

Electronic Supplementary Information

The free energy density of a crystal: calcite (CaCO_3) as a case of study

Marco Bruno

Dipartimento di Scienze della Terra - Università di Torino, via Valperga Caluso 35, I-10125 Torino, Italy.

Table S1. Static energy density at 0K (E_j^0), and vibrational energy (E_j^{vib}), vibrational entropy (S_j^{vib}), vibrational free energy (A_j^{vib}) and free energy (A_j) density at 298.15K of the layers forming the (10.4) slab and (10.4) twinned slab of calcite. The percentage contribution of A_j^{vib} to A_j for each layer is also reported, $\Delta(\%) = |A_j^{vib} / A_j| \times 100$.

layer	distance (m) $\times 10^{-9}$	E_j^0 (J m $^{-3}$) $\times 10^{11}$	E_j^{vib} (J m $^{-3}$) $\times 10^9$	S_j^{vib} (J m $^{-3}$ K $^{-1}$) $\times 10^6$	A_j^{vib} (J m $^{-3}$) $\times 10^8$	A_j (J m $^{-3}$) $\times 10^{11}$	$\Delta(\%)$
(10.4) calcite							
10	2.7304	-1.08627913	1.66648400	2.76693830	8.41521370	-1.07786392	0.78
9	2.4270	-1.10410384	1.67381310	2.50661250	9.26466600	-1.09483917	0.85
8	2.1236	-1.10327687	1.66833300	2.48556120	9.27262930	-1.09400424	0.85
7	1.8203	-1.10381440	1.66883890	2.48152070	9.28973490	-1.09452467	0.85
6	1.5169	-1.10370121	1.66798850	2.48023590	9.28506220	-1.09441614	0.85
5	1.2135	-1.10367507	1.66782980	2.48037920	9.28304720	-1.09439202	0.85
4	0.9101	-1.10381002	1.66862050	2.48190990	9.28639130	-1.09452363	0.85
3	0.6068	-1.10327669	1.66816690	2.48572210	9.27048900	-1.09400620	0.85
2	0.3034	-1.10410628	1.67369350	2.50672630	9.26313010	-1.09484315	0.85
1	0.0000	-1.08627627	1.66638960	2.76699810	8.41409090	-1.07786218	0.78
(10.4) twin calcite							
24	6.9777	-1.08618528	1.65386561	2.77957296	8.25135960	-1.07793392	0.77
23	6.6743	-1.10419944	1.66921711	2.49797880	9.24444744	-1.09495499	0.84
22	6.3709	-1.10316257	1.66461259	2.47740912	9.25973088	-1.09390284	0.85
21	6.0675	-1.10392802	1.66696946	2.47180704	9.30000192	-1.09462802	0.85
20	5.7641	-1.10350855	1.66553155	2.46899424	9.29400912	-1.09421454	0.85
19	5.4608	-1.10392174	1.66846003	2.46894576	9.32343840	-1.09459830	0.85
18	5.1574	-1.10345208	1.66724110	2.46740232	9.31585104	-1.09413623	0.85
17	4.8540	-1.10396653	1.67211955	2.46909312	9.35959416	-1.09460694	0.86
16	4.5506	-1.10332392	1.67094420	2.46954936	9.34648032	-1.09397744	0.85
15	4.2473	-1.10396728	1.67988118	2.47537272	9.41848824	-1.09454880	0.86
14	3.9439	-1.10326491	1.67804436	2.48993928	9.35668944	-1.09390822	0.86
13	3.6405	-1.10231481	1.69743746	2.53939896	9.40315680	-1.09291166	0.86
12	3.3371	-1.10114900	1.70312242	2.56774032	9.37550616	-1.09177350	0.86
11	3.0338	-1.10331296	1.68166020	2.49500136	9.37775544	-1.09393520	0.86
10	2.7304	-1.10393582	1.68459778	2.48027088	9.45105048	-1.09448477	0.86
9	2.4270	-1.10323849	1.67313494	2.47169928	9.36197832	-1.09387651	0.86
8	2.1236	-1.10398275	1.67441383	2.47022736	9.37915584	-1.09460360	0.86
7	1.8203	-1.10341126	1.66832508	2.46778440	9.32555208	-1.09408571	0.85
6	1.5169	-1.10393764	1.66950542	2.46913176	9.33333768	-1.09460430	0.85
5	1.2135	-1.10348907	1.66598789	2.46892872	9.29876808	-1.09419030	0.85
4	0.9101	-1.10393618	1.66734840	2.47202184	9.30315072	-1.09463303	0.85
3	0.6068	-1.10314830	1.66463138	2.47767552	9.25912416	-1.09388917	0.85
2	0.3034	-1.10421076	1.66899295	2.49962760	9.23728944	-1.09497347	0.84
1	0.0000	-1.08617138	1.65284431	2.79444816	8.19679584	-1.07797458	0.76

Table S2. Static energy density at 0K (E_j^0), and vibrational energy (E_j^{vib}), vibrational entropy (S_j^{vib}), vibrational free energy (A_j^{vib}) and free energy (A_j) density at 298.15K of the layers forming the (10.0) and (01.2) slabs of calcite. The percentage contribution of A_j^{vib} to A_j for each layer is also reported, $\Delta(\%) = |A_j^{vib} / A_j| \times 100$.

layer	distance (m) $\times 10^{-9}$	E_j^0 (J m ⁻³) $\times 10^{11}$	E_j^{vib} (J m ⁻³) $\times 10^9$	S_j^{vib} (J m ⁻³ K ⁻¹) $\times 10^6$	A_j^{vib} (J m ⁻³) $\times 10^8$	A_j (J m ⁻³) $\times 10^{11}$	$\Delta(\%)$
(10.0) calcite							
15	6.0396	-1.08758566	1.71698130	2.56216725	9.53071125	-1.07805495	0.88
14	5.6082	-1.10283636	1.67282340	2.51254755	9.23707305	-1.09359929	0.84
13	5.1768	-1.10432252	1.67372220	2.48931855	9.31531800	-1.09500720	0.85
12	4.7454	-1.10364477	1.67317290	2.48824335	9.31303140	-1.09433174	0.85
11	4.3140	-1.10368145	1.67309145	2.48624130	9.31818570	-1.09436326	0.85
10	3.8826	-1.10373614	1.67298090	2.48674305	9.31558470	-1.09442055	0.85
9	3.4512	-1.10370042	1.67298675	2.48610255	9.31755210	-1.09438287	0.85
8	3.0198	-1.10373810	1.67293875	2.48659875	9.31559340	-1.09442250	0.85
7	2.5884	-1.10369871	1.67298660	2.48610525	9.31754325	-1.09438117	0.85
6	2.1570	-1.10373709	1.67298075	2.48674815	9.31556850	-1.09442152	0.85
5	1.7256	-1.10368001	1.67309130	2.48624910	9.31816140	-1.09436185	0.85
4	1.2942	-1.10364363	1.67317275	2.48825385	9.31299840	-1.09433063	0.85
3	0.8628	-1.10432207	1.67372190	2.48933235	9.31527420	-1.09500680	0.85
2	0.4314	-1.10283769	1.67282295	2.51256600	9.23701395	-1.09360068	0.84
1	0.0000	-1.08758666	1.71698085	2.56219395	9.53062650	-1.07805604	0.88
(01.2) calcite							
12	4.2351	-1.08337845	1.69848072	2.57346060	9.31203480	-1.07406642	0.87
11	3.8501	-1.10417181	1.67263656	2.50128888	9.26877276	-1.09490304	0.85
10	3.4651	-1.10439124	1.67364972	2.49064680	9.31063428	-1.09508061	0.85
9	3.0801	-1.10367954	1.67300292	2.48944932	9.30773652	-1.09437180	0.85
8	2.6951	-1.10375655	1.67317584	2.48810484	9.31347360	-1.09444307	0.85
7	2.3101	-1.10371379	1.67307252	2.48794440	9.31291932	-1.09440087	0.85
6	1.9250	-1.10370004	1.67308332	2.48795592	9.31299216	-1.09438705	0.85
5	1.5400	-1.10373861	1.67318376	2.48808732	9.31360536	-1.09442500	0.85
4	1.1550	-1.10366524	1.67300856	2.48943384	9.30783804	-1.09435740	0.85
3	0.7700	-1.10452138	1.67365368	2.49064368	9.31068240	-1.09521070	0.85
2	0.3850	-1.10415941	1.67263920	2.50129644	9.26877648	-1.09489063	0.85
1	0.0000	-1.08336646	1.69847868	2.57349252	9.31191864	-1.07405454	0.87