Supporting Information

Rare Earth Core/Shell Nanobarcodes for Multiplexed Trace Biodetection**

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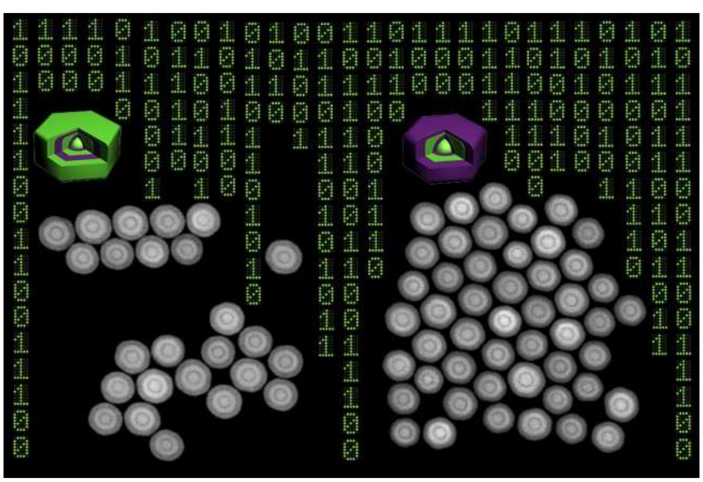


Table S1：DNA sequences

Number	DNA name	Sequences (5'-3')
1	Capture DNA_1	GTA CCA CTA CGA GAC ACT GCC TGA ACT GTA
2	Capture DNA_2	CCT ATC GAC CAT GCT CCA GCG AGA AAA TCT
3	Report DNA_1	TAA CAA CGA TCC CTC TTTTT AAAAA AAAAA AAAAA AAAAA
4	Report DNA_2	TGT AAA GCT AGC TAT TTTTT AAAAA AAAAA AAAAA AAAAA
5	Target DNA_1	GAG GGA TCG TTG TTA TAC AGT TCA GGC AGT GTC TCG TAG TGG
6	Target DNA_2	ATA GCT AGC TTT ACA AGA TTT TCT CGC TGG AGC ATG GTC GAT
7	Noncomplementary DNA_1	ATA GCT AGC TTT ACA AGA TTT TCT CGC TGG AGC ATG GTC GAT
8	Noncomplementary DNA_2	GAG GGA TCG TTG TTA TAC AGT TCA GGC AGT GTC TCG TAG TGG

Table S2. Calculated Number of Available Striping Patterns for a Given Number of Metals and Equal Length Stripes[a]

[a]The number of combinations available where M is the number of distinct materials, n is the number of stripes, and the rod is inherently asymmetric is M^n.

No. of stripes (n)	No. of matrixes (M)	
	2	3
1	2	3
2	4	9
3	8	27
4	16	81
5	32	243
6	64	729
7	128	2187
8	256	6561
9	512	13122
10	1024	26244

Table S3. Standard atomic spacings for different kinds of $NaLnF_4$ with their respective *hkl* indexs from PCPDFWIN database, and the lattice mismatch data between the $NaLnF_4$ materials along different directions.

Hexagonal phase					
hkl	100	110	101	111	201
$NaGdF_4$	5.210	3.010	2.976	2.314	2.115
$NaYbF_4$	5.130	2.960	2.870	2.252	2.063
$NaYF_4$	5.170	2.980	2.900	2.270	2.080
Mismatch					
$NaGdF_4$-$NaYbF_4$	1.54%	1.66%	3.56%	2.68%	2.46%
$NaGdF_4$- $NaYF_4$	0.77%	1.00%	2.55%	1.90%	1.65%

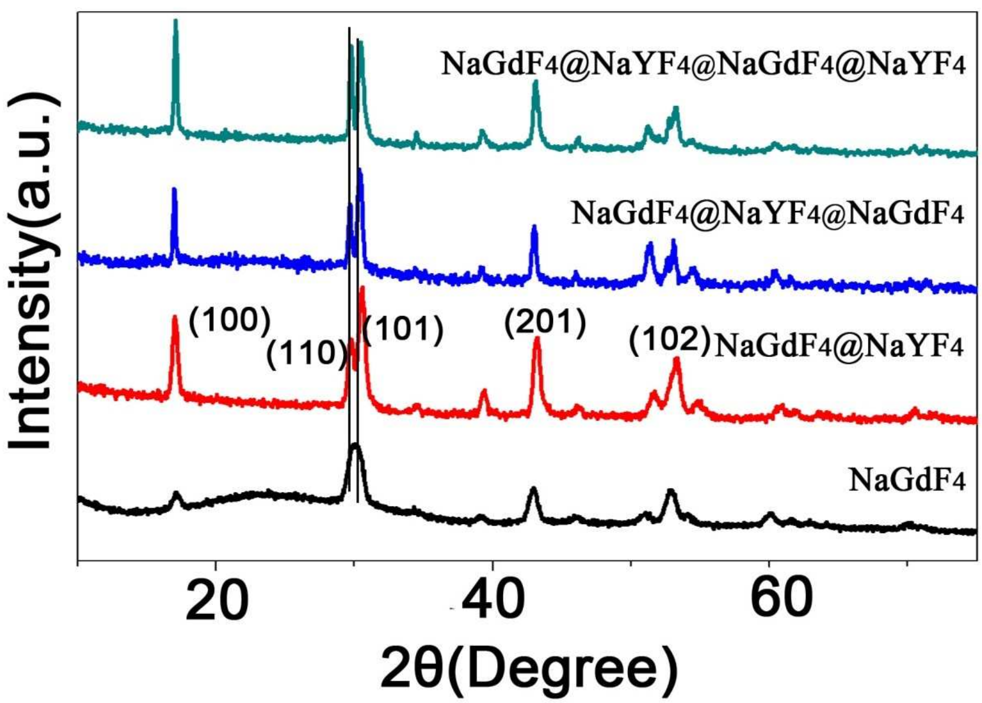
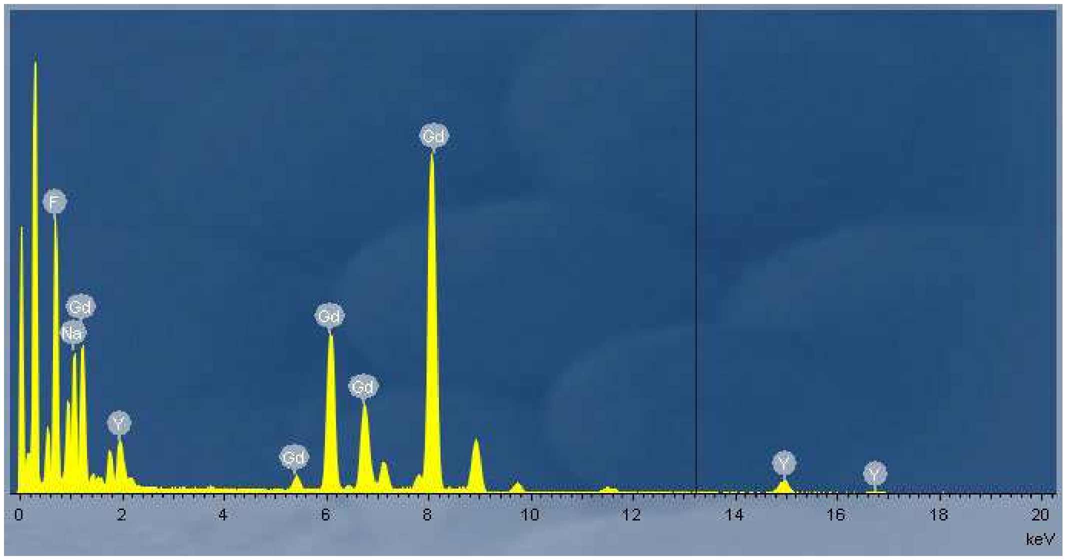


Figure S1. XRD patterns of the $NaGdF_4$ core (black line), $NaGdF_4/NaYF_4$ (C/S1) (red line), $NaGdF_4/NaYF_4/NaGdF_4$(C/S1/S2) (blue line) and $NaGdF_4/NaYF_4/NaGdF_4/NaYF_4$ (C/S1/S2/S3) NBs (green line). It is demonstrated that the crystal structures of the obtained core/shell NPs are all consistent with the structure of the core NPs. The XRD diffraction peaks narrow gradually with the growth of different layer shells, indicating that the crystalline domain size increases for the core/shell NPs. In addition, small shift of the diffraction peaks can also be detected because of the small lattice mismatch (~ 3.8%) between β-$NaGdF_4$ and β-$NaYbF_4$. These results further demonstrate the epitaxial seeded growth approach.



FigureS2. EDAX spectra of the $NaGdF_4/NaYF_4/NaGdF_4/NaYF_4$ (C/S1/S2/S3) NBs. The existence of Na, F, Gd, Yb, Y can be confirmed, further demonstrating the multilayer core/shell nanostructure with different components.

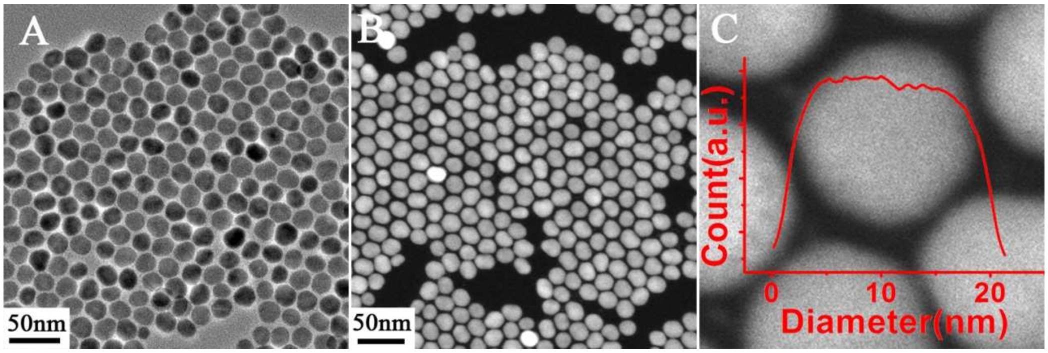


Figure S3. TEM (A) and HAADF-STEM (B, C) images of $NaGdF_4$/$NaYbF_4$ core/shell nanoparticles.

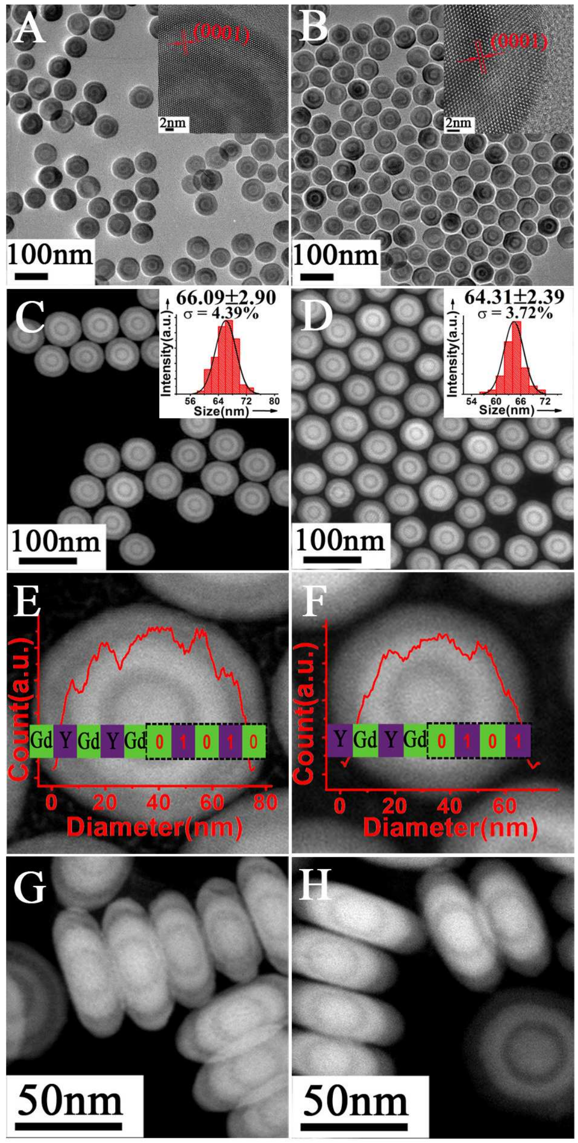


Figure S4. TEM (A,B) and HAADF-STEM (C-H) images of $NaGdF_4$/$NaYF_4$/$NaGdF_4$/$NaYF_4$/$NaGdF_4$ (NBs_{01010}) five layer (A, C, E) and $NaGdF_4$/$NaYF_4$/$NaGdF_4$/$NaYF_4$ (NBs_{0101}) four layer (B, D, F) NBs and DNA conjugated NBs_{01010}(G) and NBs_{0101}(H) NBs in water. It's worth noting that the discernible contrast not only can be confirmed from the [0001] direction (Figure E, F) but also the $[10\bar{1}0]$ direction (Figure G, H).

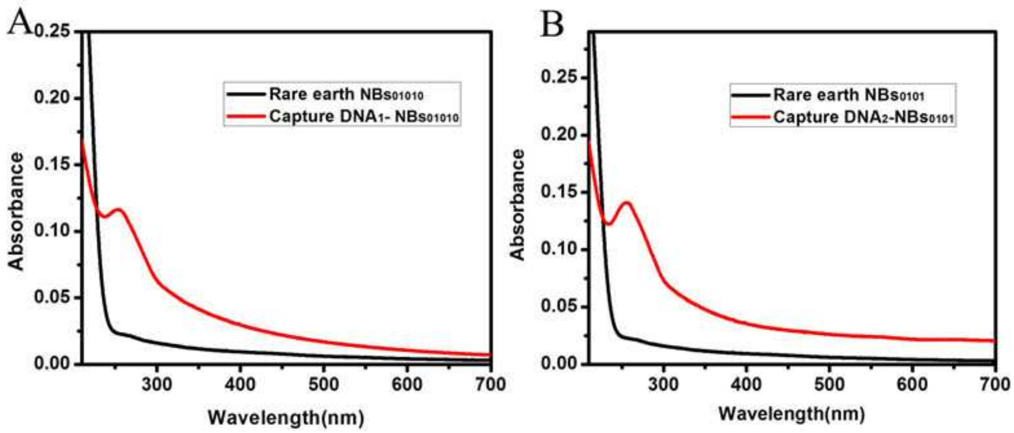


Figure S5. The UV/Vis spectrum of A) the NBs_{01010} in chloroform and capture DNA_1-NBs_{01010} in water; B) the NBs_{0101} in chloroform and capture DNA_2-NBs_{0101} in water.

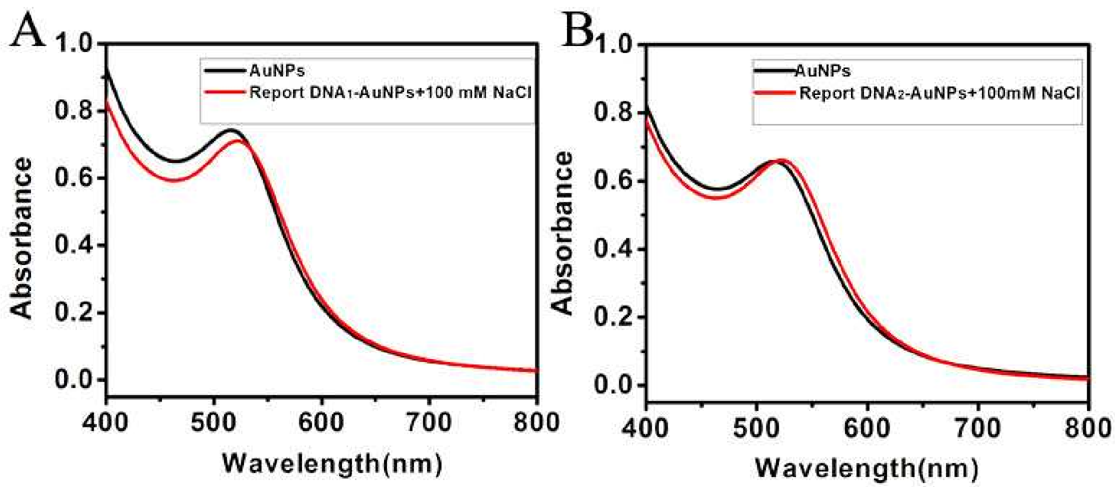


Figure S6. UV-vis spectra of A) AuNPs and report DNA_1-AuNPs sample in solutions with 100 mM NaCl and B) AuNPs and report DNA_2-AuNPs sample in solutions with 100 mM NaCl.

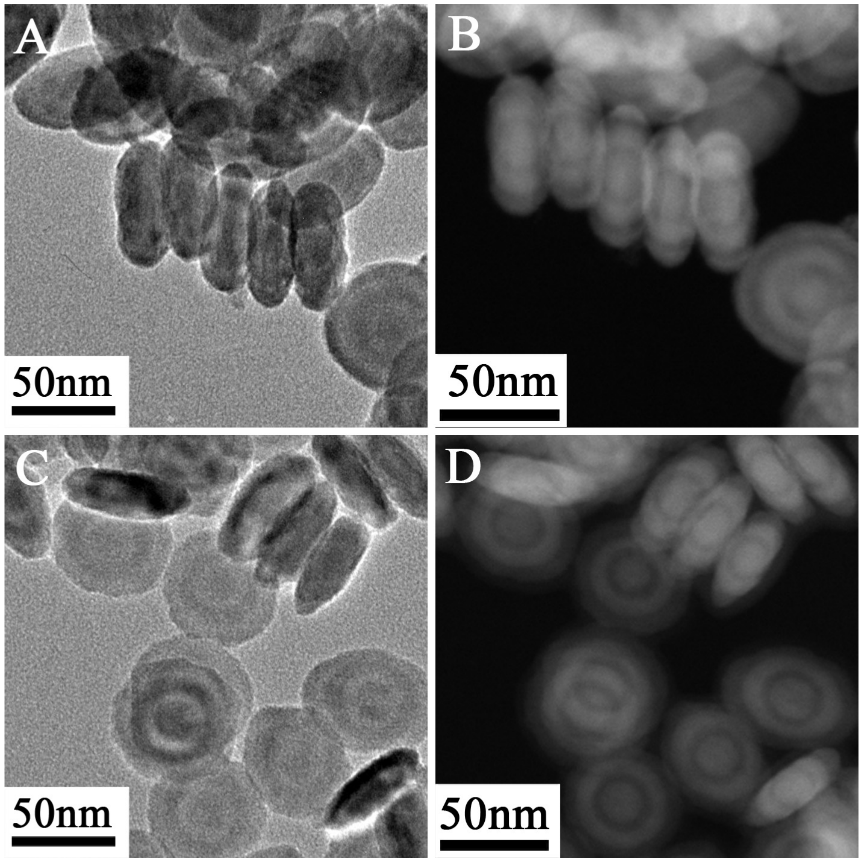


Figure S7. TEM (A, C) and HAADF-STEM (B, D) images of the $NaGdF_4/NaYF_4/NaGdF_4/NaYF_4/NaGdF_4$ (NBs_{01010}) with addition of noncomplementary target DNA_2 (A, B) and $NaGdF_4/NaYF_4/NaGdF_4/NaYF_4$ (NBs_{0101}) with addition of noncomplementary target DNA_1 (C, D).

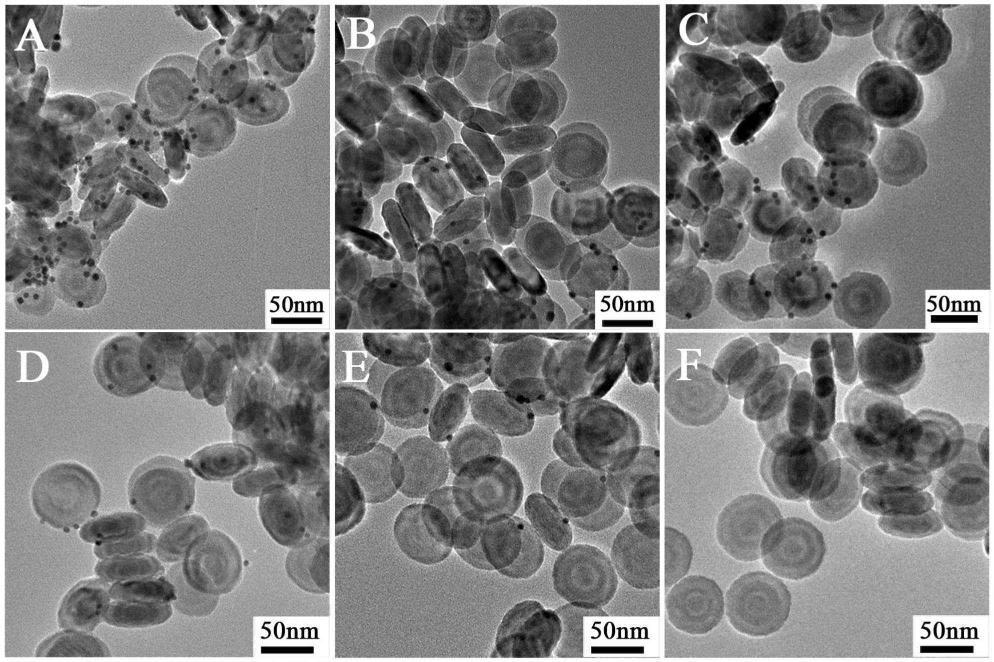


Figure S8. TEM images of the $NaGdF_4/NaYF_4/NaGdF_4/NaYF_4$ (NBs_{0101}) and $NaGdF_4/NaYF_4/NaGdF_4/NaYF_4/NaGdF_4$ (NBs_{01010}) with (A) both target DNA_1 and target DNA_2; (B) only target DNA_1; (C) only target DNA_2. TEM images of the five layer NBs_{01010} for DNA trace detection at target concentration of (D) 60 pM DNA_2, (E) 6 pM DNA_2 and (F) no target DNA.