

Supplementary Information

Table 1. Interatomic potentials employed in the modelling studies.

buckingham	A[eV]	C [eV Å⁶]	r [Å]	Y	K [eV Å⁻²]
Ge core O shell	1497.3996	0.325646	16.00	4.00	
La core O shell	4579.23	0.30437	0.00	3.00	
Ba core O shell	931.7	0.39	0	2	
O shell O shell	22764.0	0.1490	27.879	-2.9	74.92

Table 2. Comparison of observed and calculated lattice parameters for La₈Ba₂Ge₆O₂₆ and La₉BaGe₆O_{26.5}.

Cell Parameters	La ₈ Ba ₂ Ge ₆ O ₂₆			La ₉ BaGe ₆ O _{26.5}		
	<i>Experimental</i>	<i>Calculated</i>	<i>Difference [%]</i>	<i>Experimental</i>	<i>Calculated</i>	<i>Difference [%]</i>
a	9.9828	10.0906	1.0800	9.9303	9.9014	-0.2910
b	9.9828	10.0906	1.0800	9.9303	10.0326	1.0302
c	7.4046	7.2564	-2.0000	7.3615	7.1764	-2.5144
alpha	90.0000	90.0000		90.0000	91.73	1.9222
beta	90.0000	90.0000		90.0000	88.76	-1.3778
gamma	120.0000	120.0000		120.0000	119.15	-0.7083
Vol.	639.0531	639.8603	0.1300	628.685	622.29	1.0172
Lattice Energy		-1317.30			-1347.46	

Table 3. Calculated and experimentally observed Raman active frequencies for the stoichiometric composition $\text{La}_8\text{Ba}_2(\text{GeO}_4)_6\text{O}_2$. For $\text{La}_9\text{Ba}(\text{GeO}_4)_6\text{O}_{2.5}$, the modelling predicts an extra mode in the gap region between bending and stretching modes at 675cm^{-1} (experimentally observed at 645cm^{-1})

	Lattice and Bending internal modes	Stretching internal modes
v observed [cm^{-1}]	77, 83, 91, 99, 140, 160, 169, 207, 241, 261, 339, 386, 421, 476	707, 724, 766, 775
v calculated [cm^{-1}]	43.6, 94.3, 101.5, 112.7, 119.9, 155, 160, 161.6, 184.6, 187.2, 193.4, 203.7, 218.3, 236, 246.2, 276.4, 302.7, 360.4, 363.4, 375.7, 451.1, 495.7, 499.4, 510.2, 520.1, 545	758.8, 763.6, 771.8, 792.9, 806.8, 812.5, 842.2