

Supplementary Materials

Vibrational Dynamics and the Interplay of Li-ion Mobility and Thermal Transport in LiAlSiO₄

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Table S1: Refined structural parameters of LiAlSiO₄ as obtained from the Rietveld refinement of XRD data recorded at ambient temperature

LiAlSiO ₄ ; Hexagonal Space Group: P6 ₄ 22 (No. 181) a = 10.5014(1) Å, c = 11.2047(1) Å, V = 1070.10(1) Å ³ (Rp = 2.49 %, Rwp = 4.02 %, χ^2 = 5.34, R _B : = 3.04 %)					
Atoms	Wyckoff sites	x	y	z	Occupancy
Li1	3b	0	0	0.5	3.8(1.0)
Li2	3c	0.5	0	0	2.6(1.0)
Li3	6f	0.5	0	0.3129(3)	1.4(8)
Al1	6h	0.2494(2)	0	0.5	1.1(2)
Al2	6j	0.2499(1)	0.4998(2)	0.5	0.6(2)
Si1	6g	0.2504(2)	0	0	0.8(2)
Si2	6i	0.2493(1)	0.4985(2)	0	1.4(2)
O1	12k	0.0891(3)	0.2054(3)	0.2411(4)	0.8(3)
O2	12k	0.6035(4)	0.6940(3)	0.2711(4)	0.1(2)
O3	12k	0.0978(4)	0.7023(3)	0.2509(4)	1.2(2)
O4	12k	0.5952(4)	0.2036(2)	0.2502(4)	1.5(3)

Table S2: The calculated bottleneck dimensions along the c-axis formed by the SiO₄ and AlO₄ polyhedral network at equilibrated and saddle point configurations using NEB images.

	Bottleneck dimension (Rmin/Rmax) (Å)
At Equilibrium	0.92/1.50
At Saddle point	0.93/1.50

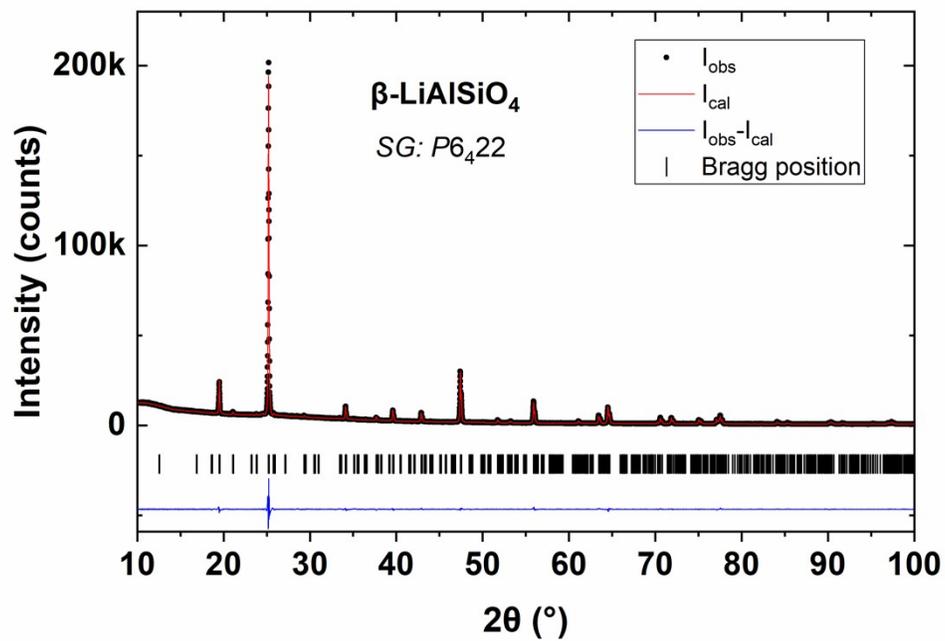


Figure S1 Rietveld refinement plot for the powder XRD data of β -eucryptite (LiAlSiO₄). Structural details reported by Pillars and Peacor¹ are used as model for refinement of the XRD data and refined parameters are given in table S1.

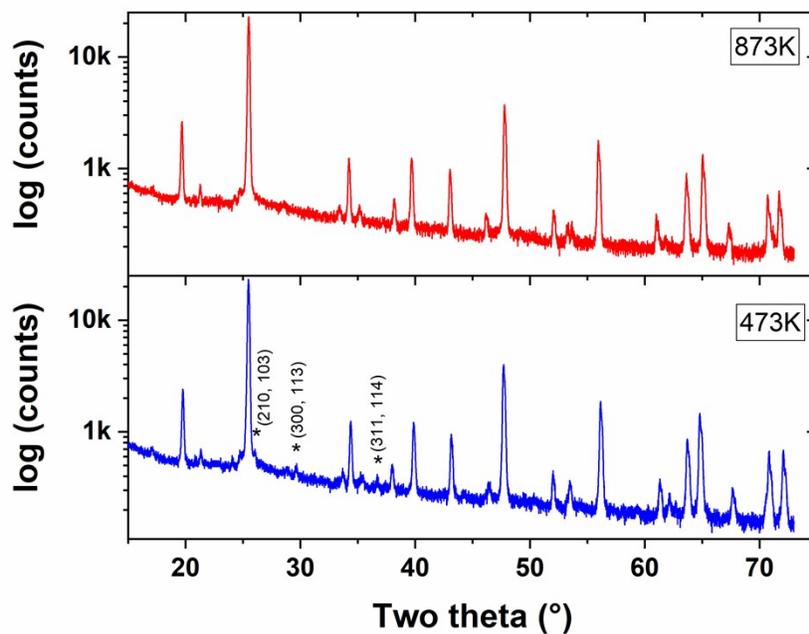


Figure S2: Powder XRD pattern of LiAlSiO₄ at 473 and 873 K. The super structure peaks are marked in the XRD pattern recorded at 473 K.

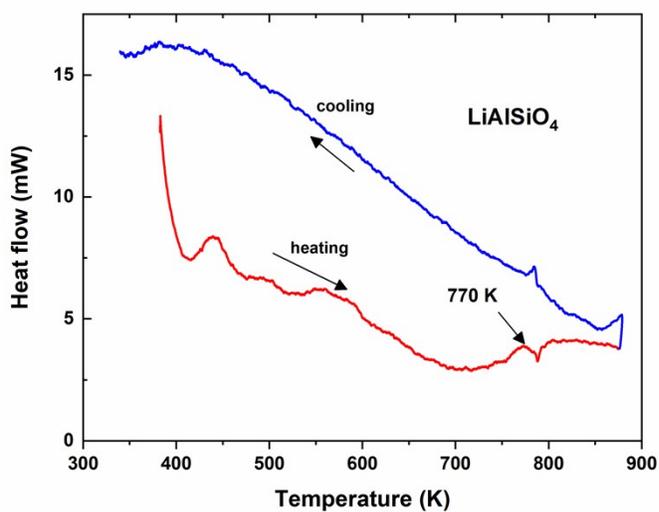


Figure S3: DSC measurement of LiAlSiO₄ recorded while heating from 373 K to 873K and then cooling in flowing air atmosphere. The heating and cooling rates for this study was 5°C/min in.

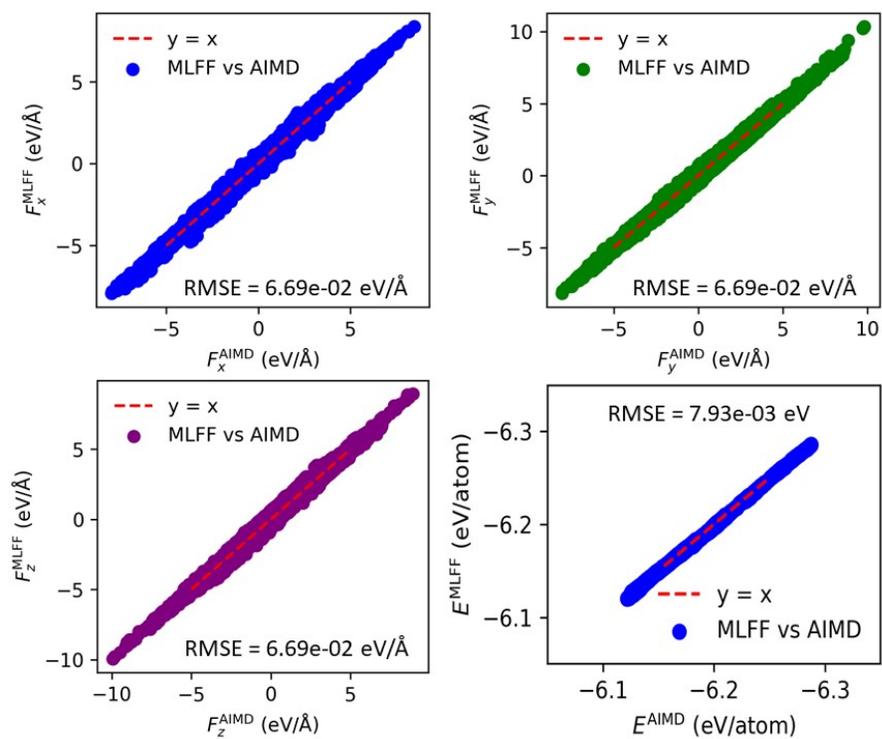


Fig S4 Validation of MLMD force-field against AIMD computed forces and energies.

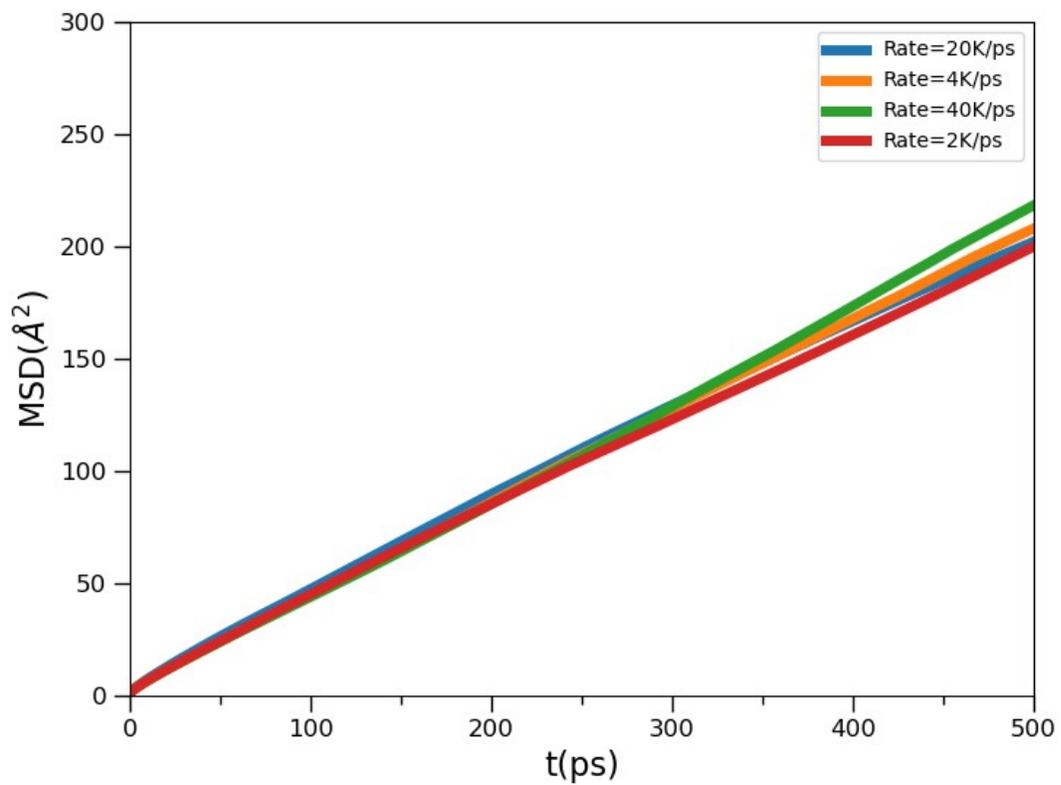


Fig S5 Calculated MSD of Li using the amorphous structure obtained using different cooling rates from a melt configuration.

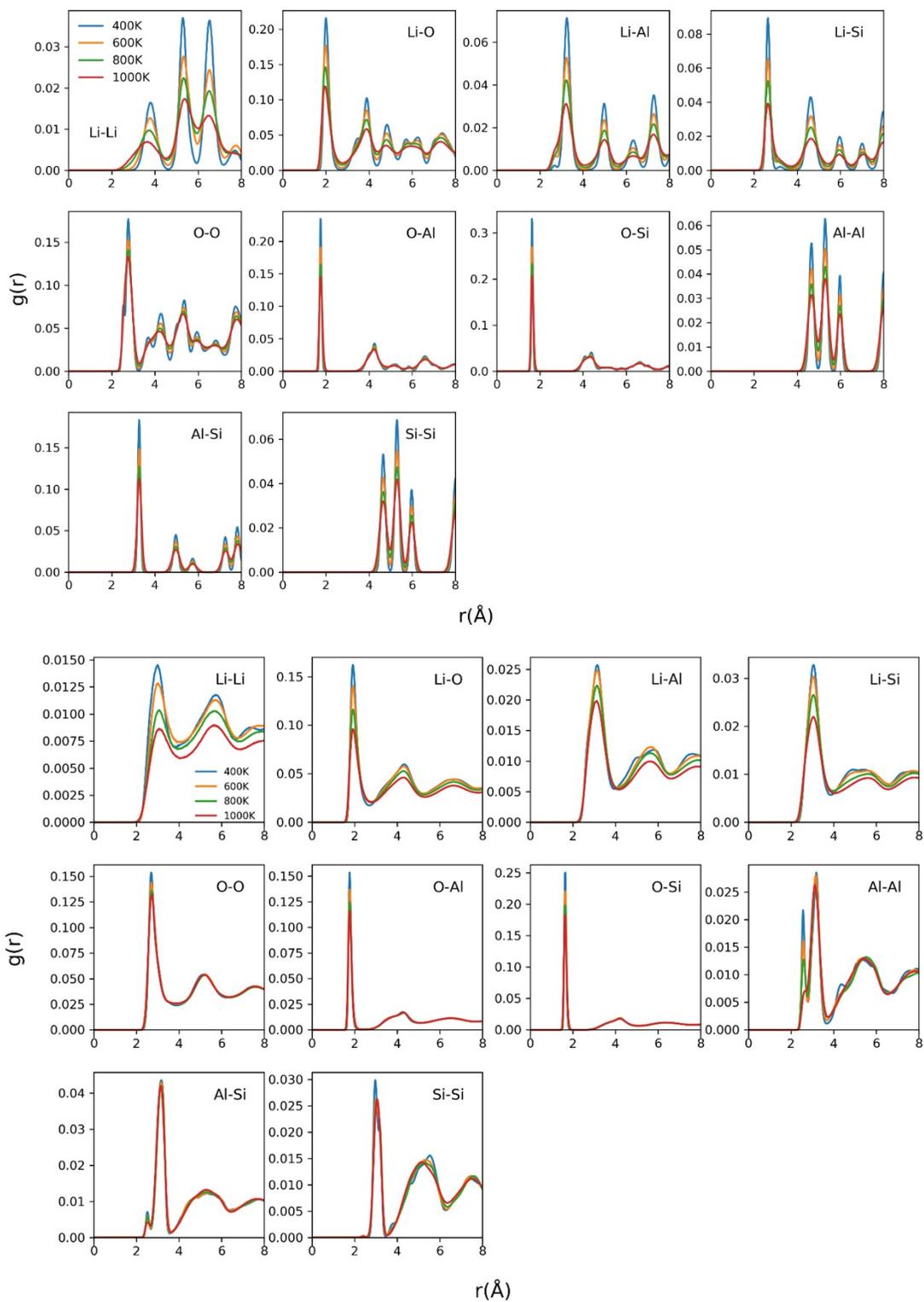


Fig S6: Radial Distribution Function of all pairs in LiAlSiO₄ for the Crystalline and Amorphous cases.

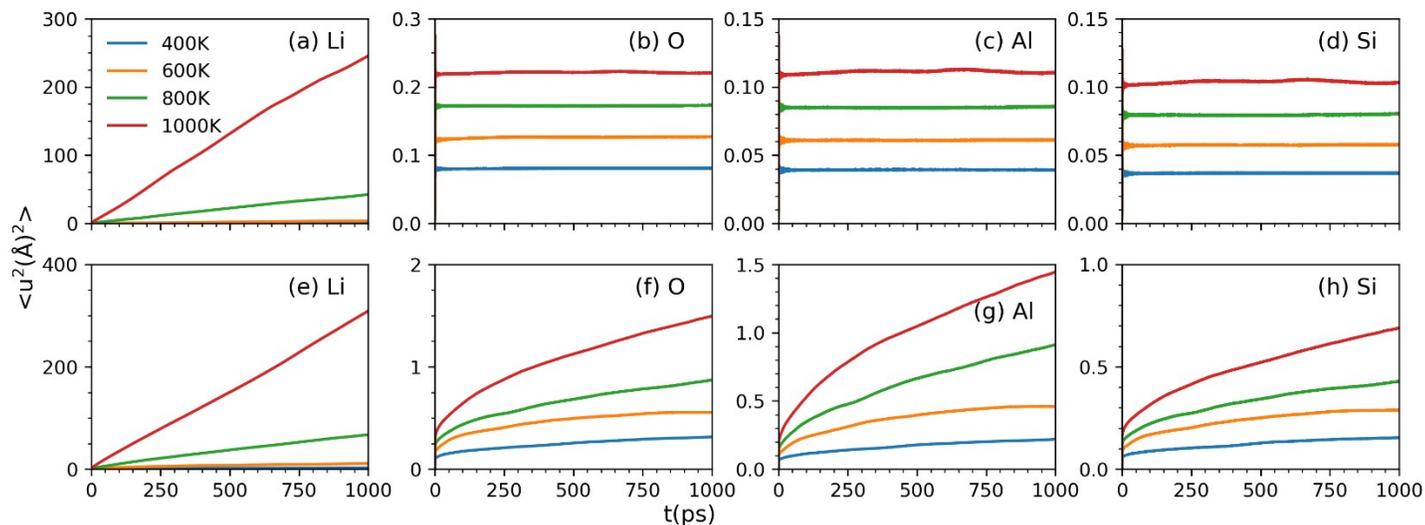


Fig S7: Mean Square Displacement (MSD) of (a, e) Li, (b, f) O, (c, g) Al, and (d, h) Si for crystalline and amorphous LiAlSiO₄, respectively.

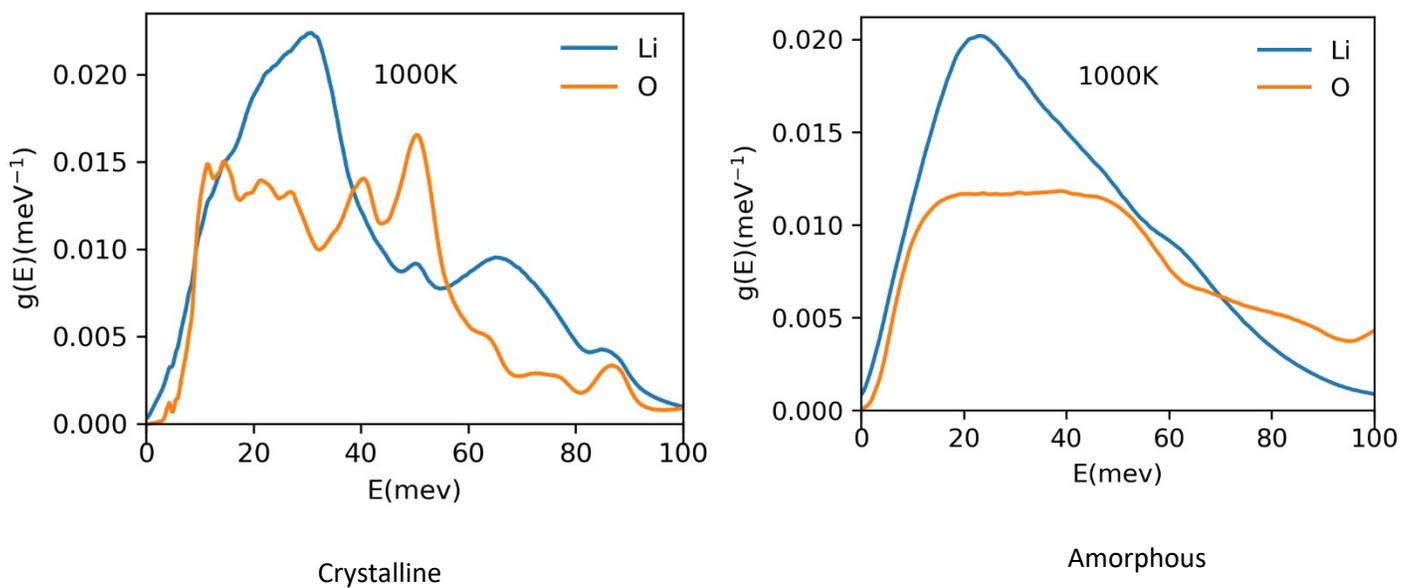


Fig S8: Li and O PDOS at 1000 K for crystalline and amorphous LiAlSiO₄.

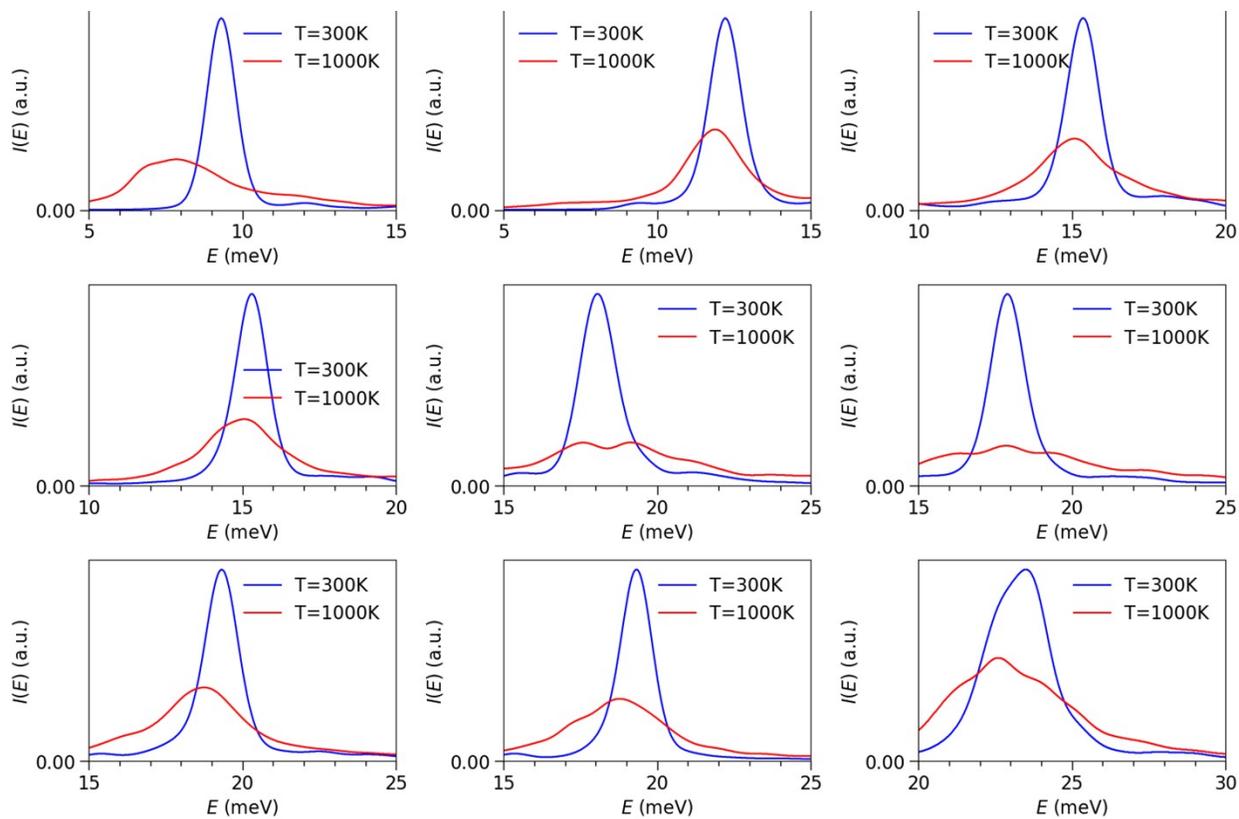


Figure S9: The phonon spectral function of first nine low-E modes at K-point at 300 K and 1000 K.

1. W. W. Pillars and D. R. Peacor, *American Mineralogist*, 1973, **58**, 681-690.