

## Supporting Information

### Intramolecular-locked strategy for design nonlinear optics material with the remarkable first hyperpolarizability

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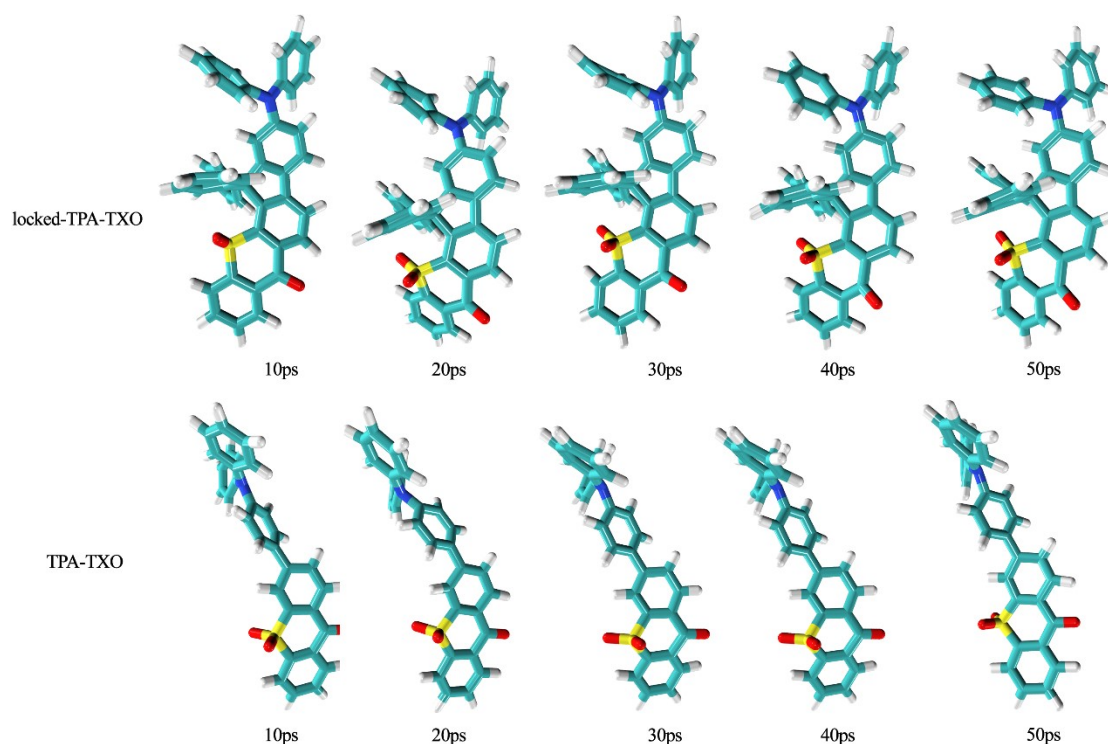


Fig. S1 The geometries TPA-TXO and locked-TPA-TXO of ab initio molecular dynamics stimulations for total time of 50 ps and temperature of 298.15 K.

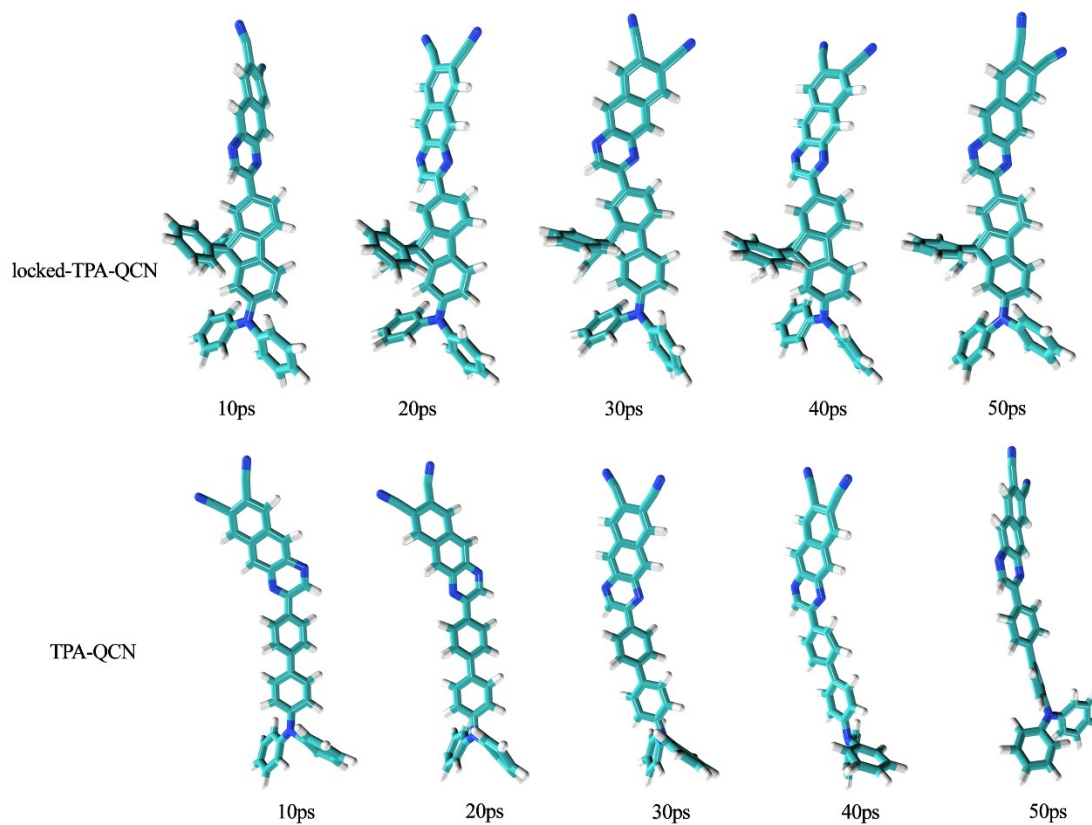


Fig. S2 The geometries TPA-QCN and locked-TPA-QCN of ab initio molecular dynamics stimulations for total time of 50 ps and temperature of 298.15 K.

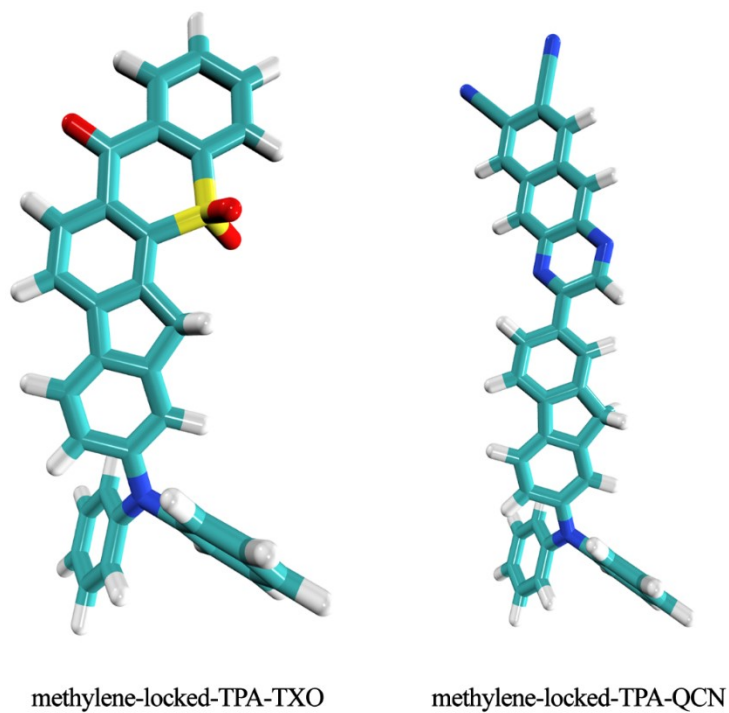


Fig. S3 The methylene linker locked structures of TPA-TXO and TPA-QCN at M06-2X-D3/def2-TZVP level.

The  $\omega$  values of  $\omega$ B97XD are 0.1323 Bohr<sup>-1</sup> and 0.1265 Bohr<sup>-1</sup> for methylene-locked-TPA-TXO and methylene-locked-TPA-QCN, respectively.