Supporting Information

Influence of original and simulated microscopic units on SHG response in semiorganic NLO materials

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Detail parameters setting of calculating SKSC2B and SRSC2B.

Fig.S1 Microscopic unit and sturcture of RSCB

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Detail parameters setting of calculating SKSC2B and SRSC2B

Geometry optimization, electronic structures and optical properties were performed by the density functional theory (DFT) method within the generalized gradient approximation (GGA) in the scheme of Perdew−Burke−Ernzerhof (PBE) functional, implemented in the CASTEP package. And the norm-conserving pseudopotential (NCP) was adopted. The number of plane waves included in the basis was established by a cutoff energy of 950 eV in calculating crystal properties. Meanwhile, the Monk-horst-Pack k-point scheme of SKSC2B and SRSC2B are 5×5×5 in the irreducible Brillouin zone.

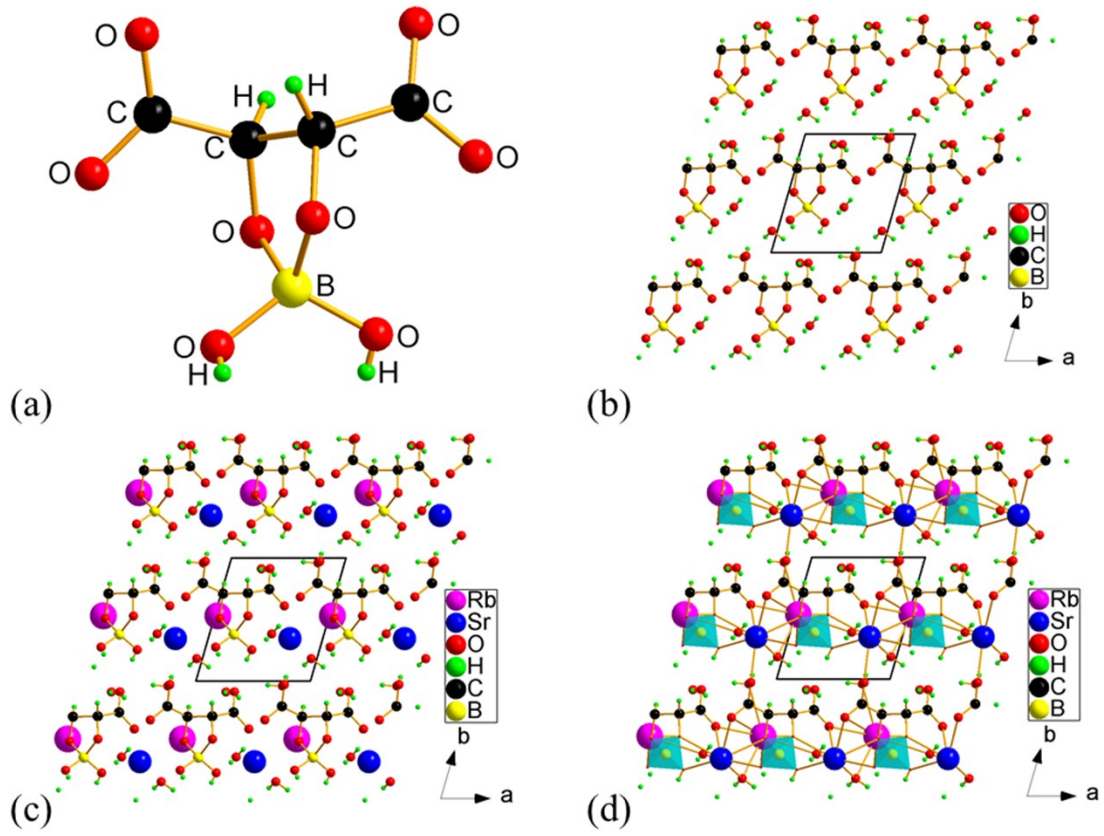

Fig.S1 (a) Microscopic unit (MU) $[C4H2O6B(OH)2]^{3-}$ in RSCB; (b) MU aligned in RSCB crystal viewed down the c axis; (c) MU with metal cations and water molecules; (d) KSCB crystal with BO_4 polyhedron viewed down the c axis.

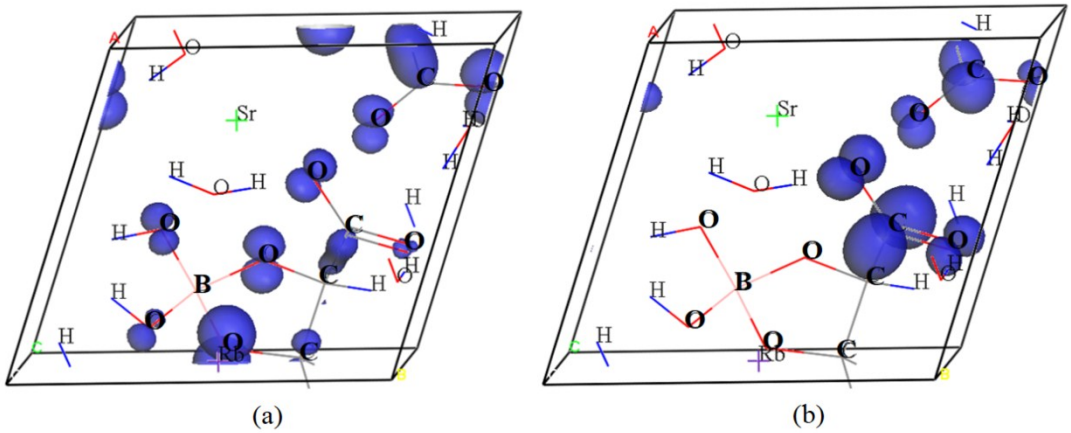

Fig.S2 The frontier molecular orbital of RSCB **(**a, HOMO; b, LUMO).

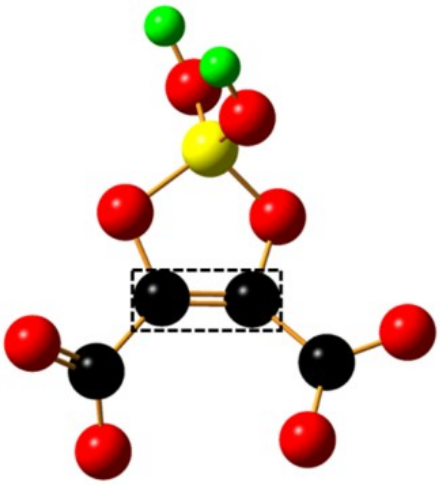

Fig.S3 Simulated microscopic unit (SMU)

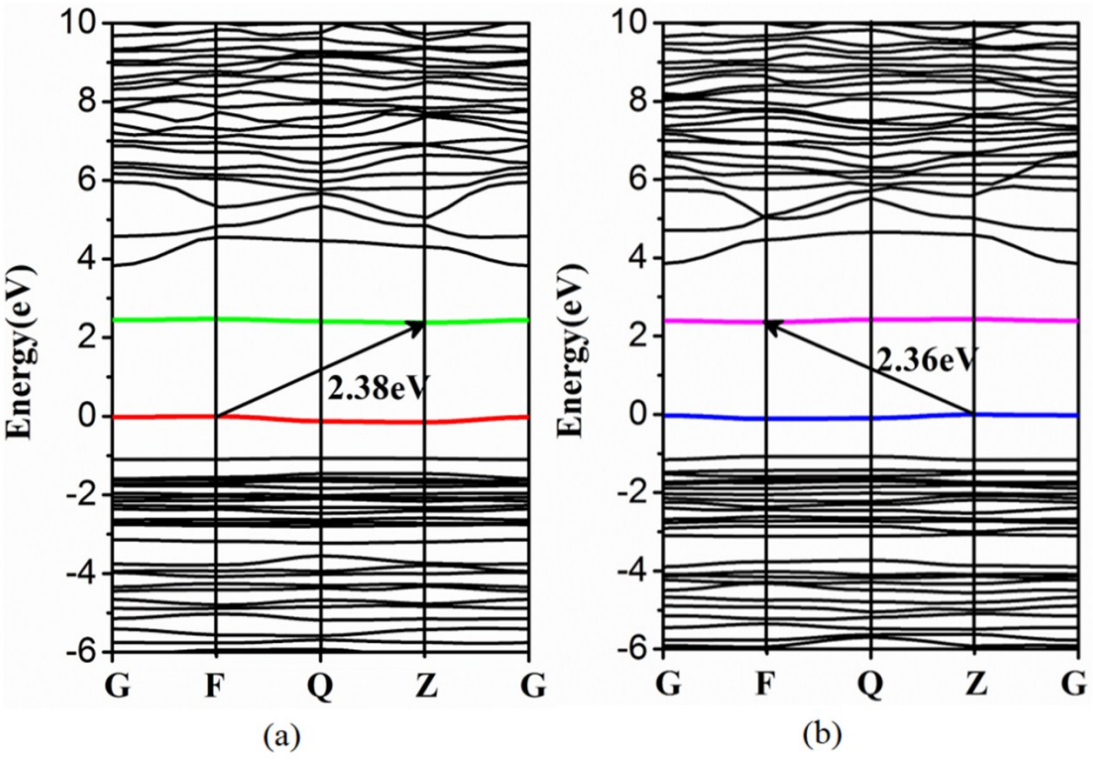

Fig.S4 Band structures of SKSC2B (a) and SRSC2B (b).

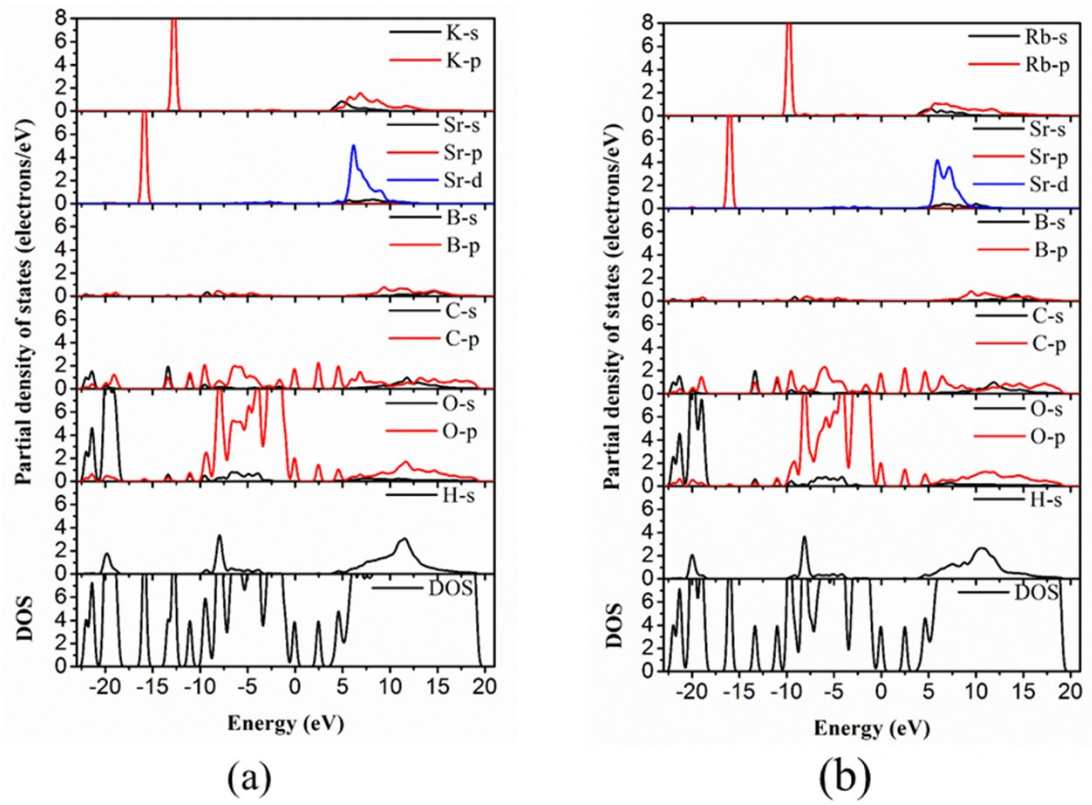

Fig.S5 Partial density of states of SKSC2B (a) and SRSC2B (b).

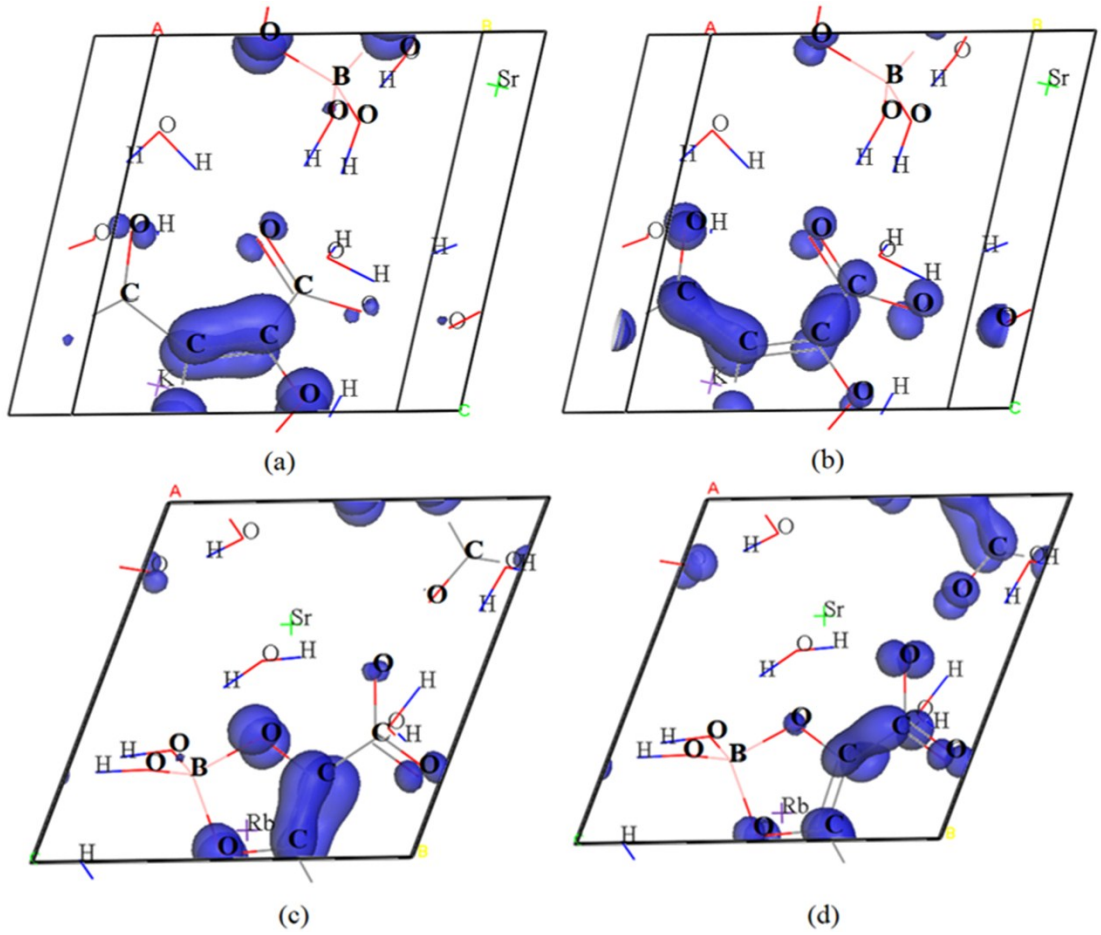

Fig.S6 The frontier molecular orbital of SKSC2B (a, HOMO; b, LUMO) and SRSC2B (c, HOMO; d, LUMO).

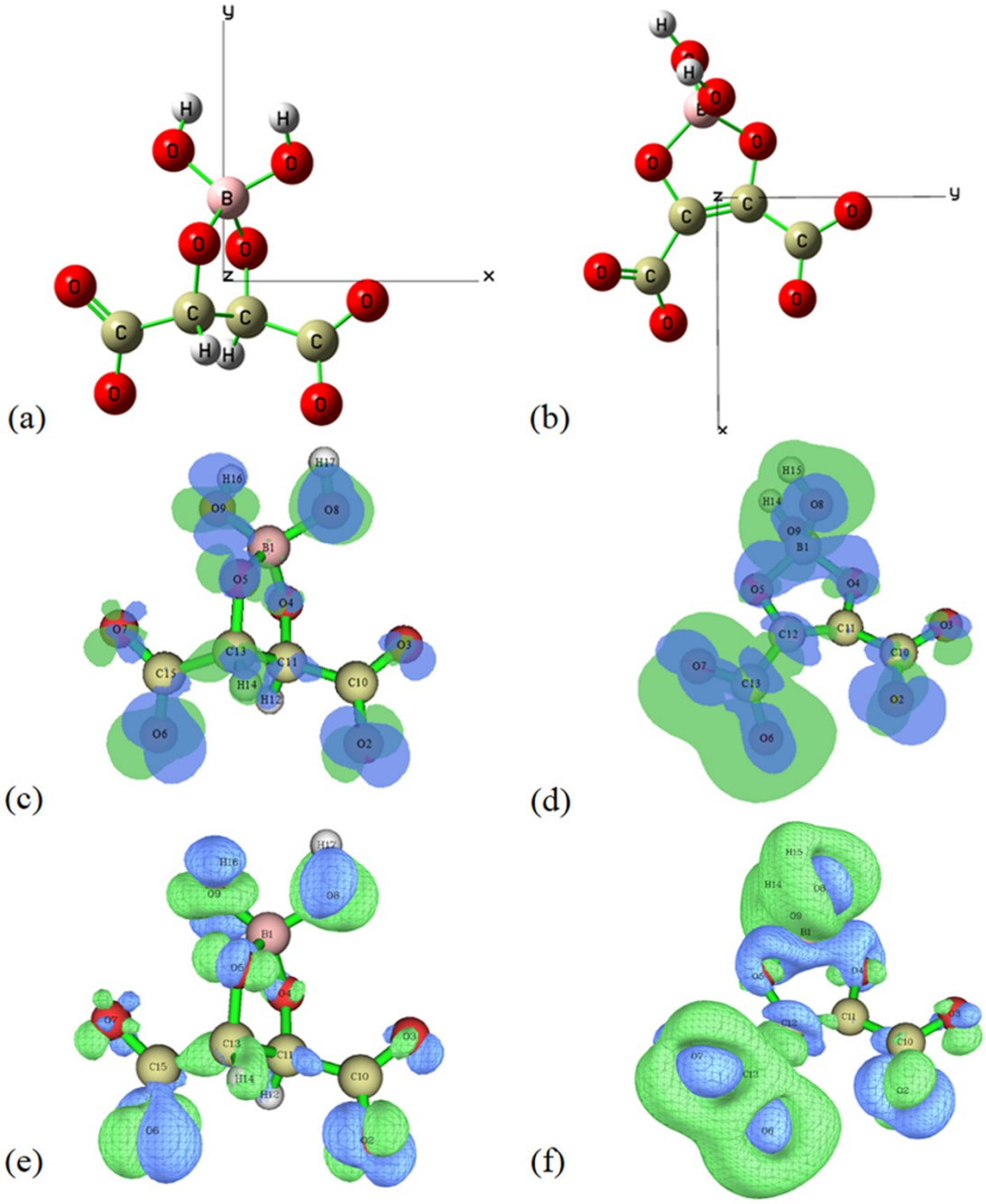

Fig.S7 Hyperpolarizability density of MU and SMU. The picture of (a), (c) and (e) are MU, which (b), (d) and (f) are SMU. The picture of (a) and (b) are the original structure without external electronic field. For (c), (d), (e) and (f) are the picture under an external electronic field, which (c) and (d) are transparent face and (e) and (f) are solid face and mesh. The standard orientation of MU and SMU are shown in (a) and (b). The

direction of external electronic field of MU is y, while for SMU is –x.