Electronic Supplementary Information:

Changing the shape of chromophores from "H-type" to "Star-type": increasing the macroscopic NLO effects in a large degree

Wenbo Wu, Can Wang, Cheng Zhong,^a Cheng Ye, Guofu Qiu, Jingui Qin, and Zhen Li*



Chart S1. The structure of polyurethanes embedded with "H-type" chromophores.



Chart S2. The structure of "H-sharp" NLO polymers PS3 and PS4.



Fig. S1. The FT-IR spectra of polymers P1-P3.



Fig. S2. The FT-IR spectra of monomers M1-M3.





Fig. S4. ¹³C NMR spectrum of monomer M1 in chloroform-*d*.

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Fig. S5. ¹H NMR spectrum of chromophore S5 in chloroform-*d*.



Fig. S6. ¹³C NMR spectrum of chromophore S5 in chloroform-*d*.



Fig. S7. ¹H NMR spectrum of monomer M2 in chloroform-*d*.



Fig. S8. ¹³C NMR spectrum of monomer M2 in chloroform-*d*.



Fig. S9. ¹H NMR spectrum of monomer M3 in chloroform-*d*.



Fig. S10. ¹³C NMR spectrum of monomer M3 in chloroform-*d*.



Fig. S11. ¹H NMR spectrum of chromophore S6 in chloroform-*d*.



Fig. S12. ¹³C NMR spectrum of chromophore S6 in chloroform-*d*.



Fig. S13. ¹H NMR spectrum of polymer P1 in chloroform-*d*.



Fig. S14. ¹³C NMR spectrum of polymer P1 in chloroform-*d*.



Fig. S15. ¹H NMR spectrum of polymer P2 in chloroform-*d*.



Fig. S16. ¹³C NMR spectrum of polymer P2 in chloroform-*d*.



Fig. S17. ¹H NMR spectrum of polymer P3 in chloroform-*d*.



Fig. S18. ¹³C NMR spectrum of polymer P3 in chloroform-*d*.



Fig. S19. UV-Vis spectra of P1 in different solutions. (0.02 mg/mL).



Fig. S20. UV-Vis spectra of P2 in different solutions. (0.02 mg/mL).



Fig. S21. UV-Vis spectra of P3 in different solutions. (0.02 mg/mL).

| | THF | 1,4-dioxane | chloroform | dichloromethane | DMF | DMSO | film |
|-----------|-----|-------------|------------|-----------------|-----|------|------|
| P1 | 496 | 487 | 508 | 507 | 512 | 520 | 509 |
| P2 | 496 | 487 | 507 | 505 | 509 | 511 | 500 |
| P3 | 502 | 495 | 509 | 512 | 516 | 519 | 510 |

Table S1. The maximum absorption of polymers (λ_{max} , nm).^{*a*}

 $^{\it a}$ The maximum absorption wavelength of polymer solutions with the concentrations fixed at 0.02 mg/mL.



Fig. S22. Absorption spectra of the film of P3 before and after poling.