

Supplementary Information

Light-induced contraction and extension of single macromolecules on a modified graphite surface

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1) Preservation of the lamellar structure of ODA monolayer after deposition of **P1** as well as after UV- and visible-light irradiation

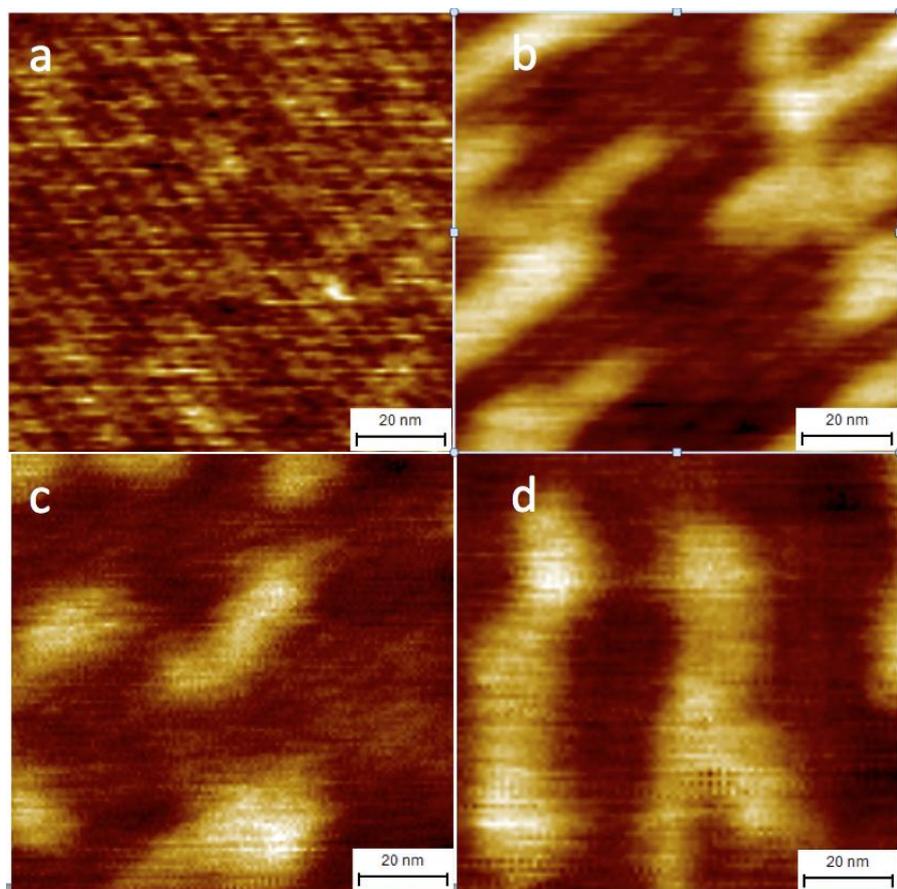
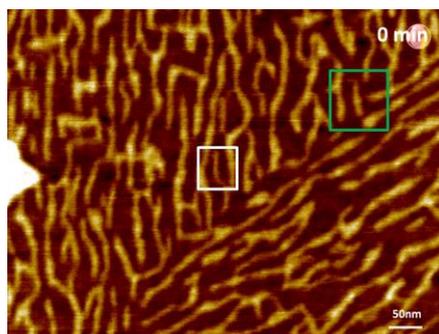


Figure S1. AFM images (100 nm × 100 nm) of (a) ODA lamellar structure formed on graphite with width $w = 6 \pm 0.2$ nm, measured by image Fourier transform. (b) After deposition of **P1** polymer, ODA lamellae were preserved with width $w = 6.3 \pm 0.3$ nm. (c) After 19 minutes of UV irradiation, lamellae remained observable with width $w = 5.8 \pm 0.3$ nm. (d) Finally, after 10 minutes of 436 nm irradiation, lamellae remained observable with width $w = 5.7 \pm 0.3$ nm. The morphology of lamellar structure remained straight and undistorted even in the regions close to **P1** polymers.

It was observed that the morphology as well as the width of the ODA lamellae were not substantially altered by irradiation with light of different wavelengths (the variations lie within error of measurement). Therefore, it is assumed that ODA lamellae remain well adsorbed on the graphite surface.

2) Snapshots of **P1** polymers undergoing contractions and extensions



Movie S1. In this movie, the SFM images were taken after irradiation at $\lambda = 365$ nm for 30 s, 2 min, 4 min, 9 min, 19 min, and then at $\lambda = 436$ nm for 10 min. The white squares are shown in the manuscript in Figure 3, the green squares in Figure 4, and the red squares (appearing at 0'28" in the movie) in Figure 5.

3) Shrinking of **P1** polymers upon UV and blue light irradiation

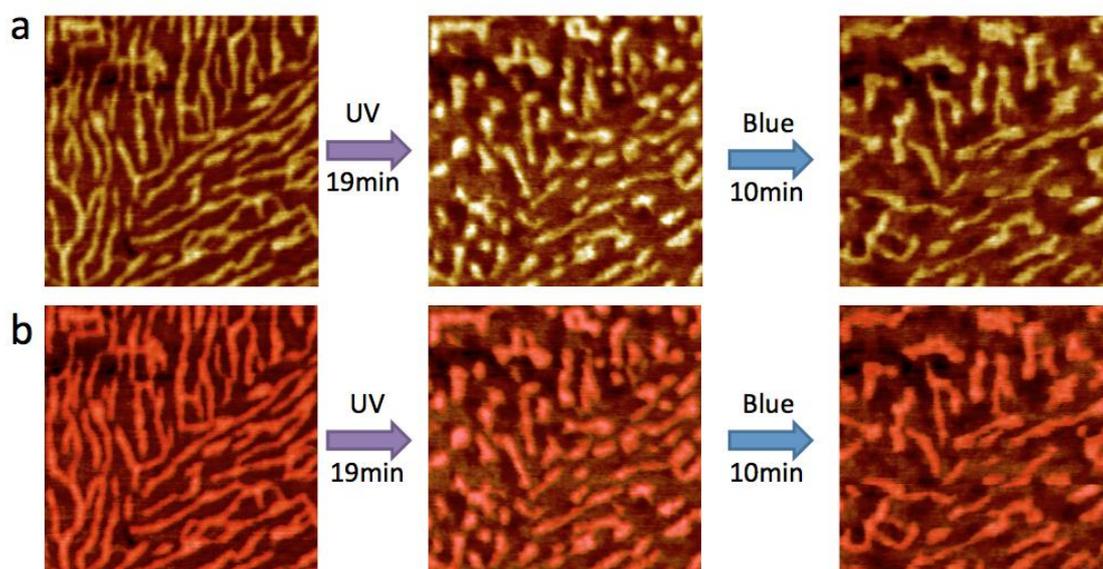


Figure S2. (a) SFM images of the same area ($475 \text{ nm} \times 475 \text{ nm}$) before and after UV-light irradiation, as well as after back switching upon blue-light irradiation, and (b) the corresponding **P1** areas marked in red with a setting height threshold of 0.3 nm.

Two SFM images of the same area ($475 \text{ nm} \times 475 \text{ nm}$) are extracted from **Movie S1** at 0 min and 19 min. Polymer areas are calculated by setting the height threshold to 0.3 nm. The **P1** areas before and after UV-light irradiation amount to 93.1 nm^2 and 71.5 nm^2 , respectively,

resulting in an average area shrinkage of 23.2%. After reverse *cis* → *trans* isomerization, which was induced by irradiation with blue light at 436 nm, several isolated molecules were found to stretch back. However, since at the same time many molecules also aggregate with nearest neighbors, we refrain from discussing average numbers.