Supporting Information

Beyond the Helix Pitch: Direct Visualization of Native DNA in Aqueous Solution

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Supporting Information Figures S1–S11

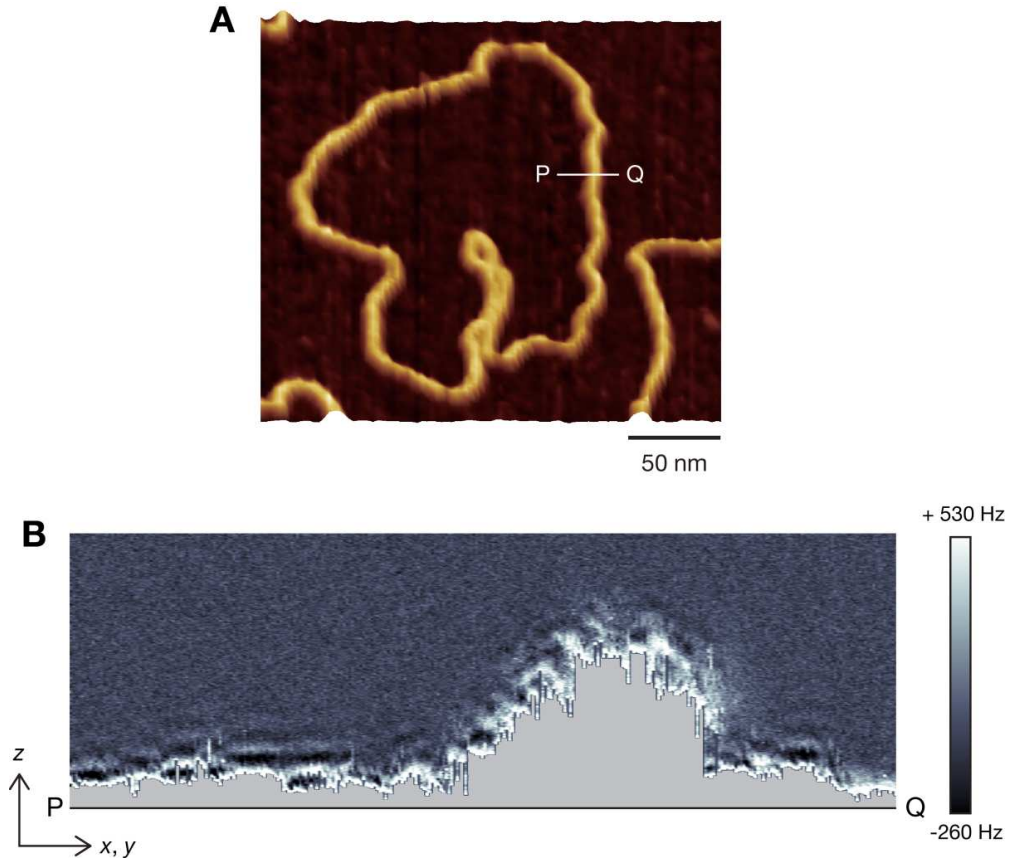

Figure S1. Two-dimensional (2D) frequency shift (Δ*f*) mapping of the plasmid DNA (pUC18) in aqueous solution by FM-AFM. The method of the 2D-Δ*f* mapping by FM-AFM has been described elsewhere.[1–2] (A) FM-AFM image of the plasmid DNA in 50 mM $NiCl_2$ solution. 2D-Δ*f* mapped area is indicated by the P–Q line. (B) 2D-Δ*f* image in a vertical cross-section along the P–Q line in (A). An Si cantilever with an Au backside coating (Nanosensors, PPP-NCHAuD) was used (k = 42 N/m, amplitude of oscillation (a): 0.15 nm). Image size is 33.6 nm × 4.5 nm.

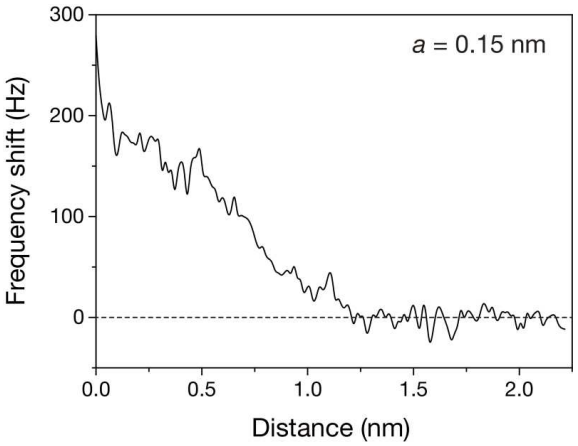

Figure S2. Averaged one-dimensional (1D) Δf versus distance curve (1D-Δf curve) on the plasmid DNA (pUC18) in aqueous solution (50 mM $NiCl_2$) extracted from the 2D-Δf map in Figure S1(B).

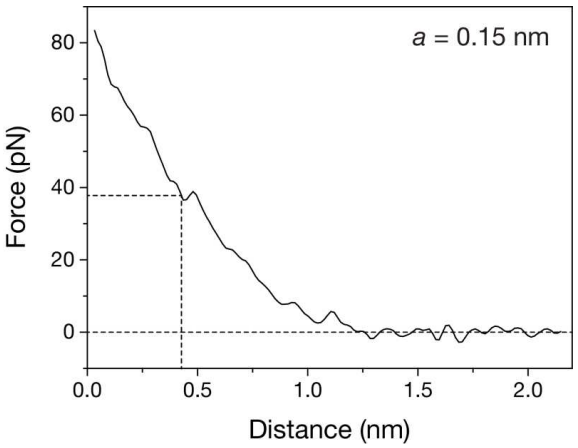

Figure S3. Force versus distance curve (force curve) on the plasmid DNA (pUC18) by FM-AFM in aqueous solution (50 mM $NiCl_2$). The formula proposed by J. E. Sader and S. P. Jarvis[3] was used to convert the 1D-Δf curve (Figure S1) into the force curve.

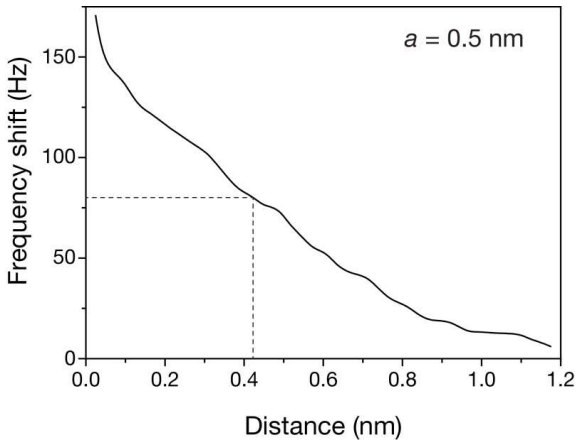

Figure S4. Estimated Δf curve on the plasmid DNA (pUC18) in aqueous solution. The formula derived by F. J. Giessibl[4] was used to reconvert the force curve (Figure S2) into the 1D-Δf curve based on the experimental conditions used in this study (a = 0.5 nm). The tip–sample interaction force in this study (Δf = +80 Hz) was estimated to be about +40 pN (see also Figures S2 and S3).

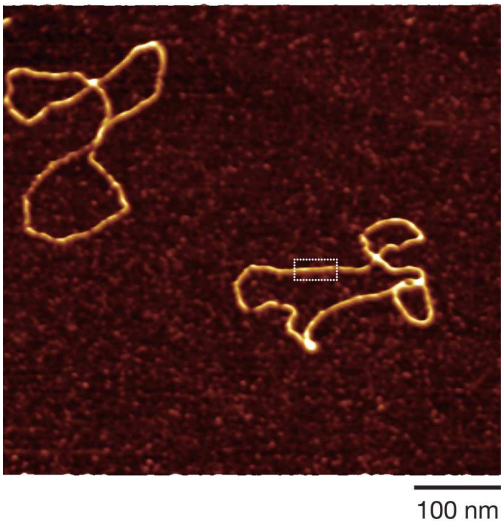

Figure S5. Large area FM-AFM topographic image of the plasmid DNA (pUC18) in aqueous solution (50 mM $NiCl_2$). The white rectangle shows the area where the high-resolution FM-AFM image (Figure 1(B)) was imaged. Contour length of the plasmid DNA was roughly estimated at about 950 nm (axial rise: 0.35 nm/bp).

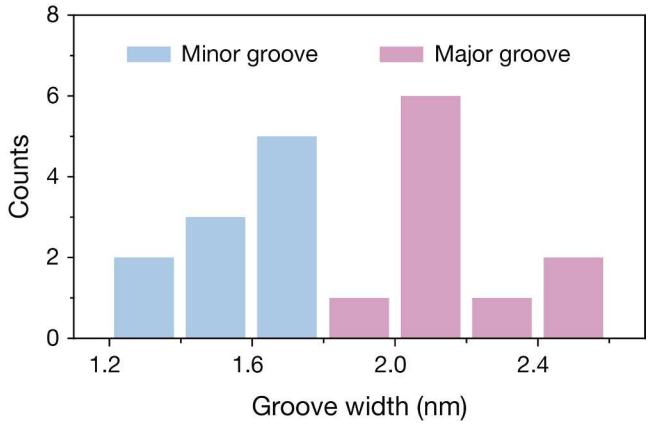

Figure S6. Histogram of the groove widths of B-DNA measured in Fig. 1(B).

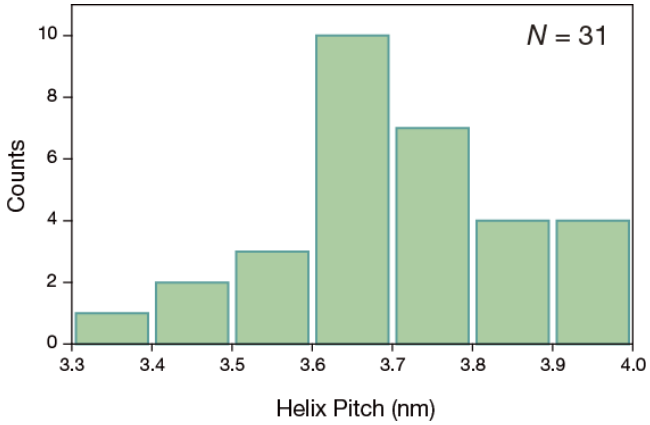

Figure S7. Histogram of the helix pitch of B-DNA measured in the high-resolution FM-AFM image of the plasmid DNA shown in Figure 1(B).

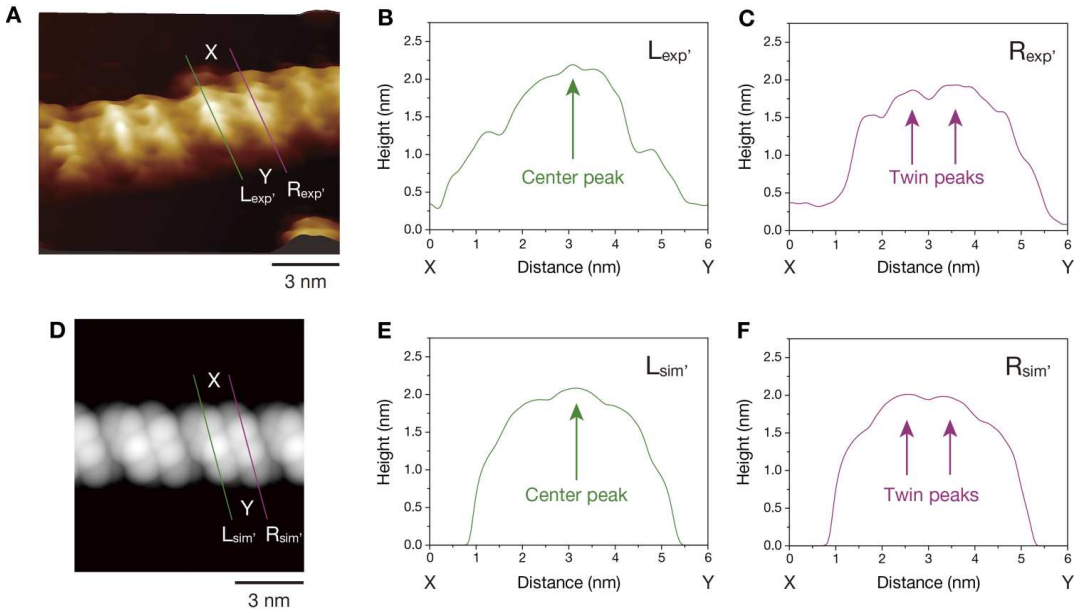

Figure S8. Comparison of cross-sectional profiles along adjacent DNA backbones with a minor groove in between extracted from FM-AFM and simulated AFM images. (A) FM-AFM image of the plasmid DNA in aqueous solution (Figure 2(A)). (B and C) Cross-sectional profiles along the green ($L_{exp'}$) and purple ($R_{exp'}$) lines in (A), respectively. (D) Simulated AFM images of B-DNA (Figure 2(D)). (E and F) Cross-sectional profiles along the green ($L_{sim'}$) and purple ($R_{sim'}$) lines in (D), respectively. In (B), a center peak is observed, whereas twin peaks are observed in (c). These two type of corrugations reflect different positions of phosphate groups on the DNA backbones as observed in (E) and (F).

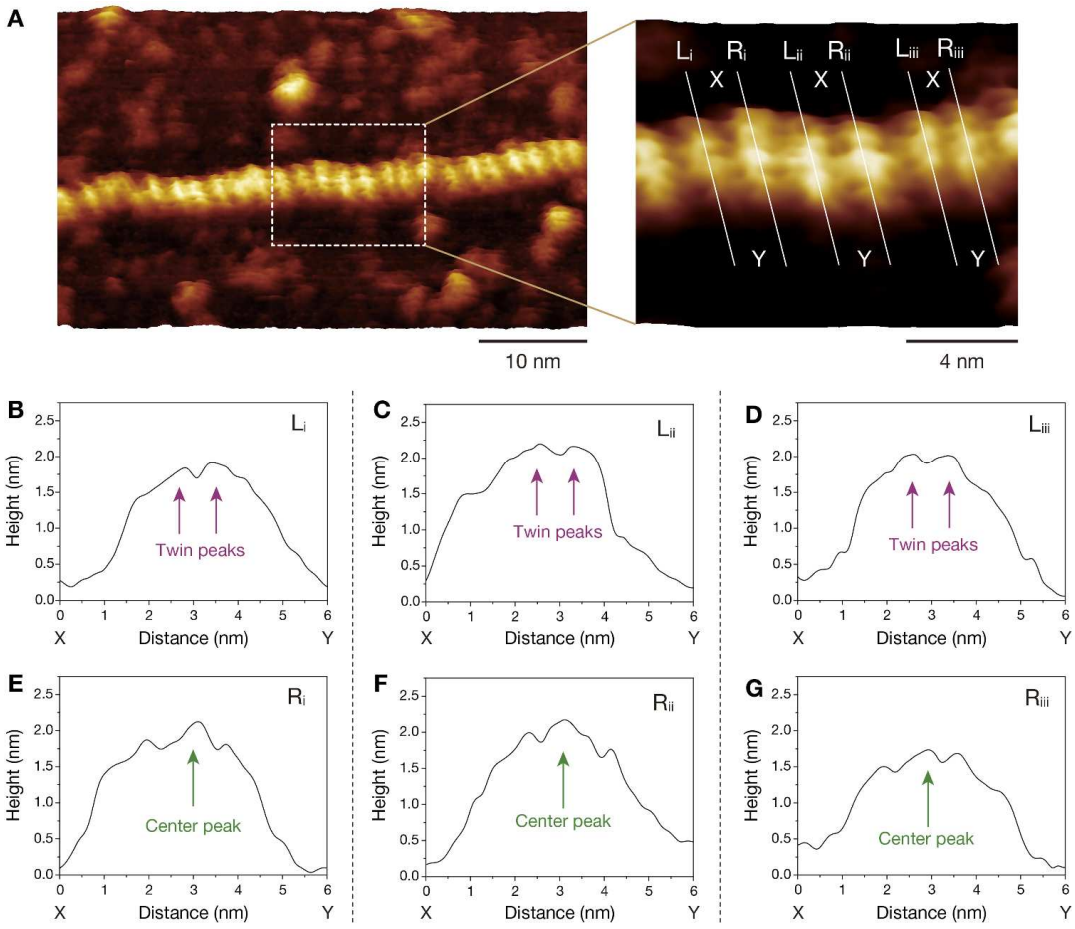

Figure S9. Additional cross-sectional profiles along three sets of two DNA backbones with a minor groove in between extracted from the FM-AFM image. (A) FM-AFM image of the plasmid DNA in aqueous solution (Figure 1(B)). (B–D) Cross-sectional profiles along the L_i, L_{ii}, and L_{iii} lines in (A), respectively. (E–G) FM-AFM image of the plasmid DNA in aqueous solution (Figure 1(B)). (B–D) Cross-sectional profiles along the R_i, R_{ii}, and R_{iii} lines in (A), respectively. In (B–D), twin peaks are observed, whereas a center peak is observed in (E–G). These results indicate that the shift of corrugation phase on the DNA backbones is non-sequence-specific (Figure S10).

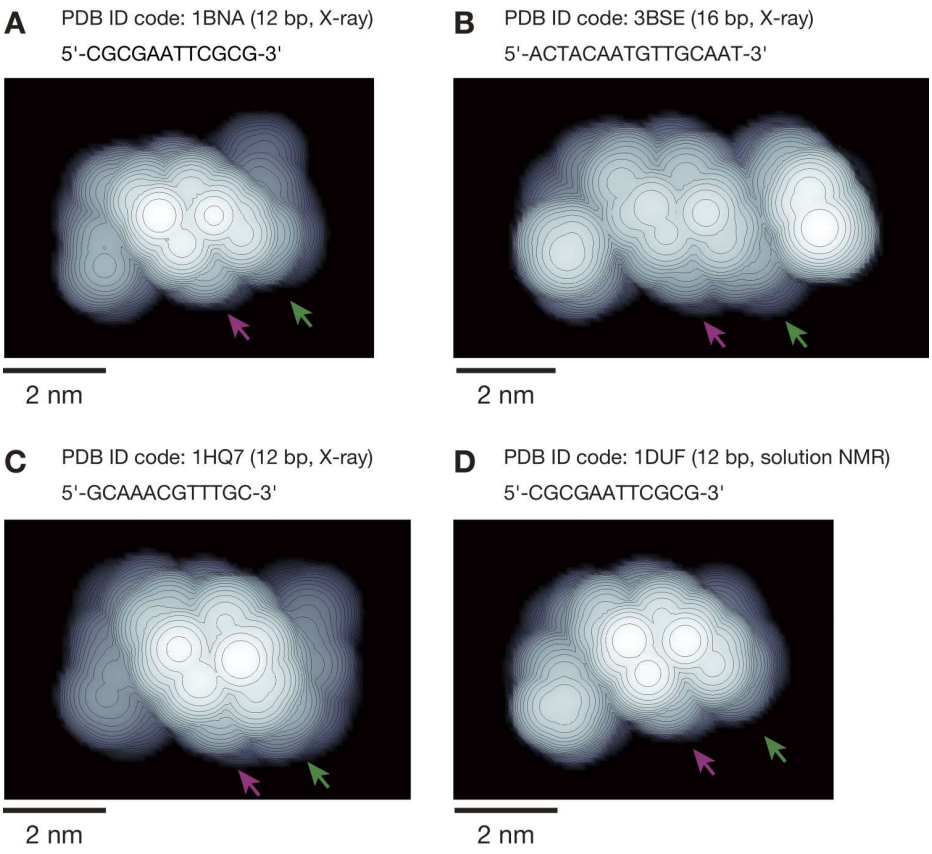

Figure S10. Simulated AFM images of B-DNA assuming tip radius of 1.0 nm. Atomic coordinate data obtained from the Protein Data Bank were used for the simulation. (A) PDB ID code: 1BNA.[5] (B) PDB ID code: 3BSE.[6] (C) PDB ID code: 1HQ7.[7] (D) PDB ID code: 1DUF.[8] Green and purple arrows indicate the backbones with a center peak and with twin peaks, respectively.

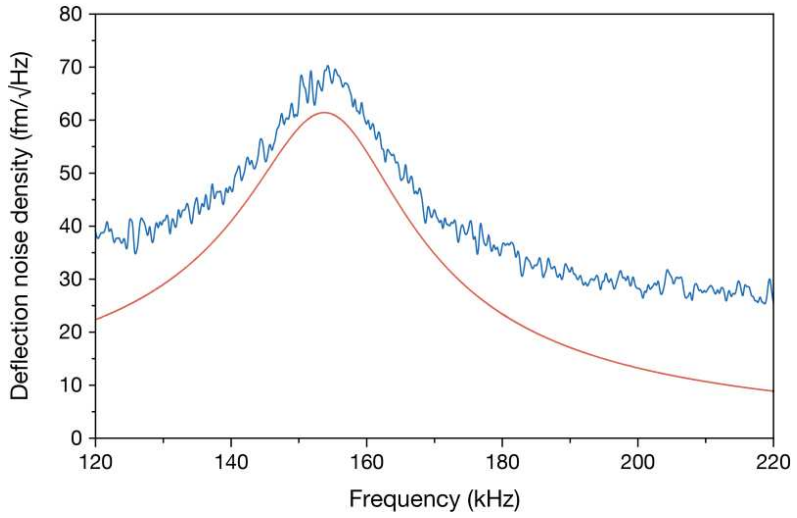

Figure S11. Typical frequency spectrum of the cantilever Brownian motion in water (Q = 6.6, f_0 = 154.7 kHz). The blue curve shows experimentally measured values, whereas the red curve shows theoretically calculated values.[9] An Si cantilever with an Au backside coating (Nanosensors, PPP-NCHAuD) was used (k = 42 N/m). In this experimental condition, the noise-equivalent displacement density was estimated to 20 fm/√Hz.

Supplementary References 1–9

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