Supporting information

Origin and Mechanism of Piezoelectricity in α-Quartz-type $M^{III}X^{V}O_4$ Compounds (M = B, Al, Ga; X = P, As)

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Table S1: Calculated contributions of the E-modes to the e_{11}^{str} piezoelectric internal-strain in $M^{III}X^{V}O_4$ compounds (M = Al, Ga; X = P, As). See Eqs. (2) and (3) for the definition of p_α^m and g_j^m, respectively.

$AlPO_4$			$AlAsO_4$			$GaPO_4$			$GaAsO_4$		
ω_m (cm^{-1})	$p_1 \cdot g_1$ ($\times 10^{-6}$ a.u)	e_{11}^{str} (C/m^2)	ω_m (cm^{-1})	$p_1 \cdot g_1$ ($\times 10^{-6}$ a.u)	e_{11}^{str} (C/m^2)	ω_m (cm^{-1})	$p_1 \cdot g_1$ ($\times 10^{-6}$ a.u)	e_{11}^{str} (C/m^2)	ω_m (cm^{-1})	$p_1 \cdot g_1$ ($\times 10^{-6}$ a.u)	e_{11}^{str} (C/m^2)
95	-0.07	-0.01	94	-0.32	-0.05	98	-0.43	-0.07	82	-0.15	-0.04
106	0.06	0.01	115	0.13	0.02	111	0.42	0.06	106	-0.17	-0.02
157	0.32	0.02	128	0.02	0.00	135	0.28	0.03	122	0.03	0.00
180	-0.20	-0.02	180	-0.14	-0.01	176	-0.79	-0.04	167	0.38	0.02
281	-0.01	0.00	229	0.46	0.01	**227**	**-2.88**	**-0.10**	**204**	**-3.48**	**-0.13**
360	**-7.78**	**-0.10**	**284**	**-6.31**	**-0.12**	278	1.64	0.04	215	1.00	0.03
392	**6.62**	**0.07**	312	-1.82	-0.03	**345**	**17.86**	**0.25**	269	-1.84	-0.04
428	-1.97	-0.02	**334**	**4.47**	**0.06**	382	-5.97	-0.07	**280**	**5.42**	**0.11**
445	**7.21**	**0.06**	358	2.20	0.03	431	-2.48	-0.02	309	-1.98	-0.03
531	0.56	0.00	419	1.30	0.01	440	1.51	0.01	407	0.31	0.00
619	2.79	0.01	**530**	**17.93**	**0.10**	570	1.69	0.01	**462**	**19.25**	**0.14**
662	4.09	0.01	574	-2.82	-0.01	624	-0.30	0.00	517	0.32	0.00
715	-0.13	0.00	590	2.62	0.01	639	-2.31	-0.01	529	-0.14	0.00
1059	**79.20**	**0.12**	**856**	**126.25**	**0.27**	**1002**	**69.95**	**0.11**	**786**	**111.61**	**0.28**
1067	33.95	0.05	866	5.31	0.01	1012	-0.92	0.00	799	19.37	0.05
1095	27.60	0.04	929	16.46	0.03	**1022**	**122.21**	**0.20**	851	8.41	0.02
1192	5.09	0.01	962	-0.57	0.00	1146	6.11	0.01	896	4.95	0.01

Table S2: Calculated contributions of the E-modes to the e_{14}^{str} piezoelectric internal-strain in $M^{III}X^{V}O_4$ compounds (M = Al, Ga; X = P, As). See Eqs. (2) and (3) for the definition of p_α^m and g_j^m, respectively.

$AlPO_4$			$AlAsO_4$			$GaPO_4$			$GaAsO_4$		
ω_m (cm^{-1})	$p_1 \cdot g_4$ ($\times 10^{-6}$ a.u)	e_{14}^{str} (C/m^2)	ω_m (cm^{-1})	$p_1 \cdot g_4$ ($\times 10^{-6}$ a.u)	e_{14}^{str} (C/m^2)	ω_m (cm^{-1})	$p_1 \cdot g_4$ ($\times 10^{-6}$ a.u)	e_{14}^{str} (C/m^2)	ω_m (cm^{-1})	$p_1 \cdot g_4$ ($\times 10^{-6}$ a.u)	e_{14}^{str} (C/m^2)
95	0.15	0.03	94	0.30	0.05	98	0.03	0.00	82	0.05	0.01
106	-0.08	-0.01	115	-0.09	-0.01	111	-0.38	-0.05	106	0.14	0.02
157	-1.14	-0.07	128	-0.05	0.00	135	-0.48	-0.05	122	-0.04	0.00
180	0.96	0.05	180	1.26	0.06	176	3.39	0.19	167	3.10	0.18
281	-0.09	0.00	229	2.86	0.09	227	7.43	0.25	204	4.83	0.18
360	15.93	0.20	284	4.93	0.10	278	2.57	0.06	215	2.88	0.10
392	1.69	0.02	312	4.23	0.06	345	-19.00	-0.27	269	-3.38	-0.07
428	4.80	0.04	334	-10.73	-0.15	382	-0.63	-0.01	280	-5.40	-0.11
445	-29.38	-0.25	358	-2.24	-0.03	431	0.78	0.01	309	-2.63	-0.04
531	-0.01	0.00	419	-0.87	-0.01	440	-0.25	0.00	407	0.05	0.00
619	-0.31	0.00	530	-1.76	-0.01	570	-3.96	-0.02	462	-9.06	-0.07
662	-17.75	-0.07	574	-29.72	-0.14	624	-4.04	-0.02	517	-15.38	-0.09
715	-10.72	-0.03	590	-10.55	-0.05	639	-11.98	-0.05	529	-3.98	-0.02
1059	195.07	0.29	856	122.36	0.26	1002	141.64	0.24	786	114.23	0.29
1067	-84.41	-0.12	866	-15.10	-0.03	1012	-3.60	-0.01	799	-24.68	-0.06
1095	-3.49	0.00	929	-5.14	-0.01	1022	-36.89	-0.06	851	-1.22	0.00
1192	-1.61	0.00	962	0.40	0.00	1146	-3.45	0.00	896	-2.85	-0.01

Table S3: Calculated elastic, compliance and dielectric constants in MXO_4 compounds (M = Al, Ga, B; X = P, As). Experimental values (between brackets) for $AlPO_4$, $GaPO_4$ and $GaAsO_4$ are from Bailey *et al.*,[1] Wallnöfer *et al.*[2] and Bhalerao *et al.*,[3] respectively.

	$AlPO_4$	$AlAsO_4$	$GaPO_4$	$GaAsO_4$	BPO_4	$BAsO_4$
Elastic Constants under zero electric field (GPa)						
$C_{11}^{(\mathscr{E})}$	66.07 (69.3)	55.53	61.91 (66.58)	48.94	164.40	128.13
$C_{12}^{(\mathscr{E})}$	2.91 (10.5)	14.99	17.28 (21.81)	18.76	27.76	37.84
$C_{13}^{(\mathscr{E})}$	8.93 (13.5)	17.73	20.75 (24.87)	21.24	41.38	44.99
$C_{14}^{(\mathscr{E})}$	13.69 (13.0)	4.89	5.29 (3.91)	0.10 (1.7)	13.76	-2.07
$C_{33}^{(\mathscr{E})}$	73.64 (88.6)	79.57	93.44 (102.13)	87.73 (103.5)	211.62	191.83
$C_{44}^{(\mathscr{E})}$	40.55 (43.0)	34.41	37.29 (37.66)	27.11 (30.7)	109.44	72.61
Compliance Constants under zero electric field (TPa^{-1})						
$S_{11}^{(\mathscr{E})}$	16.76 (16.4)	20.76	18.74 (17.93)	25.28	6.60	8.99
$S_{12}^{(\mathscr{E})}$	-1.80 (-3.2)	-4.78	-4.45 (-4.82)	-7.86	-0.91	-2.09
$S_{13}^{(\mathscr{E})}$	-1.82 (-2.0)	-3.56	-3.17 (-3.19)	-4.22	-1.11	-1.62
$S_{14}^{(\mathscr{E})}$	-6.27 (-5.9)	-3.63	-3.29 (-2.36)	0.12	-0.94	0.31
$S_{33}^{(\mathscr{E})}$	14.02 (11.9)	14.16	12.11 (11.35)	13.44	5.16	5.98
$S_{44}^{(\mathscr{E})}$	28.90 (26.8)	30.09	27.75 (27.04)	36.89	9.37	13.79
Free-stress Dielectric Constants						
$\varepsilon_{11}^{(\sigma)}$	4.66 (4.73)	5.37	6.08 (5.2)	6.93	5.86	7.27
$\varepsilon_{33}^{(\sigma)}$	4.64	5.31	5.77	6.38	6.15	7.48
Fixed-strain Dielectric Constants						
$\varepsilon_{11}^{(\eta)}$	4.56	5.19	5.82	6.45	5.84	7.19
$\varepsilon_{33}^{(\eta)}$	4.64	5.31	5.77	6.38	6.15	7.48

References

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