

APPENDIX A

Calculated largest NLO tensor coefficients d_{ij}^{\max} of the selected borate crystals, together with the recommended bond-valence parameters d_0 for the corresponding configuration of borate FBBs. Experimental data are in italics

Crystal	FBB	d_{ij}^{\max} (pm/V) [†]	d_0 (Å)	Note‡
$\text{Ca}_4\text{GdO}(\text{BO}_3)_3$	2T:T	$d_{11} (-4.38)^{\text{a}}$	1.378	A
$\text{NdAl}_3(\text{BO}_3)_4$	2T:T	$d_{11} (-2.43; 1.71)^{\text{b}}$	1.378	A
$\beta\text{-BaB}_2\text{O}_4$	3T:<3T>	$d_{22} (1.86; \pm(1.93 \pm 0.13))^{\text{c}}$	1.373	A
LiB_3O_5	2T1F:<2TF>	$d_{31} (-0.93; -(1.05 \pm 0.13))^{\text{a}}$	1.367	A
CsB_3O_5	2T1F:<2TF>	$d_{36} (1.17; 1.04(1 \pm 0.2))^{\text{d}}$	1.367	A
$\text{CsLiB}_6\text{O}_{10}$	2T1F:<2TF>	$d_{36} (1.09; 0.95)^{\text{d}}$	1.367	A
$\text{K}[\text{B}_5\text{O}_6(\text{OH})_4] \cdot 2\text{H}_2\text{O}$	4T1F:<<2TF>-<F2T>>	$d_{33} (-0.43)^{\text{a}}$	1.364	H
$\text{Eu}_2\text{B}_5\text{O}_9\text{Cl}$	2T3F:<<T2F>-<2FT>>	$d_{31} (0.40)^{\text{a}}$	1.372	A, X
$\text{Ca}_2\text{B}_5\text{O}_9\text{Cl}$	2T3F:<<T2F>-<2FT>>	$d_{31} (2.29)^{\text{e}}$	1.372	A, X
$\text{Eu}_2\text{B}_5\text{O}_9\text{Br}$	2T3F:<T2F>FT	$d_{33} (-0.90)^{\text{e}}$	1.364	A, X
$\text{Ca}_2\text{B}_5\text{O}_9\text{Br}$	2T3F:<T2F>FT	$d_{33} (-0.16)^{\text{a}}$	1.364	A, X

† d_{ij}^{\max} represents the largest d_{ij} tensor coefficient among all d_{ij} tensors of the corresponding crystals, the data are taken from the following references: ^a Xue, D. *et al.* (2000); ^b Xue, D. & Zhang, S. (1996); ^c Xue, D. & Zhang, S. (1998); ^d Xue, D. *et al.* (2002); ^e Xue, D. *et al.* (2000).

‡ Letters A, H and X represent respectively anhydrous, hydrated structures and halogen ions contained in borates.