Supporting materials

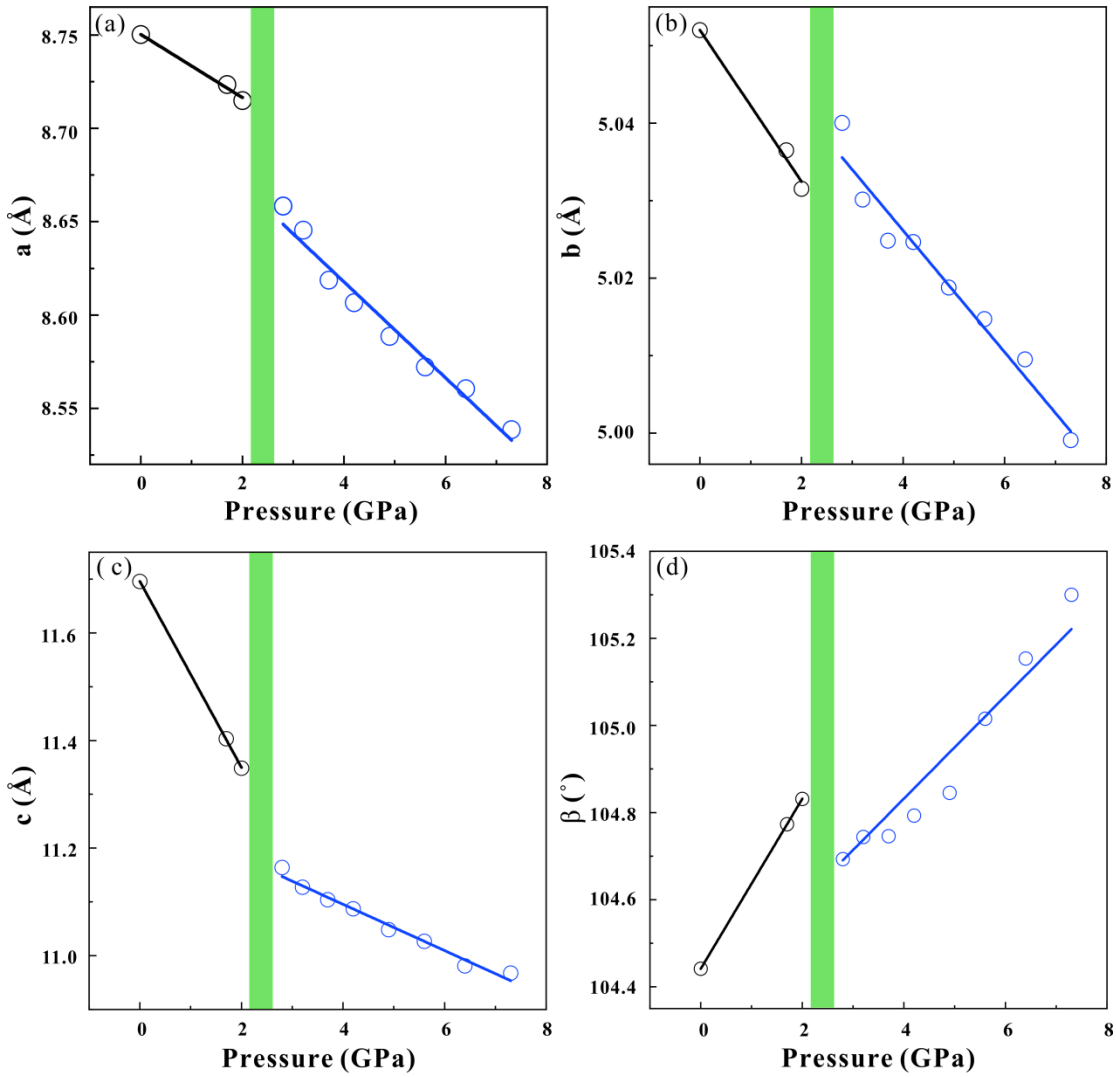

Figure S1. Isothermal compressibility of the *a*-axis (values for the α-phase correspond to a/√3) (**a**), *b*-axis (**b**), *c*-axis (values for the α-phase correspond to c/3sinβ) (**c**), β (values for the α-phase are calculated for an equivalent monoclinic cell setting) (**d**) at room temperature up to ~7.3 GPa. The green region marks a phase transition from α-phase (black) to γ-phase (blue) occurs around 2.8 GPa. The experimental error is claimed to be smaller than the symbol size.

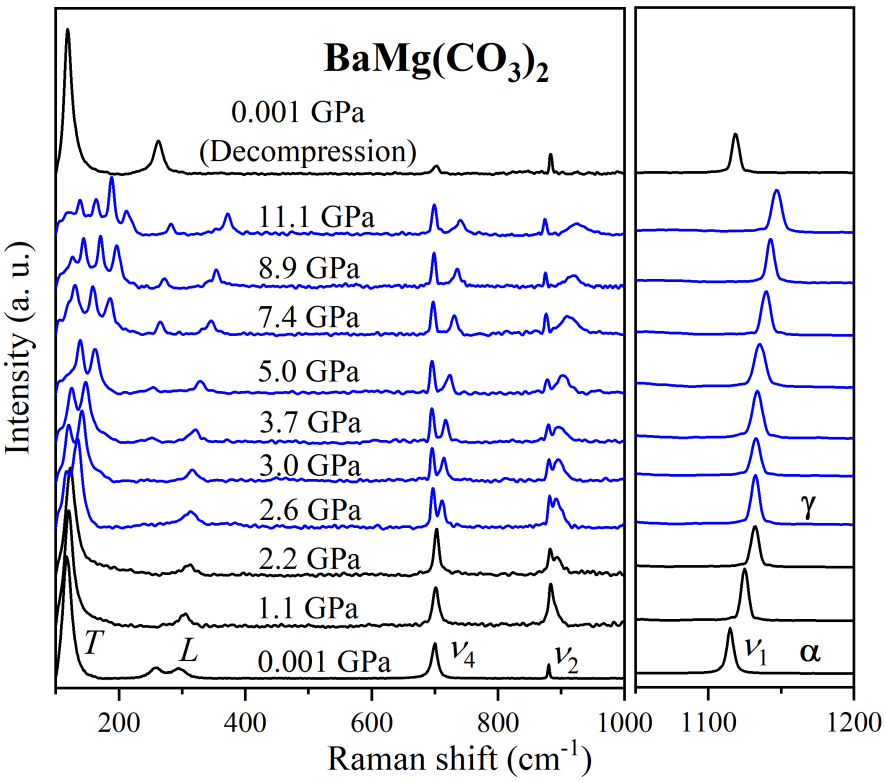

Figure S2. Representative Raman spectra of $BaMg(CO_3)_2$ at high pressures and room temperature. The black and blue solid lines represent the α- and γ-$BaMg(CO_3)_2$ phase.

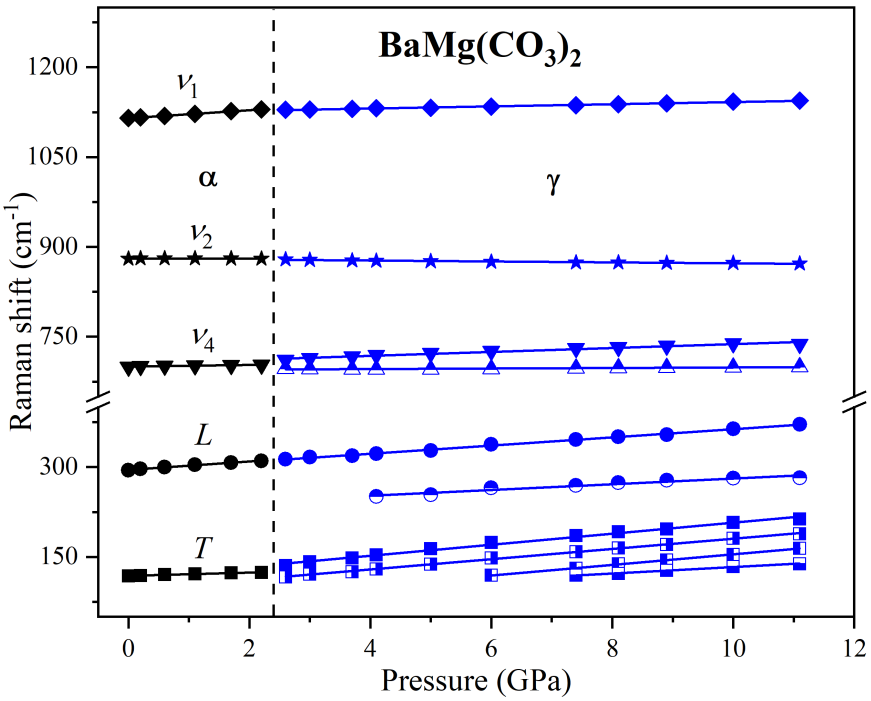

Figure S3. Representative Raman shifts of $BaMg(CO_3)_2$ at high pressures and room temperature. Error bars smaller than symbols are not shown for clarity.

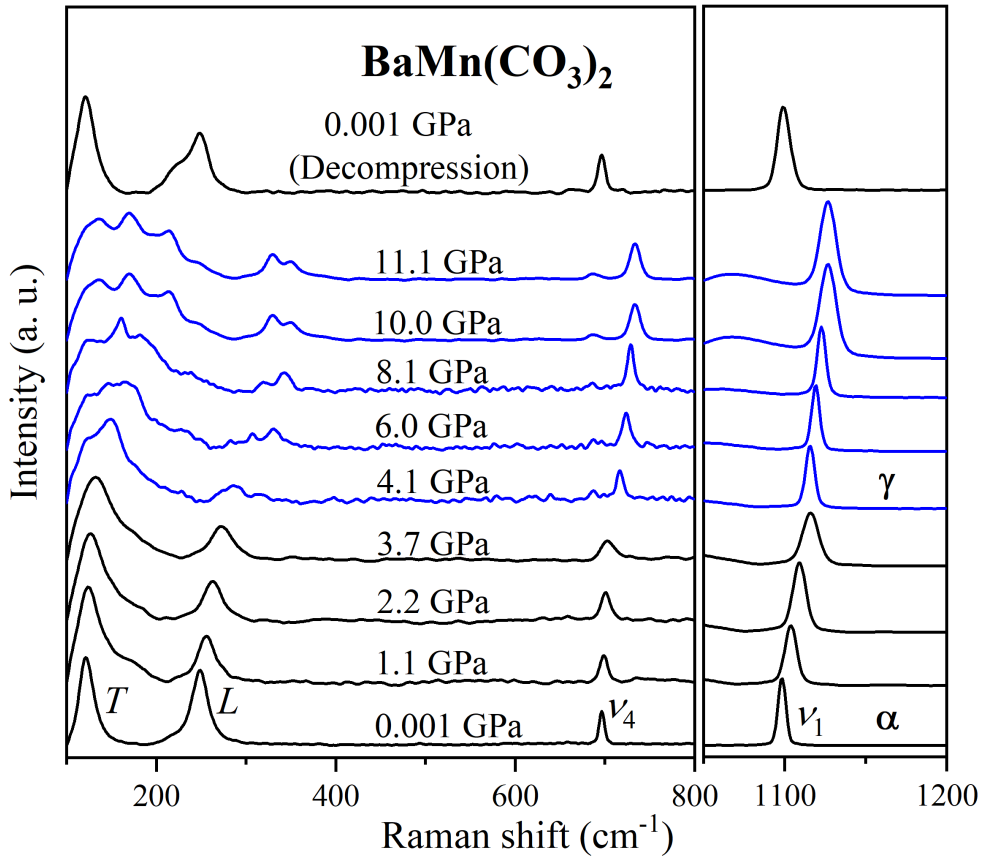

Figure S4. Representative Raman spectra of $BaMn(CO_3)_2$ at high pressures and room temperature. The black and blue solid lines represent the α- and γ-$BaMn(CO_3)_2$ phase.

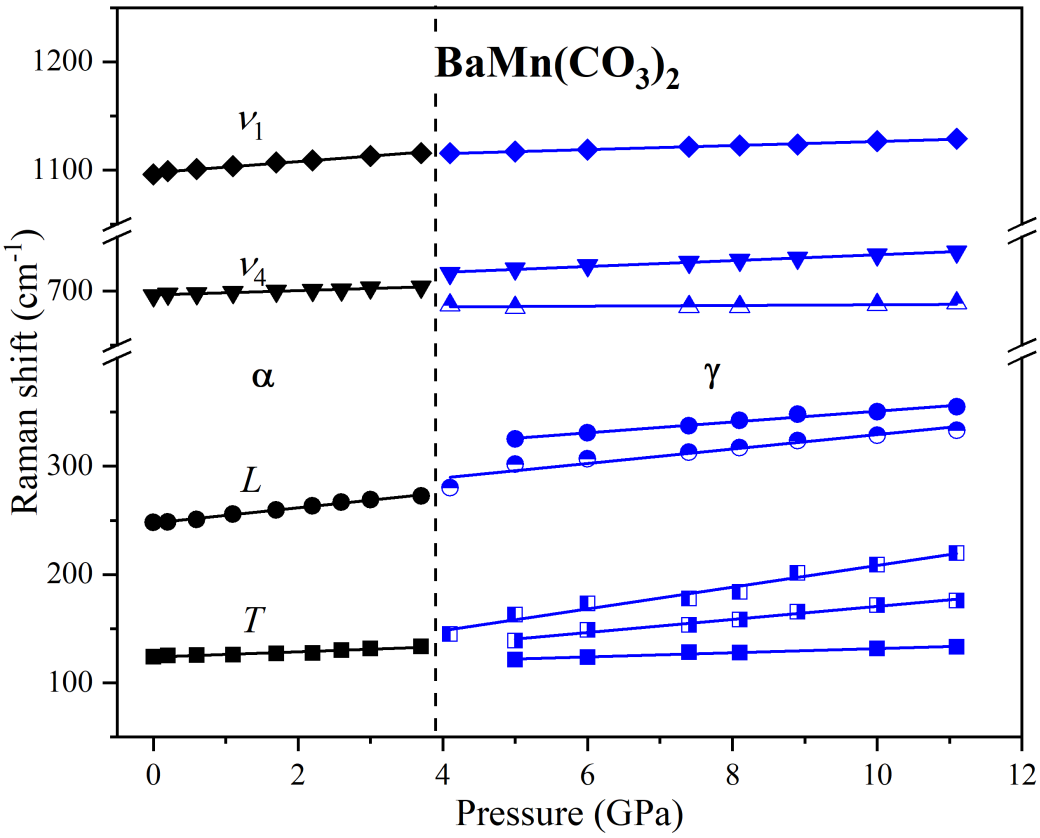

Figure S5. Representative Raman shifts of $BaMn(CO_3)_2$ at high pressures and room temperature. Error bars smaller than symbols are not shown for clarity.

Table S1. Vibrational parameters of $BaMg(CO_3)_2$ at high pressures and room temperature.

Raman modes	**α-$BaMg(CO_3)_2$** (0–2.6 GPa)			**γ-$BaMg(CO_3)_2$** (2.6–11.1 GPa)			**α-phase** 0 GPa	**α-phase** 0 GPa	**α-phase** 0 GPa
	v_{0i}	dv_i/dP	γ	v_{0i}	dv_i/dP	γ	v_{0i}	v_{0i}	v_{0i}
T	118	2.75(8)	1.54	79	5.34(3)	1.88		122	120
				66	8.78(1)	3.09			
				95	8.42(2)	3.03			
				115	9.13(9)	2.82			
L	259			233	4.92(2)	0.82	294	265	270
	295	8.15(4)	1.83	295	6.91(6)	0.92			
v_4	700	1.21(2)	0.11	694	0.56(7)	0.03	701	702	700
				705	3.20(5)	0.19			
v_2	880	0.33(1)	0.02	880	-0.88(8	-0.04	882	883	880
v_1	1115	6.60(1)	0.39	1124	1.88(4)	0.07	1118	1117	1120
References	This study						Böttcher et al. (1997)	Schmidt et al. (2013)	Pippinger et al. (2014)

Notes: v_{0i} are in cm^{-1}, dv_i/dP are in cm^{-1}/GPa. α-phase denotes α-$BaMg(CO_3)_2$. The reference frequency at room pressure (v_{i0}) and pressure coefficients, $\partial v_i/\partial P$, were used to calculate the mode Grüneisen parameters (γ_i) using the fitted K_{T0} values obtained from Pippinger et al. (2014): K_0=66.2(23) GPa for α-$BaMg(CO_3)_2$, K_0=41.9(4) GPa for γ-$BaMg(CO_3)_2$.

Table S2. Vibrational parameters of $BaMn(CO_3)_2$ at high pressures and room temperature.

Raman modes	**α-$BaMn(CO_3)_2$** (0–3.7 GPa)		**γ-$BaMn(CO_3)_2$** (3.7–11.1 GPa)		**α-$BaMn(CO_3)_2$** (0–3.9 GPa)		**γ-$BaMn(CO_3)_2$** (3.9–10.0 GPa)		**α-phase** 0 GPa
	v_{0i}	dv_i/dP	v_{0i}	dv_i/dP	v_{0i}	dv_i/dP	v_{0i}	dv_i/dP	v_{0i}
T	124	2.36(4)	112	1.95(6)	123	2.60(5)	94	4.90(10)	124
			110	6.16(2)			123	4.90(6)	
			108	10.57(5)			116	10.19(5)	
L	248	7.74(3)	262	6.67(1)	248	8.40(5)	268	7.02(2)	249
			301	5.00(1)			312	3.49(21)	
v_4	696	2.13(4)	684	0.38(19)	695	2.97(2)			697
			707	2.67(1)			709	2.46(1)	
v_1	1097	5.35(3)	1107	1.81(4)	1096	6.30(2)	1109	1.58(1)	1097
References	This study				Liang et al. (2021)				Schmidt et al. (2013)

Notes: v_{0i} are in cm^{-1}, dv_i/dP are in cm^{-1}/GPa. α-phase denotes α-$BaMn(CO_3)_2$.