Table S1. Sequences of ssDNA for the preparation of crosslinkers and spacers.

Name	Sequence (5′ ---> 3′)
Y_1	CGATTGACTCTCCACGCTGTCCTAAC-CATGACCGTCGAAG
Y_2	CGATTGACTCTCCTTCGAC-GGTCATGTACTAGATCAGAGG
Y_3	CGATTGACTCTCCCTCTGATCTAGTAG-TTAGGACAGCGTG
X_1	CGATTGACTCTCCGACCGATGAA-TAGCGGTCAGATCCGTACCTACTCG
X_2	CGATTGACTCTCCGAGTAGGTAC-GGATCTGCGTATTGCGAACGACTCG
X_3	CGATTGACTCTCCGAGTCGTTCGCAA-TACGGCTGTACGTATGGTCTCG
X_4	CGATTGACTCTCCGAGAC-CATACGTACAGCACCGC-TATTCATCGGTCG
C_1	GTGTGTGTGTGTGTGTGTGTGTGTGTGTGTGTGTGTGTGTGATGCGAGGCTATTCT
C_2	CGATTGACTCTCAGAATAGCCTCGCAT
G	CGATTGACTCTCAAAAAAAAAAAAA
Ss_1	GAGAGTCAATCGTCTATTCGAATTCAC-CGTAAG
Ss_2	GAGAGTCAATCGCTTAC-GGTGAATTCGAATAGA
Sm_1	GAGAGTCAATCGTCTATTCGCATGA-GAATTCCATTCACCGTAAG
Sm_2	GAGAGTCAATCGCTTAC-GGTGAATGGAATTCTCATGCGAATAGA
Sl_1	GAGAGTCAATCGTCTATACGCAGGA-GACCAGGAATTCGAAATCATACACCG-TAAG
Sl_2	GAGAGTCAATCGCTTACGGTGTATGAT-TTCGAATTCCTGGTCTCCTGCGTATAGA

Sticky ends are underlined in S1.

Table S2. Concentrations and ratios of crosslinkers and spacers to prepare for hydrogel.

Name	Concentrations of strands with sticky end in the stock solutions	Ratios of final concentrations in the hydrogels
Spacers (S)	1.5 mM	N/A
Y-shaped DNA tile (Y)	1 mM	Y:S = 2:3
X-shaped DNA tile (X)	1 mM	X:S = 1:2
DNA-CNT conjugates	1 mM	DNA-CNT:S = 2:1
DNA-AuNP conjugates	16 μM	DNA-AuNP:S = 1:10

CNT dispersions

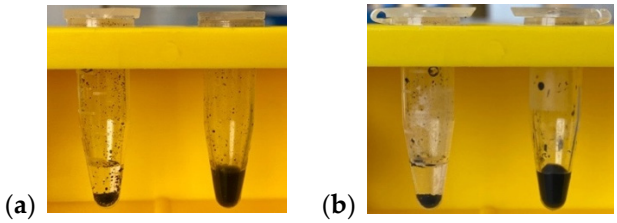

Figure S1. Photos of **(a)** SWNT and **(b)** MWNT dispersions in deionized (DI) water without (left) and with (right) DNA modification. SWNTs and MWNTs without DNA modification settle at the bottom within 15 min after sonication.

Photos of hydrogels

Figure S2. Photos of pure and DNA hydrogel composites constructed by Sl with (from left to right) Y-shaped DNA tiles, X-shaped DNA tiles, DNA–SWNT conjugates, DNA–MWNT conjugates, and DNA–AuNP conjugates.

UV-Vis spectroscopy for AuNPs

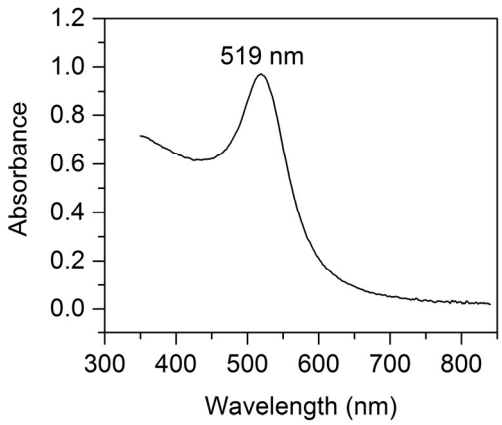

Figure S3. UV–Vis spectrum of as-synthesized AuNPs. A peak λ_{max} at 519 nm wavelength proves that these are well-formed AuNPs with an average diameter of 13 nm [1]. UV–Vis spectrum was recorded on a Thermo Scientific NanoDrop 2000c Spectrophotometer (Waltham, USA).

Gel electrophoresis for AuNPs and DNA-AuNP conjugates

Gel electrophoresis (Agarose, 1%) was carried out in 0.5x TBE buffer (Tris-Borate-EDTA buffer) [2]. First, 1 g of agarose powder was added to 100 mL of 0.5x TBE buffer, and the solution was heated in the microwave for 60 seconds until it became homogeneous and transparent. Then, the agarose solution was casted in a gel tray (Bio-Rad Sub-Cell GT, Hercules, USA) until solid. To prepare for the samples, glycerol (final content 10%) was added to AuNP and DNA-AuNP conjugate stocks, then 10 μL of each of the stocks was loaded in a well in the agarose gel. The gel was run for 1 hour at 100 V in the 0.5x TBE buffer using Bio-Rad PowerPac™ Basic Power Supply. The corresponded electric field

was 9.9 V/cm. After gel electrophoresis, a cell phone was used to take a digital picture of the gel to visualize the mobilities of AuNP and DNA-AuNP conjugate samples in the gel.

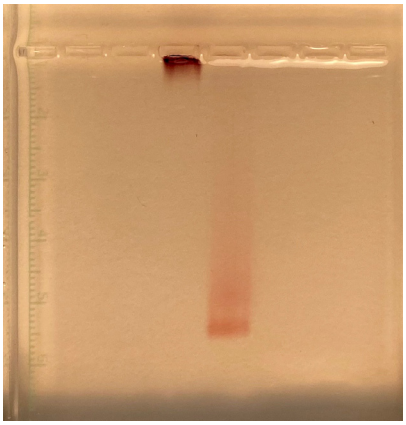

Figure S4. The band of pure AuNPs (left) appears violet under visible light, while the band of DNA–AuNP conjugates (right) retains the red color and shows a much better mobility.

Electrical measurement

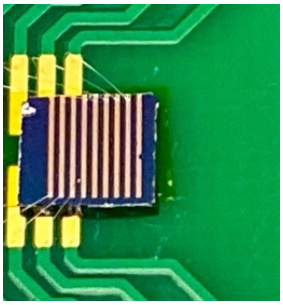

Figure S5. Gold electrodes of 200 μm spacing for electrical characterization.

Supplementary References

1. Hill, H.D.; Mirkin, C.A. The Bio-Barcode Assay for the Detection of Protein and Nucleic Acid Targets using DTT-Induced Ligand Exchange. *Nat. Protoc.* **2006**, *1*, 324-36. doi:10.1038/nprot.2006.51.
2. Pellegrino, T.; Sperling, R.A.; Alivisatos, A.P.; Parak, W.J. Gel Electrophoresis of Gold-DNA Nanoconjugates. *J. Biomed. Biotechnol.* **2007**, *2007*, 26796; doi:10.1155/2007/26796.