Electronic Supporting Information

The tellurium-oxygen groups enhanced birefringence in tellurium phosphates: A first-principles investigation

Pengyun Jin[1], Xuerui Shi[1], Xiuhua Cui[1*], Yi Jiang[1], Qun Jing[1], Ming-Hsien Lee[2], Mengqiu Long[1,3], Haibin Cao[4], Hanqin Ding[1*]

[1]Institute of Low-dimensional Quantum Materials and Devices, School of Physical Science and Technology, Xinjiang University, 666 Shengli Road, Urumqi 830046, China

[2]Department of Physics, Tamkang University, New Taipei City 25137, Taiwan

[3]Hunan Key laboratory of Super Micro-structure and Ultrafast Process, Central South University, Changsha 410083, China

[4]Department of Physics, College of Sciences, Shihezi University, Shihezi 832000, China

*To whom should be correspond: xjcxh0991@xju.edu.cn (Xiuhua Cui); dinghq@xju.edu.cn (Hanqin Ding)

Table S1. The crystal data of $Te_2P_2O_9$, $Ba_2TeO(PO_4)_2$, and $Te_3O_3(PO_4)_2$.

Compound	Space group	Lattice Parameters	Basic building groups
$Te_2P_2O_9$	*Cc*	*a* = 5.361 Å *b* = 13.631 Å *c* = 9.532 Å *β* = 103.697°	PO_4 tetrahedron TeO_5 square pyramid
$Ba_2TeO(PO_4)_2$	$P\bar{1}$	*a* = 6.946 Å *b* = 7.397 Å *c* = 8.887 Å *α* = 76.843° *β* = 79.933° *γ* = 75.688°	PO_4 tetrahedron TeO_5 square pyramid BaO_8 and BaO_{10}
$Te_3O_3(PO_4)_2$	$P2_1/c$	*a* = 12.375 Å *b* = 7.317 Å *c* = 9.834 Å *β* = 98.04°	PO_4 tetrahedron TeO_5 and TeO_4

Table S2. The obtained results using different functionals.

Method	Calculated SHG response（×KDP）*	E_g(eV)	Birefringence
GGA-PBE	2.23	3.43	0.12496-0.09236 @ 404.65-1013.61 nm
LDA-CA-PZ	2.19	3.78	0.16533-0.10913 @ 404.65-1013.61 nm
GGA-PW91	2.30	3.83	0.17191-0.11397 @ 404.65-1013.61 nm
GGA-RPBE	2.37	3.84	0.17474-0.11603 @ 404.65-1013.61 nm
experimental Δn =0.13786- 0.10615 @ 404.66-1013.98 nm			
experimental E_g^{exp} =4.3eV			
experimental PSHG intensity =1.3×KDP			

* The SHG response was calculated used the method suggested by Kurtz-Perry[1], Cyvin[2], and Cheng[3] *et al*.

[1] S. K. Kurtz, T. T. Perry, *J. App. Phys.*, 1968, 39, 3798-3813

[2] S. J. Cyvin, J. E. Rauch, J. C. Decius, *J. Chem. Phys.*, 1965, 43, 4083-4095

[3] X. Cheng, M. Whangbo, G. Guo, M. Hong, S. Deng, *Angew. Chem.* 2018, 130, 3997-4001

Table S3. The obtained Born effective charges of $Te_2P_2O_9$.

Te_1	4.34265	1.29741	-0.25903
	0.81526	4.65936	0.27256
	-0.34231	0.25305	6.63644
Te_2	6.42180	0.38026	-0.71374
	-0.18403	5.38842	0.34935
	-0.51613	0.73740	3.37218
Te_3	4.34265	-1.29741	-0.25903
	-0.81526	4.65936	-0.27256
	-0.34231	-0.25305	6.63644
Te_4	6.42180	-0.38026	-0.71374
	0.18403	5.38842	-0.34935
	-0.51613	-0.73740	3.37218
P_1	4.40285	-0.42392	-0.34998
	-0.30103	3.49299	1.12633
	-1.60626	-0.39242	4.36297
P_2	4.70919	-0.67166	-0.35318
	1.05416	4.86415	1.08782
	0.51145	-1.41008	3.77507
P_3	4.40285	0.42392	-0.34998
	0.30103	3.49299	-1.12633
	-1.60626	0.39242	4.36297
P_4	4.70919	0.67166	-0.35318
	-1.05416	4.86415	-1.08782
	0.51145	1.41008	3.77507
O_1	-1.78151	0.39504	1.14214
	0.48735	-1.84853	-1.32589
	1.24655	-0.79784	-2.74356
O_2	-3.28440	-1.23170	0.27685
	-1.75774	-2.12701	-0.04874
	0.65756	0.13560	-1.33367
O_3	-2.85789	1.11864	0.65349
	1.30695	-1.94646	-0.79139
	0.45728	-0.31034	-1.32288
O_4	-1.40561	-0.01706	0.66896
	-0.08619	-0.89945	0.38375
	1.50189	0.67420	-3.71391
O_5	-1.30274	0.75298	-0.51761
	0.74658	-2.25599	1.10197
	-0.59401	1.14779	-1.88828
O_6	-1.56074	-0.82169	-0.62558
	-1.01824	-3.25254	-1.55641

	-0.76077	-1.23740	-1.86712
O_7	-1.54143	0.67062	-0.77852
	0.48474	-1.92023	0.78002
	-1.19561	1.36410	-2.76231
O_8	-3.00828	-1.62644	0.32490
	-1.50450	-2.10674	0.17400
	0.09419	0.45970	-1.13507
O_9	-3.13389	1.73005	0.53131
	1.23971	-2.04796	-0.54343
	0.54617	-0.43220	-1.37986
O_{10}	-1.78151	-0.39504	1.14214
	-0.48735	-1.84853	1.32589
	1.24655	0.79784	-2.74356
O_{11}	-3.28440	1.23170	0.27685
	1.75774	-2.12701	0.04874
	0.65756	-0.13560	-1.33367
O_{12}	-2.85789	-1.11864	0.65349
	-1.30695	-1.94646	0.79139
	0.45728	0.31034	-1.32288
O_{13}	-1.40561	0.01706	0.66896
	0.08619	-0.89945	-0.38375
	1.50189	-0.67420	-3.71391
O_{14}	-1.30274	-0.75298	-0.51761
	-0.74658	-2.25599	-1.10197
	-0.59401	-1.14779	-1.88828
O_{15}	-1.56074	0.82169	-0.62558
	1.01824	-3.25254	1.55641
	-0.76077	1.23740	-1.86712
O_{16}	-1.54143	-0.67062	-0.77852
	-0.48474	-1.92023	-0.78002
	-1.19561	-1.36410	-2.76231
O_{17}	-3.00828	1.62644	0.32490
	1.50450	-2.10674	-0.17400
	0.09419	-0.45970	-1.13507
O_{18}	-3.13389	-1.73005	0.53131
	-1.23971	-2.04796	0.54343
	0.54617	0.43220	-1.37986

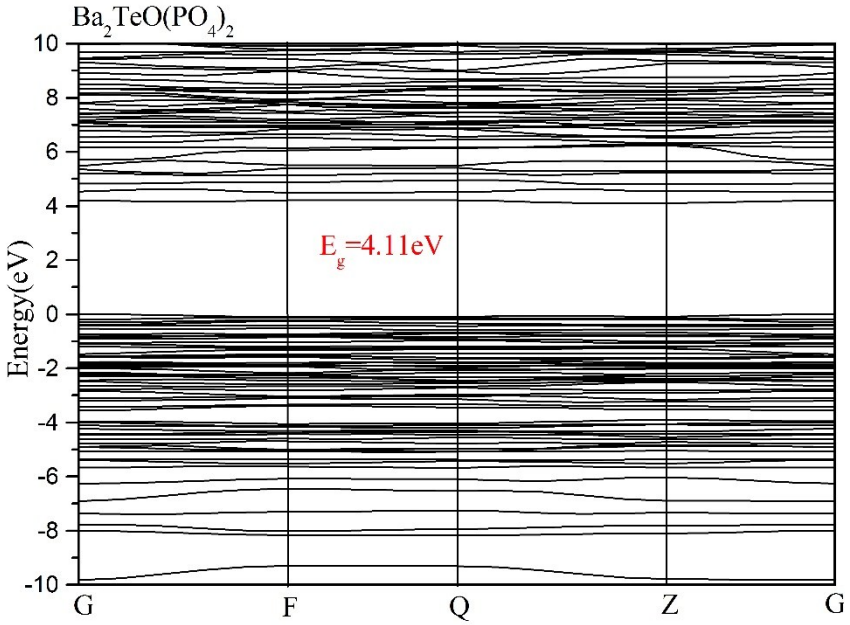

Figure S1. The band structure of $Ba_2TeO(PO_4)_2$

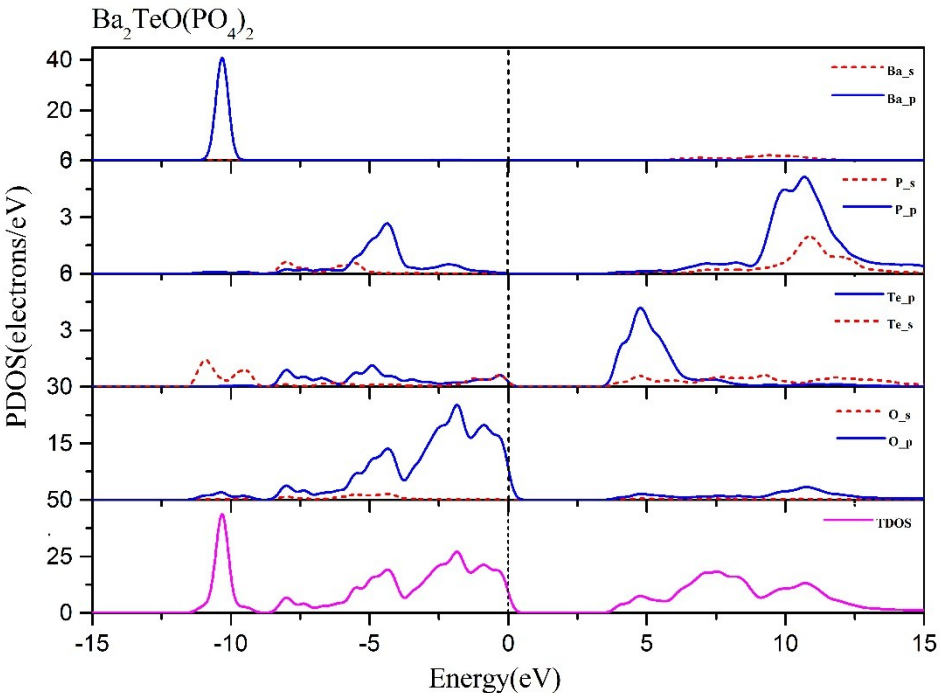

Figure S2. The projected density of states (PDOS) of $Ba_2TeO(PO_4)_2$.

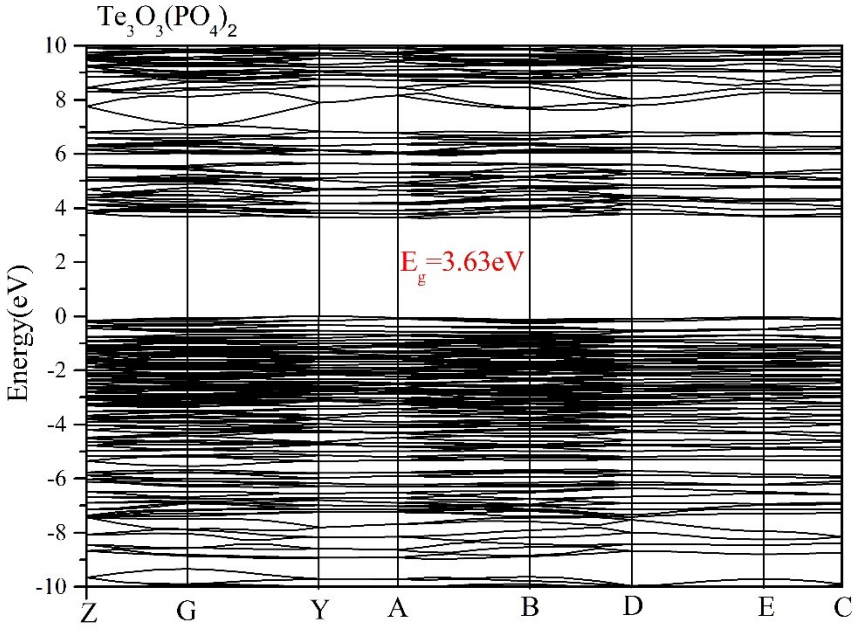

Figure S3. The band structures of $Te_3O_3(PO_4)_2$

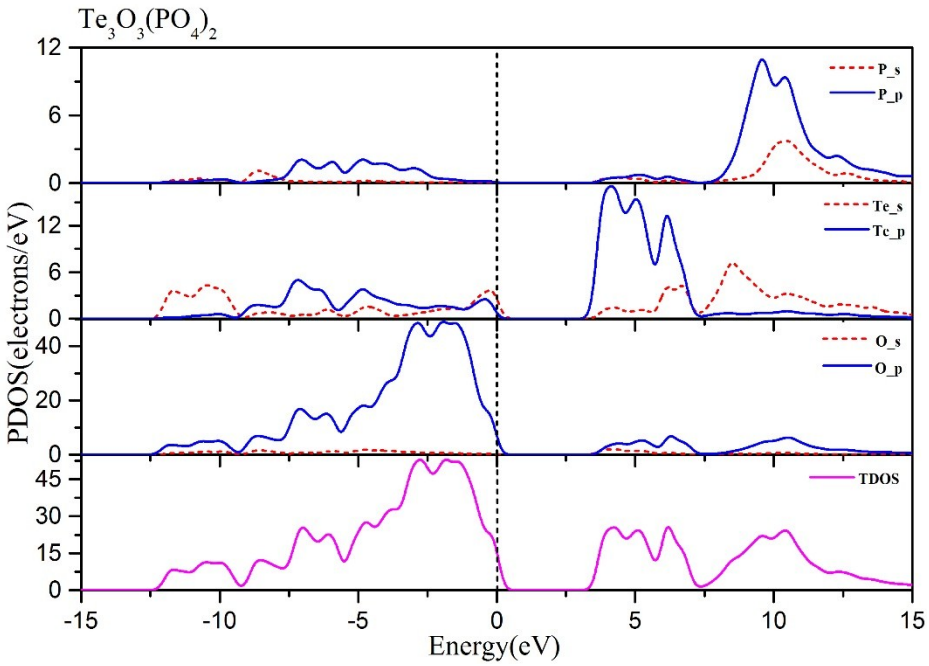

Figure S4. The projected density of states (PDOS) of $Te_3O_3(PO_4)_2$.

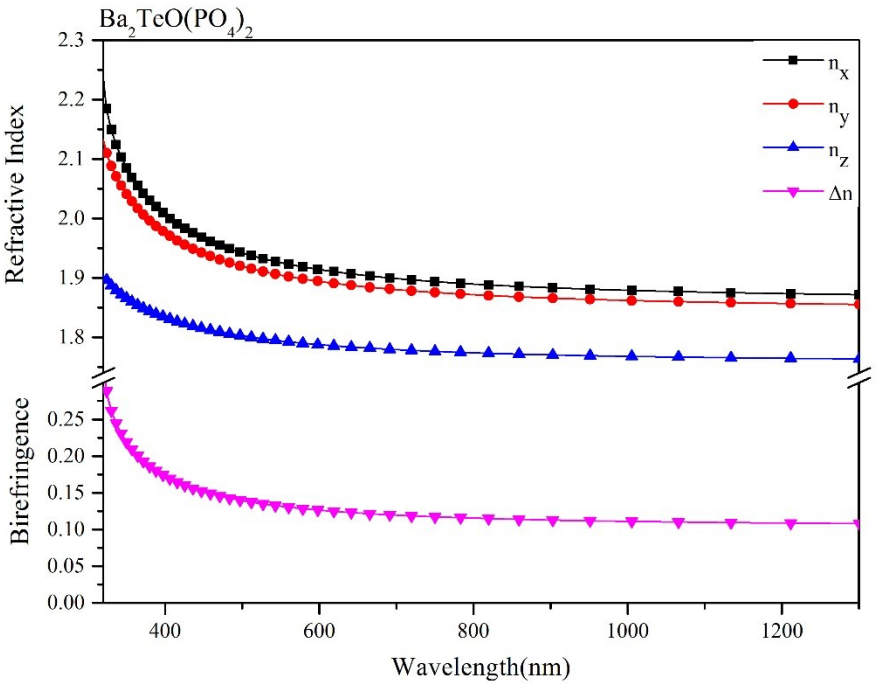

Figure S5. The obtained refractive indices and birefringence of $Ba_2TeO(PO_4)_2$ using the CASTEP and OPTADOS code.

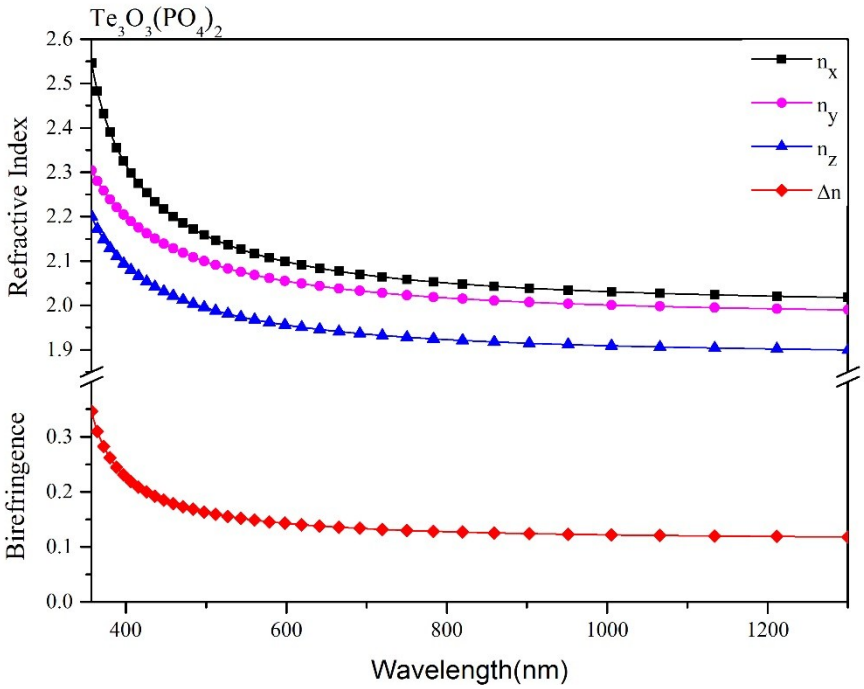

Figure S6. The obtained refractive indices and birefringence of $Te_3O_3(PO_4)_2$ using the CASTEP and OPTADOS code.

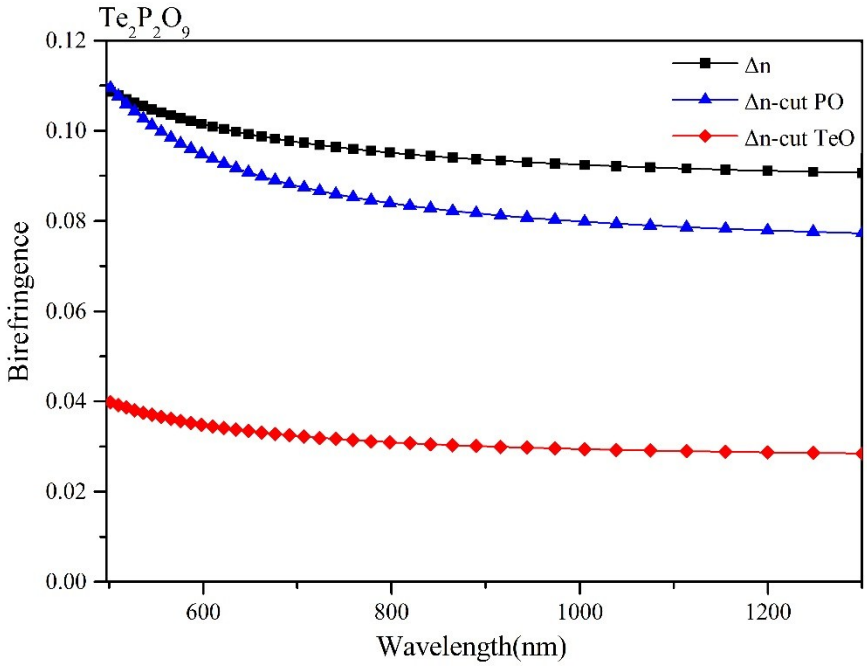

Figure S7. The obtained birefringence of $Te_2P_2O_9$ after the real-space atom-cutting method was performed.

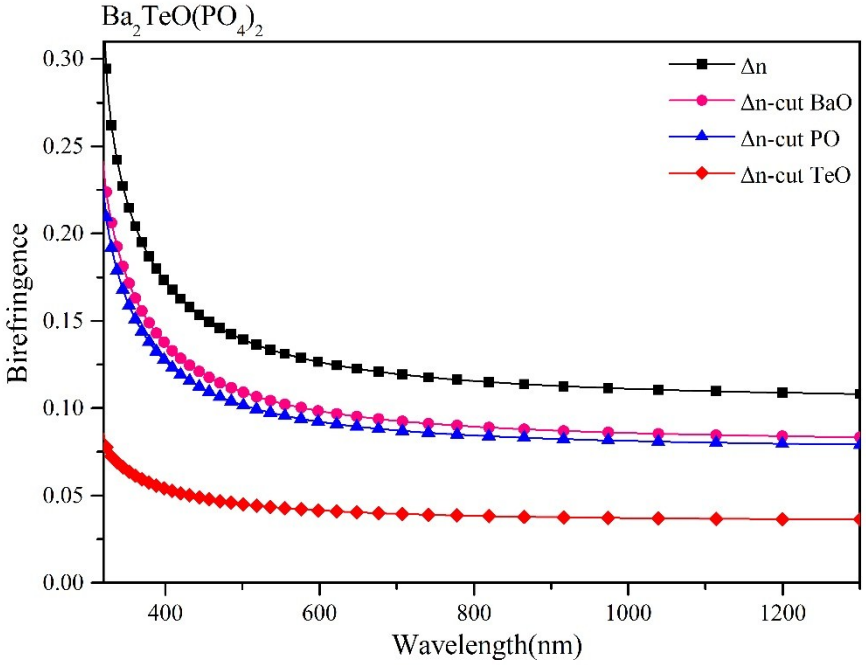

Figure S8. The obtained birefringence of $Ba_2TeO(PO_4)_2$ after the real-space atom-cutting method was performed.

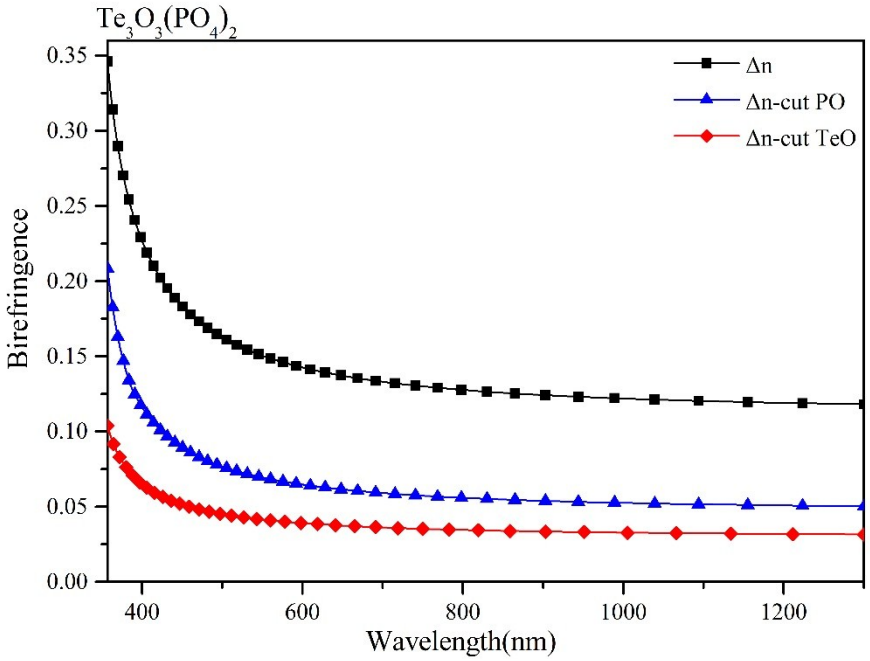

Figure S9. The obtained birefringence of $Te_3O_3(PO_4)_2$ after the real-space atom-cutting method was performed.