Rationally Designed Helical Nanofibers *via* Multiple Non-Covalent Interactions: Fabrication and Modulation

Yiyang Lin,^{*a*} Andong Wang,^{*a*} Yan Qiao,^{*a*} Chen Gao,^{*a*} Markus Drechsler,^{*b*} Jianpin Ye,^{*c*} Yun Yan^{*a*} and Jianbin Huang^{*,*a*}

^a Beijing National Laboratory for Molecular Sciences (BNLMS), (State Key Laboratory for Structural Chemistry of Unstable and Stable Species), College of Chemistry and Molecular Engineering, Peking University, Beijing 100871, China

^b Elektronenmikroskopie Labor, University of Bayreuth.

^c CAS Key Laboratory of Photochemistry, Institute of Chemistry, Chinese Academy of Sciences, Beijing 100080, People's Republic of China.



Fig. S1 Low magnified cryo-TEM image of double helix in C_4AG solution.



Fig. S2 Negative-staining TEM of helical nanofibers at 50 °C.



Fig. S3 IR absorbance of C₄AG solution in DMSO (left) and C₄AG hydrogel in D₂O (right).

The existence of hydrogen-bonding interaction was demonstrated by FT-IR (Figure S3).

- The IR spectrum of C₄AG solution in DMSO was given in Fig S3a, in which hydrogel or nanofibers are not observed. From Fig S3a, a broad intense O-H stretch at 3440 cm⁻¹ can be seen.
- (2) The IR spectrum of C₄AG solution in D₂O was given in Fig S3b, in which hydrogel or nanofibers are observed. The effect of the hydrogen-bonding interaction was demonstrated from Fig S3b, in which the O-H stretch band becoming stronger in intensity and slightly shifting to 3400 cm⁻¹. Therefore, a hydrogen bonding interaction on the sugar moieties is believed to exist.



Fig. S4 Dynamic light scattering of C₄AG solution under acid condition indicating the existence of small micelles.



Fig. S5 CD spectrum of C_4AG solution at pH 5.6 and 8.0.