

## Supporting information

### Large-Size benzo[e]indolium-salt single crystals with high optical nonlinearity

Shufang Shang,<sup>a</sup> Ruian Li,<sup>b</sup> Linjun Ping,<sup>a</sup> Wenzhen Luo,<sup>c</sup> Mingtan Hai,<sup>a</sup> Zhou Yang,<sup>\*a</sup> Dong Wang,<sup>a</sup> Hui Cao,<sup>a</sup> Wanli He<sup>a</sup>

*(<sup>a</sup> School of Materials Science and Engineering, University of Science and Technology Beijing, Beijing 100083, China E-mail: [yangz@ustb.edu.cn](mailto:yangz@ustb.edu.cn))*

*(<sup>b</sup> Beijing Key Laboratory of Energy Conversion and Storage Materials, College of Chemistry, Beijing Normal University, Beijing100875)*

*(<sup>c</sup> Key Laboratory of Terahertz Optoelectronics, Ministry of Education, Capital Normal University, Beijing100037)*

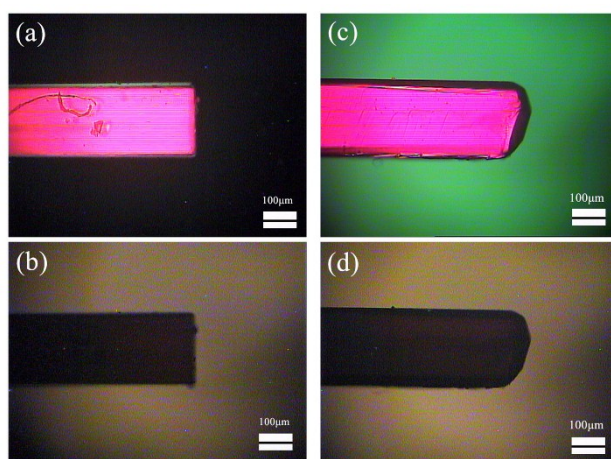


Fig.S1 Crystal of HMTB-1 and HMTB-2 between crossed polarizers in a microscope at the position of the maximum transmission (a), (c) and rotated by 45° from this position (b), (d)

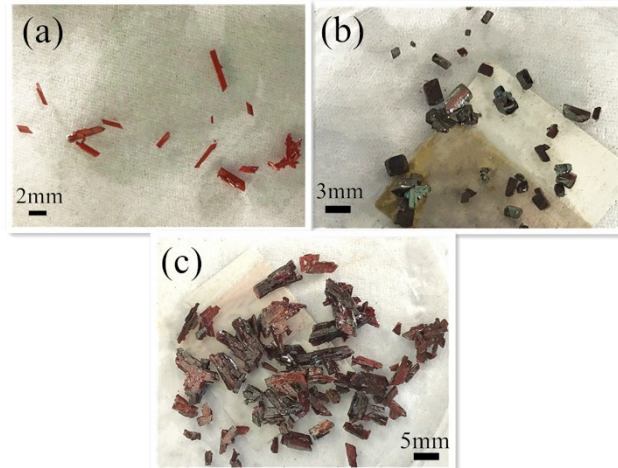


Fig.S2 (a) HMTB-1 crystals grown by evaporation of methanol at room temperature. (b) HMTB-2 crystals grown by evaporation of methanol at room temperature. (c) HMTB-1 crystals grown by evaporation of methanol/acetonitrile mixture (1:1 mol/mol) at room temperature

SHG measurements of HMTB-1 and HMTB-2 for three particle size ranges have been measured, the materials were ground and sifted to the grain size of  $<63\mu\text{m}$ ,  $63\text{-}90\mu\text{m}$  and  $>90\mu\text{m}$ , respectively. And squeezed into the sample box to obtain a constant sample thickness, and then the samples were irradiated by a laser with a wavelength  $\lambda=1570\text{nm}$ . The powder second harmonic generation efficiency of HMTB-1 and HMTB-2 were described compared with DAST. The SHG efficiencies of HMTB-1 and HMTB-2 are around 0.6 times and 0.5 times that of DAST. As the particle size decreases, the SHG intensity decreases slightly (substantially unchanged), but the relative proportion is basically consistent, which shows that HMTB-1 and HMTB-2 are phase matched (see Fig.S3).

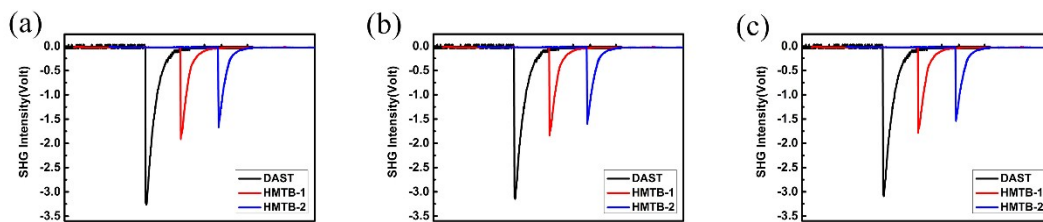


Fig. S3 SHG intensity of HMTB-1 and HMTB-2 compared with DAST. (a) The grain size is  $<63\mu\text{m}$ . (b) The grain size is  $63\text{-}90\mu\text{m}$ . (c) The grain size is  $>90\mu\text{m}$ .