM Tris base, 1 mM EDTA, supplement with 10 mM MgCl 2 ) with a total volume of $100 \mu \mathrm{~L}$. The mixed solution was slowly cooled down to room temperature $\left(24^{\circ} \mathrm{C}\right)$ in a PCR thermal cycler over 18 hrs in the following protocol: $65^{\circ} \mathrm{C}$ for $5 \mathrm{mins}, 60$ to $24^{\circ} \mathrm{C}$, at $30 \mathrm{~min} /{ }^{\circ} \mathrm{C}$.

Conjugation of DNA onto AuNPs: 50 mL of colloidal citrated 10 nm AuNP solution was added with 15 mg of Bis(p-sulfonatophenyl) phenylphosphine dihydrate dipotassium salt (BSPP) and shaken for overnight. Then NaCl was added until a color change of the solution from red to blue was observed. The AuNPs were centrifuged to the bottom at $1,000 \mathrm{rcf}$ for 30 min and the clear supernatant was removed with a pipette. The particles were dissolved in BSPP solution ( $1 \mathrm{~mL}, 2.5 \mathrm{mM}$ ) followed by addition of 3 mL of methanol. The particles were centrifuged again and re-suspended in 1 mL of BSPP. The concentration of the AuNPs was determined from the optical absorption at a wavelength of 520 nm using a Nanodrop spectrophotometer. For reducing the disulfide bonds of the thiolated ssDNA strands to monothiol, the modified strands were incubated with 100 -fold of TCEP (Tris(carboxyethyl) phosphine hydrochloride) for at least 30 mins . AuNPs and thiolated DNA were mixed in $0.5 \times$ TBE buffer at a designated ratio (1: 100). Within a period of $24 \mathrm{hrs}, \mathrm{NaCl}$ solution was added to the solution to achieve a final concentration of 300 mM . To remove the unbound DNA strands, the mixture was spun for 30 min at $18,000 \mathrm{rcf}$, and the supernatant was removed carefully without disturbing the pellet. 4 additional washing and spinning steps ( $30 \mathrm{~min}, 18,000 \mathrm{rcf}$ ) with $100 \mu \mathrm{~L}$ of $0.5 \times$ TBE buffer containing 300 mM NaCl were conducted to fully removed unbound DNA.

Attaching AuNPs onto DNA origami templates: DNA origami hexagon tile was assembled first, as described above, which was then added with 5-fold of DNA-conjugated AuNPs and incubated for at least 2 hours at room temperature under constant shaking. DNA origami hexagon monomer with 6 AuNPs were purified from agarose gel. For dimer and trimer samples, corresponding connector strands (100fold) were added to the monomer and incubated at $40^{\circ} \mathrm{C}$ overnight. These samples were then subject to agarose gel electrophoresis and corresponding bands of dimer and trimer were extracted for subsequent experiments. For 1D chain, DNA connector strands ( $100-$ fold ) were added to the purified monomer and incubated at $40^{\circ} \mathrm{C}$ overnight.

Gel electrophoresis: DNA origami with AuNPs was subject to $1 \%$ native agarose gel electrophoresis for 2 hours (gel prepared in $0.5 \times \mathrm{TBE}$ buffer supplemented with $10 \mathrm{mM} \mathrm{MgCl}_{2}$ and $0.005 \%$ (v/v) EtBr ) in an ice water bath. Then, the target gel bands were excised and placed into a Freeze ' N Squeeze column (Bio-Rad Laboratories, Inc.). The gel pieces were crushed into fine pieces by a microtube pestle in the column, and the column was then centrifuged at 7000 rcf for 5 minutes. Samples that were extracted through the column were collected for subsequent experiments.

TEM imaging: $10 \mu \mathrm{~L}$ of purified samples were deposited onto glow-discharged, carbon-coated copper TEM grids for 5-30 minutes inside a moisture chamber. Residual solution was dried by filter paper. The grids were then stained for 1 minute using a $1 \%$ aqueous uranyl formate solution containing 25 mM NaOH . Imaging was performed using a Hitachi- 7700 microscope operated at 80 kV . No staining was conducted for samples if silver growth has happened.

In-situ silver growth: $10 \mu \mathrm{~L}$ of samples were deposited onto glow-discharged, carbon-coated copper TEM grids for 5-30 minutes inside a moisture chamber. Residual solution was dried by filter paper. 10 $\mu \mathrm{L}$ of mixed solution from HQ silver enhancement kit (Nanoprobes, Inc.) was added onto the copper grids in a dark room. After 3-10 min of growth, dip the copper grids into distilled water for several times to completely wash away the silver growth solution, dry the copper grids by filter paper after the last wash.

FDTD Calculations: A commercial software package (FDTD Solutions, trial version, Lumerical Solutions Inc.) was used to calculate the localized electric field (E-field) distribution of the DNAtemplated metamolecules. The sizes and geometric parameters of gold or silver nanoparticles were set from corresponding TEM images, respectively. In the process of computational calculations, the incident light is circularly polarized with wavelength of 633 nm , which is consistent with SERS measurement in the experiment. A monitor of "frequency-domain field profile" was set up to calculate the localized Efield distributions. The obtained electromagnetic (EM) fields were normalized to the magnitude of the incident E-fields. To ensure the convergence of the calculations, a mesh size of $1 \mathrm{~nm} \times 1 \mathrm{~nm} \times 1 \mathrm{~nm}$ was chosen. Perfectly matched layer (PML) absorbing boundary were used in all directions. The dielectric constant spectra of the Ag materials were taken from the model of Palik ${ }^{4}$ and gold were taken from Johnson and Christy ${ }^{5}$, respectively.

Dark-field microscopy measurement: An inverted microscope (eclipse Ti-U, Nikon, Japan) equipped with a dark-field condenser ( $0.8<$ NA < 0.95 ), a 100 W halogen lamp, a monochromator (Acton SP2300i) equipped with a spectrograph CCD (CASCADE512B, Roper Scientific), and a true-color digital camera (Nikon DS-fi) and a grating ( 1200 grooves per mm ) were used for the dark-field spectrum measurements. The true-color images were taken using a $60 \times$ objective lens.

Surface enhanced Rahman spectroscopy measurement: TEM grid loaded DNA-templated metamolecules was immersed in $100 \mu \mathrm{M} 4-\mathrm{MBA}$ for 1 h . Then, the TEM grid was washed by water and dried by nitrogen stream. The SERS measurements were performed on a confocal Raman microscope (InVia, Renishaw, England) connected with the above-mentioned inverted microscope using a 633 nm excitation laser (we used $50 \%$ power), $60 \times$ objective lens (NA=0.70) and 3 s acquisition time. For each sample, ten SERS spots were recorded to obtain an averaged SERS spectrum and the baselines were subtracted by the software Wire 4.0.


Figure S1. TEM images of DNA origami hexagon tile decorated with six of 10 nm AuNPs.


Figure S2. Native agarose gel electrophoresis of hexagon monomer, dimer, trimer, and 1D metamolecules


Figure S3. In-situ silver growth on AuNP hexagon monomers with varied growth time.


Figure S4. FDTD simulation of 3-nanoparticle metamolecules. Size of nanoparticles are the same as 6nanoparticle metamolecules but with different interparticle gaps. FDTD revealed no observable electromagnetic fields.


Figure S5. Discrete Ag@Au core-shell nanoparticles. a, b) TEM images; c) DFM image; d) Raman spectra of 4-MBA.


Figure S6. Native agarose gel electrophoresis of AuNP Core-satellite structures.


Figure S7. AuNP Core-satellite structures prior to silver deposition.


Figure S8. Ag@Au core-satellite metamolecules.


Figure $\mathrm{S} 9 . \mathrm{Ag} @ \mathrm{Au}$ hexagon dimer metamolecules.


Figure $\mathrm{S} 10 . \mathrm{Ag} @$ Au hexagon trimer metamolecules.


Figure S11. Ag@Au hexagon 1D chain metamolecules.


Figure S12. Average Raman intensity of 4-MBA at $1585 \mathrm{~cm}^{-1}$ for complex metamolecules ( $\mathrm{N}=10$ ).

Table S1. Theoretical enhancement factors for SERS metamolecules

| Metamolecules | Monomer |  |  |  | Core-satellite | Dimer | Trimer | 1 D |  |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| Growth time | $0-\mathrm{min}$ | $3-\mathrm{min}$ | 7 -min | $10-\mathrm{min}$ | $12-\mathrm{min}$ | $7-\mathrm{min}$ | $10-\mathrm{min}$ | $10-\mathrm{min}$ | $10-\mathrm{min}$ |
| Enhancement factor | $3.6 \times 10^{1}$ | $4.5 \times 10^{1}$ | $1.3 \times 10^{2}$ | $2.4 \times 10^{3}$ | $2.6 \times 10^{6}$ | $4.1 \times 10^{4}$ | $3.2 \times 10^{4}$ | $3.9 \times 10^{4}$ | $8.2 \times 10^{4}$ |

## Sequence of p7560 scaffold DNA:

GCTTGGCACTGGCCGTCGTTTTACAACGTCGTACTGGGAAAACCCTGGCGTTACCCAACTTAATCGCCTTGCAGCACATCCCCCTTTCGCCAGCTGGCGTAATAGCGA AGAGGCCCGCACCGATCGCCCTTCCCAACAGTTGCGCAGCCTGAATGGCGAATGGCGCTTTGCCTGGTTTCCGGCACCAGAAGCGGTGCCGGAAAGCTGGCTGGAGTG CGATCTTCCTGAGGCCGATACTGTCGTCGTCCCCTCAAACTGGCAGATGCACGGTTACGATGCGCCCATCTACACCAACGTGACCTATCCCATTACGGTCAATCCGCCGT TTGTTCCCACGGAGAATCCGACGGGTTGTTACTCGCTCACATTTAATGTTGATGAAAGCTGGCTACAGGAAGGCCAGACGCGAATTATTTTTGATGGCGTTCCTATTGGTTA AAAAATGAGCTGATTTAACAAAAATTTAATGCGAATTTTAACAAAATATTAACGTTTACAATTTAAATATTTGCTTATACAATCTTCCTGTITTTGGGGCTTTTCTGATTATCAAC GGGGGTACATATGATTGACATGCTAGTTITACGAITACCGTTCATCGATTCTCTTGTTTGCTCCAGACTCTCAGGCAATGACCTGATAGCCTITGTAGATCTCTCAAAAATAG CTACCCTCTCCGGCATTAATTTATCAGCTAGAACGGTTGAATATCATATTGATGGTGATTTGACTGTCTCCGGCCTTTCTCACCCTTTTGAATCTTTACCTACACATTACTCA GGCATTGCATTTAAAATATATGAGGGTTCTAAAAATTTTTATCCTTGCGTTGAAATAAAGGCTTCTCCCGCAAAAGTATTACAGGGTCATAATGTTTTTGGTACAACCGATTTA GCTTTATGCTCTGAGGCTTTATTGCTTAATTTTGCTAATTCTTTGCCTTGCCTGTATGATTTATTGGATGTTAATGCTACTACTATTAGTAGAATTGATGCCACCTTTTCAGCT CGCGCCCCAAATGAAAATATAGCTAAACAGGTTATTGACCATTTGCGAAATGTATCTAATGGTCAAACTAAATCTACTCGTTCGCAGAATTGGGAATCAACTGTTATATGGAA TGAAACTTCCAGACACCGTACTTTAGTTGCATATTTAAAACATGTTGAGCTACAGCATTATATTCAGCAATTAAGCTCTAAGCCATCCGCAAAAATGACCTCTTATCAAAAGG AGCAATTAAAGGTACTCTCTAATCCTGACCTGTTGGAGTTTGCTTCCGGTCTGGTTCGCTTTGAAGCTCGAATTAAAACGCGATATITGAAGTCTTTCGGGCTTCCTCTTAAT CTTTTTGATGCAATCCGCTTTGCTTCTGACTATAATAGTCAGGGTAAAGACCTGATTTTTGATTTATGGTCATTCTCGTTTTCTGAACTGTTTAAAGCATTTGAGGGGGATTCA ATGAATATTTATGACGATTCCGCAGTATTGGACGCTATCCAGTCTAAACATTTTACTATTACCCCCTCTGGCAAAACTTCTTTTGCAAAAGCCTCTCGCTATTTTGGTTTTTA CGTCGTCTGGTAAACGAGGGTTATGATAGTGTTGCTCTTACTATGCCTCGTAATTCCTTTTGGCGTTATGTATCTGCATTAGTTGAATGTGGTATTCCTAAATCTCAACTGAT GAATCTTTCTACCTGTAATAATGTTGTTCCGTTAGTTCGTTTTATTAACGTAGATTTTTCTTCCCAACGTCCTGACTGGTATAATGAGCCAGTTCTTAAAATCGCATAAGGTAA TTCACAATGATTAAAGTTGAAATTAAACCATCTCAAGCCCAATTTACTACTCGTTCTGGTGTTTCTCGTCAGGGCAAGCCTTATTCACTGAATGAGCAGCTTTGTTACGTTGA TTTGGGTAATGAATATCCGGTTCTTGTCAAGATTACTCTTGATGAAGGTCAGCCAGCCTATGCGCCTGGTCTGTACACCGTTCATCTGTCCTCTTTCAAAGTTGGTCAGTTC GGTICCCTTATGATTGACCGTCTGCGCCTCGITCCGGCTAAGTAACATGGAGCAGGTCGCGGATITCGACACAATITATCAGGCGATGATACAAATCTCCGITGTACTITG TTTCGCGCTTGGTATAATCGCTGGGGGTCAAAGATGAGTGTTTTAGTGTATTCTTTTGCCTCTTTCGTTTTAGGTTGGTGCCTTCGTAGTGGCATTACGTATTTTACCCGITT AATGGAAACTTCCTCATGAAAAAGTCTTTAGTCCTCAAAGCCTCTGTAGCCGTTGCTACCCTCGTTCCGATGCTGTCTTTCGCTGCTGAGGGTGACGATCCCGCAAAAGCG GCCTTTAACTCCCTGCAAGCCTCAGCGACCGAATATATCGGTTATGCGTGGGCGATGGTTGTTGTCATTGTCGGCGCAACTATCGGTATCAAGCTGTTTAAGAAATTCACC TCGAAAGCAAGCTGATAAACCGATACAATTAAAGGCTCCTTTTGGAGCCTTTTTTTTGGAGATTTTCAACGTGAAAAAATTATTATTCGCAATTCCTTTAGTTGTTCCTTTCTA TTCTCACTCCGCTGAAACTGTTGAAAGTTGTTTAGCAAAATCCCATACAGAAAATTCATTTACTAACGTCTGGAAAGACGACAAAACTTTAGATCGTTACGCTAACTATGAGG GCTGTCTGTGGAATGCTACAGGCGTTGTAGTTTGTACTGGTGACGAAACTCAGTGTTACGGTACATGGGTTCCTATTGGGCTTGCTATCCCTGAAAATGAGGGTGGTGGC CTGAGGGTGGCGGTTCTGAGGGTGGCGGTTCTGAGGGTGGCGGTACTAAACCTCCTGAGTACGGTGATACACCTATTCCGGGCTATACTTATATCAACCCTCTCGACGGC ACTTATCCGCCTGGTACTGAGCAAAACCCCGCTAATCCTAATCCTTCTCTTGAGGAGTCTCAGCCTCTTAATACTTTCATGTTTCAGAATAATAGGTTCCGAAATAGGCAGG GGGCATTAACTGTTTATACGGGCACTGTTACTCAAGGCACTGACCCCGTTAAAACTTATTACCAGTACACTCCTGTATCATCAAAAGCCATGTATGACGCTTACTGGAACGG TAAATTCAGAGACTGCGCTTTCCATTCTGGCTTTAATGAGGATTTATTTGTTTGTGAATATCAAGGCCAATCGTCTGACCTGCCTCAACCTCCTGTCAATGCTGGCGGCGGC TCTGGTGGTGGTTCTGGTGGCGGCTCTGAGGGTGGTGGCTCTGAGGGTGGCGGTTCTGAGGGTGGCGGCTCTGAGGGAGGCGGTTCCGGTGGTGGCTCTGGTTCCGG TGATTTTGATTATGAAAAGATGGCAAACGCTAATAAGGGGGCTATGACCGAAAATGCCGATGAAAACGCGCTACAGTCTGACGCTAAAGGCAAACTTGATTCTGTCGCTAC TGATTACGGTGCTGCTATCGATGGTTTCATTGGTGACGTTTCCGGCCTTGCTAATGGTAATGGTGCTACTGGTGATTTTGCTGGCTCTAATTCCCAAATGGCTCAAGTCGGT GACGGTGATAATTCACCTTTAATGAATAATTTCCGTCAATATTTACCTTCCCTCCCTCAATCGGTTGAATGTCGCCCTTTTGTCTTTGGCGCTGGTAAACCATATGAATTTTCT ATTGATTGTGACAAAATAAACTTATTCCGTGGTGTCTTTGCGTTTCTTTTATATGTTGCCACCTTTATGTATGTATTTTCTACGTTTGCTAACATACTGCGTAATAAGGAGTCTT AATCATGCCAGTTCTTTTGGGTATTCCGTTATTATTGCGTTTCCTCGGTTTCCTTCTGGTAACTTTGTTCGGCTATCTGCTTACTTTTCTTAAAAAGGGCTTCGGTAAGATAGC TATTGCTATTTCATTGTTTCTTGCTCTTATTATTGGGCTTAACTCAATTCTTGTGGGTTATCTCTCTGATATTAGCGCTCAATTACCCTCTGACTTTGTTCAGGGTGTTCAGTTA ATTCTCCCGTCTAATGCGCTTCCCTGTTTTTATGTTATTCTCTCTGTAAAGGCTGCTATTTTCATTTTTGACGTTAAACAAAAAATCGTTTCTTATTTGGATTGGGATAAATAAT ATGGCTGTTTATTTTGTAACTGGCAAATTAGGCTCTGGAAAGACGCTCGTTAGCGTTGGTAAGATTCAGGATAAAATTGTAGCTGGGTGCAAAATAGCAACTAATCTTGATT TAAGGCTTCAAAACCTCCCGCAAGTCGGGAGGTTCGCTAAAACGCCTCGCGTTCTTAGAATACCGGATAAGCCTTCTATATCTGATTTGCTTGCTATTGGGCGCGGTAATG ATTCCTACGATGAAAATAAAAACGGCTTGCTTGTTCTCGATGAGTGCGGTACTTGGTTTAATACCCGTTCTTGGAATGATAAGGAAAGACAGCCGATTATTGATTGGTTTCTA CATGCTCGTAAATTAGGATGGGATATTATTTTTCTTGTTCAGGACTTATCTATTGTTGATAAACAGGCGCGTTCTGCATTAGCTGAACATGTTGTTTATTGTCGTCGTCTGGA CAGAATTACTTTACCTTTTGTCGGTACTTTATATTCTCTTATTACTGGCTCGAAAATGCCTCTGCCTAAATTACATGTTGGCGTTGTTAAATATGGCGATTCTCAATTAAGCCC TACTGTTGAGCGTTGGCTTTATACTGGTAAGAATTTGTATAACGCATATGATACTAAACAGGCTTTTTCTAGTAATTATGATTCCGGTGTTTATTCTTATTTAACGCCTTATTTA TCACACGGTCGGTATTTCAAACCATTAAATTTAGGTCAGAAGATGAAATTAACTAAAATATATTTGAAAAAGTTTTCTCGCGTTCTTTGTCTTGCGATTGGATTTGCATCAGCA TTTACATATAGTTATATAACCCAACCTAAGCCGGAGGTTAAAAAGGTAGTCTCTCAGACCTATGATTTTGATAAATTCACTATTGACTCTTCTCAGCGTCTTAATCTAAGCTAT CGCTATGTTTTCAAGGATTCTAAGGGAAAATTAATTAATAGCGACGATTTACAGAAGCAAGGTTATTCACTCACATATATTGATTTATGTACTGTTTCCATTAAAAAAGGTAAT TCAAATGAAATTGTTAAATGTAATTAATTTTGTTTTCTTGATGTTTGTTTCATCATCTTCTTTTGCTCAGGTAATTGAAATGAATAATTCGCCTCTGCGCGATTTTGTAACTTGG TATTCAAAGCAATCAGGCGAATCCGTTATTGTTTCTCCCGATGTAAAAGGTACTGTTACTGTATATTCATCTGACGTTAAACCTGAAAATCTACGCAATTTCTTTATTTCTGTT TTACGTGCAAATAATTTTGATATGGTAGGTTCTAACCCTTCCATTATTCAGAAGTATAATCCAAACAATCAGGATTATATTGATGAATTGCCATCATCTGATAATCAGGAATAT GATGATAATTCCGCTCCTTCTGGTGGTTTCTTTGTTCCGCAAAATGATAATGTTACTCAAACTTTTAAAATTAATAACGTTCGGGCAAAGGATTTAATACGAGTTGTCGAATTG TTTGTAAAGTCTAATACTTCTAAATCCTCAAATGTATTATCTATTGACGGCTCTAATCTATTAGTTGTTAGTGCTCCTAAAGATATTTTAGATAACCTTCCTCAATTCCTTTCAA CTGTTGATTTGCCAACTGACCAGATATTGATTGAGGGTTTGATATTTGAGGTTCAGCAAGGTGATGCTTTAGATTTTTCATTTGCTGCTGGCTCTCAGCGTGGCACTGTTGC AGGCGGTGTTAATACTGACCGCCTCACCTCTGTTTTATCTTCTGCTGGTGGTTCGTTCGGTATTTTTAATGGCGATGTTTTAGGGCTATCAGTTCGCGCATTAAAGACTAAT AGCCATTCAAAAATATTGTCTGTGCCACGTATTCTTACGCTTTCAGGTCAGAAGGGTTCTATCTCTGTTGGCCAGAATGTCCCTTTTATTACTGGTCGTGTGACTGGTGAAT CTGCCAATGTAAATAATCCATTTCAGACGATTGAGCGTCAAAATGTAGGTATTTCCATGAGCGTTTTTCCTGTTGCAATGGCTGGCGGTAATATTGTTCTGGATATTACCAGC AAGGCCGATAGTTTGAGTTCTTCTACTCAGGCAAGTGATGTTATTACTAATCAAAGAAGTATTGCTACAACGGTTAATTTGCGTGATGGACAGACTCTTTTACTCGGTGGCC TCACTGATTATAAAAACACTTCTCAGGATTCTGGCGTACCGTTCCTGTCTAAAATCCCTTTAATCGGCCTCCTGTTTAGCTCCCGCTCTGATTCTAACGAGGAAAGCACGTT ATACGTGCTCGTCAAAGCAACCATAGTACGCGCCCTGTAGCGGCGCATTAAGCGCGGCGGGTGTGGTGGTTACGCGCAGCGTGACCGCTACACTTGCCAGCGCCCTAG CGCCCGCTCCTTTCGCTTTCTTCCCTTCCTTTCTCGCCACGTTCGCCGGCTTTCCCCGTCAAGCTCTAAATCGGGGGCTCCCTTTAGGGTTCCGATTTAGTGCTTTACGGC ACCTCGACCCCAAAAAACTTGATTTGGGTGATGGTTCACGTAGTGGGCCATCGCCCTGATAGACGGTTTTTCGCCCTTTGACGTTGGAGTCCACGTTCTTTAATAGTGGAC TCTTGTTCCAAACTGGAACAACACTCAACCCTATCTCGGGCTATTCTTTTGATTTATAAGGGATTTTGCCGATTTCGGAACCACCATCAAACAGGATTTTCGCCTGCTGGGG CAAACCAGCGTGGACCGCTTGCTGCAACTCTCTCAGGGCCAGGCGGTGAAGGGCAATCAGCTGTTGCCCGTCTCACTGGTGAAAAGAAAAACCACCCTGGCGCCCAATA CGCAAACCGCCTCTCCCCGCGCGTTGGCCGATTCATTAATGCAGCTGGCACGACAGGTTTCCCGACTGGAAAGCGGGCAGTGAGCGCAACGCAATTAATGTGAGTTAGC TCACTCATTAGGCACCCCAGGCTTTACACTTTATGCTTCCGGCTCGTATGTTGTGTGGAATTGTGAGCGGATAACAATTTCACACAGGAAACAGCTATGACCATGATTACGA ATTCGAGCTCGGTACCCGGGGATCCTCCGTCTTTATCGAGGTAACAAGCACCACGTAGCTTAAGCCCTGTTTACTCATTACACCAACCAGGAGGTCAGAGTTCGGAGAAAT GATTTATGTGAAATGCGTCAGCCGATTCAAGGCCCCTATATTCGTGCCCACCGACGAGTTGCTTACAGATGGCAGGGCCGCACTGTCGGTATCATAGAGTCACTCCAGGG CGAGCGTAAATAGATTAGAAGCGGGGTTATTTTGGCGGGACATTGTCATAAGGTTGACAATTCAGCACTAAGGACACTTAAGTCGTGCGCATGAATTCACAACCACTTAGA AGAACATCCACCCTGGCTTCTCCTGAGAA

Sequence of thiolated DNA strand:
5’-ThioMC6-D/TTTTTTTTTTTTTTTTTTTT-3'.

Table S2. Sequence of DNA staple strands

| Name | Sequence |
| :---: | :---: |
| O[47] | ACAATATTTTTGAATGAGGTGAGGTCGCCATTAGCCAGCA |
| 0[79] | CCTGAAAGCGTAAGGTTTGGATTAGAGACTACCTTTTTAACCTCCGGCT |
| 0[153] | GACTCCAACGTCAAAGAAGGCGTTTTTCTGACCCCTGTTTA |
| O[185] | GAACAAGAGTCCACAAGCAAGCCCAATGAAATAGCAATAGCTATCTTAC |
| 0[259] | TCGGCAAAATCCCTTAATACATAAATTAAGACTAAGTTTA |
| 0 [291] | AAAATCCTGTTTGAGAGCCACCATAGGTGTATCACCGTACTCAGGAGGT |
| 0[365] | CCTTCACCGCCTGGCCTTCCACAGGTAACACTTCTTTCCA |
| 0[397] | ACCAGTGAGACGGGGAGGCTTTGAATTTCAACTTTAATCATTGTGAATT |
| 0 [471] | TGAATCGGCCAACGCGTGCAGATATAGAAAGAACTATCAT |
| 0 [503] | GGGAAACCTGTCGTTCCTTTTGAGTAAAGATTCAAAAGGGTGAGAAAGG |
| 0 [577] | GGGGTGCCTAATGAGTGGTAATCGCAGGTCATCAGAAAAG |
| 0[609] | CGAGCCGGAAGCATCCAGGCAAACAGGGCTTAAGCTACGTGGTGCTTGT |
| 0[636] | TATCCGCTCACACATAAATCATTTCCAGCAGA |
| 1[32] | ATAGCCCTAAAGGGTTATATAACGGCACAG |
| 1[72] | GATGCAAATAGGTCTGATACTTCTGAATAACGA |
| 1[96] | AAGAACGCGAGAAAACTTTTTCAAACCGTCTATCATCTGGCCAACAGAATTAAGAC |
| 1[138] | TAGTTAATTTCCCTTTTTAAGAAGAACGTG |
| 1[178] | CCGAACAAGCAAGAAAGTTTTTATTTTCATGCG |
| 1[202] | ACCGAGGAAACGCAATAATAACGGAAGAATAGCCCGAGATAGGGTTGAAGAGAGAT |
| 1[244] | AAAGAACTGGCCCGCCACCCTCATCCGAAA |
| 1[284] | AGAACCGAGCCCGGAACCGGAACCGCCTCCGTT |
| 1[308] | CACCCTCATTTTCAGGGATAGCAAGTTGCAGCAAGCGGTCCACGCTGGTACCAGGC |
| 1[350] | GGAACCCATGTTGCGATTTTAAGTGATTGC |
| 1[390] | ACCAGTCAGATGGTTTAGGACTAAAGACTTGAA |
| 1[414] | AAATCTACGTTAATAAAACGAACTGGCGGTTTGCGTATTGGGCGCCAGGCCCTGAC |
| 1[456] | AACATTATTACACAGTCAAATCAGCATTAA |
| 1[496] | TTCAACCAATGTGTAGTAAGAGGTCATTTTGCA |
| 1[520] | TAATGCCGGAGAGGGTAGCTATTTTCACATTAATTGCGTTGCGCTCACAATTTTTA |
| 1[562] | TCTACAAAGGCGATAAAGACGGAAAAGCCT |
| 1[602] | CGAGCTCATGAGTAAAGCGCCATTCGCCATTGA |
| 1[626] | TCATAGCTGTTTCCTGTGTAATGCGCGAACTG |
| $2[20]$ | ACGAACCACTCCGAACTCTGATCTGTAAGCAA |
| 2[48] | TAGGTTACACGGAATTCATCAATATA |
| $2[87]$ | TCAAAATCATCCAATCCCTTCTGA |
| 2[126] | GTTTGAAAGAAGAGTCAATAGTAAATCGTCGC |
| 2[154] | CGAAGCATCAAACAAGTACCGCACTC |
| 2[193] | ATAATAAGAAGTTACCCCAGTTTG |
| 2[232] | CAGTATGTACAAGAATTGAGTAGACGGGAGAA |
| 2[260] | TTAGTAATGAGGCATAATCAAAATCA |


| 2[299] | ATAAGTATCCACCCTCAGCAGGCG | core |
| :---: | :---: | :---: |
| 2[338] | CCAGTACAAGTGCCGTCGAGAAAGTATTAAGA | core |
| 2[366] | ACCTTAACCACAGAACGAGGGTAGCA | core |
| 2[405] | TGGGCTTGAGGACGTTTTCTTTTC | core |
| 2[444] | AGATTTAGACACCAGAACGAGAACCGGATATT | core |
| 2[472] | CCGGAGAGGCATGATTAGAGAGTACC | core |
| 2[511] | GCCTGAGTGTTCTAGCTTCCAGTC | core |
| 2[550] | CTGGAGCACTCATATATTTTAAAACATTATGA | core |
| 2[578] | TACCTCTATTAAGGCACCGCTTCTGG | core |
| 2[617] | TTGGTGTAGAATTCGTACAACATA | core |
| 3[21] | AGATAAAACAGGCTATTAGTCTTTGAAATTGT | core |
| 3[56] | ATCCTGATTAATACGTTATATGTAAATGCT | core |
| 3[88] | TAGCTTAGGATAGAACGCAAGACA | core |
| 3[104] | GCTGATACCGACCGTGTGATAAATGGCGAAAAATATATTT | core |
| 3[162] | ATCGAGAACTATTAAAAAGTAAGCAGATAG | core |
| 3[194] | CTAATATCGTGTTGTTAGAAGGAA | core |
| 3[210] | AACCCTAGCAAACGTAGAAAATACTAAATCAAAATACCCA | core |
| 3[268] | CCGGAACCATGGTGGTGAACCGCCACCCTC | core |
| 3[300] | TTGCTCAGTTTGCCCCAGAGCCAC | core |
| 3[316] | GGATAAACTACAACGCCTGTAGCACTGAGAGAGCCCAATA | core |
| 3[374] | ACGGCTACACAACAGCAACTGGCTCATTAT | core |
| 3[406] | TAAGGCTTGGTGGTTTGGGAAGAA | core |
| 3[422] | GAGAAGAATACCACATTCAACTAACGGGGAGAAACGGAAC | core |
| 3[480] | TTTAATTGCGCCAGCTCCATCAATATGATA | core |
| 3[512] | AGGATAAATGCCCGCTTGATAAAT | core |
| 3[528] | GAACCAACAAGAGAATCGATGAACGAGCTAACTTGAGAGA | core |
| 3[586] | TGCCGGAAAAAAGTGTGGATCCCCGGGTAC | core |
| 3[618] | CGCATTTCAATTCCACAATCATGG | core |
| 4[71] | TATCATCATCATTTGATCTGGTCAGTTGGC | core |
| 4[177] | TTTCCTTATTGTCCAGATTGAGAATCGCCA | core |
| 4[283] | CATAGCCCCATCACCATTCAACCGATTGAG | core |
| 4[389] | ATCGTCACCTTATCAGGAGTGAGAATAGAA | core |
| 4[495] | AACCAGACCAGAAAACTTTGCCAGAGGGGG | core |
| 4[601] | CCTCAGGAATAAATGTAAATTCGCATTAAA | core |
| 5[40] | GCAAATGACAGTTGAATTAGAGCCGTCAATA | core |
| 5[72] | CTCAAATATCAAACAACACCAGAAGAAAACAGA | core |
| 5[146] | GTATCATACAACGCCACCGACAAAAGGTAAA | core |
| 5[178] | AAGCCAACGCTCAATAAATCAATAAATAGAAGG | core |
| 5[252] | TTTTGTCAAGGTAAATTTTGGGAATTAGAGC | core |
| 5[284] | CCAGCGCCAAAGACCCGTCATCGGCCACCCTCA | core |
| 5[358] | GACGTTAGACTAAAGGAGGAGCCTTTAATTG | core |
| 5[390] | CTAAACAACTTTCAACAAAGGCCGCGCCACTAC | core |
| 5[464] | AACCCTCGAAAATGTTCCTCAAATGCTTTAA | core |
| 5[496] | TAGCGAGAGGCTTTCGGATTCGAGCGCTCAACA | core |
| 5[570] | CCCCAAAATAAATCAGTGTAGCCAGCTTTCA | core |


| 5[602] | AAATTGTAAACGTTATAGACGACGATCTTCGCT | core |
| :---: | :---: | :---: |
| 6[55] | AAATCAAAAAATCTAATATCAGATGATGGCTCA | re |
| 6[161] | TATTTAATGCGTTATAAACGGGTATTAAACTAA | ore |
| 6[267] | GGAGGGACAATCAATAGTTTGCCATCTTTTTGG | ore |
| 6[373] | AGGAACATAAATGAATGAAAGACAGCATCGGCC | ore |
| 6[479] | TAATAGTTTTACCAGACTCCAACAGGTCAGAAC | ore |
| 6[585] | TTTTTGTACAGGAAGACCAGCCAGCTTTCCAAC | ore |
| 7[21] | AGCACTAACAAACCGCCTGCAACATGGGCACG | ore |
| 7[56] | GATAATAATTCCTGATAGCATCACCTTGCTGAAC | ore |
| 7[84] | TATTAGACTTTAAAGAAACCGTTATTAATCAGATGA | ore |
| 7[127] | AATAAGAGAATACCGGAATCATAAAATTTTCC | ore |
| 7[162] | GTAATTCCATTCCAAGCAAATTCTTACCAGTATA | ore |
| 7[190] | AAACAACATGTTAGAAACCAGTCCTGAAGCGAACCT | ore |
| 7[233] | CCGTCACCGACAAAAGAAACGCAAACACCCTG | core |
| 7[268] | CAGCAAACTTATTAGCGAAAATTCATATGGTTTA | core |
| 7[296] | ACCATTAGCAAGGCGCGTTTTAATCAGTCAGCATTG | core |
| 7[339] | CCAAAAAAAAGTAGCGTAACGATCCTCCTCAA | re |
| 7[374] | TATCGGTCTCAGCAGCTTTCTGTATGGGATTTTG | ore |
| 7[402] | GGTGAATTTCTTGGGAGTTAACCATCGCATACACTA | re |
| 7[445] | TAAATATTCATGAATTACGAGGCAAAATCAAC | ore |
| 7[480] | ACAGTTCGGAAGCAAACGACGATAAAAACCAAAA | re |
| 7[508] | TAAATCAAAAATGCGTTTTAATTGCATCAAGTTTCA | core |
| 7[551] | AATTCGCGTCTCAATCATATGTACTACTTTTG | core |
| 7[586] | TCAACATGATCGCACTTTGTATAAGCAAATATTT | core |
| 7[614] | AACCCGTCGGATTTTGAGGGGGTCACGTAGGCGATT | core |
| 8[71] | CCTACCATAAACAATTGAATACCAAGTTAC | core |
| 8[106] | CTTAGAATCCTGGAAACAGTACATGAGCCAGT | core |
| 8[177] | CCGCGCCCAAACGATTACAATTTTATCCTG | core |
| 8[212] | AACAAAGTCAGAATAGCAGCCTTTAATTATCA | core |
| 8[283] | CCTCAGAACCAGTGCCTAAAGCCAGAATGG | core |
| 8[318] | GAGAAGGATTACAGTTAATGCCCCGAAAATCT | core |
| 8[389] | TTCCATTAATTTGAAATGTATCATCGCCTG | core |
| 8[424] | GTAACAAAGCTACCAGGCGCATAGAATCGTCA | core |
| 8[495] | AGCTTAATTAATCATACATTTCGCAAATGG | core |
| 8[530] | CGGGAGAAGCCAATTAAGCAATAATCAAAAAT | core |
| 8[601] | TGTTGGGAACGCCAAACTTTCTCAGGAGAA | core |
| 8[636] | AATATAGGGGCACGCTCGCCCTGGTCTTTAGG | core |
| 9[40] | ATATACAGCGCAGAGGGAAAACAA | core |
| 9[64] | CGGGAGAAACAATAACGGATTCGCCTTAGTGGAAGGGTTAGAA | core |
| 9[107] | TATTAATTTTACTAGAAAAAGTAAATTTAATG | core |
| 9[146] | CCCGACTTCCAACGCTATTTATCC | core |
| 9[170] | TTAAATCAAGATTAGTTGCTATTGGATGACGTAGGAATCATTA | core |
| 9[213] | TTAACTGAAGACACCACGGAATCCTTATTACG | core |
| 9[252] | ACAGGAGGAGTCTCTGTGGTAATA | core |
| 9[276] | TTGGCCTTGATATTCACAAACAACTGGGGCTCAGAGCCGCCAC | core |


| 9[319] | GGCTGAGATAAAGTTTTGTCGGAGTTTCGTCA | core |
| :---: | :---: | :---: |
| 9[358] | AAACACTCGTGTCGAATAAGGGAA | core |
| 9[382] | TTATACCAAGCGCGAAACAAAGTATCAGTTTTCATGAGGAAGT | core |
| 9[425] | CATTACCCTAGTAAGAGCAACTTCATCAGTTG | core |
| 9[464] | TTCCATATCCTGTTTAGTAGTAGC | core |
| 9[488] | CTGCGAACGAGTAGATTTAGTTTGGTAACTGCGGATGGCTTAG | core |
| 9[531] | CCCTGTAACCCGGTTGATAATTGCCTGAGAGT | core |
| 9[570] | AAGTTGGGTGGATGTTTGTCAACC | core |
| 9[594] | AGTCACGACGTTGTAAAACGACGCGGGGCTCAGGCTGCGCAAC | core |
| 9[637] | CTCGTCGGGTGCCACGCTGAGAAAAATACCGA | core |
| 10[55] | AAAATCGTAACAGTACTTTGCACGTGAGCGGAAT | core |
| 10[95] | TGAGTGAATAACCTGATTGCTTTTCATTTG | core |
| 10[161] | AATCTTAGCGGGAGGTAAATCAGATTCGGCTGTC | core |
| 10[201] | CATAAAAACAGTTGCACCCAGCTTTTTGTT | core |
| 10[267] | AAAGCGCTTGAGGCAGCAGAGCCACATTTTCGGT | core |
| 10[307] | AACCTATTATTATAAATCCTCATTTGAGTA | core |
| 10[373] | ATAAATTATCTTTGACATACGTAATTTTTGCGGG | core |
| 10[413] | CATCAAGAGTAACAACGGAGATTGAGGACA | core |
| 10[479] | TCAATAAAACAGTTGAAATGCTGTATTCAAAGCG | core |
| 10[519] | AAAGCTAAATCGACCATTAGATACAGGCAA | core |
| 10[585] | GCCAGGGTAACGCCAGGTGCGGGCCCAGTATCGG | core |
| 10[625] | TACCGACAGTGGCCAGTGCCAAGATAACCC | core |
| 11[48] | AATTAATTACATTTTCAAAATTACTTTTACAT | core |
| 11[80] | AATTACCTTTTTTAATTGAAAACATGCTTCTGTGAATTTA | core |
| 11[154] | CAATCCAAATAAGAATAGCAAGCTTTGAAGCC | core |
| 11[186] | TAACGTCAAAAATGAAAGGGTAATAGCGCATTTAAGCCCA | core |
| 11[260] | AGTTTTAACGGGGTCGCCACCCTGTCAGACGA | core |
| 11[292] | ACAGTGCCCGTATAAAGGATTAGCAAACATGAGGGTTGAT | core |
| 11[366] | CCGAACTGACCAACACGGGTAAACCCCAGCGA | core |
| 11[398] | GATGAACGGTGTACAGGCTCATTCTTGACAAGTAGTAAAT | core |
| 11[472] | ATTAACATCCAATAGCTGAATATTTCCCAATT | core |
| 11[504] | GGCAAAGAATTAGCAATTTATTTCTGTACCAAAATGCAAT | core |
| 11[578] | TTATGACAATGTCCGGGCGATCGGGTTTTCCC | core |
| 11[610] | CGCTTCTAATCTATTTCTTGAATCCCCTGCCACCTCCTGG | core |
| 4[39] | GTATTAACCTAATAGAAGGAATTG | AuNP Capture |
| 4[145] | GAATAAACATAAAGTAACATGTAA | AuNP Capture |
| 4[251] | CAACATATTTGAGCCAATTGACGG | AuNP Capture |
| 4[357] | CTCATAGTGCTCCAAAAATTGCGA | AuNP Capture |
| 4[463] | GCCAAAAGTGAATCCCTAGACTGG | AuNP Capture |
| 4[569] | TAGCATGTGGCCTTCCCTCATTTT | AuNP Capture |
| 6[31] | AGGAAGGTTATCTAAAATAAGTGACTCTATGA | AuNP Capture |
| 6[137] | TTTAGGCAGAGGCATTTTCAAATCAATATATG | AuNP Capture |
| 6[243] | AAATTATTCATTAAAGGTGACAGAGAGAATAA | AuNP Capture |
| 6[349] | ATAATAATTTTTTCACGTTCTGCCTATTTCGG | AuNP Capture |


| 6[455] | ATAGCGTCCAATACTGCGGGCTGGCTGACCTT | AuNP Capture |
| :---: | :---: | :---: |
| 6[561] | TTAACCAATAGGAACGCCAAGCCTCAGAGCAT | AuNP Capture |
| 4[109] | CATTTTGCGGAACACAAACAATTCGACAAAAG | Dimer connector |
| 5[110] | GTAACTTCAGGTTTAACGTTTTAAAAGTTTGA | Dimer connector |
| 8[39] | AATAAAGAAACATCAACGAATTATTCATTTCAATTACC | Dimer connector |
| 10[17] | TGAGCAACTCGTATTAAATCCTTTGCCCGCCTCAATCAATAGGATTTAGAAG | Dimer connector |
| 11[18] | AAGATGATGAAACAAATTGCGTAGATTATTAT | Dimer connector |
| 4[109] | CATTTTGCGGAACACAAACAATTCGACGCCAG | Trimer connector |
| 4[215] | ATTTACGAGCATGTCAGCTAATGCAGAAAAAG | Trimer connector |
| 5[110] | GTAACGCGAGGCGTTTTACAAGAAAAATAATA | Trimer connector |
| 5[216] | TCCCATTCAGGTTTAACGTTTTAAAAGTTTGA | Trimer connector |
| 8[39] | AATAAAGAAACATCAACGAATTATTCATTTCAATTACC | Trimer connector |
| 8[145] | CTTATCCGGCCATATTAACGAGCGTCTTTCCAGAGCCT | Trimer connector |
| 10[17] | TGAGCACGCGCCTGTTTATCAACAATAGACAGTAGGGCTTAACGACGACAAT | Trimer connector |
| 10[123] | AATTTAACTCGTATTAAATCCTTTGCCCGCCTCAATCAATAGGATTTAGAAG | Trimer connector |
| 11[18] | AAGATGATGAAACAAATTGCGTAGATTTCCTA | Trimer connector |
| 11[124] | TTACAAAATAAACAGTATTCTAAGAACATTAT | Trimer connector |
| 4[109] | CATTTTGCGGAACACAAACAATTCGACATGGC | Tetramer connector |
| 4[321] | AGCGTCAGACTGTAGCCGGAAACGTCAAAAAG | Tetramer connector |
| 5[110] | GTAACCCAGAGCCGCCGCAGCGACAGAATCAA | Tetramer connector |
| 5[322] | GTTTGTTCAGGTTTAACGTTTTAAAAGTTTGA | Tetramer connector |
| 8[39] | AATAAAGAAACATCAACGAATTATTCATTTCAATTACC | Tetramer connector |
| 8[251] | GAGCCGCCGAGTGTACAATTTACCGTTCCAGTAAGCGT | Tetramer connector |
| 10[17] | TGAGCCCAATGAAACCATCGATAGCAGCAAAAAGGGCGACAGTAGCACCATT | Tetramer connector |
| 10[229] | CATACAACTCGTATTAAATCCTTTGCCCGCCTCAATCAATAGGATTTAGAAG | Tetramer connector |
| 11[18] | AAGATGATGAAACAAATTGCGTAGATTCCTTT | Tetramer connector |
| 11[230] | TTTTGATGATACAGACCAGAACCACCAATTAT | Tetramer connector |
| 4[109] | CATTTTGCGGAACACAAACAATTCGACCCGGA | 1D connector |
| 4[427] | CGCTGAGGCTTGCAAAACAGCTTGATAAAAAG | 1D connector |
| 5[110] | GTAACAAGAGGCAAAAGACCACGCATAACCGA | 1D connector |
| 5[428] | TATATTTCAGGTTTAACGTTTTAAAAGTTTGA | 1D connector |
| 8[39] | AATAAAGAAACATCAACGAATTATTCATTTCAATTACC | 1D connector |
| 8[357] | GAAGGCACGTCAATCAATCCGCGACCTGCTCCATGTTA | 1D connector |
| 10[17] | TGAGCCCGATAGTTGCGCCGACAATGACAACAGTTTCAGCGCTTGCTTTCGA | 1D connector |
| 10[335] | CTTAGAACTCGTATTAAATCCTTTGCCCGCCTCAATCAATAGGATTTAGAAG | 1D connector |
| 11[18] | AAGATGATGAAACAAATTGCGTAGATTTCGGT | 1D connector |
| 11[336] | ACGAGGCGCAGACGCAACCTAAAACGAATTAT | 1D connector |

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