**Supporting Information**

**Table S1. Solubility of Poly(1a)−Poly(3)** [a]

| | poly(**1a**) | poly(**1b**) | poly(**2a**) | poly(**2b**) | poly(**3**) |
|---|---|---|---|---|---|
| hexane | − | − | − | − | − |
| toluene | ± | + | ± | ± | + |
| $CHCl_3$ | + | + | + | + | + |
| THF | + | + | + | + | + |
| MeOH | − | − | − | − | − |
| DMF | ± | ± | + | + | + |
| DMSO | − | − | − | − | − |

[a] +, soluble; ±, partly soluble; −, insoluble.

**Table S2. IR Spectroscopic Data of 1a–3 and Poly(1a)–Poly(3)** [a]

| compound | amide I ($cm^{-1}$) | amide II ($cm^{-1}$) | compound | amide I ($cm^{-1}$) | amide II ($cm^{-1}$) |
|---|---|---|---|---|---|
| **1a** | 1683 | 1521 | poly(**1a**) | 1650 | 1545 |
| **1b** | 1683 | 1521 | poly(**1b**) | 1650 | 1547 |
| **2a** | 1681 | 1520 | poly(**2a**) | 1649 | 1541 |
| **2b** | 1682 | 1520 | poly(**2b**) | 1656 | 1542 |
| **3** | 1685 | 1515 | poly(**3**) | 1661 | 1531 |

[a] Measured in $CHCl_3$ ($c$ = 50 mM).

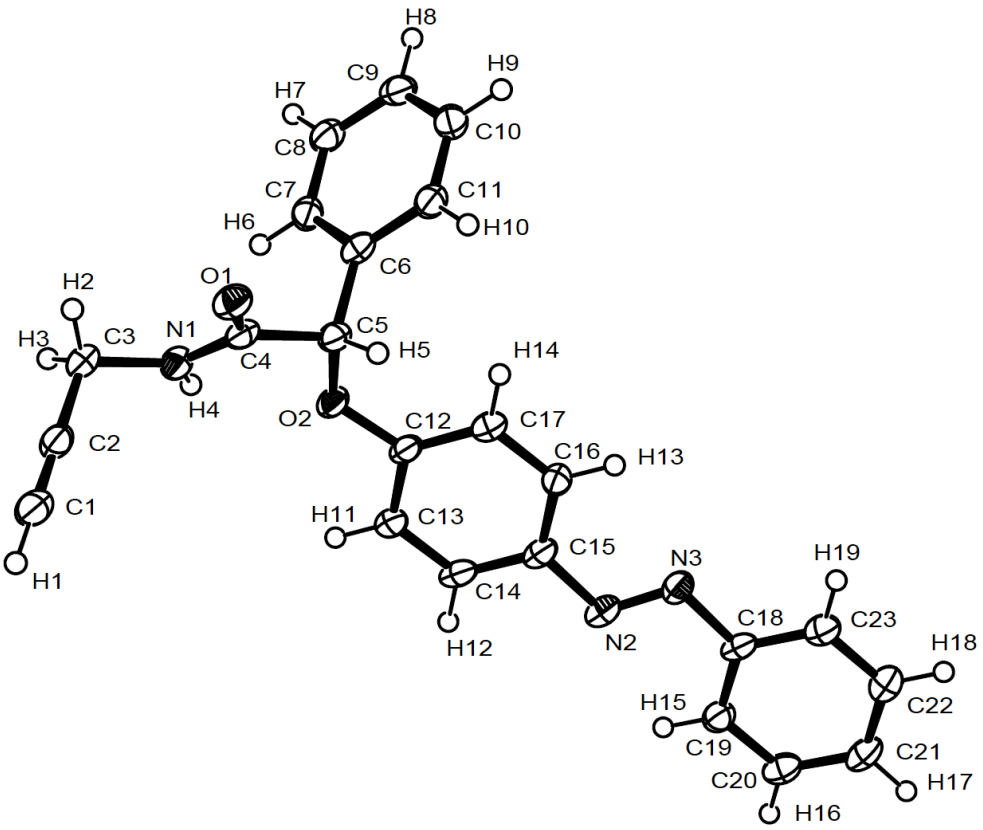


**Figure S1.** ORTEP drawing of the structure of **3'**.* Ellipsoid are given with 50% probability.

*This compound was synthesized from (*R*)-(–)-mandelic acid as a starting material. Consequently, **3'** should be the enantiomeric isomer of **3**, which was synthesized from (*S*)-(+)-mandelic acid as described in the experimental section. This figure confirms that **3'** is *S* form, whose absolute configuration is inverted from that of (*R*)-(–)-mandelic acid.

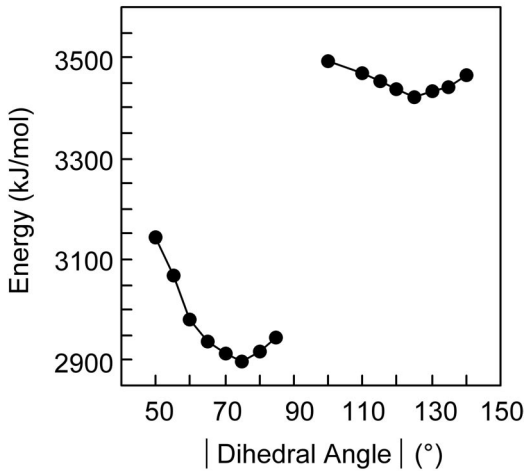


**Figure S2.** Relationship between the dihederal angle at the single bond of the main chain of a 18-mer of **1b** and the energy calculated by MMFF94.

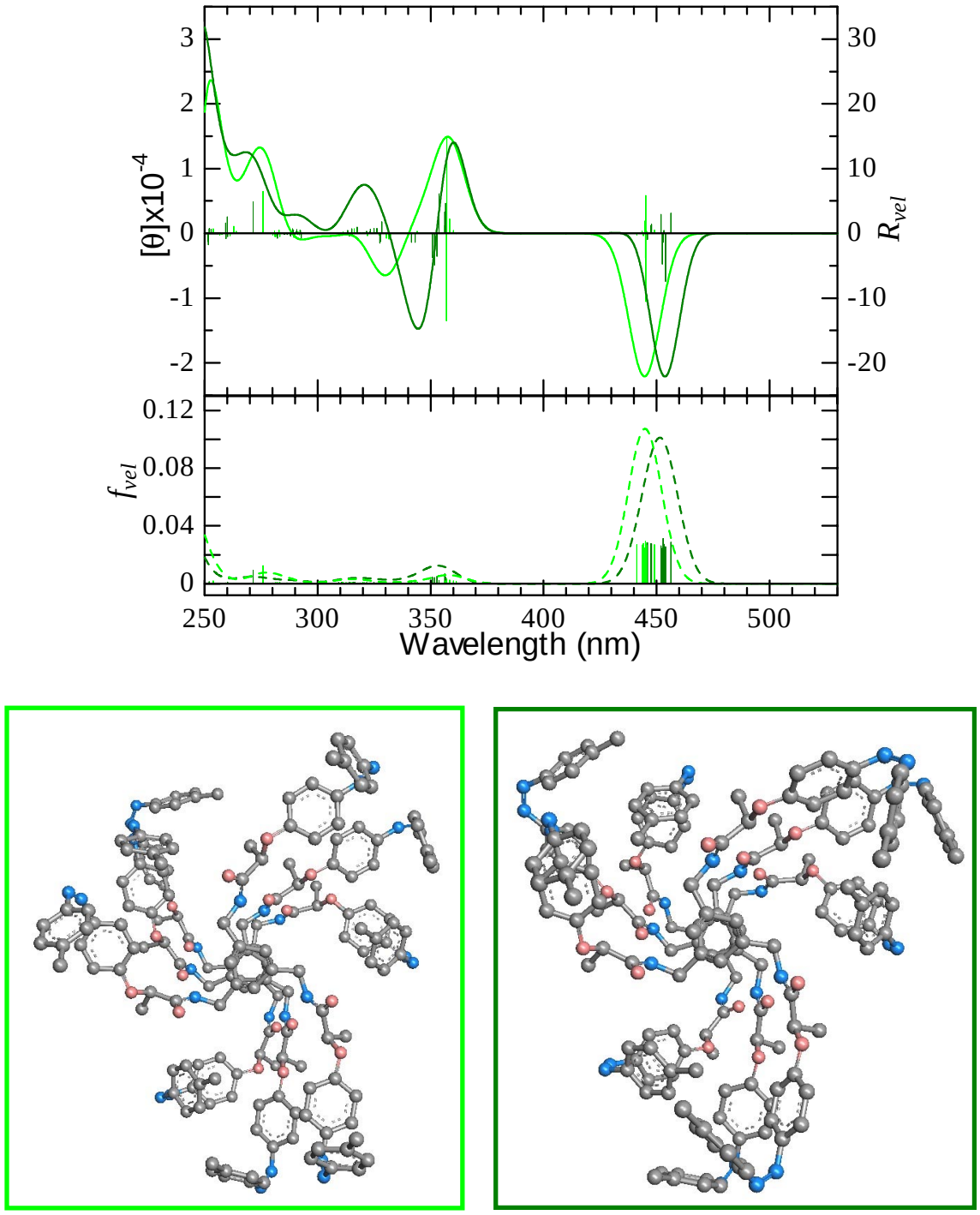


**Figure S3.** Simulated CD spectra of two 9-mers of **1b** with *cis*-azobenzene in helical conformations: the actual hexyl group in each side chain was replaced by methyl group. [θ], $R_{vel}$, and $f_{vel}$ are expressed per monomer unit. Light and deep green lines correspond to the structures in the bottom.

**The full list of authors of reference 21a**

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