

## Electronic Supplementary Information

### A methanol VOC sensor using divalent metal ion-modified 2D DNA lattices†

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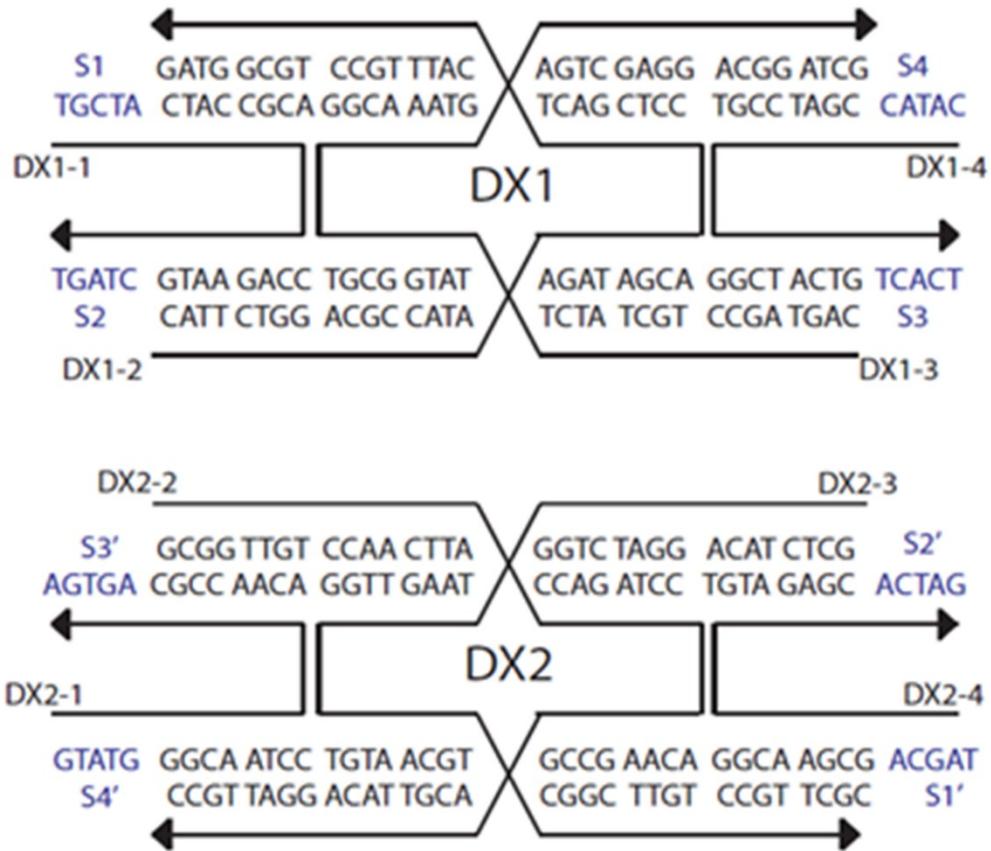
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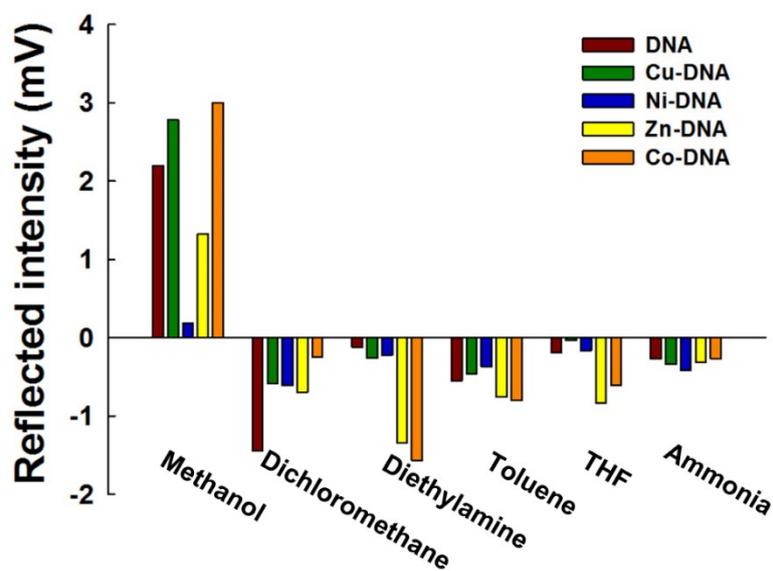
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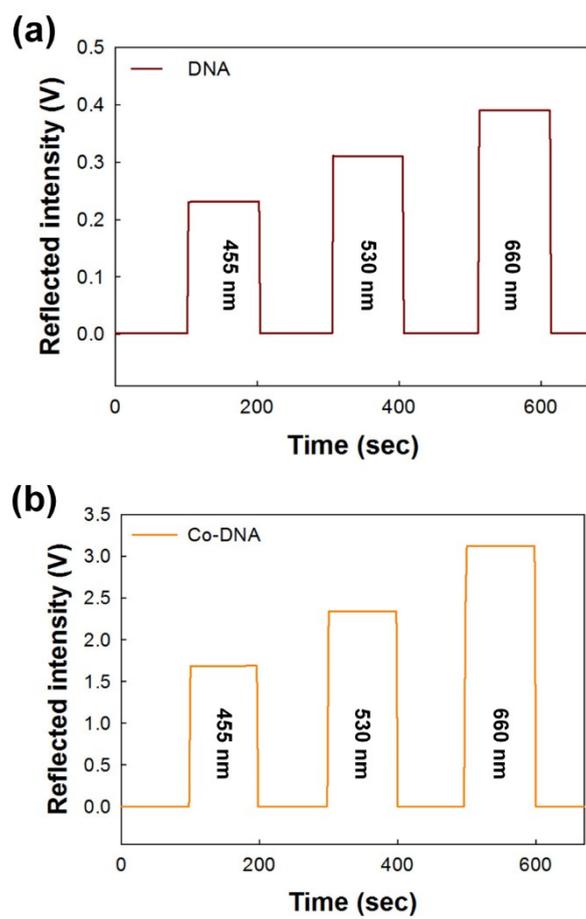
**Fig. S1.** A schematic diagram of the double-crossover (DX) [DX = (DX1) + (DX2)] tiles. Each tile consists of four strands DX1-1, DX1-2, DX1-3, and DX1-4 for (DX1) tile and DX2-1, DX2-2, DX2-3, and DX2-4 for (DX2). The complementary sticky end pairs are shown as S# and S#' in sequence drawings (blue).



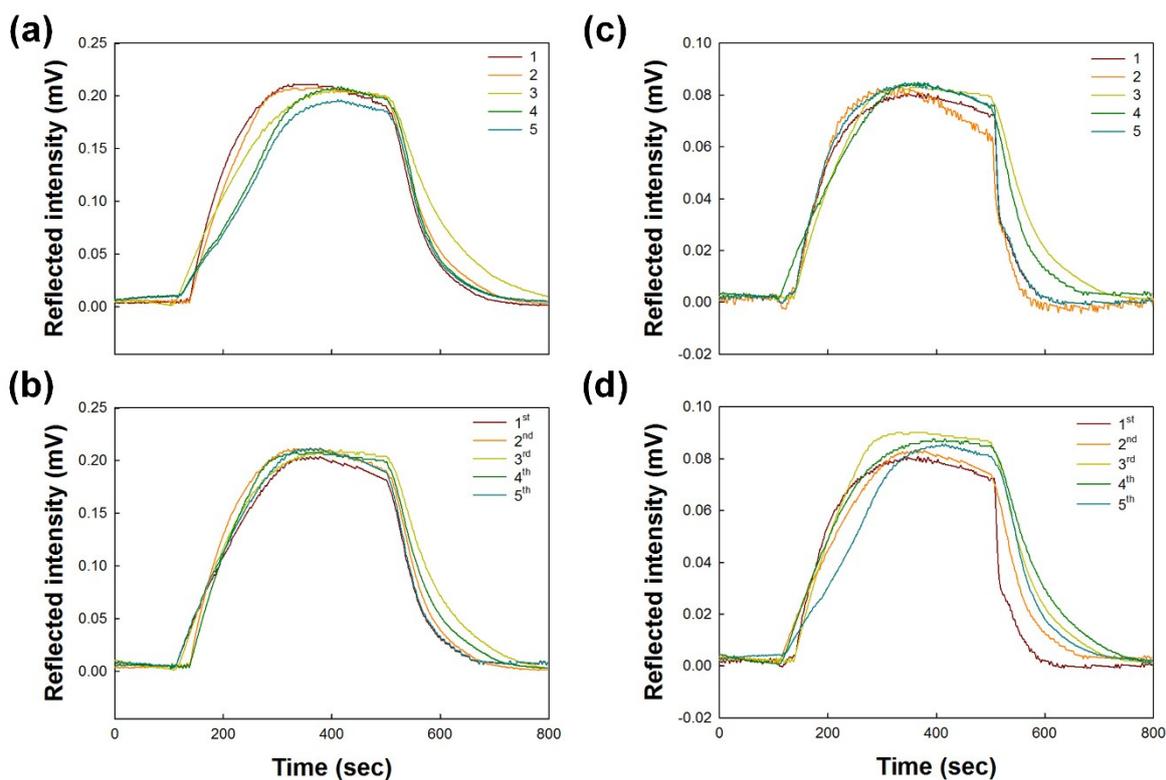
**Fig. S2.** Comparative plots of the reflected intensity to DNA DX lattice and four M-DNA DX lattice with six different VOCs.

Band positions	Intensity changes following VOCs (%)		Band assignment
	DNA	Co-DNA	
740	68.9	73.8	Adenine
1095	94.1	79.4	Phosphate Backbone
1240	74.5	72.5	Cytosine, Adenine
1336	70.0	70.6	Adenine, Guanine
1375	74.1	76.5	Thymine, Adenine, Guanine
1576	61.5	69.7	Adenine, Guanine

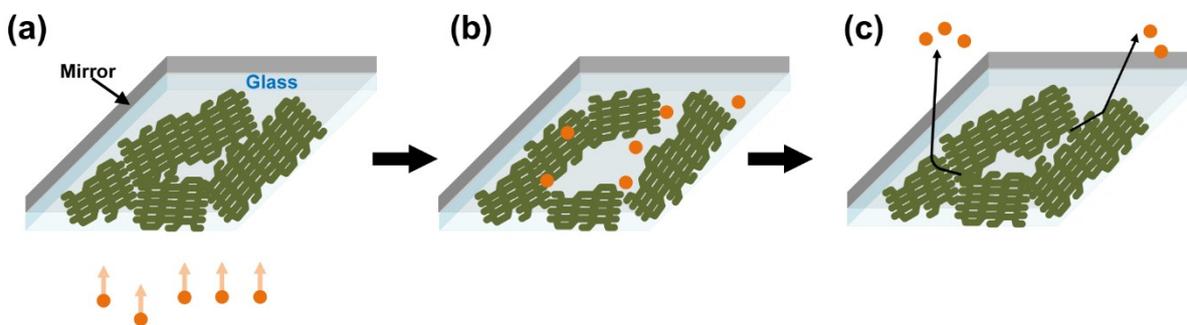
**Table S1.** Assignment of Raman bands of DNA and Co-DNA DX lattices and corresponding changes of reflected light intensities after methanol vapor exposure.



**Fig. S3.** Variation of reflected intensities of (a) pristine DNA and (b) Co-DNA DX lattices with 3 different light sources ( $\lambda = 455, 530$  and  $660$  nm) at fixed optical power ( $60 \mu\text{W}$ )



**Fig. S4.** Verifications of repeatability and reproducibility of Co-DNA and pristine DNA lattices under the influence of methanol with 95 ppm. (a, b) Five consecutive measurements for sensing methanol using single Co-DNA lattice sample and 5 different Co-DNA lattice samples, respectively. (c, d) Five consecutive measurements for sensing methanol using single DNA lattice sample and 5 different DNA lattice samples, respectively.



**Fig. S5.** Schematic representation of the topological changes (eventually affected to changing reflected intensity) of DNA DX lattices (a) during exposure of VOCs to DNA DX lattices, (b) after completion of exposure of VOCs, and (c) after injection of nitrogen gas on DNA DX lattices.