Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Tunable Optical Activity of Plasmonic Dimers Assembled by DNA Origami

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Experimental Methods

1. Chemicals

$HAuCl_4 \cdot 3H_2O$, Hexadecyltrimethyl ammonium bromide (CTAB), Sodium dodecyl sulfate (SDS), Tris (2-carboxyethyl) phosphine hydrochloride (TCEP), Bis (p-sulfonatophenyl) phenyl phosphine dihydrate dipotassium salt (BSPP), Silver nitrate ($AgNO_3$), Sodium borohydride ($NaBH_4$), L-ascorbic acid, Trisodium citrate dehydrate, and Citric acid were purchased from Sigma-Aldrich. All chemicals were used as received without further purifications. All oligonucleotides were purchased from Invitrogen.

2. Preparation of DNA origami structures

Rectangular origami templates were prepared according to reference 1 with several changes. All of the side staples were left out to prevent aggregation via helix stacking interactions between adjacent origami. Additionally, several groups of different capture strands were designed to assemble AuNRs on the origami templates. These capture strands were extended on the 5' end by 15 bases with specific sequences. The rectangular DNA origami was annealed at a ratio of 1:5:8 for the M13 strand, capture strands, and staples strands. The DNA origami was subsequently assembled in a 1×TAE/Mg^{2+} buffer (Tris, 40 mM; Acetic acid, 20 mM; EDTA, 2 mM; and Magnesium acetate, 12.5 mM; pH 8.0) by cooling slowly from 90 °C to room temperature. The DNA origami was then filtered with 100 kDa (MWCO) centrifuge filters to remove the extra capture strands and staple strands for three times.

3. Preparation and functionalization of the AuNRs

a. A two-step seed-mediated growth method was used to synthesize AuNRs according to reference 2.

Synthesis of the AuNR seeds: 50 μL of 2% (w/v) $HAuCl_4$ solution and 7.5 mL of 100 mM CTAB solution were added to a 25 mL flask. A 600 μL of ice-cold $NaBH_4$ solution (10 mM) was added to the flask under vigorous stirring. The color of the solution changed quickly to yellowish brown. The resulting solution acted as nucleation seeds for the AuNR synthesis in the next step.

Growth of AuNRs: An 80 μL of 2% (w/v) $HAuCl_4$ solution was added to a 10 mL of 100 mM CTAB solution. 75μL of 10 mM $AgNO_3$ solution and 50 μL of 100 mM L-ascorbic acid solution were added to the flask in order. A 20 μL seed solution was added to the mixture solution under stirring. The final solution was kept undisturbed at 28 °C about 5 hours after being stirred for 1 minute.

b. Purification of the AuNRs

A 10 mL AuNR solution was centrifuged at 8000 rpm for 30 minutes and then the supernatant was removed with a pipette. The pellet was suspended in 10 mL water. The solution was centrifuged at 3000 rpm for 25 minutes and the supernatant was collected. The AuNR solution was then centrifuged at 8000 rpm for 30 minutes. The supernatant was discarded. The pellet was suspended in 100 μL water. The concentration was measured by a UV spectrometer.

c. Preparation of the AuNRs modified with thiolated DNA

The disulfide bonds in the thiolated oligonucleotides were reduced to monothiol using TCEP (20mM, 1h) in water. The oligonucleotides were purified using size exclusion columns (G-25, GE Healthcare) to remove small molecules. The purified DNA was added to the AuNR solution (OD~1) containing 0.01% (w/v) SDS with a molecular ratio of 3000: 1. The mixture solution was incubated for 6 hours at 28 °C. Then the buffer concentration was adjusted to 1× TBE (89 mM Tris, 89 mM boric acid, 2 mM EDTA, pH 8.0) from a 10× TBE buffer. After that, a 5M NaCl solution was slowly added until the final concentration of NaCl reached 500 mM in 24 hours. Subsequently, the AuNR-DNA conjugates were centrifuged at 8000 rpm for 25 minutes. The pellet was suspended in a 1 mL 0.5× TBE buffer containing 200 mM NaCl while the supernatant was discarded. The same centrifugation procedure was repeated three times to completely remove the excess thiolated DNA.

4. Preparation and functionalization of the AuNSs

a. Synthesis of the AuNSs.

AuNSs were synthesized by a one-step method. A 1.25 mL $HAuCl_4$ solution (0.2%, w/v) was diluted in 25 mL double-distilled water and heated to boiling. A 1 mL sodium citric solution (1%, w/v; containing 0.05% critic acid) was added to the flask under vigorous stirring. The solution in the flask was kept boiling for 5 minutes under stirring and then cooled at room temperature.

b. Surface modification of the AuNSs with BSPP.

BSPP (15 mg) was added to the Au colloidal solution (50 mL) and the mixture was shaken overnight at room temperature. Sodium Chloride (solid) was added slowly to this mixture solution while stirring until the solution color was changed from deep burgundy to light purple. The resulting mixture was centrifuged at 3000 rpm for 30 min and the supernatant was carefully removed with a pipette. AuNSs were then resuspended in a 1 mL BSPP solution (2.5 mM). Upon mixing with 1mL methanol, the mixture was centrifuged. The supernatant was removed and the AuNSs were resuspended again in a 1 mL BSPP solution (2.5 mM). The concentration of the AuNSs was estimated according to the optical absorption at ~ 523 nm. Phosphine coating increases the amount of negative charges on the AuNS surface and therefore stabilizes the AuNSs at high electrolyte concentrations.

c. Preparation of AuNS-DNA conjugates.

AuNS-DNA conjugates were prepared following a regular protocol. The disulfide bond in the thiol-modified oligonucleotides was reduced to monothiol using TCEP (20 mM, 1 hour) in water. The oligonucleotides were purified using size exclusion columns (G-25, GE Healthcare) to remove small molecules. Monothiol modified oligonucleotides and BSPP modified AuNSs were then incubated in a DNA to Au molar ratio of more than 300:1 in a 1 × TBE buffer solution (89 mM Tris, 89 mM boric acid, 2 mM EDTA, pH 8.0, containing 50 mM NaCl) for 20 hours at room temperature. The concentration of NaCl was slowly increased to 300 mM in the subsequent 20 hours in order to increase the binding density of thiolated DNA on AuNSs. AuNS-DNA conjugates were then washed using a 1×TBE buffer solution in 100 kDa (MWCO) centrifuge filters to remove the extra free oligonucleotides. The concentration of the AuNS-DNA conjugates was estimated according to the optical absorption at ~ 523 nm. Freshly prepared, fully coated AuNSs do not precipitate in a 1×TAE/Mg^{2+} buffer, preferable for the formation of DNA origami.

4. Self-assembly of the AuNRs/AuNSs on the DNA origami templates and electrophoretic gel purification

The purified rectangular DNA origami structures were mixed with the AuNR/AuNS-DNA conjugates. The mixture was annealed from 45 °C to 25 °C for 30 cycles over 48 hours. The annealed product of the DNA origami assembled AuNRs was loaded in a 1% Ethidium Bromide stained agarose gel (running buffer 1× TAE/Mg^{2+}, loading buffer 60% glycerol, 15 V/cm). Selected bands were cut out and the DNA origami assembled AuNRs were extracted from the gel with Freeze-Squeeze columns (Bio-Rad) at 4 °C or by electro-elution with dialysis tubing membranes (MWCO: 50K).

5. TEM characterization of the DNA origami assembled AuNRs/AuNSs

The samples for TEM imaging were prepared by depositing a 3 µL of the sample solution on a carbon-coated TEM grid. After 15 min, the excess solution was wicked from the grid using filter paper. To remove the deposited salt, the grid was washed with a droplet of water and the excess water was wicked away using filter paper. A droplet of a 1% uranyl acetate solution was used to treat the grid and the excess solution was wicked away. Then, the grid was treated with a second droplet of the uranyl acetate solution for 40 seconds and the excess solution was removed using filter paper. All the grids were kept at room temperature. TEM studies were conducted using a Tecnei G2-20S TWIN (200 kV). AuNRs in a few of the dimers had deformation, probably because of the drying process on the TEM grids.

6. Circular Dichroism measurements

The CD spectra were measured by a Jasco J-815 spectropolarimeter using the following parameters: wavelength range, 200 nm to 900 nm; optical length, 0.5 cm; scanning speed, 200 nm/min.

Additional Figures

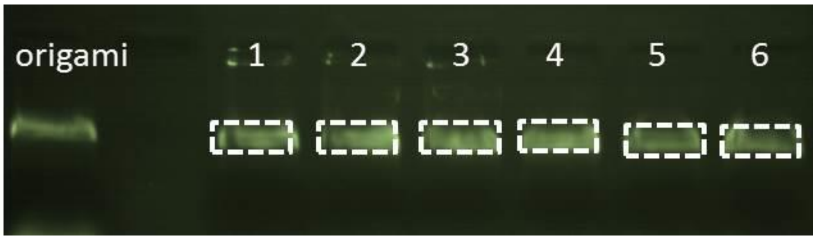
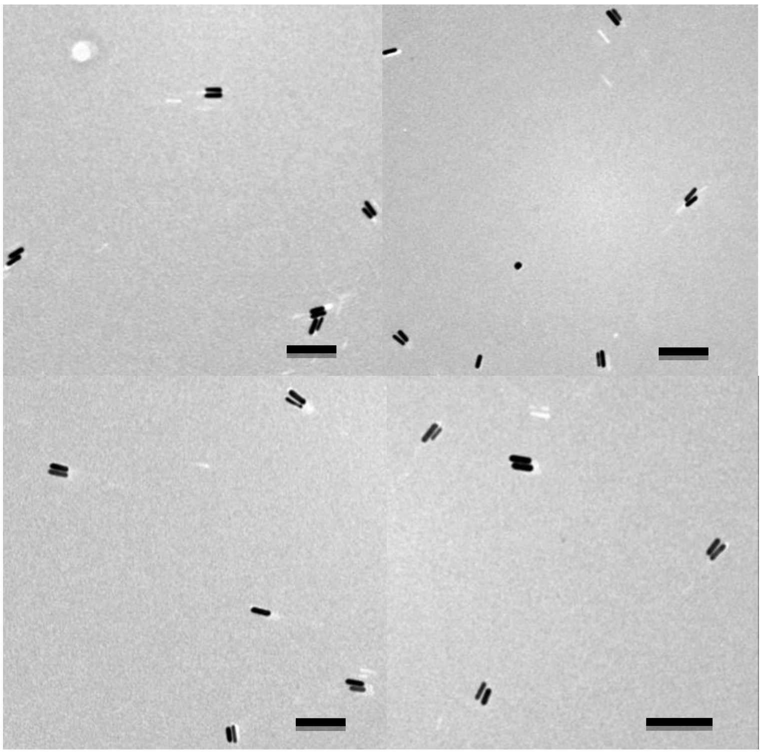
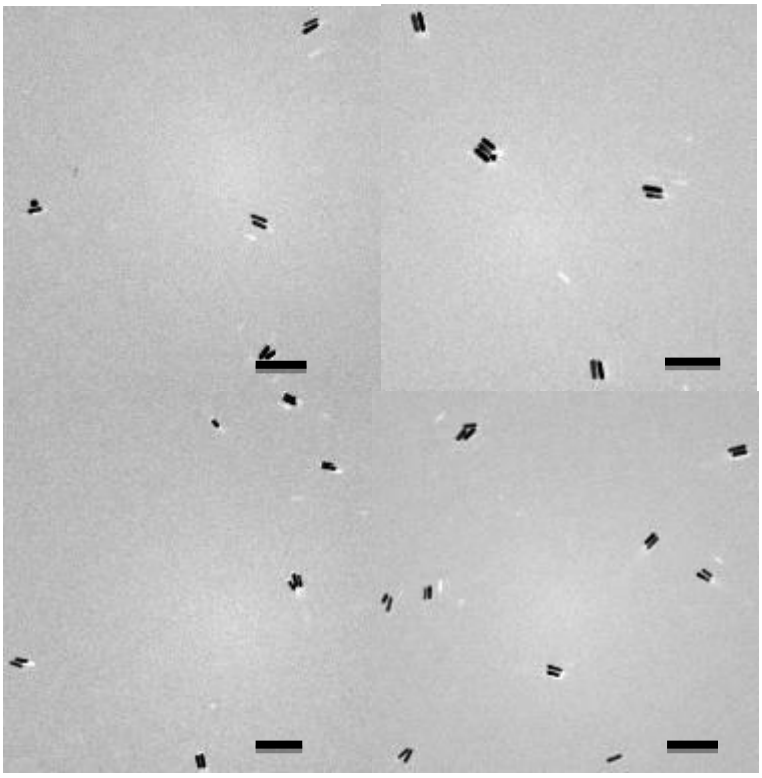


Figure S1 1% Ethidium Bromide stained agarose gel image of each design. The leftmost Lane is origami only. Others are the AuNR dimers assembled on rectangular origami, which correspond to nanoarchitectures (1)-(6) in Figure 1b. The target products are contained in the bands highlighted by the white dashed lines.

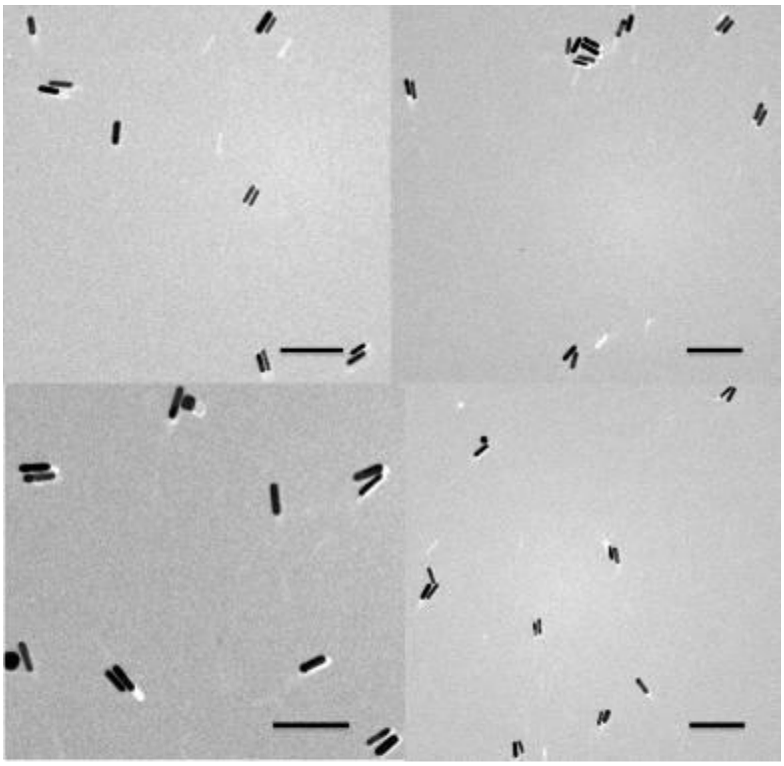
Additional TEM images of the AuNR dimer (1)



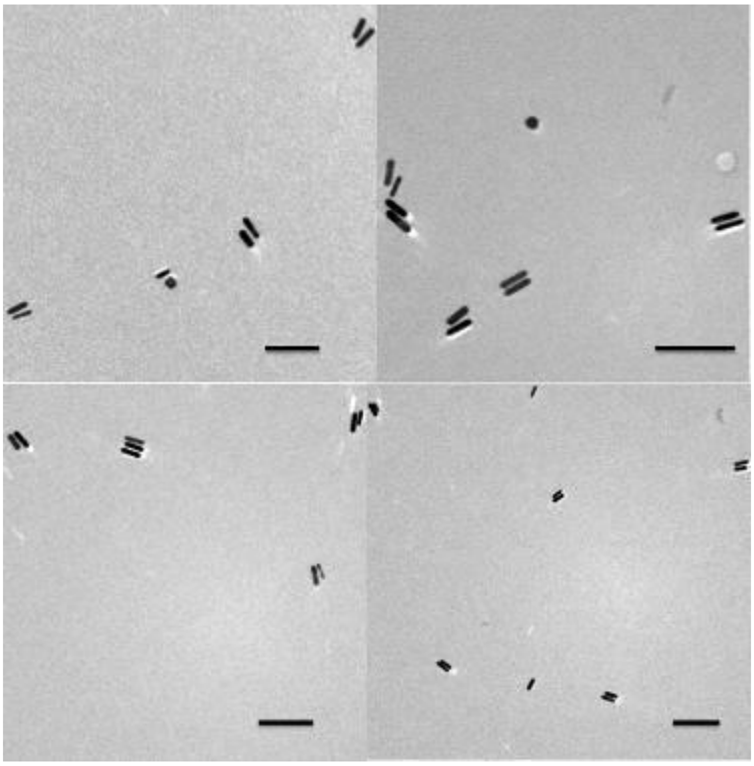
Additional TEM images of the AuNR dimer (2)



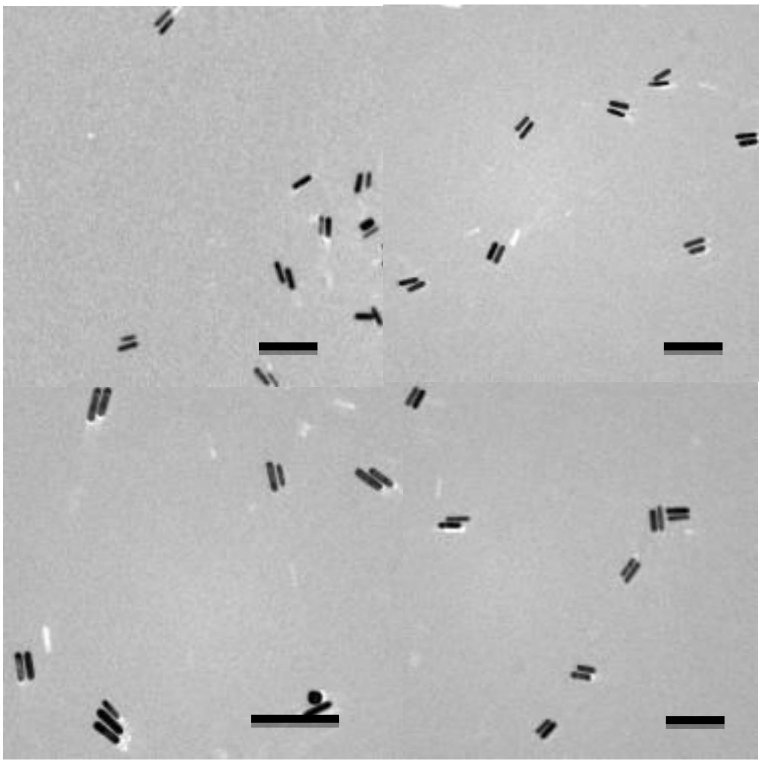
Additional TEM images of the AuNR dimer (3)



Additional TEM images of the AuNR dimer (4)



Additional TEM images of the AuNR dimer (5)



Additional TEM images of the AuNR dimer (6)

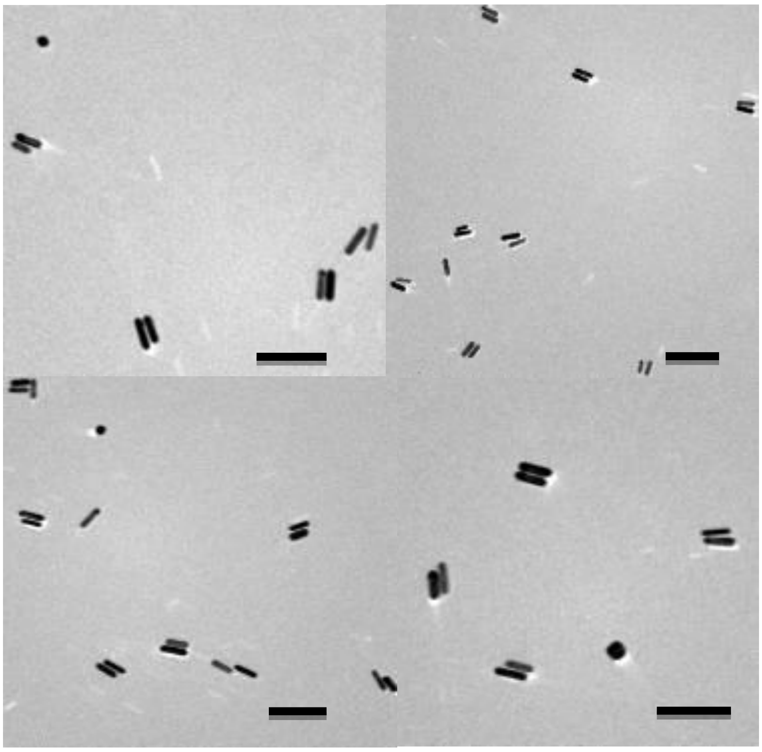


Figure S2 Additional TEM images of the AuNR dimers correspond to nanoarchitectures (1)-(6) in Figure 1b (scale bar: 100 nm).

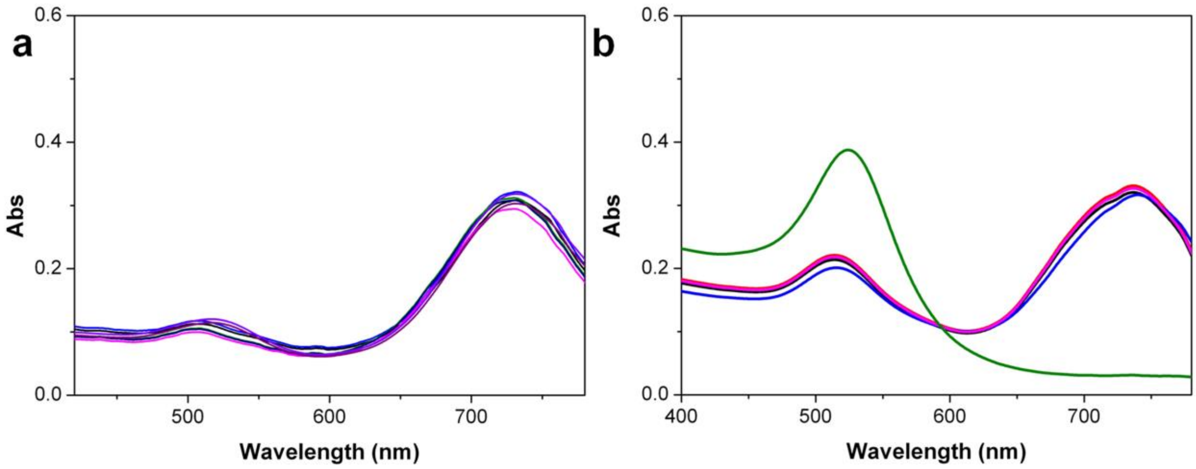


Figure S3 (a)Absorption spectra of six types of AuNR dimer nanoarchitectures. Different colors correspond to nanoarchitectures shown in Figure 3. (b)Absorption spectra of the four types of AuNR-AuNS heterodimers and the AuNS. Different colors correspond to the nanoarchitectures shown in Figure 4.

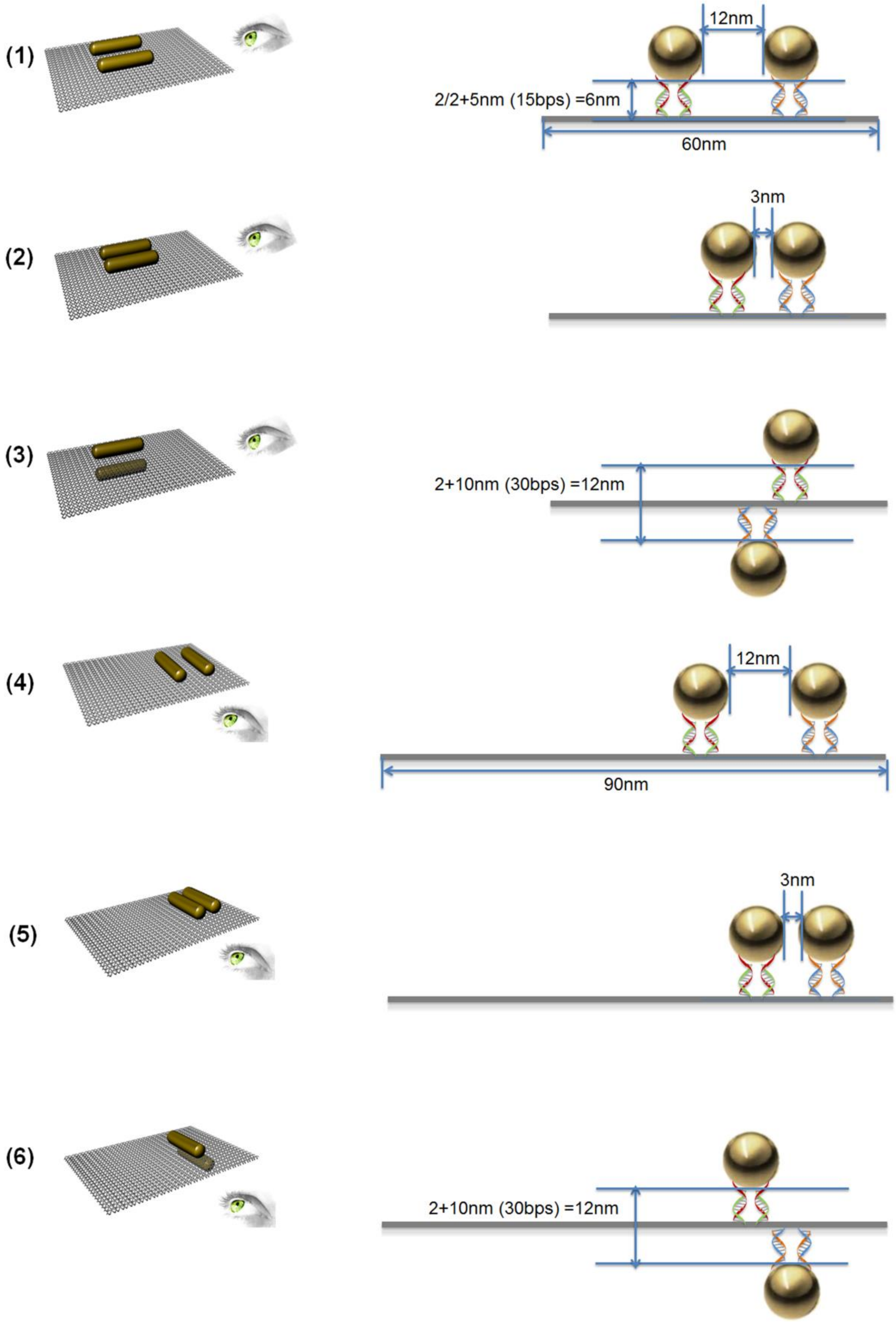
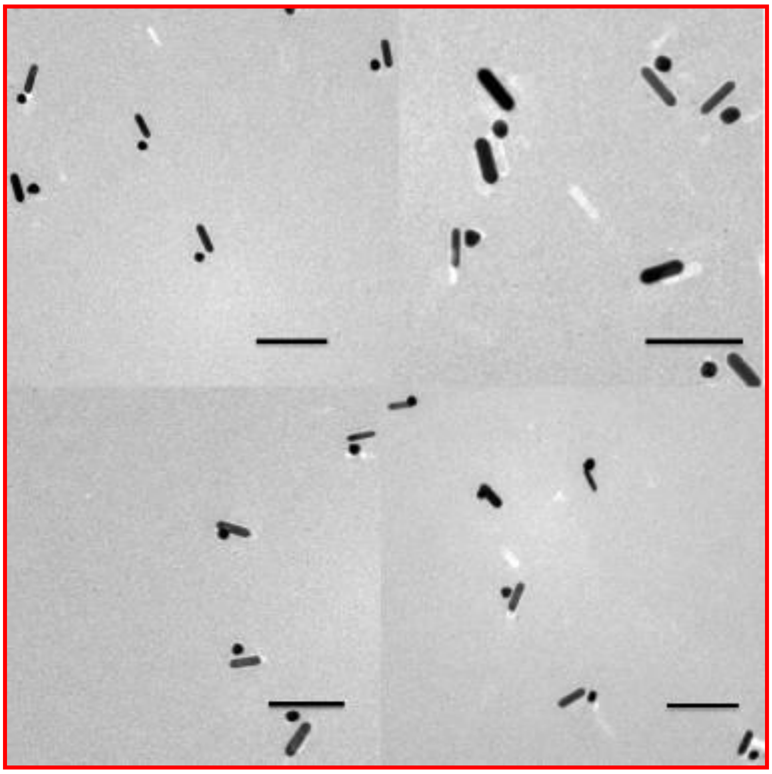
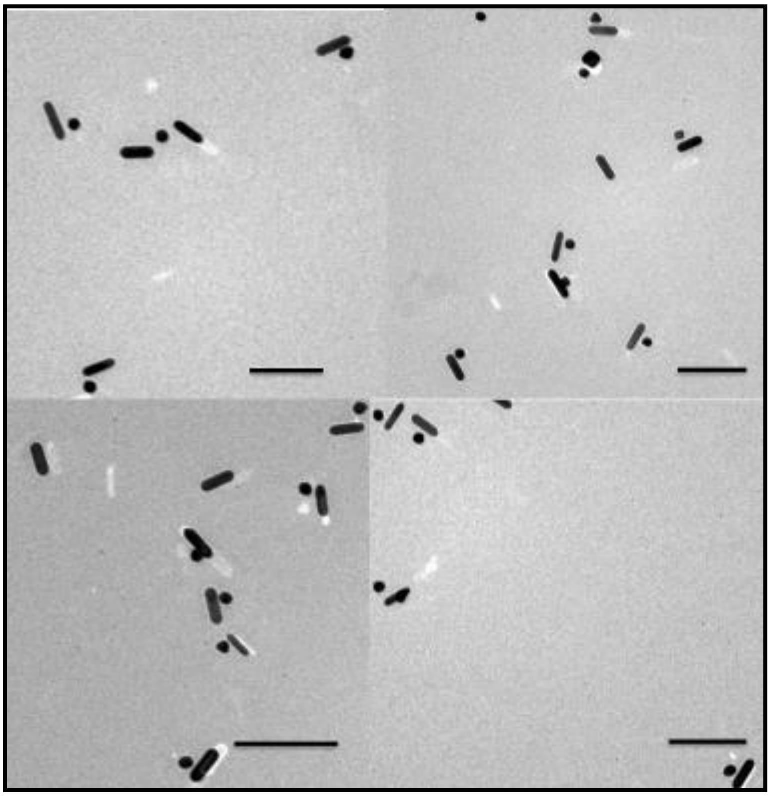
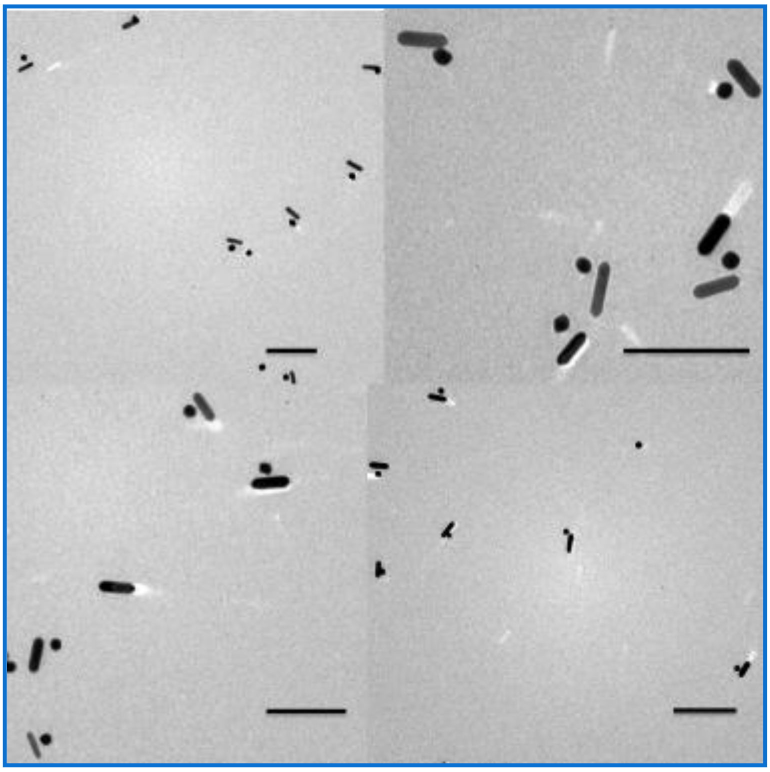


Figure S4 Diagram of six types of AuNR dimers assembled in different configuration







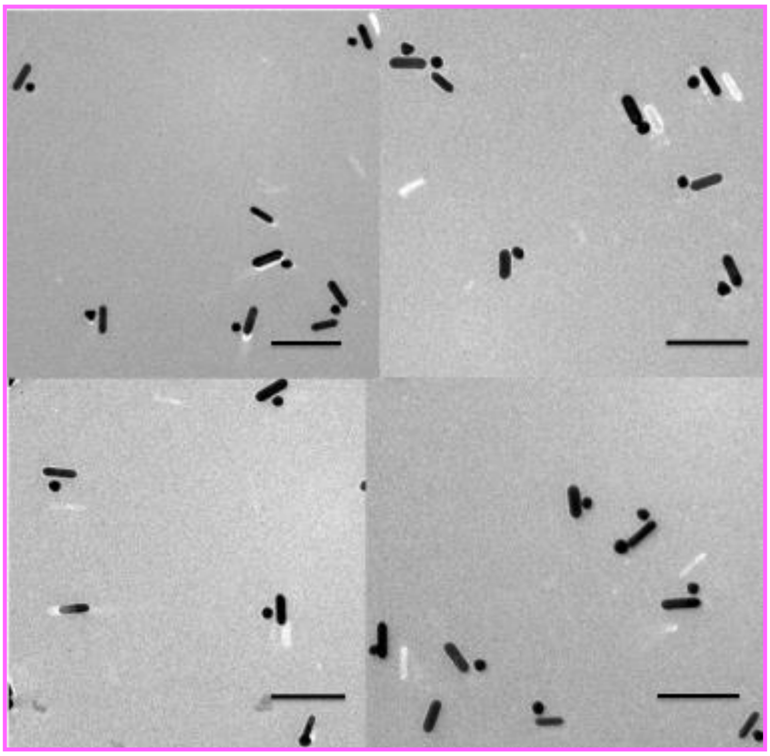


Figure S5 Additional TEM images of the AuNS-AuNR nanoarchitectures (scale bar: 100 nm). Different colors correspond to the nanoarchitectures shown in Figure 4.

Theoretical Methods

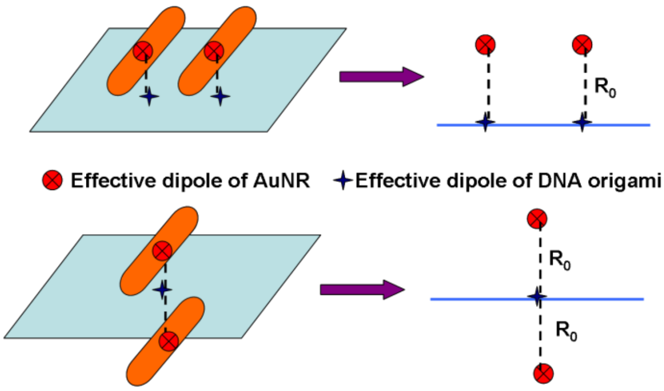


Figure S6 Schematic diagram of the theoretical model.

The optical response of the nanocomplexes is determined by the interactions among the effective dipoles (see figure S6). In our system, the AuNR/AuNS is represented by a dipole at its center with dipole moment

$\vec{p}_M = \alpha_M \vec{E}_0$, and polarizability

$$\alpha_M = V_{AuNR} \frac{\varepsilon_M - \varepsilon_0}{\varepsilon_0 + L_3(\varepsilon_M - \varepsilon_0)}, \qquad \text{(S1)}$$

where ε_0 and ε_M the dielectric constants for the background and for the bulk Au and

$$L_3 = \frac{1-e^2}{e^2}\left(\frac{1}{2e}\ln\frac{1+e}{1-e} - 1\right), \qquad \text{(S2)}$$

the geometrical factor of AuNR viewed as ellipsoid with $e = \sqrt{1-(b/a)^2}$, a , b the length of long and short axis. The interaction between two AuNRs (with polarizabilities α_{M1} and α_{M2}) leads to the effective dipole with renormalized polarizabilities $\alpha_{M1}^e = \alpha_{M1}\frac{1-\alpha_{M2}T_0}{1-\alpha_{M1}\alpha_{M2}T_0^2}$,

$\alpha_{M2}^e = \alpha_{M2}\frac{1-\alpha_{M1}T_0}{1-\alpha_{M1}\alpha_{M2}T_0^2}$ (T_0 the interaction strength). In the following, we neglect the index e .

The DNA origami may be viewed as effective dipole(s) at positions right opposite to the center of AuNR(s) /AuNS(s). The response of DNA origami/effective dipole(s) can be described by the polarizability tensor defined by

$$\vec{p}_D = \hat{\alpha}_D \cdot \vec{E}_D, \qquad \text{(S3)}$$

where $\vec{E}_D$ the electric field on the dipole(s) with dipole moment $\vec{p}_D$, and the polarizability tensor

$$\hat{\alpha}_D = \begin{pmatrix} \alpha_x & -iG_z & iG_y \\ iG_z & \alpha_y & -iG_x \\ -iG_y & iG_x & \alpha_z \end{pmatrix}. \qquad \text{(S4)}$$

From the basic structure and anisotropic property of DNA origami, we may have $G_z = 0$, $\alpha_z = 0$ and $G_y = -G_x$. The electric field on the effective dipole of DNA origami is the summation of the incident field $\vec{E}_0$ and the field from AuNR/AuNS

$$\frac{q}{R^3}[3\vec{n}(\vec{n}\cdot\vec{p}_M) - \vec{p}_M], \qquad \text{(S5)}$$

where R the distance between the effective dipoles (Figure S6), $\vec{n}$ the unit vector along the centers between the DNA origami effective dipoles and the center of the AuNR/AuNS, $\vec{p}_M = \alpha_M \vec{E}_0$, and we have added a factor q to take the multipole effects into account. The dominant contribution of circular dichroism (CD) at visible light regime is the origami induced and AuNR amplified absorption difference between left and right circularly polarized light, i.e.,

$$CD = Q_L - Q_R \qquad \text{(S6)}$$

$$Q_{L/R} = \mathrm{Im}(\alpha_M)\cdot\omega\cdot|\vec{E}^{L/R^*}\cdot\vec{E}^{L/R}|^2 \qquad \text{(S7)}$$

$$\vec{E}^{L/R} = \vec{E}_0^{L/R} + \frac{1}{R^3}[3\vec{n}(\vec{n}\cdot\vec{p}_D^{L/R}) - \vec{p}_D^{L/R}] \;, \qquad \text{(S8)}$$

where $\vec{E}_0^{L/R}$ and $\vec{p}_D^{L/R}$ are the left/right circularly polarized incident field and the induced dipole of DNA origami in the presence of incident field. The total CD of nanorod/nanosphere is $CD = CD_1 + CD_2$, CD_i ($i = 1,2$) the CD for the i-th nanorod/nanosphere,

$$\begin{aligned} CD_i = {} & 2G_yV_i\,\mathrm{Im}(\alpha_z^i)/R_0^3\,\mathrm{Re}(K_x - K_y) - G_yV_i\,\mathrm{Im}(\alpha_z^i)/R_0^3\,\mathrm{Re}\{[1-\alpha_xK_x/R_0^3]K_z^*\} \\ & + G_yV_i\,\mathrm{Im}(\alpha_y^i)/R_0^3\,\mathrm{Re}\{[1-\alpha_yK_y/R_0^3]K_z^*\}\,|\vec{E}_0|^2, \end{aligned} \qquad \text{(S9)}$$

where $K_x = 1 - q\alpha_x^1/R_0^3 - q\alpha_x^2/R_0^3$, $K_y = 1 - q\alpha_y^1/R_0^3 - q\alpha_y^2/R_0^3$, $K_z = 1 + 2q\alpha_z^1/R_0^3 + 2q\alpha_z^2/R_0^3$, R_0 the distance from AuNR/AuNS center to the DNA origami surface.

In the numerical calculation, we have used L_3=0.0985 for AuNR (L_3=1/3 for AuNS) from equation (S2) based on 2a=39nm, 2b=12nm. We have used a Gaussian distribution of L_3 with width 0.024 to take into account the shape fluctuation of nanorods/nanoparticles. We have chosen $q = 5.18$, α_x =10.0, α_y =10.0, $\varepsilon_0 = 2.32$ and taken the values of ε_M from reference 3.

DNA Strands Used in the Experiment

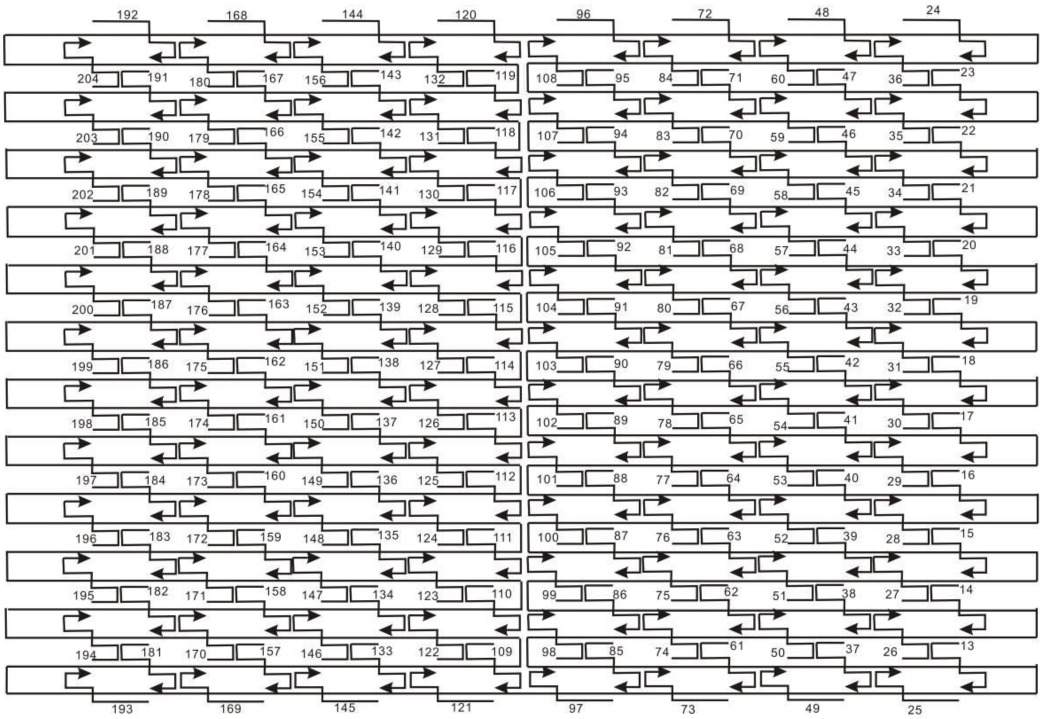


Figure S7 The basic rectangular DNA origami used in present study without being functionalized with capture strands.

1. Sequences used in the assembly of the DNA origami template (left to right 5'-3')

13 TGGTTTTTAACGTCAAAGGGCGAAGAACCATC
14 CTTGCATGCATTAATGAATCGGCCCGCCAGGG
15 TAGATGGGGGGTAACGCCAGGGTTGTGCCAAG
16 CATGTCAAGATTCTCCGTGGGAACCGTTGGTG
17 CTGTAATATTGCCTGAGAGTCTGGAAAACTAG
18 TGCAACTAAGCAATAAAGCCTCAGTTATGACC
19 AAACAGTTGATGGCTTAGAGCTTATTTAAATA
20 ACGAACTAGCGTCCAATACTGCGGAATGCTTT
21 CTTTGAAAAGAACTGGCTCATTATTTAATAAA
22 ACGGCTACTTACTTAGCCGGAACGCTGACCAA
23 GAGAATAGCTTTTGCGGGATCGTCGGGTAGCA
24 ACGTTAGTAAATGAATTTTCTGTAAGCGGAGT
25 ACCCAAATCAAGTTTTTTGGGGTCAAAGAACG
26 TGGACTCCCTTTTCACCAGTGAGACCTGTCGT
27 GCCAGCTGCCTGCAGGTCGACTCTGCAAGGCG

28 ATTAAGTTCGCATCGTAACCGTGCGAGTAACA
29 ACCCGTCGTCATATGTACCCCGGTAAAGGCTA
30 TCAGGTCACTTTTGCGGGAGAAGCAGAATTAG
31 CAAAATTAAAGTACGGTGTCTGGAAGAGGTCA
32 TTTTTGCGCAGAAAACGAGAATGAATGTTTAG
33 ACTGGATAACGGAACAACATTATTACCTTATG
34 CGATTTTAGAGGACAG ATGAACGGCGCGACCT
35 GCTCCATGAGAGGCTT TGAGGACTAGGGAGTT
36 AAAGGCCGAAAGGAACAACTAAAGCTTTCCAG
37 AGCTGATTACAAGAGTCCACTATTGAGGTGCC
38 CCCGGGTACTTTCCAGTCGGGAAACGGGCAAC
39 GTTTGAGGGAAAGGGGGATGTGCTAGAGGATC
40 AGAAAAGCAACATTAAATGTGAGCATCTGCCA
41 CAACGCAATTTTTGAGAGATCTACTGATAATC
42 TCCATATACATACAGGCAAGGCAACTTTATTT
43 CAAAAATCATTGCTCCTTTTGATAAGTTTCAT
44 AAAGATTCAGGGGGTAATAGTAAACCATAAAT
45 CCAGGCGCTTAATCATTGTGAATTACAGGTAG
46 TTTCATGAAAATTGTGTCGAAATCTGTACAGA
47 AATAATAAGGTCGCTGAGGCTTGCAAAGACTT
48 CGTAACGATCTAAAGTTTTGTCGTGAATTGCG
49 GTAAAGCACTAAATCGGAACCCTAGTTGTTCC
50 AGTTTGGAGCCCTTCACCGCCTGGTTGCGCTC
51 ACTGCCCGCCGAGCTCGAATTCGTTATTACGC
52 CAGCTGGCGGACGACGACAGTATCGTAGCCAG
53 CTTTCATCCCCAAAAACAGGAAGACCGGAGAG
54 GGTAGCTAGGATAAAAATTTTTAGTTAACATC
55 CAATAAATACAGTTGATTCCCAATTTAGAGAG
56 TACCTTTAAGGTCTTTACCCTGACAAAGAAGT
57 TTTGCCAGATCAGTTGAGATTTAGTGGTTTAA
58 TTTCAACTATAGGCTGGCTGACCTTGTATCAT
59 CGCCTGATGGAAGTTTCCATTAAACATAACCG
60 ATATATTCTTTTTTCACGTTGAAAATAGTTAG
61 GAGTTGCACGAGATAGGGTTGAGTAAGGGAGC
62 TCATAGCTACTCACATTAATTGCGCCCTGAGA
63 GAAGATCGGTGCGGGCCTCTTCGCAATCATGG
64 GCAAATATCGCGTCTGGCCTTCCTGGCCTCAG
65 TATATTTTAGCTGATAAATTAATGTTGTATAA
66 CGAGTAGAACTAATAGTAGTAGCAAACCCTCA
67 TCAGAAGCCTCCAACAGGTCAGGATCTGCGAA
68 CATTCAACGCGAGAGGCTTTTGCATATTATAG
69 AGTAATCTTAAATTGGGCTTGAGAGAATACCA
70 ATACGTAAAAGTACAACGGAGATTTCATCAAG
71 AAAAAAGGACAACCATCGCCCACGCGGTAAA

72 TGTAGCATTCCACAGACAGCCCTCATCTCCAA
73 CCCCGATTTAGAGCTTGACGGGGAAATCAAAA
74 GAATAGCCGCAAGCGGTCCACGCTCCTAATGA
75 GTGAGCTAGTTTCCTGTGTGAAATTTGGGAAG
76 GGCGATCGCACTCCAGCCAGCTTTGCCATCAA
77 AAATAATTTTAAATTGTAAACGTTGATATTCA
78 ACCGTTCTAAATGCAATGCCTGAGAGGTGGCA
79 TCAATTCTTTTAGTTTGACCATTACCAGACCG
80 GAAGCAAAAAAGCGGATTGCATCAGATAAAAA
81 CCAAAATATAATGCAGATACATAAACACCAGA
82 ACGAGTAGTGACAAGAACCGGATATACCAAGC
83 GCGAAACATGCCACTACGAAGGCATGCGCCGA
84 CAATGACACTCCAAAAGGAGCCTTACAACGCC
85 CCAGCAGGGGCAAAATCCCTTATAAAGCCGGC
86 GCTCACAATGTAAAGCCTGGGGTGGGTTTGCC
87 GCTTCTGGTCAGGCTGCGCAACTGTGTTATCC
88 GTTAAAATTTTAACCAATAGGAACCCGGCACC
89 AGGTAAAGAAATCACCATCAATATAATATTTT
90 TCGCAAATGGGGCGCGAGCTGAAATAATGTGT
91 AAGAGGAACGAGCTTCAAAGCGAAGATACATT
92 GGAATTACTCGTTTACCAGACGACAAAAGATT
93 CCAAATCACTTGCCCTGACGAGAACGCCAAAA
94 AAACGAAATGACCCCCAGCGATTATTCATTAC
95 TCGGTTTAGCTTGATACCGATAGTCCAACCTA
96 TGAGTTTCGTCACCAGTACAAACTTAATTGTA
97 GAACGTGGCGAGAAAGGAAGGGAACAAACTAT
98 CCGAAATCCGAAAATCCTGTTTGAAGCCGGAA
99 GCATAAAGTTCCACACAACATACGAAGCGCCA
100 TTCGCCATTGCCGGAAACCAGGCATTAAATCA
101 GCTCATTTTCGCATTAAATTTTTGAGCTTAGA
102 AGACAGTCATTCAAAAGGGTGAGAAGCTATAT
103 TTTCATTTGGTCAATAACCTGTTTATATCGCG
104 TTTTAATTGCCCGAAAGACTTCAAAACACTAT
105 CATAACCCGAGGCATAGTAAGAGCTTTTTAAG
106 GAATAAGGACGTAACAAAGCTGCTCTAAAACA
107 CTCATCTTGAGGCAAAAGAATACAGTGAATTT
108 CTTAAACATCAGCTTGCTTTCGAGCGTAACAC
109 ACGAACCAAAACATCGCCATTAAATGGTGGTT
110 CGACAACTAAGTATTAGACTTTACAATACCGA
111 CTTTTACACAGATGAATATACAGTAAACAATT
112 TTAAGACGTTGAAAACATAGCGATAACAGTAC
113 GCGTTATAGAAAAAGCCTGTTTAGAAGGCCGG
114 ATCGGCTGCGAGCATGTAGAAACCTATCATAT
115 CCTAATTTACGCTAACGAGCGTCTAATCAATA

116 AAAAGTAATATCTTACCGAAGCCCTTCCAGAG
117 TTATTCATAGGGAAGGTAAATATTCATTCAGT
118 GAGCCGCCCCACCACCGGAACCGCGACGGAAA
119 AATGCCCCGTAACAGTGCCCGTATCTCCCTCA
120 CAAGCCCAATAGGAACCCATGTACAAACAGTT
121 CGGCCTTGCTGGTAATATCCAGAACGAACTGA
122 TAGCCCTACCAGCAGAAGATAAAAACATTTGA
123 GGATTTAGCGTATTAAATCCTTTGTTTTCAGG
124 TTTAACGTTCGGGAGAAACAATAATTTTCCCT
125 TAGAATCCCTGAGAAGAGTCAATAGGAATCAT
126 AATTACTACAAATTCTTACCAGTAATCCCATC
127 CTAATTTATCTTTCCTTATCATTCATCCTGAA
128 TCTTACCAGCCAGTTACAAAATAAATGAAATA
129 GCAATAGCGCAGATAGCCGAACAATTCAACCG
130 ATTGAGGGTAAAGGTGAATTATCAATCACCGG
131 AACCAGAGACCCTCAGAACCGCCAGGGGTCAG
132 TGCCTTGACTGCCTATTTCGGAACAGGGATAG
133 AGGCGGTCATTAGTCTTTAATGCGCAATATTA
134 TTATTAATGCCGTCAATAGATAATCAGAGGTG
135 CCTGATTGAAAGAAATTGCGTAGACCCGAACG
136 ATCAAAATCGTCGCTATTAATTAACGGATTCG
137 ACGCTCAAAATAAGAATAAACACCGTGAATTT
138 GGTATTAAGAACAAGAAAAATAATTAAAGCCA
139 ATTATTTAACCCAGCTACAATTTTCAAGAACG
140 GAAGGAAAATAAGAGCAAGAAACAACAGCCAT
141 GACTTGAGAGACAAAAGGGCGACAAGTTACCA
142 GCCACCACTCTTTTCATAATCAAACCGTCACC
143 CTGAAACAGGTAATAAGTTTTAACCCCTCAGA
144 CTCAGAGCCACCACCCTCATTTTCCTATTATT
145 CCGCCAGCCATTGCAACAGGAAAAATATTTTT
146 GAATGGCTAGTATTAACACCGCCTCAACTAAT
147 AGATTAGATTTAAAAGTTTGAGTACACGTAAA
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151 TAAGTCCTACCAAGTACCGCACTCTTAGTTGC
152 TATTTTGCTCCCAATCCAAATAAGTGAGTTAA
153 GCCCAATACCGAGGAAACGCAATAGGTTTACC
154 AGCGCCAACCATTTGGGAATTAGATTATTAGC
155 GTTTGCCACCTCAGAGCCGCCACCGATACAGG
156 AGTGTACTTGAAAGTATTAAGAGGCCGCCACC
157 GCCACGCTATACGTGGCACAGACAACGCTCAT
158 ATTTTGCGTCTTTAGGAGCACTAAGCAACAGT
159 GCGCAGAGATATCAAAATTATTTGACATTATC

160 TAACCTCCATATGTGAGTGAATAAACAAAATC
161 CATATTTAGAAATACCGACCGTGTTACCTTTT
162 CAAGCAAGACGCGCCTGTTTATCAAGAATCGC
163 TTTTGTTTAAGCCTTAAATCAAGAATCGAGAA
164 ATACCCAAGATAACCCACAAGAATAAACGATT
165 AATCACCAAATAGAAAATTCATATATAACGGA
166 CACCAGAGTTCGGTCATAGCCCCCGCCAGCAA
167 CCTCAAGAATACATGGCTTTTGATAGAACCAC
168 CCCTCAGAACCGCCACCCTCAGAACTGAGACT
169 GGAAATACCTACATTTTGACGCTCACCTGAAA
170 GCGTAAGAGAGAGCCAGCAGCAAAAAGGTTAT
171 CTAAAATAGAACAAAGAAACCACCAGGGTTAG
172 AACCTACCGCGAATTATTCATTTCCAGTACAT
173 AAATCAATGGCTTAGGTTGGGTTACTAAATTT
174 AATGGTTTACAACGCCAACATGTAGTTCAGCT
175 AATGCAGACCGTTTTTATTTTCATCTTGCGGG
176 AGGTTTTGAACGTCAAAAATGAAAGCGCTAAT
177 ATCAGAGAAAGAACTGGCATGATTTTATTTTG
178 TCACAATCGTAGCACCATTACCATCGTTTTCA
179 TCGGCATTCCGCCGCCAGCATTGACGTTCCAG
180 TAAGCGTCGAAGGATTAGGATTAGTACCGCCA
181 CTAAAGCAAGATAGAACCCTTCTGAATCGTCT
182 CGGAATTATTGAAAGGAATTGAGGTGAAAAAT
183 GAGCAAAAACTTCTGAATAATGGAAGAAGGAG
184 TATGTAAACCTTTTTTAATGGAAAAATTACCT
185 AGAGGCATAATTTCATCTTCTGACTATAACTA
186 TCATTACCCGACAATAAACAACATATTTAGGC
187 CTTTACAGTTAGCGAACCTCCCGACGTAGGAA
188 TTATTACGGTCAGAGGGTAATTGAATAGCAGC
189 CCGGAAACACACCACGGAATAAGTAAGACTCC
190 TGAGGCAGGCGTCAGACTGTAGCGTAGCAAGG
191 TGCTCAGTCAGTCTCTGAATTTACCAGGAGGT
192 TATCACCGTACTCAGGAGGTTTAGCGGGGTTT
193 GAAATGGATTATTTACATTGGCAGACATTCTG
194 GCCAACAGTCACCTTGCTGAACCTGTTGGCAA
195 ATCAACAGTCATCATATTCCTGATTGATTGTT
196 TGGATTATGAAGATGATGAAACAAAATTTCAT
197 TTGAATTATGCTGATGCAAATCCACAAATATA
198 TTTTAGTTTTTCGAGCCAGTAATAAATTCTGT
199 CCAGACGAGCGCCCAATAGCAAGCAAGAACGC
200 GAGGCGTTAGAGAATAACATAAAAGAACACCC
201 TGAACAAACAGTATGTTAGCAAACTAAAAGAA
202 ACGCAAAGGTCACCAATGAAACCAATCAAGTT
203 TGCCTTTAGTCAGACGATTGGCCTGCCAGAAT

204 GGAAAGCGACCAGGCGGATAAGTGAATAGGTG

The sequence of the single stranded circular M13mp18 viral DNA can be found at http://www.neb.com/nebecomm/tech_reference/restriction_enzyme/sequences/13mp18.txt

2. Capture strand sequences (left to right 5’-3’)

For assembly of AuNR dimers 1

For the first AuNR

176 AAAAAAAAAAAAAAA AGGTTTTGAACGTCAAAAATGAAAGCGCTAAT
177 AAAAAAAAAAAAAAA ATCAGAGAAAGAACTGGCATGATTTTATTTTG
152 AAAAAAAAAAAAAAA TATTTTGCTCCCAATCCAAATAAGTGAGTTAA
153 AAAAAAAAAAAAAAA GCCCAATACCGAGGAAACGCAATAGGTTTACC
128 AAAAAAAAAAAAAAA TCTTACCAGCCAGTTACAAAATAAATGAAATA
129 AAAAAAAAAAAAAAA GCAATAGCGCAGATAGCCGAACAATTCAACCG
104 AAAAAAAAAAAAAAA TTTTAATTGCCCGAAAGACTTCAAAACACTAT
105 AAAAAAAAAAAAAAA CATAACCCGAGGCATAGTAAGAGCTTTTTAAG
80 AAAAAAAAAAAAAAA GAAGCAAAAAAGCGGATTGCATCAGATAAAAA
81 AAAAAAAAAAAAAAA CCAAAATATAATGCAGATACATAAACACCAGA

For the second AuNR

76 ATAATAATAATAATA GGCGATCGCACTCCAGCCAGCTTTGCCATCAA
77 ATAATAATAATAATA AAATAATTTTAAATTGTAAACGTTGATATTCA
100 ATAATAATAATAATA TTCGCCATTGCCGGAAACCAGGCATTAAATCA
101 ATAATAATAATAATA GCTCATTTTCGCATTAAATTTTTGAGCTTAGA
124 ATAATAATAATAATA TTTAACGTTCGGGAGAAACAATAATTTTCCCT
125 ATAATAATAATAATA TAGAATCCCTGAGAAGAGTCAATAGGAATCAT
148 ATAATAATAATAATA ACAGAAATCTTTGAATACCAAGTTCCTTGCTT
149 ATAATAATAATAATA CTGTAAATCATAGGTCTGAGAGACGATAAATA
172 ATAATAATAATAATA AACCTACCGCGAATTATTCATTTCCAGTACAT
173 ATAATAATAATAATA AAATCAATGGCTTAGGTTGGGTTACTAAATTT

For assembly of AuNR dimers 2

For the first AuNR

28 AAAAAAAAAAAAAAA ATTAAGTTCGCATCGTAACCGTGCGAGTAACA
29 AAAAAAAAAAAAAAA ACCCGTCGTCATATGTACCCCGGTAAAGGCTA
32 AAAAAAAAAAAAAAA TTTTTGCGCAGAAAACGAGAATGAATGTTTAG
33 AAAAAAAAAAAAAAA ACTGGATAACGGAACAACATTATTACCTTATG
34 AAAAAAAAAAAAAAA CGATTTTAGAGGACAG ATGAACGGCGCGACCT
39 AAAAAAAAAAAAAAA GTTTGAGGGAAAGGGGGATGTGCTAGAGGATC
40 AAAAAAAAAAAAAAA AGAAAAGCAACATTAAATGTGAGCATCTGCCA
43 AAAAAAAAAAAAAAA CAAAAATCATTGCTCCTTTTGATAAGTTTCAT
44 AAAAAAAAAAAAAAA AAAGATTCAGGGGGTAATAGTAAACCATAAAT
45 AAAAAAAAAAAAAAA CCAGGCGCTTAATCATTGTGAATTACAGGTAG

For the second AuNR

76 ATAATAATAATAATA GGCGATCGCACTCCAGCCAGCTTTGCCATCAA
77 ATAATAATAATAATA AAATAATTTTAAATTGTAAACGTTGATATTCA
79 ATAATAATAATAATA TCAATTCTTTTAGTTTGACCATTACCAGACCG
80 ATAATAATAATAATA GAAGCAAAAAAGCGGATTGCATCAGATAAAAA

81 ATAATAATAATAATA CCAAAATATAATGCAGATACATAAACACCAGA
82 ATAATAATAATAATA AGTAATCTTAAATTGGGCTTGAGAGAATACCA
87 ATAATAATAATAATA GCTTCTGGTCAGGCTGCGCAACTGTGTTATCC
88 ATAATAATAATAATA GTTAAAATTTTAACCAATAGGAACCCGGCACC
90 ATAATAATAATAATA TCGCAAATGGGGCGCGAGCTGAAATAATGTGT

For assembly of AuNR dimers 3

For the first AuNR

176 AAAAAAAAAAAAAAA AGGTTTTGAACGTCAAAAATGAAAGCGCTAAT
177 AAAAAAAAAAAAAAA ATCAGAGAAAGAACTGGCATGATTTTATTTTG
152 AAAAAAAAAAAAAAA TATTTTGCTCCCAATCCAAATAAGTGAGTTAA
153 AAAAAAAAAAAAAAA GCCCAATACCGAGGAAACGCAATAGGTTTACC
128 AAAAAAAAAAAAAAA TCTTACCAGCCAGTTACAAAATAAATGAAATA
129 AAAAAAAAAAAAAAA GCAATAGCGCAGATAGCCGAACAATTCAACCG
104 AAAAAAAAAAAAAAA TTTTAATTGCCCGAAAGACTTCAAAACACTAT
105 AAAAAAAAAAAAAAA CATAACCCGAGGCATAGTAAGAGCTTTTTAAG
80 AAAAAAAAAAAAAAA GAAGCAAAAAAGCGGATTGCATCAGATAAAAA
81 AAAAAAAAAAAAAAA CCAAAATATAATGCAGATACATAAACACCAGA

For the second AuNR/AuNS

78 ATAATAATAATAATA ACCGTTCTAAATGCAATGCCTGAGAGGTGGCA
79 ATAATAATAATAATA TCAATTCTTTTAGTTTGACCATTACCAGACCG
102 ATAATAATAATAATA AGACAGTCATTCAAAAGGGTGAGAAGCTATAT
103 ATAATAATAATAATA TTTCATTTGGTCAATAACCTGTTTATATCGCG
126 ATAATAATAATAATA AATTACTACAAATTCTTACCAGTAATCCCATC
127 ATAATAATAATAATA CTAATTTATCTTTCCTTATCATTCATCCTGAA
150 ATAATAATAATAATA AGGCGTTACAGTAGGGCTTAATTGACAATAGA
151 ATAATAATAATAATA TAAGTCCTACCAAGTACCGCACTCTTAGTTGC
174 ATAATAATAATAATA AATGGTTTACAACGCCAACATGTAGTTCAGCT
175 ATAATAATAATAATA AATGCAGACCGTTTTTATTTTCATCTTGCGGG

For assembly of AuNR dimers 4

For the first AuNR

28 AAAAAAAAAAAAAAA ATTAAGTTCGCATCGTAACCGTGCGAGTAACA
29 AAAAAAAAAAAAAAA ACCCGTCGTCATATGTACCCCGGTAAAGGCTA
32 AAAAAAAAAAAAAAA TTTTTGCGCAGAAAACGAGAATGAATGTTTAG
33 AAAAAAAAAAAAAAA ACTGGATAACGGAACAACATTATTACCTTATG
34 AAAAAAAAAAAAAAA CGATTTTAGAGGACAGATGAACGGCGCGACCT
39 AAAAAAAAAAAAAAA GTTTGAGGGAAAGGGGGATGTGCTAGAGGATC
40 AAAAAAAAAAAAAAA AGAAAAGCAACATTAAATGTGAGCATCTGCCA
43 AAAAAAAAAAAAAAA CAAAAATCATTGCTCCTTTTGATAAGTTTCAT
44 AAAAAAAAAAAAAAA AAAGATTCAGGGGGTAATAGTAAACCATAAAT
45 AAAAAAAAAAAAAAA CCAGGCGCTTAATCATTGTGAATTACAGGTAG

For the second AuNR/AuNS

63 ATAATAATAATAATA GAAGATCGGTGCGGGCCTCTTCGCAATCATGG
64 ATAATAATAATAATA GCAAATATCGCGTCTGGCCTTCCTGGCCTCAG
66 ATAATAATAATAATA CGAGTAGAACTAATAGTAGTAGCAAACCCTCA

67 ATAATAATAATAATA TCAGAAGCCTCCAACAGGTCAGGATCTGCGAA
68 ATAATAATAATAATA CATTCAACGCGAGAGGCTTTTGCATATTATAG
69 ATAATAATAATAATA AGTAATCTTAAATTGGGCTTGAGAGAATACCA
52 ATAATAATAATAATA CAGCTGGCGGACGACGACAGTATCGTAGCCAG
53 ATAATAATAATAATA CTTTCATCCCCAAAAACAGGAAGACCGGAGAG
55 ATAATAATAATAATA CAATAAATACAGTTGATTCCCAATTTAGAGAG
56 ATAATAATAATAATA TACCTTTAAGGTCTTTACCCTGACAAAGAAGT
57 ATAATAATAATAATA TTTGCCAGATCAGTTGAGATTTAGTGGTTTAA
58 ATAATAATAATAATA TTTCAACTATAGGCTGGCTGACCTTGTATCAT

For assembly of AuNR dimers 5

For the first AuNR

80 AAAAAAAAAAAAAAA GAAGCAAAAAAGCGGATTGCATCAGATAAAAA
81 AAAAAAAAAAAAAAA CCAAAATATAATGCAGATACATAAACACCAGA
104 AAAAAAAAAAAAAAA TTTTAATTGCCCGAAAGACTTCAAAACACTAT
105 AAAAAAAAAAAAAAA CATAACCCGAGGCATAGTAAGAGCTTTTTAAG
128 AAAAAAAAAAAAAAA TCTTACCAGCCAGTTACAAAATAAATGAAATA
129 AAAAAAAAAAAAAAA GCAATAGCGCAGATAGCCGAACAATTCAACCG
152 AAAAAAAAAAAAAAA TATTTTGCTCCCAATCCAAATAAGTGAGTTAA
153 AAAAAAAAAAAAAAA GCCCAATACCGAGGAAACGCAATAGGTTTACC
176 AAAAAAAAAAAAAAA AGGTTTTGAACGTCAAAAATGAAAGCGCTAAT
177 AAAAAAAAAAAAAAA ATCAGAGAAAGAACTGGCATGATTTTATTTTG

For the second AuNR/AuNS

89 AGCTGAAATAATGTGT AGGTAAAGAAATCACCATCAATATAATATTTT
91 ATAATAATAATAATA AAAGCGAAGATACATT TCGCAAATGGGGCGCG
92 ATAATAATAATAATA CAGACGACAAAAGATTAAGAGGAACGAGCTTC
93 CCAAATCACTTGCCCTGACGAGAACGCCAAAAGGAATTACTCGTTTAC
113 TAGAAACCTATCATAT GCGTTATAGAAAAAGCCTGTTTAGAAGGCCGG
115 ATAATAATAATAATA GAGCGTCTAATCAATAATCGGCTGCGAGCATG
116 ATAATAATAATAATA CGAAGCCCTTCCAGAGCCTAATTTACGCTAAC
117 TTATTCATAGGGAAGGTAAATATTCATTCAGTAAAAGTAATATCTTAC
137 AAAATAATTAAAGCCA ACGCTCAAAATAAGAATAAACACCGTGAATTT
139 ATAATAATAATAATA ACAATTTTCAAGAACGGGTATTAAGAACAAGA
140 ATAATAATAATAATA AAGAAACAACAGCCATATTATTTAACCCAGCT
141 GACTTGAGAGACAAAAGGGCGACAAGTTACCAGAAGGAAAATAAGAGC
161 GTTTATCAAGAATCGC CATATTTAGAAATACCGACCGTGTTACCTTTT
163 ATAATAATAATAATA AATCAAGAATCGAGAACAAGCAAGACGCGCCT
164 ATAATAATAATAATA ACAAGAATAAACGATT TTTTGTTTAAGCCTTA
165 AATCACCAAATAGAAAATTCATATATAACGGAATACCCAAGATAACCC
185 AACAACATATTTAGGC AGAGGCATAATTTCATCTTCTGACTATAACTA
187 ATAATAATAATAATA CCTCCCGACGTAGGAATCATTACCCGACAATA
188 ATAATAATAATAATA GTAATTGAATAGCAGCCTTTACAGTTAGCGAA
189 CCGGAAACACACCACGGAATAAGTAAGACTCCTTATTACGGTCAGAGG

For assembly of AuNR dimers 6

For the first AuNR

28 AAAAAAAAAAAAAAA ATTAAGTTCGCATCGTAACCGTGCGAGTAACA
29 AAAAAAAAAAAAAAA ACCCGTCGTCATATGTACCCCGGTAAAGGCTA
32 AAAAAAAAAAAAAAA TTTTTGCGCAGAAAACGAGAATGAATGTTTAG
33 AAAAAAAAAAAAAAA ACTGGATAACGGAACAACATTATTACCTTATG
34 AAAAAAAAAAAAAAA CGATTTTAGAGGACAG ATGAACGGCGCGACCT
39 AAAAAAAAAAAAAAA GTTTGAGGGAAAGGGGGATGTGCTAGAGGATC
40 AAAAAAAAAAAAAAA AGAAAAGCAACATTAAATGTGAGCATCTGCCA
43 AAAAAAAAAAAAAAA CAAAAATCATTGCTCCTTTTGATAAGTTTCAT
44 AAAAAAAAAAAAAAA AAAGATTCAGGGGGTAATAGTAAACCATAAAT
45 AAAAAAAAAAAAAAA CCAGGCGCTTAATCATTGTGAATTACAGGTAG
For the second AuNR/AuNS
51 ACTGCCCGCCGAGCTCGAATTCGTTATTACGCCAGCTGGCGGACGACG
52 ATAATAATAATAATA ACAGTATCGTAGCCAGCTTTCATCCCCAAAAA
53 ATAATAATAATAATA CAGGAAGACCGGAGAGGGTAGCTAGGATAAAA
55 ATAATAATAATAATA TTCCCAATTTAGAGAGTACCTTTAAGGTCTTT
56 ATAATAATAATAATA ACCCTGACAAAGAAGTTTTGCCAGATCAGTTG
57 ATAATAATAATAATA AGATTTAGTGGTTTAATTTCAACTATAGGCTG
58 ATAATAATAATAATA GCTGACCTTGTATCATCGCCTGATGGAAGTTT
60 CCATTAAACATAACCG ATATATTCTTTTTTCACGTTGAAAATAGTTAG
61 TAATTGCGCCCTGAGA GAGTTGCACGAGATAGGGTTGAGTAAGGGAGC
63 ATAATAATAATAATA CTCTTCGCAATCATGGTCATAGCTACTCACAT
64 ATAATAATAATAATA GCCTTCCTGGCCTCAGGAAGATCGGTGCGGGC
66 ATAATAATAATAATA TAGTAGCAAACCCTCATATATTTTAGCTGATA
67 ATAATAATAATAATA GGTCAGGATCTGCGAACGAGTAGAACTAATAG
68 ATAATAATAATAATA CTTTTGCATATTATAGTCAGAAGCCTCCAACA
69 ATAATAATAATAATA CTTGAGAGAATACCACATTCAACGCGAGAGG
70 ATACGTAAAAGTACAACGGAGATTTCATCAAGAGTAATCTTAAATTGG

3. Thiolated Capture strand sequences

Thiolated S15 5'-TTTTTTTTTTTTTTT GAGC-SH-3'
Thiolated S10 5'-TATTATTATTATTAT TTTTT-SH-3'

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