

## Supplementary Information

Synthesis, crystal growth, and second-order nonlinear optical properties of new configurationally locked polyene derivatives

Xinyuan Zhang,<sup>a, b</sup> Xingxing Jiang,<sup>a, b</sup> Yin Li,<sup>\*a</sup> Zheshuai Lin,<sup>a</sup> Guochun Zhang,<sup>a</sup>  
and Yicheng Wu<sup>a</sup>

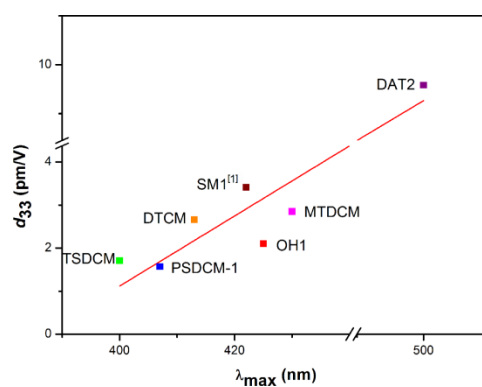


Fig. S1 Relationship between the simulation results of SHG coefficient  $d_{33}$  (pm/V) of investigated crystals and corresponding maximum absorption wavelengths  $\lambda_{\max}$  (nm).

The difference between PyT1<sup>2</sup> and PSDCM

- (1) The electron donor groups in PyT1 and PSDCM are totally different; in the former molecule the electron donor is pyrrolidinyll group, while in the latter molecule it is phenyl group. The difference in the structure results in conformation variation, e.g., in PyT1(I) and PSDCM-(I) the angle between donor group plane and molecular principle plane (constructed by hexene rings, dicyanomethylidene acceptors and benzene rings) is 8.3° and 7.9°, respectively. In addition, the powder SHG effect in PSDCM-(I) is more than 1.6 times as large as that in PyT1(I).
- (2) The single-crystal growth methods for PyT1 and PSDCM are different; the two phases of PyT1 crystal were obtained from methanol solution in the absence and presence of the PyM1 auxiliary (PyM1 is a similar compound as PyT1), respectively, while the two phases of PSDCM were grown in two different solvents (dichloromethane and chloroform) without any auxiliary compound.
- (3) The physiochemical properties of PyT1 and PSDCM are different; for instance, DSC measurements revealed that the platelike PyT1(I) crystals exhibit a reversible enantiotropic phase transition at  $T_{tr}= 85\text{ }^{\circ}\text{C}$  and PyT1(II) crystals do not show any phase transition up to the melting point at about 180 °C, while the reversible enantiotropic phase transition were not observed in both PSDCM phases until melting point.

In conclusion, the two polymorphs for PSDCM, centrosymmetric and acentric, are coincidentally similar to those for PyT1.

## References

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2. O. P. Kwon, S. J. Kwon, M. Jazbinsek, A. Choubey, P. A. Losio, V. Gramlich and P. Günter, *Cryst. Growth Des.*, 2006, **6**, 2327.