## **Supporting Information**

## **Legends for Supplementary Movies**

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**Supplementary Movie 1.** Single-molecule imaging of a photodegradation reaction of a chiral helical  $\pi$ -conjugated polymer, (–)-poly(MtOCAPA), on a mica substrate under n-octylbenzene at room temperature. A violet laser of 405 nm wavelength was irradiated since 8.4 s. In the movie, the violet triangle shows a region during a violet laser irradiation. XY: 150 nm x 150 nm, Z: 10 nm. Frame rate: 12.4 fps. Laser power: 0.2 mW (as the outgoing ray from the objective). [1]

**Supplementary Movie 2.** Single-molecules imaging of micro-Brownian movement in a chiral helical  $\pi$ -conjugated polymer chain, (+)-poly(ChOCAPA), on an APS-coated mica under n-octylbenzene at 25 ± 1 °C. XY: 500 nm x 375 nm, Z: 16.8 nm, Frame rate: 5.0 fps (x4 play = 20 fps). [2]

**Supplementary Movie 3.** Single-molecule imaging of a macromolecular walking along a chiral helical  $\pi$ -conjugated polymer chain, (–)-poly(ChOCPA), on an APS-coated mica under n-octylbenzene at 25 ± 1 °C. XY: 250 nm x 188 nm, Z: 8.4 nm, Frame rate: 5.0 fps (x3 play = 15 fps). [2]

## **References:**

- 1. K. Shinohara et al., J. Polym. Sci. Part A: Polym. Chem. 48, 4103–4107 (2010)
- 2. K. Shinohara et al., *Polymer Preprints, Japan* **62,** 2474-2475 (2013)