Electronic Supplementary Material (ESI) for ChemComm.

Supporting information

Molecular Encoder - Decoder Based on Assembly of Graphene with Dye-labeled DNA

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Experimental Section

1. Chemicals and Apparatus

Tris(hydroxymethyl)aminomethane (Tris), hydrochloric acid (HCl), and sodium chloride (NaCl) were obtained from Sinopharm Chemical Reagent Co.,Ltd (Shanghai, China). Graphene oxide (GO) was synthesized from graphite (Shanghai, China) by a modified Hummers method[1] and characterized by. DNA oligonucleotides were synthesized and purified by Sangon Biotechnology Co., Ltd. (Shanghai, China). The sequences of these oligonucleotides are shown in Table S1. Ultrapure water was prepared by a Millipore Milli-Q system and used throughout.

Table S1 DNA sequences and modification

DNA	Sequences (5′ to 3′)
P1	FAM-CAGAGGCAGTAACCA
P2	ROX-CCTGGTGCCGTAGAT
P3	Cy5-CCCTAATCCGCCCAC
T1	TGGTTACTGCCTCTG
T2	ATCTACGGCACCAGG
T3	GTGGGCGGATTAGGG
R	GCAGAGCCAGTTCCT
T1-T2	TGGTTACTGCCTCTGATCTACGGCACCAGG
T1-T3	TGGTTACTGCCTCTGGTGGGCGGATTAGGG
T2-T3	GTGGGCGGATTAGGGATCTACGGCACCAGG
T1-T2-T3	TGGTTACTGCCTCTGGTGGGCGGATTAGGG ATCTACGGCACCAGG
P-C-P1	TGGTTACTGCCTCTGGCATA ATCTACGGCACCAGG
T4	TGGTTACTGCCTCTGGTGGGCGGATTAGGGTATGCCAGAGGCAGTAACCA

UV-Vis detection was carried out on a UV-Vis spectrophotometer (Agilent 8453, USA). AFM image was taken by using an Innova microscope. Raman spectra were recorded with a LabRamHR equipped by an Ar^+ laser giving the excitation line of 514.5 nm. Fluorescence spectra were measured on a model F-7000 spectrofluorometer (Hitachi, Japan).

2. Fabrication of Molecular Encoder and Decoder

2.1 2-to-1 encoder

10 μL of 1 μM P1 was prepared in Tris-HCl buffer (20 mM, pH 7.4, containing 0.1M NaCl, 5 mM KCl, and 5 mM $MgCl_2$) and mixed with 170 μL of 0.5 mg/mL GO for 5 min. Then 20 μL of 1 μM T1 or 1 μM

R as two possible inputs was added into the mixture solution. After 60 min incubation at room temperature, the fluorescence of the mixture solution was detected.

2.2 4-to-2 encoder

10 μL of 1μM P1 and 10 μL of 50 μM P2 were prepared in Tris-HCl buffer (20 mM, pH 7.4, containing 0.1M NaCl, 5 mM KCl, and 5 mM $MgCl_2$) and mixed with 160 μL of 0.5 mg/mL GO for 5 min, followed by the addition of 20 μL of 1 μM four possible DNA inputs (R, T1, T2, or T1-T2) to the mixture solution, respectively. After allowing the mixture solution to incubate for 60 min at room temperature, the fluorescence of the mixture solution was detected.

2.3 8-to-3 encoder

10 μL of 10 μM P1, 10 μL of 40 μM P2, and 10 μL of 10 μM P3 were prepared in Tris-HCl buffer (20 mM, pH 7.4, containing 0.1M NaCl, 5 mM KCl, and 5 mM $MgCl_2$) and mixed with 150 μL of 0.5 mg/mL GO for 5 min at room temperature, followed by the addition of 20 μL of 1 μM eight possible DNA inputs (R, T1, T2, T1-T2, T1-T3, T2-T3, T1-T2, or T1-T2-T3) to the mixture solution, respectively. After allowing the mixture solution to incubate for 60 min at room temperature, the fluorescence of the mixture solution was detected.

2.4 1-to-2 decoder

10 μL of 1μM P1, 10 μL of 10 μM P2, 10 μL of P-C-P1 were prepared in Tris-HCl buffer (20 mM, pH 7.4, containing 0.1M NaCl, 5 mM KCl, and 5 mM $MgCl_2$) and mixed with 150 μL of 0.5 mg/mL GO for 5 min at room temperature, followed by the addition of two possible DNA inputs (T4 or R) to the mixture solution. After 60 min incubation at room temperature, the fluorescence of the mixture solution was detected.

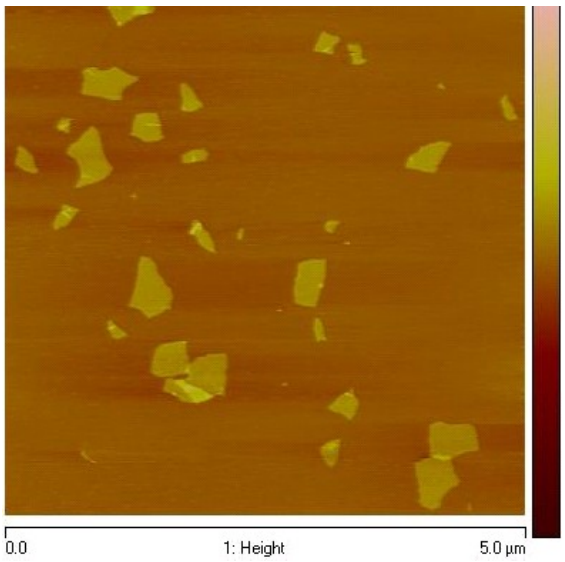


Fig. S1 AFM image of the obtained GO

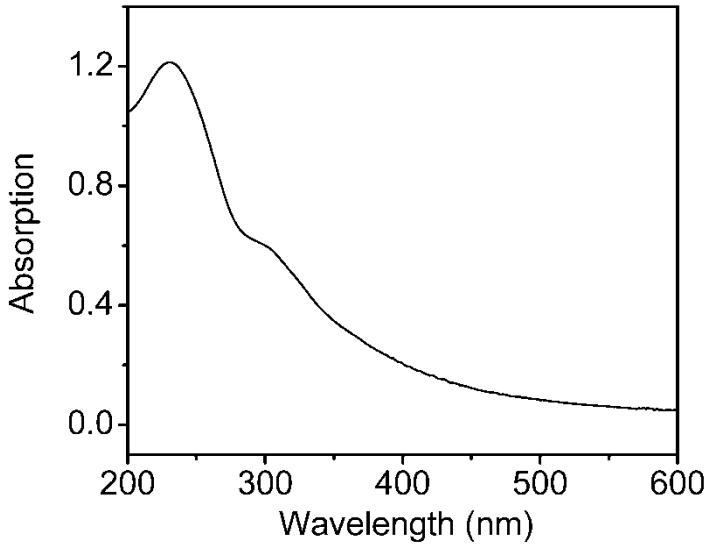


Fig. S2 UV-Vis absorption spectra of the obtained GO

The as-prepared GO was characterized by atomic force microscopy (AFM) and UV-Vis spectra. As shown in Fig. S1, the size of the obtained GO ranged from about 100 to 630 nm. The aqueous solution of the obtained GO displayed a strong absorption peak at 230 nm and a shoulder at ~290-300 nm (Fig. S2), which correspond to π - π^* of C=C and n-π^* transition of C=O band.

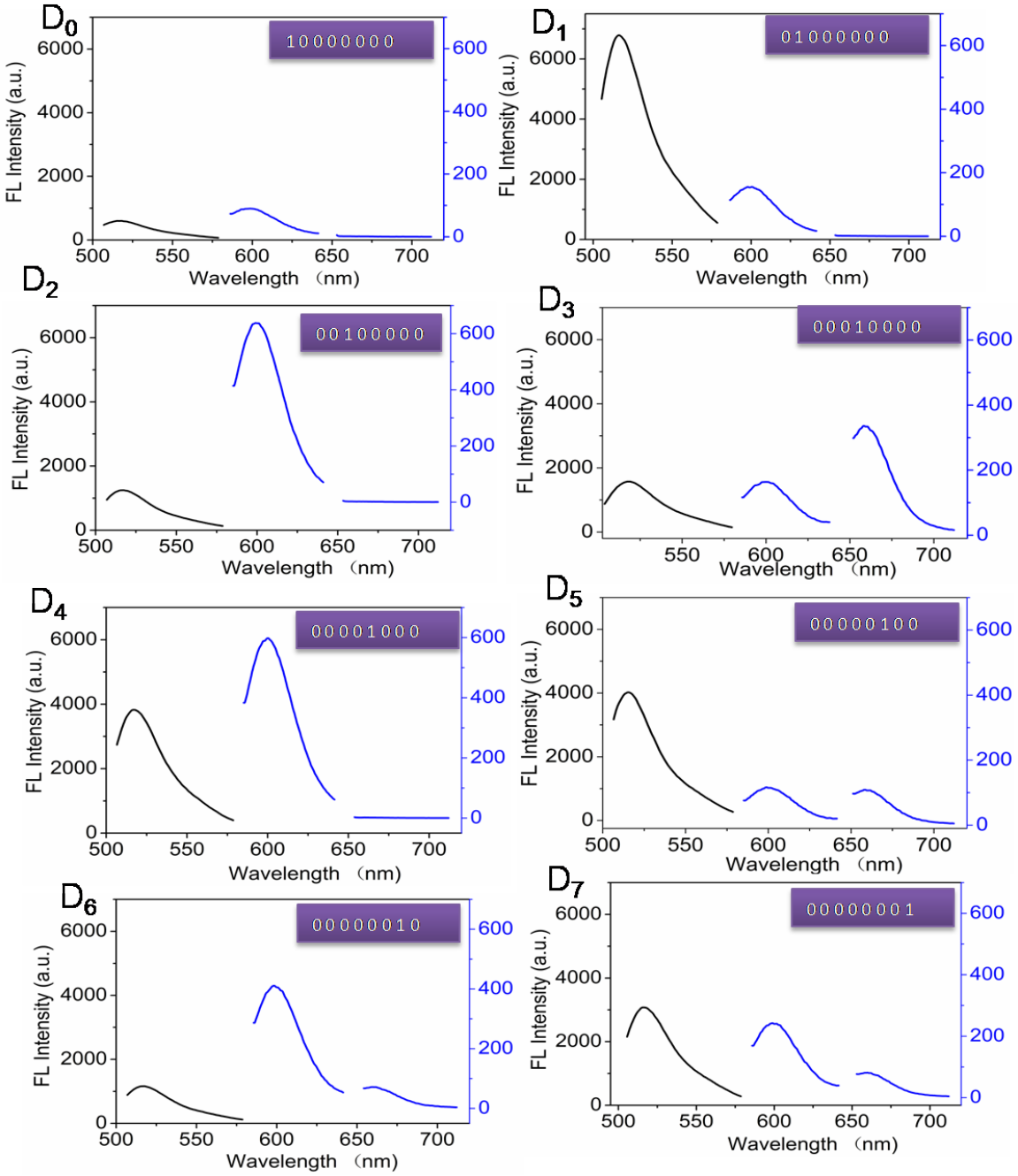
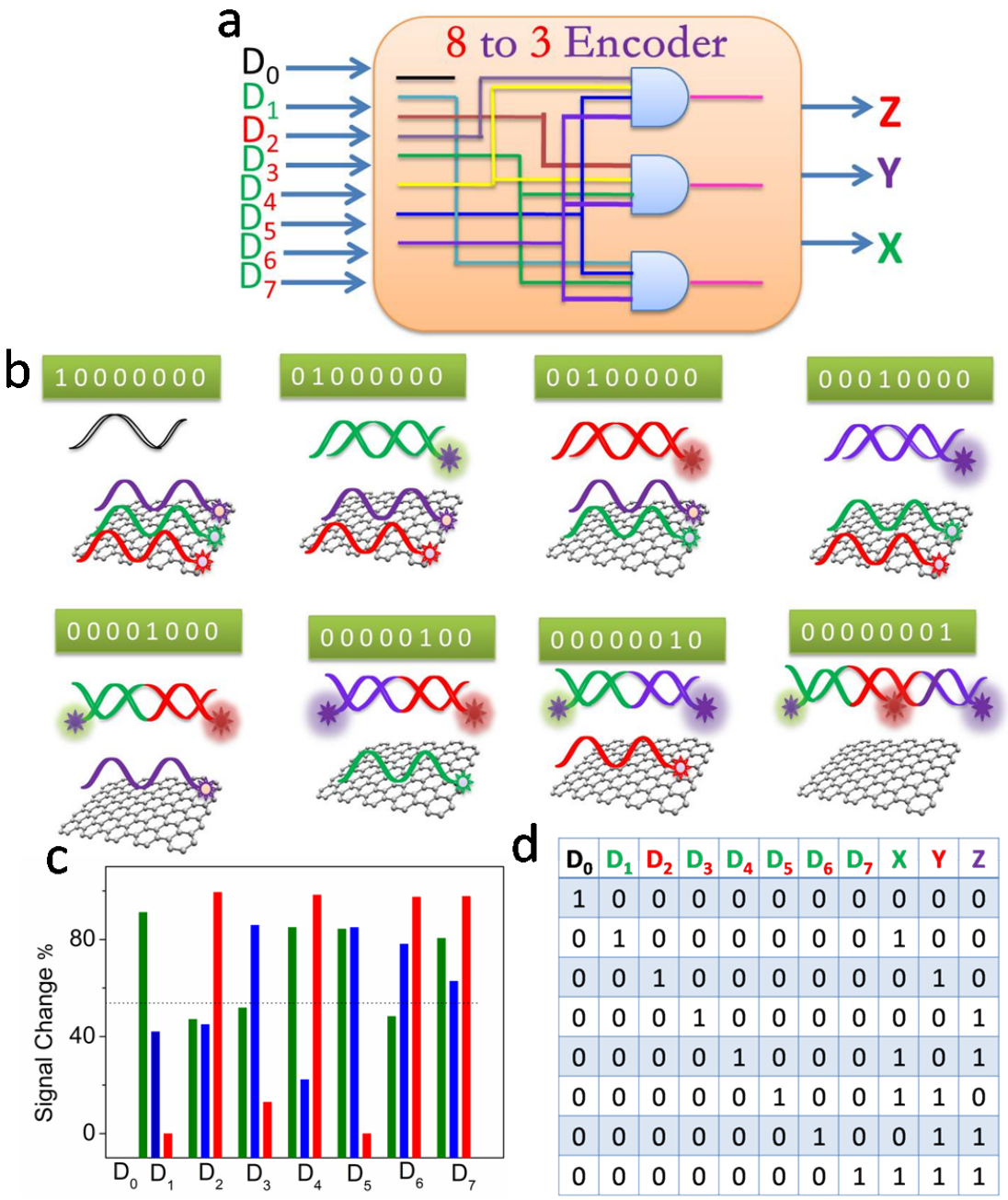


Fig. S3 Fluorescence spectra of the system of GO, P1, P2 and P3 in the presence of different inputs (D_0-D_7) with excitation wavelength of 492, 575, and 643 nm, respectively.



D_0	D_1	D_2	D_3	D_4	D_5	D_6	D_7	X	Y	Z
1	0	0	0	0	0	0	0	0	0	0
0	1	0	0	0	0	0	0	1	0	0
0	0	1	0	0	0	0	0	0	1	0
0	0	0	1	0	0	0	0	0	0	1
0	0	0	0	1	0	0	0	1	0	1
0	0	0	0	0	1	0	0	1	1	0
0	0	0	0	0	0	1	0	0	1	1
0	0	0	0	0	0	0	1	1	1	1

Fig. S4. Schematic representation (a-b) for 8-to-3 encoder and fluorescence change (c) in presence of different input signals. Truth table (d) for 8-to-3 encoder.

It is noted that the fluorescence change in the case of 8-to-3 encorder is smaller than that in the case of 2-to1 or 4-to-2 encorders. This phenomenon is explained as follows. Compared with 2-to-1 and 4-to-2 encoders, 8-to-3 encorder has much more DNA sequence inputs and signal reporters, which might generate the cross reactivity[2, 3], leading to that the fluorescence change of 8-to-3 encorder is smaller than that of 2-to-1 or 4-to-2 encorders.

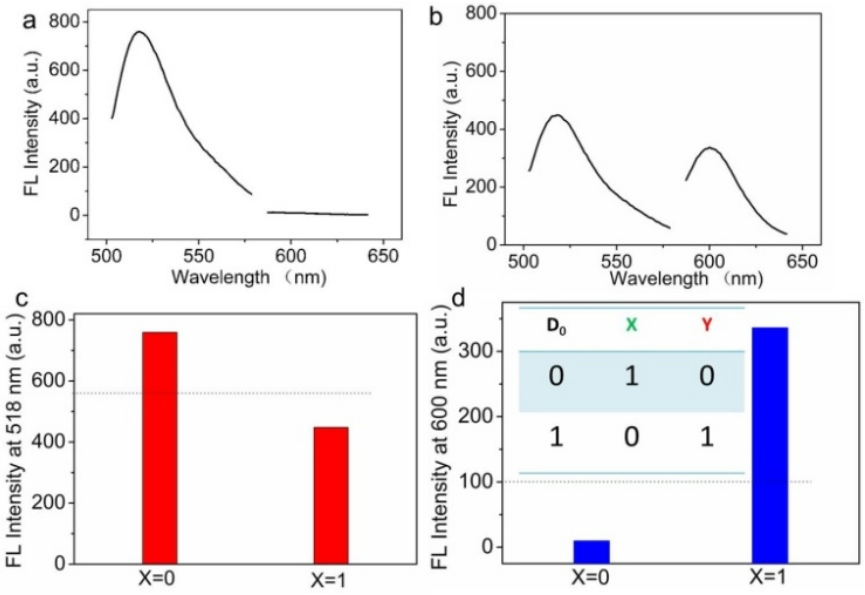


Fig. S5. Fluorescence spectra of 1-to-2 decoder in the absence (a) and presence (b) of input with excitation wavelength of 492 and 575 nm, respectively. Fluorescence intensity of FAM (c) and ROX (d) in the absence and presence of input. The inset displays truth table of 1-to-2 decoder.

Reference

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