Supporting Information

High-Security Data Encryption Enabled by DNA Multi-Strand Solid-Phase Hybridization and Displacement in Inkjet-Printed Microarrays

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Note1：

When multiple inkjet droplets fall in the same location, they merge to form a larger droplet, as shown in Figure 2d of the main text. In the second, fifth, and seventh columns, single droplets are observed, while the first, third, and sixth columns display double droplet fusion, and the fourth column shows triple droplet fusion. According to statistical analysis, the diameters of droplets formed by one, two, and three small droplets are 34.51 ± 0.93 μm, 45.11 ± 1.64 μm, and 50.85 ± 1.06 μm, respectively. The volume of the droplets is related to their diameter by the formula $V=4\pi d^3(1-\cos^2(A))$, where V is the droplet volume, d is the droplet diameter, and A is the contact angle of the droplet's surface. Thus, the number of fused droplets is proportional to the cube of the diameter. Based on the experimental results, the following relationship was fitted: $d^3=691.05\times n$ ($R^2 = 0.9992$). Therefore, when n=13, the droplet diameter is 83.15 μm.

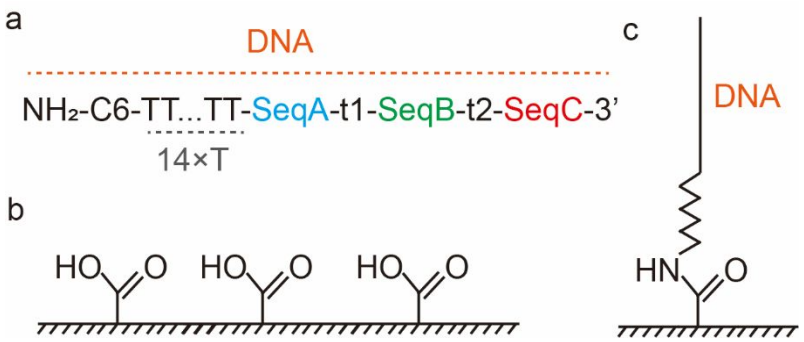

Figure S1. Support DNA strand structure and its connection to the glass substrate. a) The structure of the support DNA strand. It consists of an amino-modified group, a spacer of six carbon atoms and 14 thymine (T) nucleotide, followed by hybridization sequences A, toe sequences t1, hybridization sequence B, toe sequence t2, and hybridization sequence C. b) The carboxyl-modified glass substrate. c) The DNA is connected to the activated carboxyl group on the glass substrate through the formation of an amide bond, facilitated by the amino modification.

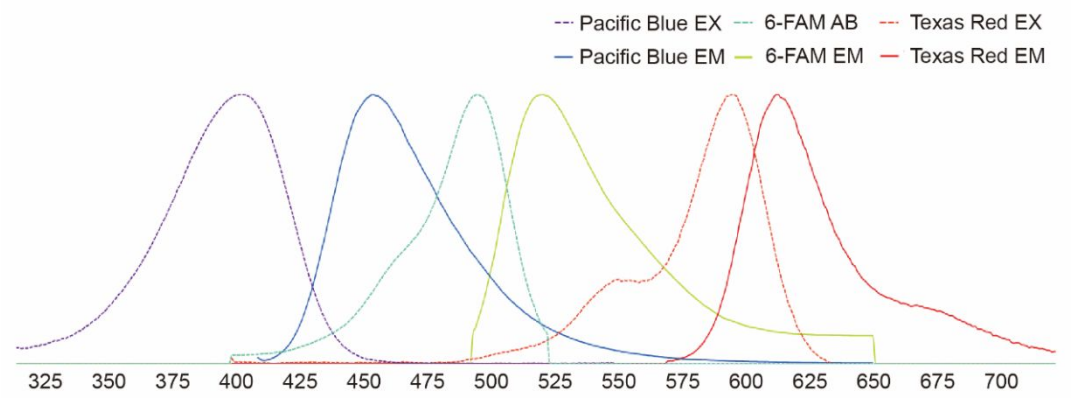

Figure S2. Fluorescence Excitation and Emission Spectra. For DNA strand A*, Pacific Blue is modified at its 5' end, emitting blue fluorescence upon excitation at 405 nm. For DNA strand B*, 6-FAM is modified at its 5' end, emitting green fluorescence upon excitation at 488 nm. For DNA strand C*, Texas Red is modified at its 5' end, emitting red fluorescence upon excitation at 594 nm.

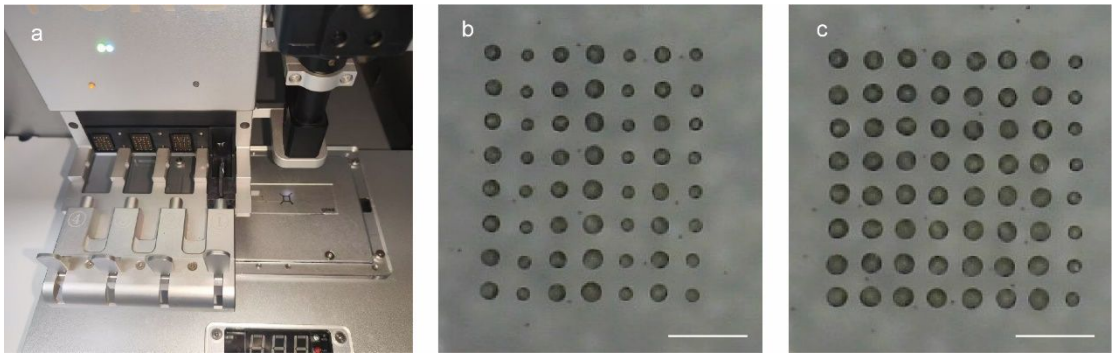

Figure S3. Inkjet printer and microscopic images of droplets formed by inkjet printing. a) The picture of the inkjet printer. b) Microscopic image of droplets formed by directly printing three different data DNA strands. Small droplets formed by single drop, medium droplets formed by two drops merging, and large droplets formed by three drops merging are observed. c) Additional buffer solution is printed onto small and medium droplets to make them form larger droplets, ensuring uniformity of printed dots. A column of buffer solution is printed on the far-right side as a blank fluorescence control.

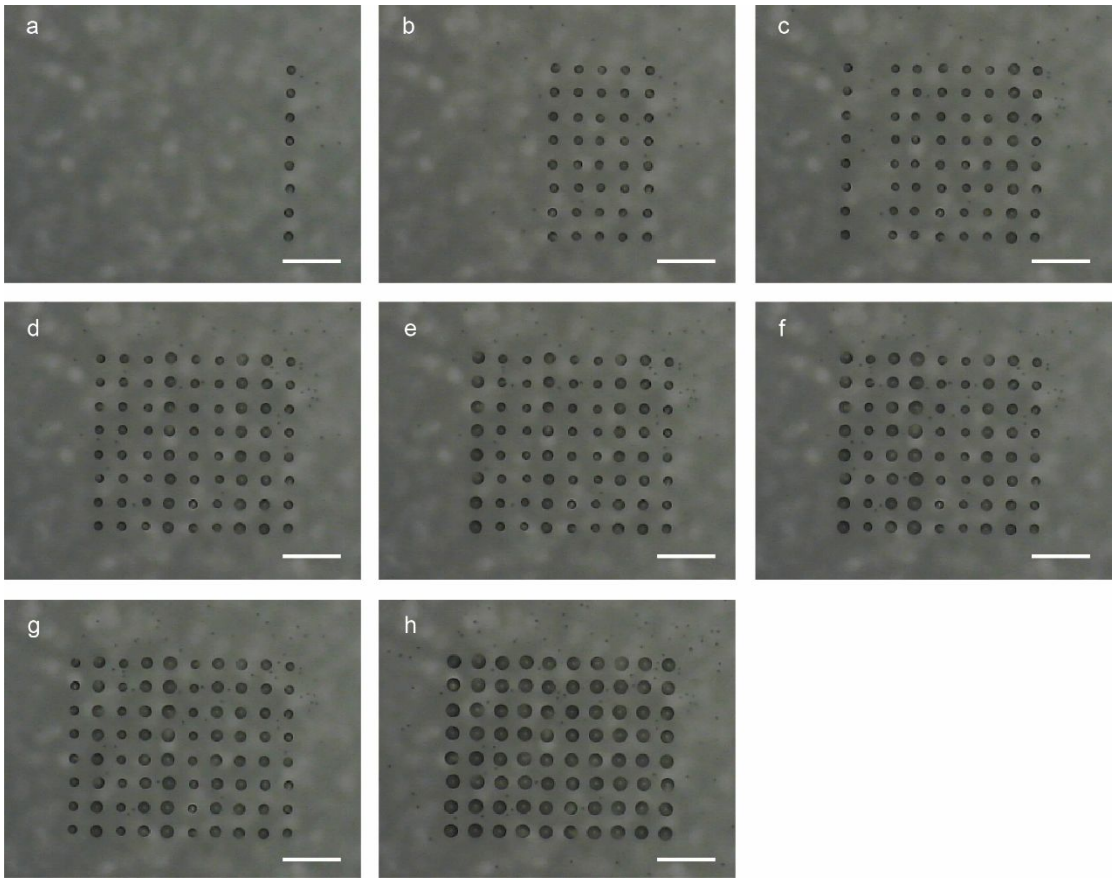

Figure S4. Micrographs showing the printing of data strands and interference strands to form droplets. a) First, droplets of data strand with blue fluorescence were printed. b) Next, droplets of data strand with green fluorescence were printed. c) Droplets of data strand with red fluorescence were printed. d) Droplets of interference strand with blue fluorescence were printed. e) Droplets of interference strand with green fluorescence were printed. f) Droplets of a protective strand that constructs a secondary structure were printed. g) Droplets of interference strand with red fluorescence were printed. h) Droplets of buffer solution to ensure uniform droplet size and

concentration across the pattern were printed.

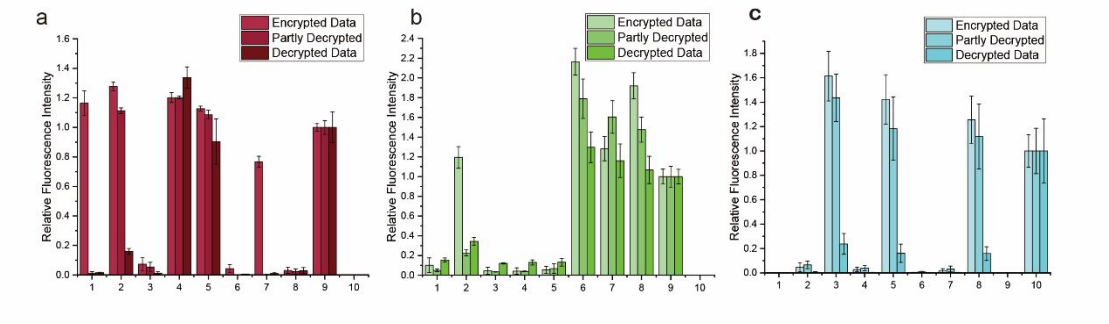

Figure S5. Relative Fluorescence Intensity Results for Encryption demonstration (n=4). a) Fluorescence changes of different red fluorescence after decryption, where the 9th position serves as the standard value. b). Fluorescence changes of different green fluorescence after decryption, where the 9th position serves as the standard value. c) Fluorescence changes of different blue fluorescence after decryption, where the 10th position serves as the standard value.

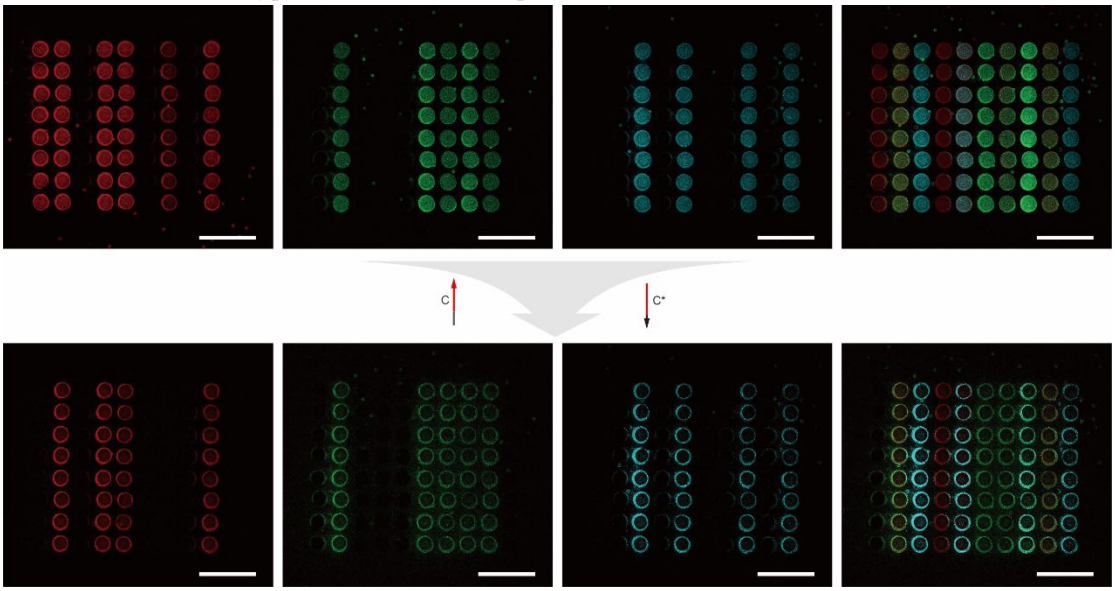

Figure S6. Decryption demonstration with only the red decryption strand. If only the red decryption strand is added in two steps, the interference strands in the first and seventh columns are removed, while the interference strand in the second column remains unaffected.

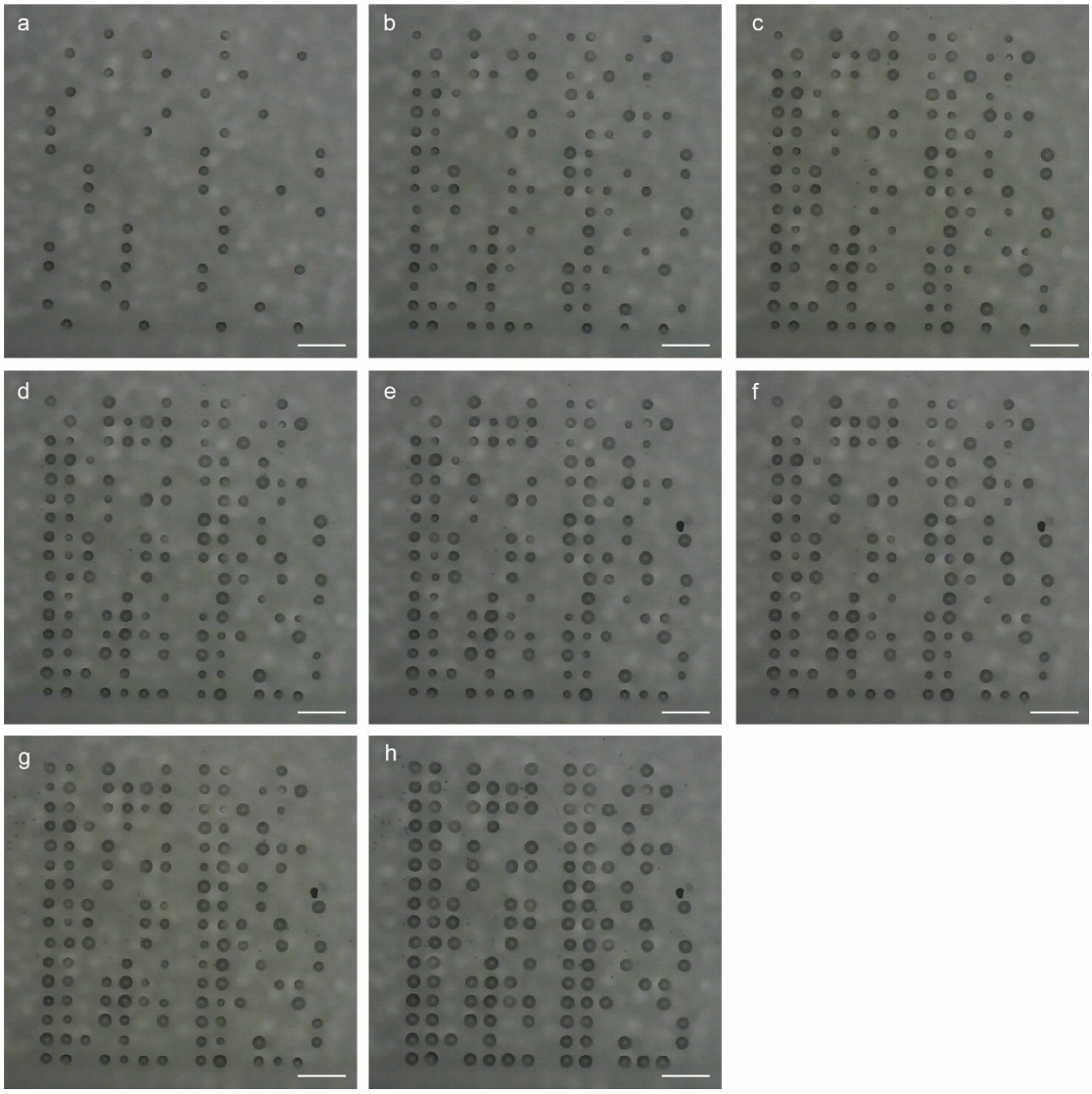

Figure S7. Data array micrographs showing the printing of data strands and encryption strands to form droplets. a) First, droplets of data strand with blue fluorescence were printed. b) Next, droplets of data strand with green fluorescence were printed. c) Droplets of data strand with red fluorescence were printed. d) Droplets of interference strand with blue fluorescence were printed. e) Droplets of interference strand with green fluorescence were printed. f) Droplets of a protective strand that constructs a secondary structure were printed. g) Droplets of interference strand with red fluorescence were printed. h) Droplets of buffer solution to ensure uniform droplet size and concentration across the pattern were printed.

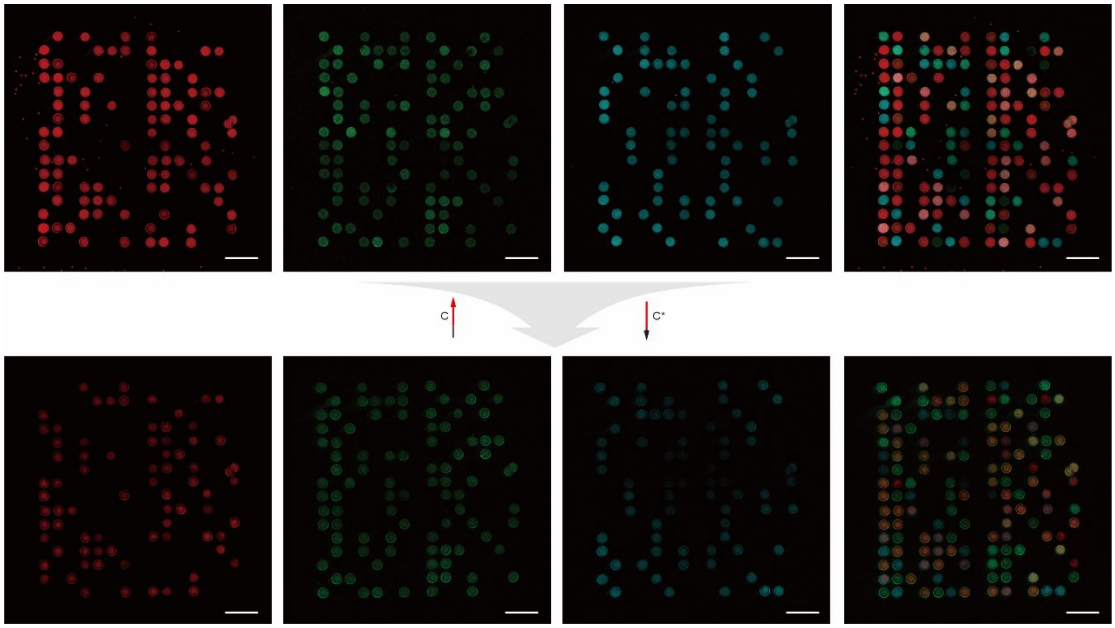

Figure S8. Data decryption effect with only the red decryption strand. When only the red decryption strand is added in two steps, part of the red interference strands is removed. This results in an incomplete decryption, and the red portion is not correctly decrypted.

Table S1 Table of DNA sequences

Support strand	NH2-C6-5‘– TTTTTTTTTTTTTTTTATTCATTTTGGTTTTAACGTGCA TTGTTACGCAAACGTGGGACAGACAGG
Red data strand	Texas Red-5‘-CCTGTCTGTCC
Green data strand 1	6-FAM-5‘-TGCGTAACAATG
Green data strand 2	6-FAM-5‘-CACGTTTGCGTAACAATG
Blue data strand	Pacific Blue-5‘-AAAACCAAAATGAATAA
Red interference strand 1	Texas Red-5‘-CCTGTCTGTCCTTGACC
Red interference strand 2	Texas Red-5‘-CCTGTCTGTCCGCGATT
Red interference strand 2	6-FAM-5‘-CACGTTTGCGTAACAATGATGCTC
Blue interference strand	Pacific Blue-5‘-AAAACCAAAATGAATAACCTTAG
Red decryption strand 1	5‘-GGTCAAGGACAGACAGG
Red decryption strand 1	5‘- AATCGCGGACAGACAGG
Red decryption strand 3	5‘- CCTGTCTGTCCCACGTT

Green protection strand	5‘-CACGTTTGCGTAACAATG
Green decryption strand 3	5‘- GAGCATCATTGTTACGCAAACGTG
Blue decryption strand 3	5‘-CTAAGGTTATTCATTTTGGTTTT