Supporting Information: Dynamic Actuation of DNA-Assembled Plasmonic Nanostructures in Microfluidic Cell-Sized Compartments

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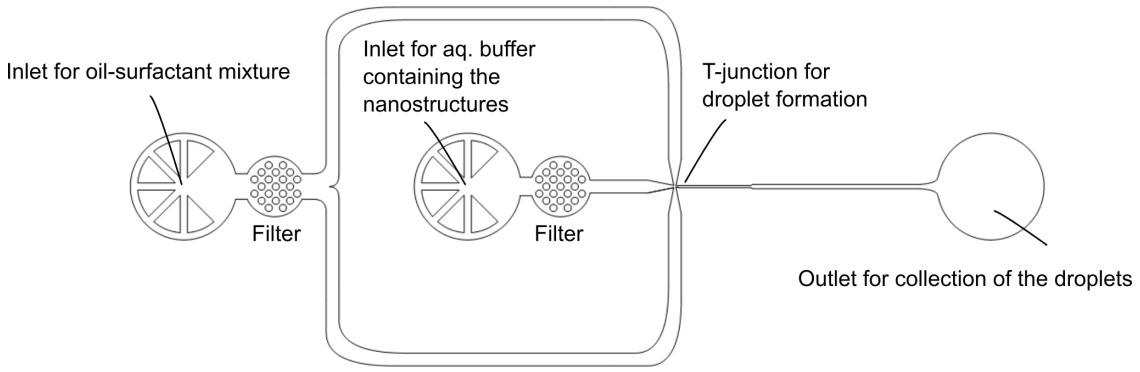
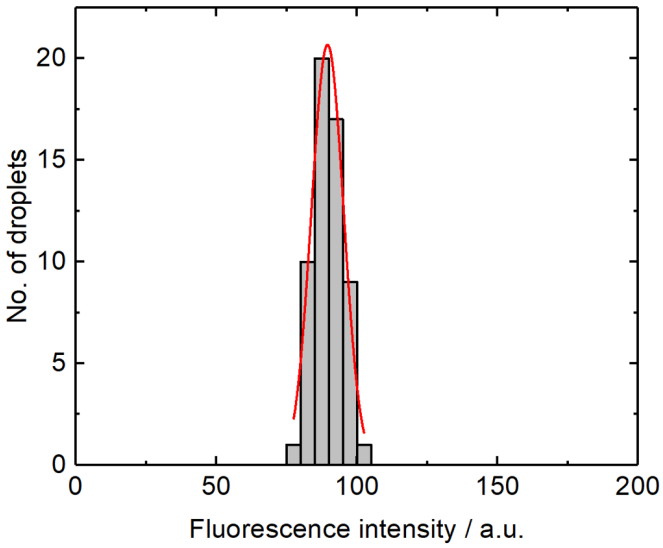
Figure S1: Layout of the microfluidic droplet production device



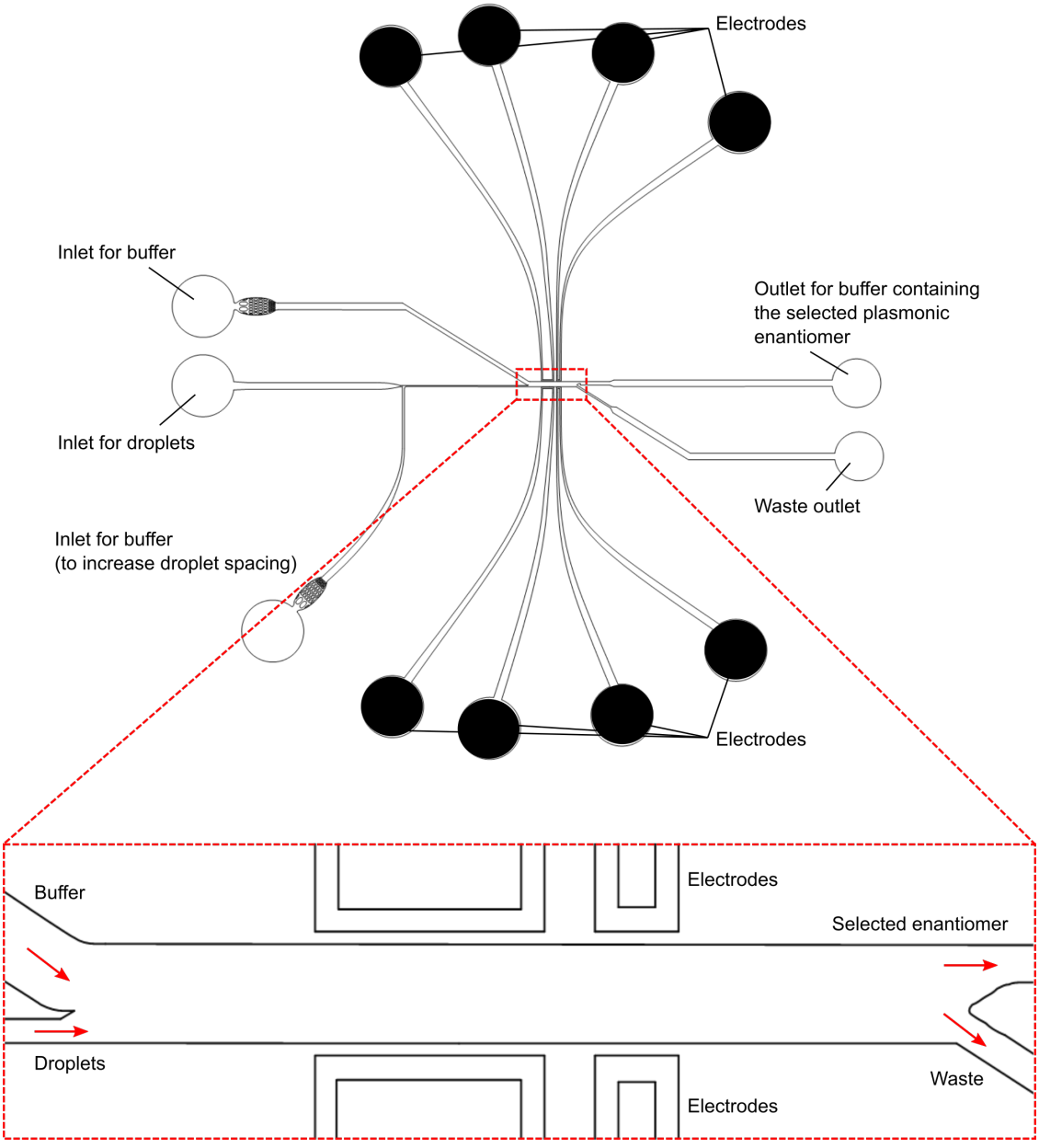
Supplementary Figure 1: Layout of the microfluidic droplet production device for the encapsulation of DNA-assembled plasmonic nanostructures. All fluids are injected via the inlet channels. Droplets are produced at the T-juction, where the flow of the aqueous buffer (containing the nanostructures) is intercepted by an oil-flow. Droplets are collected from the outlet for subsequent experiments.

Figure S2: Homogeneity of the encapsulation of DNA nanostuctures



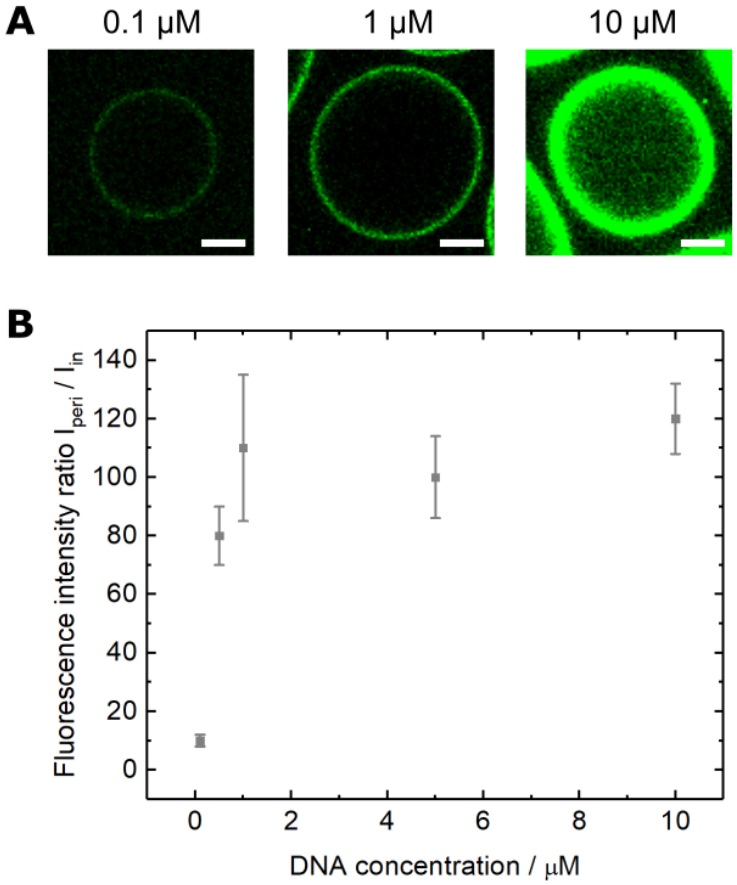
Supplementary Figure 2: Distribution of the fluorescence intensity of microfluidic droplets encapsulating the DNA-assembled plasmonic nanostructures. The narrow distribution confirms the uniform encapsulation of the nanostructures during the droplet formation process.

Figure S3: Layout of the microfluidic device for plasmonic enantiomer separation



Supplementary Figure 3: Layout of the microfluidic device for the separation of plasmonic enantiomers. Droplets containing a racemic mixture are supplied via the droplet inlet. When exposed to an electric field (see zoom), the droplets fuse to the co-flowing buffer phase, releasing their content, which can then be collected from the outlet. Note that only the electrode facing the aqueous side (top electrode) was turned on for the release (500 V, 0.5 kHz).

Figure S4: Attachment efficiency of cholesterol-tagged DNA to the droplet periphery



Supplementary Figure 4: A) Confocal fluorescence images showing the attachment of cholesterol-tagged DNA at different DNA concentrations. The images are taken with the same imaging settings. Scale bars: $20\,\mu m$. B) Fluorescence intensity ratio as a function of DNA concentration, error bars correspond to the standard deviation (n > 10). The relative fluorescence intensities at the droplet periphery (I_{peri}) compared to the droplet lumen (I_{in}) for a broad range of different DNA concentrations (100 nm, 500 nm, $1\,\mu m$, $5\,\mu m$, $10\,\mu m$) serves a direct measure of the attachment efficiency. We can conclude that there is no strong correlation between the attachment efficiency of the cholesterol-tagged DNA and the DNA concentration. The seemingly lower attachment efficiency at low concentrations of cholesterol-tagged DNA is likely due to the weak signal-to-noise ratio as can be appreciated from the confocal fluorescence images.

Text S1: Density of cholesterol-tagged DNA on the droplet periphery

As visible in Figure S4, the cholesterol-tagged DNA accumulates at the droplet periphery even at high concentrations ($10\,\mu m$). Let's consider the density of the surface coverage assuming a droplet with a radius of $r = 20\,\mu m$. This droplet has a volume V of

$$V = 4/3\pi \cdot r^3 \approx 33\,\mathrm{pL}; \tag{1}$$

and a surface area A of

$$A = 4\pi \cdot r^2 \approx 5 \cdot 10^9 \mathrm{nm}^2. \tag{2}$$

At the maximum DNA concentration of $c = 10\,\mu m$, it would encapsulate

$$N = \mathrm{N_A} \cdot c \cdot V = 2 \cdot 10^8 \tag{3}$$

DNA strands. Hence, an area of $25\,\mathrm{nm}^2$ is available per DNA strand. This is significantly more space than the hydrodynamic radius of the short DNA oligomer itself (about 3 nm). This explains why the concentration of the DNA handles has no effect on the filtration efficiency, as long as they are supplied in excess compared to the plasmonic DNA nanostructures. The experimentally achievable concentration of the plasmonic DNA nanostructures is limited to about 10 nM. Diluting it for the experiments does not make much sense since it would decrease the signal-to-noise ratio for fluorescence detection as well as CD spectroscopy. Therefore, we chose to work with the maximum concentration of plasmonic DNA nanostructures and a 100x excess of the cholesterol-tagged DNA handles to ensure efficient binding. Additionally we have designed highly polyvalent attachment sites between the DNA origami and the cholesterol anchors (7 handles per origami) to ensure stable binding.

Table S1: Thermal annealing protocol

Supplementary Table 1: Thermal annealing protocol for the assembly of the cross-shaped DNA nanostructures.[1]

Temperature, °C	**Time, min**
80	15
79 - 71	1
70 - 68	5
67 - 63	10
62	15
61	20
60	30
59 - 38	60
37 - 36	45
35	30
34	20
33 - 32	10
31 - 25	5
24 - 21	2
20	Hold

Supplementary Table 2: List of DNA 'core' sequences and their start and end points in caDNAno.[2] Adapted from Kuzyk *et al.*[1]

DNA Sequence	**Start**	**End**
AACGAGCGTCTTTCGCCATATTTGTTTAGAAGCGCATTAGAC	4[202]	7[202]
AGGTTTAACGTCGACACCGCCTGGAAGGGTTAGAAGATTTTC	22[62]	22[63]
TACTTTTTCTAATCTATTTACAGAAAGGTGCCTGAAACAGTGA GGCCAC	15[196]	25[202]
AAGAAGTCCAAAATTAGAGAGCTTAATT	4[118]	13[118]
TTTGAGATGTTTTTATAATCGCATTCGC	19[196]	23[195]
AATTTTAATTTTCAGCAAGGCAAAGAAT	26[146]	16[133]
TCGACAAATTATCAGTCAATATAGGTCTGAGAGACGTGAATT ACGAATA	25[119]	14[112]
ACTCGTCAATCGGAGAGTAACCTCGTATGAGCACTGGAAGGT AACCTCA	14[132]	22[126]
CTCATAATGTTTAGACACCCCCAGCGATAAGAGGA	5[84]	9[90]
CATAACCTTATTACCCCTCAGAACCGCCCCAGGTCATTTTTG	2[97]	13[97]
CCGGCTTAGACGCTCGGCTGACGCATTTCCCAAATGAGCGG ATGGAGGA	16[97]	25[90]
TGTCCATCACGCAAATTAACTTTTAGACAATAGGAACCCGTC GGCAAAT	25[217]	22[224]
AATTAATTTTCCCTCTCCTGGTTGGTGTGTGGACT	15[42]	27[48]
AGGGAGCGCCATCTAAAGCCTCAGAGCA	27[140]	15[153]
GTCAATACTAACTTTTTCAAATTAACACGCAAATGAAAAATC	24[104]	21[118]
TATTCTGAAATAAAGGTGGAATAAATTGAGGTTAAAGGT AGCAAG	0[261]	9[258]
TATACTTAGATACATTTCATCACAGAGGTCACCTTGCAGACA	18[139]	21[139]
GGCAATTTTCTGAATAATGGCAGTTACAAAAATTAATTACAT	25[63]	18[49]
GAGATTTACCTAAAACGAAAGGCCACTAAATCATTTTGGGA AGAATTCC	10[55]	2[49]
CCAAAGAAGAAAATACAAACAACTGGTAATAAGTTCAGTGC CTCAAGAG	11[224]	0[217]
GTGGTTGTGGGAAGGGAAACCAGGCAAA	15[252]	17[265]
CATACAGTTTGGGGAGTATCATAATGGTGAACCACTGAATGG	16[153]	21[153]
AACCGCCGAGGCAGGTCAGACGATTGACCACGCCACCCTCAG	5[224]	5[223]

Supplementary Table 3: List of DNA 'core' sequences (continued).

DNA Sequence	**Start**	**End**
GTTTGATGGTGGTTCCGTAGCCCGTGTTATCCTGTA AATCTCCCCGGGT	25[25]	16[24]
AACGGCTAGGGATAAAACTACAACGCCT	13[35]	1[48]
GGAGGTTACCCTCAACCACCGTAGCGCGTTTTCATA TCGATAAGCACCA	4[237]	9[244]
CGAGTAAGTAATAAAGGGTGATATTTTAAATGCAA	25[203]	16[196]
AGAATCCAAATCAGTTGTAAACGTTAATCCCCAAAA ACTAGCATGTCAA	24[216]	20[196]
ACCATCACACATAAGATAGCTTAGATTAAGGTTGG	27[77]	16[84]
GAGGTTTTTTTAGCGCGGGGTTTTGCTCAGAACTG	2[202]	13[195]
AATATCAAACCCTCTTGAAAGGAATTGAAACAACTT AGTTAATTTCGCA	22[125]	18[119]
GAAACACCAGAACGAGTAGTAGTGAGAA	4[62]	5[48]
AGCTAAACAGGAGGAATTCGCATTAAAT	24[265]	22[252]
AATTGCTGACGACGATAAAAATTTGCCAAGCGTCCACAGTTC	12[104]	6[105]
CTTGATATTCAACCTGAGAGTCTGGACCAAGGGATTTGACGA	18[244]	25[244]
AAAGAAATTACATCGGGAGACGCTGAGACATAAAACAGAAAT	22[83]	22[84]
AAGATTAGTATTCTGGCGGATAAGTGCCACTAATAACGGAAT	2[181]	13[181]
TTTGCCCGAACGCCCTGAAATGGATTTACTAAATTTATGCGT	25[140]	18[140]
AAGGATTCCTTATTGTCAGATAGCCGAACTGAATCCTTGCGG	0[216]	2[203]
CCTCATTTTGTCACAGAAAATACATACACATGAAAAACAGTT	3[238]	1[251]
GCATAGTATCAGTTTCAGGAGGTTTAGTCTTAGAGTACCTTT	2[118]	12[105]
ATTTAAACTCATTTTTTAACCAGGAACGCATTGCCGTTCTAG	22[223]	18[224]
CGTAATCAGTAGCGAGAGCCAGCAAAATTTGAGCCA TTTGGGACCAGCG	8[230]	11[223]
ATTAACGAGGCGCAAACGGTGATCAAGA	8[90]	6[77]
GAGGCTGCAAACGTAATCAATCAAAAGGCACCGACC ACCAGTGCAGCAC	0[237]	8[231]
AAACAGGACGTCAAAAATGAATTACAAATAGCCATAT TTAACCGGAGCG	7[182]	9[174]
AATACGTGGCACTGTATCTAAAAATTTACATTGGCGCGTAAG	22[146]	22[147]

Supplementary Table 4: List of DNA 'core' sequences (continued).

DNA Sequence	**Start**	**End**
TTAACGATTGCCTTGATATTCTCATATGACGCAGT	2[216]	13[223]
TTCGAGGAATTGTAAGGAATTGCGAATAACAGTTTAC CATCGGCTTGCA	8[41]	11[34]
AAGACGGAGGAGTAAACAGGGCTTAAGCTA	15[22]	14[18]
GAGGAAACGCAAGGTAATTGAGAATCGCTATCTTAGGAAACC	12[181]	12[182]
ACAGACATTAATTTCAACTTTCGAAGGCCTTTGAG	2[48]	13[55]
ATGTTAGAGACTCCTTGAGTAACAGTGCGGAGTGTAATAAAT	13[224]	3[237]
CAATATCCTATGGTGTGTAGCGGTCACGGTGCTGACCTCTAA	17[238]	15[251]
AATAAATTCGAGCTCAGGTCAGGACGAGTAGATTTAGCCAGT	10[125]	10[126]
CGGAGACTACTTCTCGGGCGCTAGGGCGGACAATGA CGCAAGATGTG	17[217]	36[226]
TATTACCGCCTTGCTGAAAAGAACATCC	25[161]	16[161]
TTTGGGGCTTGAATGAGAAGAGTCAATATACCTTTTTAACCT	27[98]	16[98]
CATTTATATTATTTGCACGGTTGGCAAA	24[90]	23[104]
ACTAATCGGTTTATTGACCAACTTTGAACCGGATAAGGAACA	6[46]	6[47]
TGATTGCAGTAACAGTACCTTTTGCGTACCTACCA	20[69]	23[76]
GCACGTAGCGCGTATGGTGCCGGCGATCGGTGCGGGC CTCTATCACCAT	25[245]	17[237]
ACCAGAACCGAAGCCAATGAAATAACCCACCCTGAACAAAGT	12[195]	8[182]
GGGAACAAGTAACAACGCCATCAAAAATCCGATTAAG TTTGAGGCACCG	21[238]	18[245]
ACCCAAAAGTACCAAAGAACGCGAGGCGTGAAGCCGCTACAA	13[182]	3[195]
TTATCAATCCCATCCTCATTGAATCCCCAGGTCTTTACCTGT	7[140]	7[139]
AGTAATAAAAGGGACTCATGGGCGTTAAATAAGTT	23[175]	19[195]
TTAACCTGCCTAATGAGTGCACCTGAGACAAAATCCCATGAT	18[48]	25[62]
CTATTAGCCTGAAAAGATTCACCAGTCATGACGCTACC GACCGAAAAAG	21[154]	18[161]
AGAGCCAAATCTACGTTAATACAGGACGGTGAATTCTGACGA	0[62]	4[63]
GGGAGTTAAAGGCAGTATCATCGCCTCACAGCGGAA ATTGGGCTCTGTA	11[35]	4[28]

Supplementary Table 5: List of DNA 'core' sequences (continued).

DNA Sequence	**Start**	**End**
GTTATTACATAAATACATAGCATCATTTCTCCGAAGGC CCACCCTGATT	16[83]	26[63]
CTTAAGTGTCCTTACTGCGCGCTACAGGTAACGTG	14[265]	25[258]
TTTGCGGAGATGGTGCCCTCATAGTTAGCCAGTACGC AAGCCCAATAGG	12[41]	0[21]
GTGTGATTCGCCATTGCGCGAACTGATAACAGAGATAGAACC	19[168]	22[168]
AATGCCCTACATGGGAATGGAGAGCCGC	1[252]	4[252]
AGCACTAGGTGGGCTATCAAAATAAATTAAGCAATGTAAGCA	27[119]	14[133]
CAGAGGGTAATTACAATAAACTGTACAGAGAGAATAACATAA	8[181]	7[181]
CATTTGAATAAATCCCAACGTGGGCGATCTCTGACTAGAATC	17[49]	15[62]
AATAATACAATAGATAAGTCCAACATGTTTCTGTC	6[153]	9[160]
GCTGAATACCGTACGAGATTTAGGAATAAAAGGAAGGGCACT	13[119]	3[132]
TATCAAATGTTTGGCGAATTATTCATTTCCAAACAATAACGG	23[77]	20[77]
GGGAGAGTAAGAAACGATTTTTATTTATCAAGAAACCTTTTT	7[203]	11[202]
ATTAAATTTTTGTTTGAGAAGGATCTACCCGGAGAGG GTAGCATTCAAA	23[196]	17[202]
GTAGCAACCACCCTCATTTTCACAGAGGACCCGCT	1[49]	12[42]
TGAAACCCGGCATTCCGGAACCAGAGCCGAGCCACTC ACCGTGCGACAT	8[251]	11[244]
AACGCCACCACATTGGAATAGGTGTATC	2[139]	0[126]
TAGAGCCTCAACAGAATCAATATCTGGTGCCAGCACGCCTGC	24[111]	20[98]
AGAAAAGGCTGGCTGACCTTCTACAGACCAGGCGCCAAAAAG	6[104]	8[91]
ATCATTCTTTTATTAAGTTTCATTCCATCCAACGCTCAACAG	4[160]	11[160]
GCTTGCCACCTTATGTAAAATTCATGAG	4[76]	13[76]
TAATAGTTTTACCACCTTTTGATAAGATCACTCATCTT TGCGGAAGCAA	4[97]	11[104]
CAGACGAAACGCCAATCAATAATCGGAGAATAGCAG CCTTAACAAGAAA	9[161]	6[154]
ATCAGTTATTACCTTGTGAGTGAATAACCTTGCTTCGAATTT	26[62]	17[48]
TAGAATTCATTACCCAAATCATGACAAGAAAGAGG	5[49]	7[69]

Supplementary Table 6: List of DNA 'core' sequences (continued).

DNA Sequence	**Start**	**End**
CAGATATGGGTTGATATAAGTTTTTAAATATGCAA	1[154]	13[160]
ACTCACAACGCGCGGGAGATGAATATACTTTGAATACAGCTA	20[48]	20[49]
CAAAAGGAGCCTTTTGAATTTCAATGACAAGATAAATT GTGTGCTTGCT	7[21]	8[42]
TCAACCGGTTTATTAAAGCCACTTTTGATGATACAC CGTATAGTATTAA	11[245]	0[238]
CATCGAGTTGATTCTGTAGCTCAACATGATAGCCCCAA CTAATGCAAAT	3[133]	1[153]
TTTTATCCAAAGTTGCATGATTAAGACTAGGATTAG AACCTCCCGGGGT	3[196]	1[216]
AGAAAAGATTTTGTTAAAATTCTGGCCAGCCGGTTGATAATC	21[196]	21[195]
AAAGTAATCAGCTAATGCAGAACGCGCCTACCGACAAAAGGT	9[147]	9[146]
AAAAGAAAAATCGGGAGTTGCTGCCCTTGAGGCGGTT TGCGTTCGGC	26[48]	21[39]
AGCTGATAGCAAGCCTGGGGTCACAATTCCACACAACAT	23[35]	18[24]
ACTCCAATCAAAGCCAAATATAAAGCGGATTGCATA TACGAGAATGACC	11[105]	7[111]
ATAAATCCAGAAGCCGCGTTTTAGAGAATATAAAGCTGACTA	7[112]	8[126]
GCTTTCCGGGGACGCGTAACCGATTGACCGTAATG	18[258]	21[265]
ACAGATGGACGGTCTGTTACTTAGCCGGTATACCAAAAGCTG	7[70]	5[83]
AAGAAAAGTAAGTTAACAATAATAAGAGCCCAATCCAACGCT	11[203]	4[203]
CAGACTGGAACCGCCTCCCTCAGAGCAATTTGCCTTTAGCGT	7[224]	7[223]
AGAGCCGAAAATCATTCGGTCATAGCCC	5[252]	7[265]
TTACCATTGAATTACACCCTCCGCCAGCATTGACA	9[245]	4[238]
AAACCAAGTACCCTGTCATAAATATTAA	4[139]	5[139]
ATAATCCATTATCAAGATAACTATATGTAAGATGA	25[77]	18[84]
GCTGCATTAATGAAATTGGGCGGGCAAC	21[21]	23[34]
TCGGAACGAACCCTCACGCTGAGCCCACGCATA	13[19]	10[18]
TACCAGTAAACGAAGAACCGCCACCCTC	2[76]	0[63]
TAAAGCATGAGGCGGTCAGTATATATTTAATAGAT	21[119]	24[112]

Supplementary Table 7: List of DNA 'core' sequences (continued).

DNA Sequence	**Start**	**End**
GTGAAATAGATAGGCACTATTAAAGAACAATGAGTCGCTATT	17[28]	15[41]
ATTGTCACCAGGGTTTTCCCATGTAAAACCAGG	14[244]	37[249]
GTGAGCGAACGGCGGTGCATCTGGCAAACAAGAGATCTCCGT	22[251]	21[237]
GTAATCTACGTAACAGCGCGAAGAATACACTAAAATAAACGG	6[76]	12[77]
CTTGAAACAATATATTTTTAAAGAAAACAAATCGC	15[63]	19[69]
CCCAATAGCAAGCAAAGCCGTCAAGAACGCATGTAGAA ACCAACATGTA	2[160]	10[154]
CATTAATTGCGTTGCGGTAAAGCGGTCCACGCTGGTT	21[40]	24[21]
AATAAATTAAAGCTGACAGTGCGGCCCTCCCCGATATTTATT	16[160]	26[147]
AGATTGTATAAGATATCGATGAACGGTAATCAGGTGTACGCC	21[217]	24[217]
CGTGGCGGCTCGCCTTATGACCCTGTAATGCCTGA	27[182]	16[189]
CAGGAAAGTAGAAGACGTGTAGGTAAAGTAAATAA	25[182]	18[189]
GTAGCAAAGTCAAAAAAATTTTTAGAACCCTCATAGAAAGGC	26[230]	17[216]
GACTCTAGGAAAGCACTATCGGCCAGCCTACATTTCACGACC	14[174]	23[174]
TTGACGGTGATACCAAATCGGTTGTACCTAGCATTGT GGCATATTACTA	27[161]	18[168]
TCTAGCTGGCGAGGAGAAGCGACGGC	36[249]	38[238]
TAAATGAATTTTTGGATCGTCGGGTAGC	3[21]	13[34]
GTAATTAATAGTAGAAAAACACTGGAGT	16[188]	14[175]
CGAGCTTTCTCAAAGGGGGGATTCGCTATTACG	37[226]	15[244]
CAGTGCCGGTAACGACCTTATCTGGCAATGCCGTT	38[237]	26[231]

Supplementary Table 8: List of DNA sequences carrying poly-A-overhangs for the attachment of AuNRs (for CD spectroscopy) or fluorescent dyes (for confocal fluorescence imaging).

DNA Sequence	**Start**	**End**
CGGATGGACCGCCAAGGTAGAAAGATTCAAGAGCAACACTAT AAAAAAAAAAAAAAA	13[98]	2[98]
AACAGTGCAATTACGATAATATTTAGAA AAAAAAAAAA	20[97]	25[97]
CTAATATCAGAGAGATAGCAAATAAACACAGAGCC AAAAAAAAAAAAAAA	9[175]	4[182]
AAGACAAAGAACGCGAGAAGAGCAAAAGAAATGCTCCAGA AGCAAGTTT AAAAAAAAAAAAAAA	18[118]	27[97]
TAGGGCTTGTCTGGTTCATCGTAGGATGCCCTGTCTTTCCTT AAAAAAAAAAAAAAA	11[161]	4[161]
GTATTAGGAAACCAGATGCAAATCCAATCGCAATGTTTTGCGG CCGTAA AAAAAAAAAAAAAAA	25[98]	27[118]
GAAGTTTACCCTCACTAACGGAACAACACTACTGGCTCATTA AAAAAAAAAAAAAAA	13[77]	2[77]
CTAAAGTGTCGAGAAGAAGGCTTATCCGGTATCATTACCGCG AAAAAAAAAAAAAAA	13[161]	2[161]
TGAAACAAACATCATGGAAACTCATATTTACGTGA AAAAAAAAAAAAAAA	18[83]	27[76]
AGCCCGAAAGACTTGAACCAGACTGGATGAGGGGG AAAAAAAAAAAAAAA	9[91]	4[98]
CCTGTTTCGCGAGCTGGTAATTTAGAGC AAAAAAAAAAAAAAA	18[160]	27[160]
TTATAGTAAAAATCCTCAAATGCTTTAAAATACTGCGGAA TCTTTGCAA AAAAAAAAAAAAAAA	8[125]	4[119]
ATTCGCCGCAGAGGATTATACCATCAAT AAAAAAAAAAAAAAA	20[76]	25[76]
GCGATTTTAAGACGAATCAGTGAATAAG AAAAAAAAAAAAAAA	3[77]	4[77]
TCATATGTACCCCCTAAAACAAAATAAGAAATACCATTGCAA AAAAAAAAAAAAAAA	20[195]	25[181]
TAATTTGCTATTTTGCACCCATTAAATC AAAAAAAAAAAAAAA	4[181]	2[182]
TAGCACAACCTGTTTAGCTATAAAGTTTACCCTAA AAAAAAAAAAAAAAA	16[132]	27[139]
CTTCTGATCTTTAATAAAAATACCGAACTTGAAATCAATCGT AGAACAA AAAAAAAAAAAAAAA	22[167]	25[160]

Supplementary Table 9: List of DNA sequences carrying poly-A-overhangs (continued).

DNA Sequence	**Start**	**End**
ACACCGGAATCATACAATTCTAACTCAACGGCGAA AAAAAAAAAAAAAAA	18[188]	27[181]
ATTTTCGAGACCAGTATAAAGATAACAGAACAAGCGATACAT AAAAAAAAAAAAAAA	10[139]	2[140]
ATTTAGGCAGAGGCTTTACGAGGGTATT AAAAAA AAAAAAAAA	10[153]	4[140]
TAGGGGCTCGAGGTGAACAAAACTTTACAAACAAT AAAAAAAAAAAAAAA	14[111]	25[118]
ATAATGCCCAATTCTGCGAATAGCGAGATTACGAG AAAAAAAAAAAAAAA	13[126]	2[119]

Supplementary Table 10: List of DNA sequences for pH switching.

DNA Sequence	**Role**	**Start**	**End**
GACTAAAGACTTTTACGTAATAGGCAAA AACAAAGTGCTCCAAATCATA–TTTT–Duplex1	RH pH switch	13[56]	8[63]
Duplex2–TTTT–AGGGAACCGAACCACG AAATCCGCGACCTACAACG	RH pH switch	8[62]	10[56]
CTGATAAATTAATGAAAGGCT ATCGTAAAACAGGA–TTTT–ssDNA	RH pH switch	18[223]	21[216]
CATCACTAAGGGAAGAAAGCG CCCCGCTGCGGGAG–TTTT–Duplex1	LH pH switch	26[202]	15[209]
Duplex2–TTTT–AAGCCTTTATTTCATCCC GCCAAAATAAAAAGGAGTTGATTAAAGAGTC	LH pH switch	15[210]	25[216]
CAAATTAACTGAACACAAGAATTGAGTT AAGCCTTACAGAAT–TTTT–ssDNA	LH pH switch	8[209]	8[210]

Supplementary Table 11: List of DNA sequences carrying overhangs for complementary base pairing with cholesterol-tagged DNA. The start and end positions indicate which of the original staples have to be replaced in the LH structure for the plasmonic enantiomer separation experiment.

DNA Sequence	**Start**	**End**
TCCTTCTATGCATCA – TTTT – GCGTTCGGCCATTAATTGC GTTGCGGTAAAGCGGTCCACGCTGGTT	21[40]	24[21]
TCCTTCTATGCATCA – TTTT – GGTTTAACGTCGACACCGCC TGGAAGGGTTAGAAGATTTTC	22[62]	22[63]
TTTCCCTCTCCTGGTTGGTGTGTGGACT	15[42]	27[48]
AAAAGAAAAATCGGGAGTTGCTGCCCTTGAGGCGG	26[48]	21[39]
TCCTTCTATGCATCA – TTTT – AAGAACAATGAGTCGCTATT AATTAAT	17[28]	15[41]
TCCTTCTATGCATCA – TTTT – GTTCCGTAGCCCGTGTTATC CTGTAAATCTCCCCGGGT	25[25]	16[24]
TCCTTCTATGCATCA – TTTT – GCTGATAGCAAGCCTGGGGT CACAATTCCACACAACAT	23[35]	18[24]

Supplementary Table 12: DNA sequences carrying chemical modifications (HPLC-purified) and their function.

DNA Sequence	**Function**
Thiol–TTTTTTTTTTTTTTT	Functionalization of the AuNRs
Atto550–TTTTTTTTTTTTTTT	Fluorescence modification of the LH structure
Atto647–TTTTTTTTTTTTTTT	Fluorescence modification of the RH structure
TGATGCATAGAAGGAA–CholTEG	DNA handle for enantiomer separation

References

(1) Kuzyk, A.; Urban, M. J.; Idili, A.; Ricci, F.; Liu, N. *Science Advances* **2017**, *3*, e1602803.

(2) Douglas, S. M.; Marblestone, A. H.; Teerapittayanon, S.; Vazquez, A.; Church, G. M.; Shih, W. M. *Nucleic Acids Research* **2009**, *37*, 5001–5006.