Supplementary Materials

Soft Anharmonic Coupled Vibrations of Li and SiO_4 Enable Li-ion Diffusion in Amorphous $Li_2Si_2O_5$

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Crystalline and amorphous samples of $Li_2Si_2O_5$ were prepared by solid state reaction of stoichiometric amounts of Li_2CO_3 and SiO_2 in a procedure similar that adopted[1] for $Na_2Si_2O_5$. Li_2CO_3 was dried overnight at 150°C for 10 hours while SiO_2 was heated at 800°C for 12 hours prior to weighing for reaction. About 40 gm of $Li_2Si_2O_5$ was prepared in four batches. Desired amounts of dried Li_2CO_3 and SiO_2 for batches of about 10 gm of products were thoroughly homogenized using ethanol and then acetone as grinding media. The homogenous mixtures were pressed to pellets of 20 mm diameter and 20 mm height. The pellets were heated at 400° for 12 hours and then the temperature was raised to 800°C and soaked for 24 hours. The pellets were crushed to powder and then reorganized and again heated in similar pellet form at 950°C for 24 hours. Formation of orthorhombic $Li_2Si_2O_5$ phase was observed in all the batches of sample. All the four batches of samples were crushed and mixed together. The powder was pressed again nto pellets, and heated at 1000°C for 8 hours and then cooled to room temperature. Orthorhombic (*C*cc2) phase of $Li_2Si_2O_5$ was confirmed by powder XRD of the obtained product. All the observed peaks could be accounted to the orthorhombic lattice with unit cell parameters: a = 5.8189(5) Å, b = 14.6294(5) Å, and c = 4.7786(3) Å, which are in agreement with those reported in literature[2].

About half of the powdered sample was placed in a platinum crucible and heated to 1120°C and soaked for 2 hours. The transparent glass like solid was obtained by quenching the molten mass in liquid nitrogen. The powder XRD pattern of crystalline and amorphous $Li_2Si_2O_5$ sample are shown in Figure-S1. For preparation of $^7Li_2Si_2O_5$, the same procedure was adopted using $^7Li_2CO_3$. The powder XRD pattern of crystalline and amorphous $^7Li_2Si_2O_5$ samples are similar to those shown in Figure-S1.

Figure S1. Rietveld refinement plot of powder XRD data of crystalline and amorphous $Li_2Si_2O_5$.

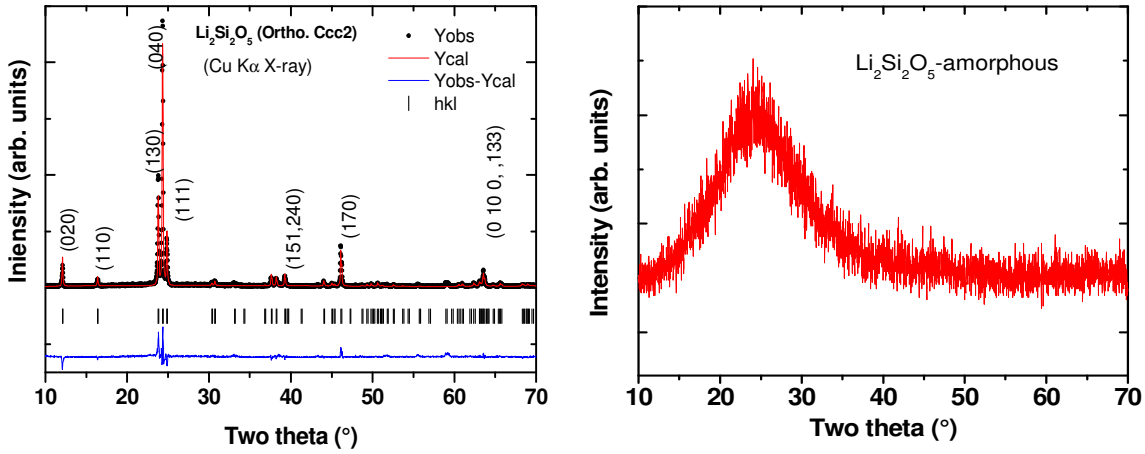


Figure S2. Neutron diffraction data of polycrystalline sample of $Li_2Si_2O_5$ in two phases (a) crystalline (*n*-LSO) (b) amorphous (*amr*-LSO). The data is measured simultaneously along while performing the phonon measurements. The Q resolution is not very good; however, it shows transformation of amorphous to crystalline transformation at 823 K.

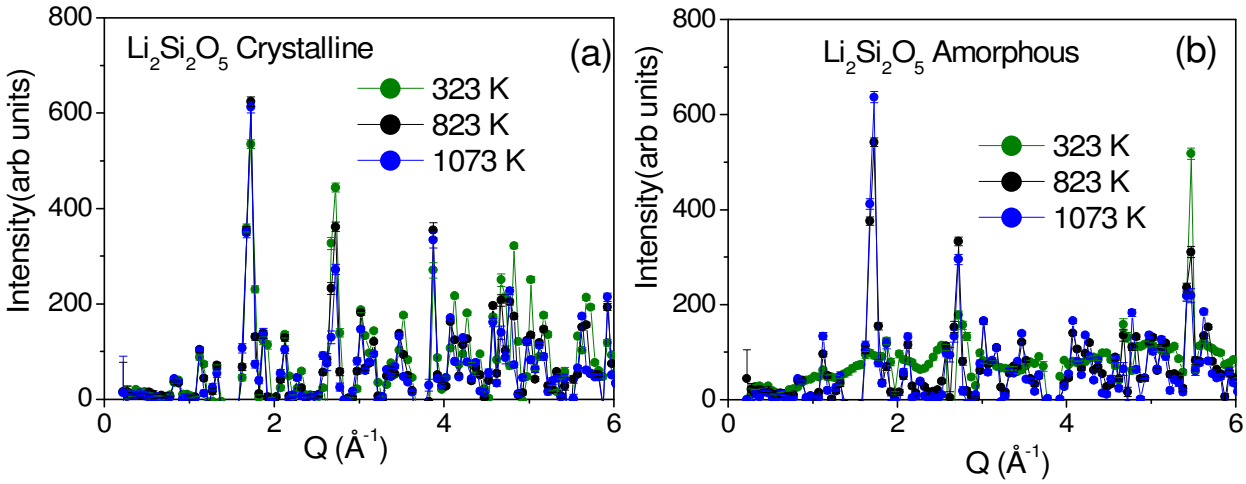


Figure S3. Differential scanning calorimetry (DSC) measurement to evaluate the transition temperature of crystallization.

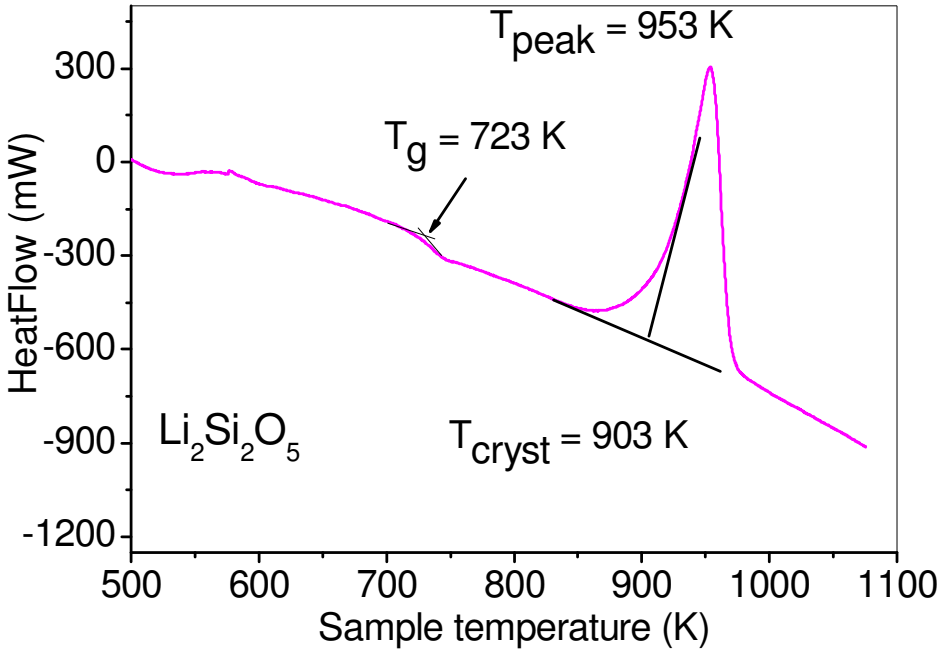


Figure S4. The measured total neutron weighted DOS in crystalline and amorphous structure of $Li_2Si_2O_5$ at various temperature with two incident energy beam E_i= 30 meV and 180 meV.

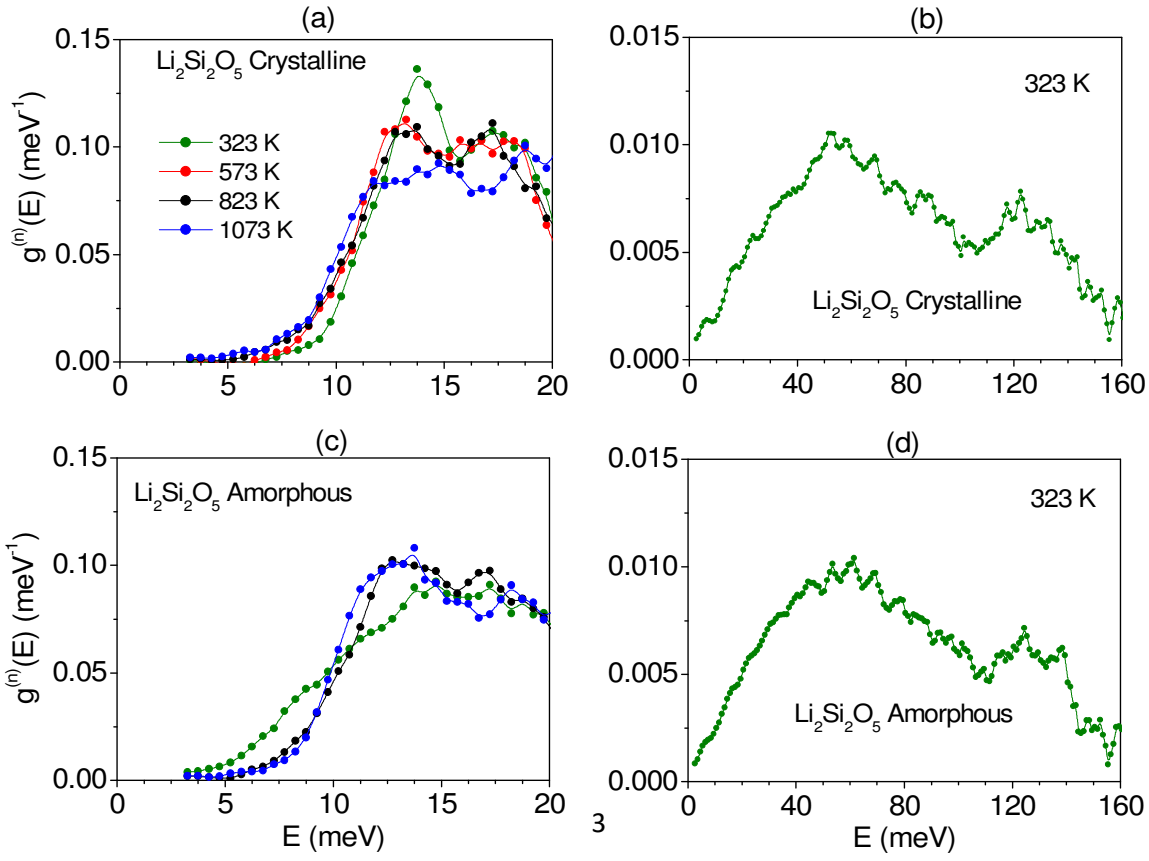


Figure S5. The calculated mean square displacement with respect to phonon energy in *n*-LSO and *amr*-LSO using lattice dynamics (LD)and ab-initio molecular dynamics (AIMD).

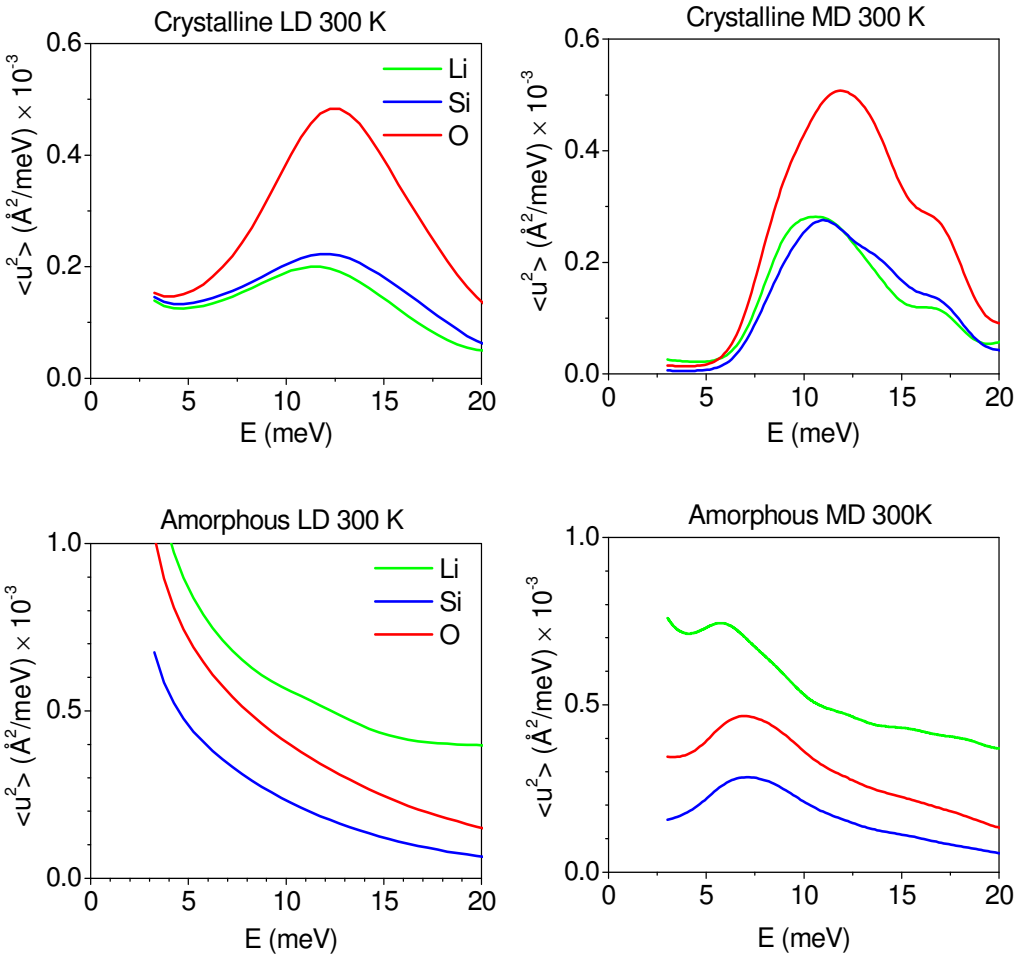


Figure S6. The calculated partial and total DOS in crystalline structure of $Li_2Si_2O_5$ at 300 K and 1000 K using AIMD

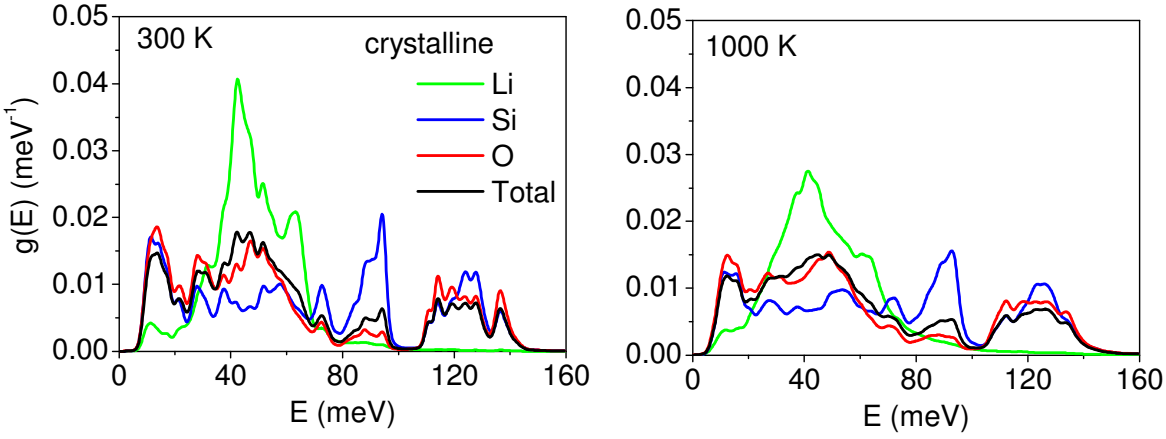


Figure S7. The calculated partial and total phonon density of states in amorphous $Li_2Si_2O_5$ at various temperature from 300 K to 1000 K.

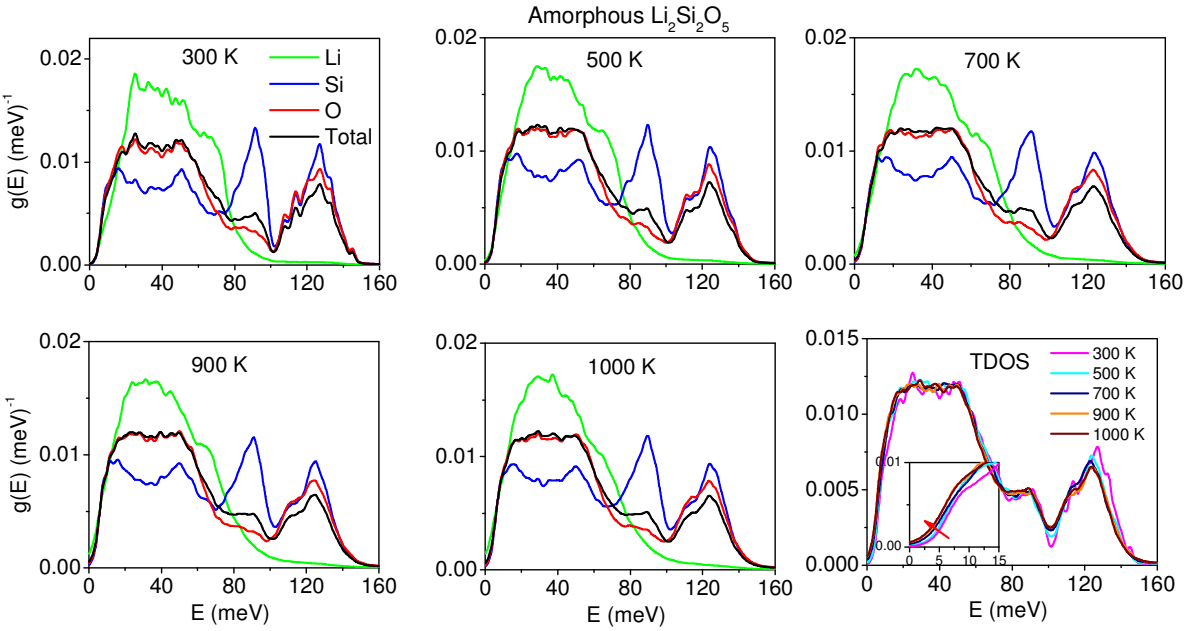


Figure S8. (a,b) The calculated contribution from low energy zone centre phonon modes. (c,d) Mean square displacement (300 K) of individual atom from phonons at zone centre in *n*-LSO and *amr*-LSO using quasiharmonic lattice dynamics calculations.

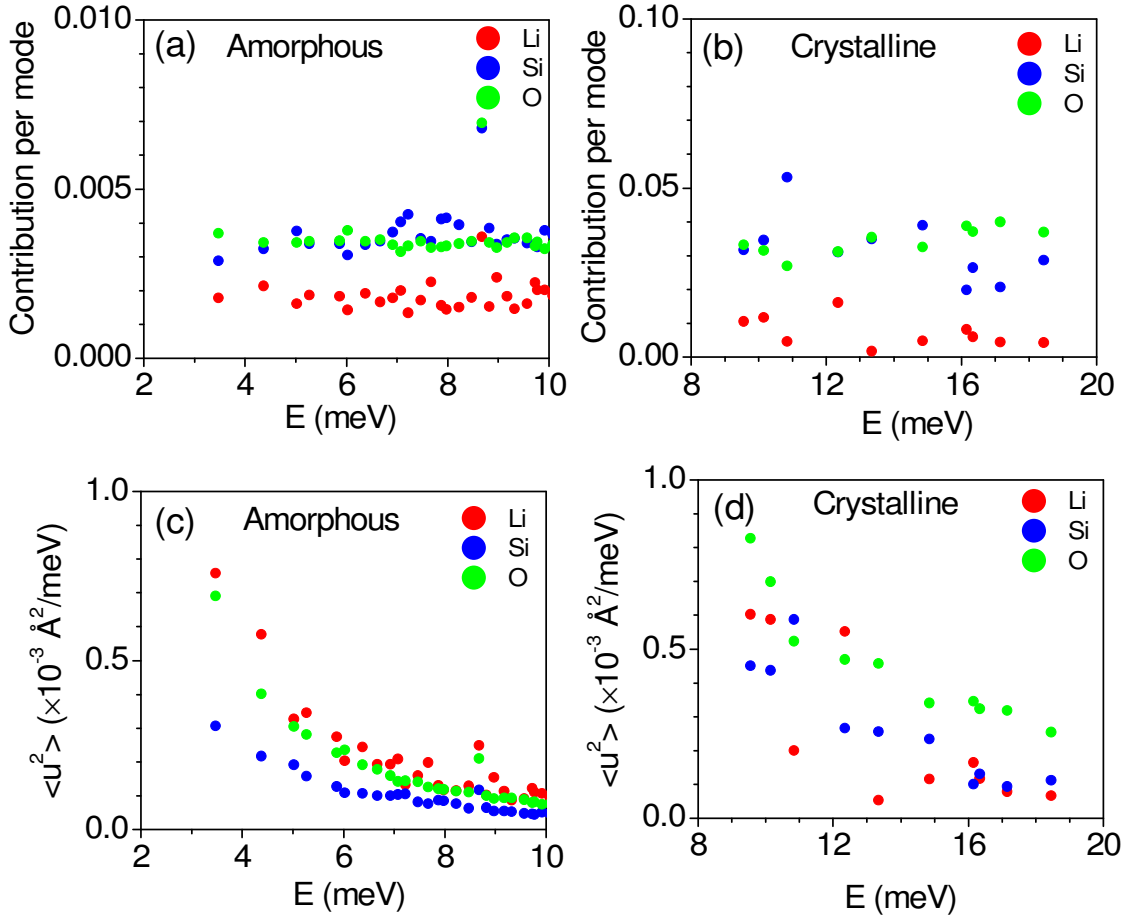


Figure S9. (a) The measured intensity at various temperature from 300 K to 900 K (b) the fitting of measured S(*Q*, *E*) data with a Lorentzian function , Gaussian function and flat background. (b) The estimated Lorentz and Gauss area at 725 K.

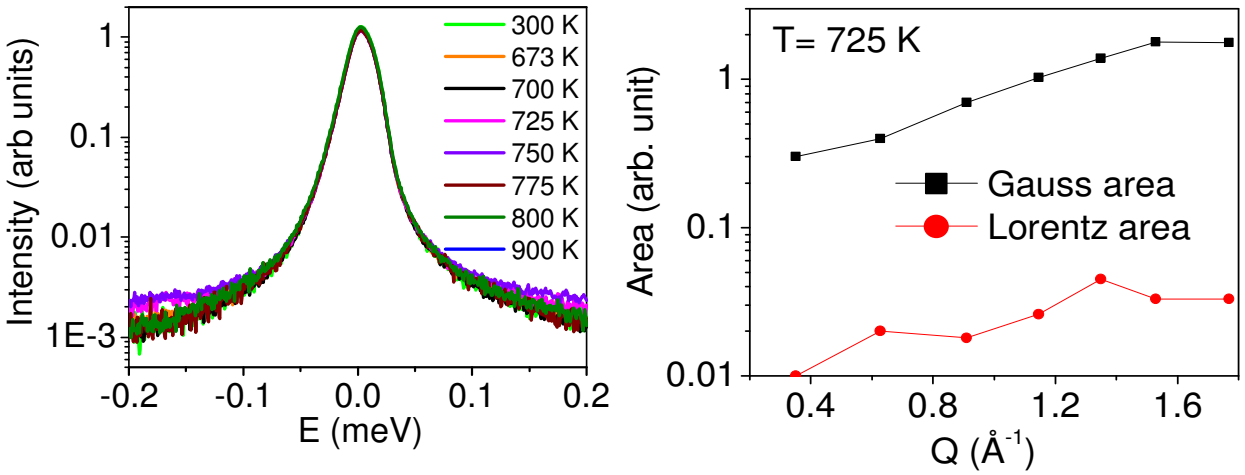


Figure S10. The calculated nuetron weighted dynamical structure factor S(Q, E) of contituent atom (Li, O, and Si) and neutron weighted total S(Q, E) at 700 K for *amr*-LSO using AIMD[3, 4]. The S(Q, E) is fitted (solid line) with single lorentzian to estimate the half width at half maximum (Γ).

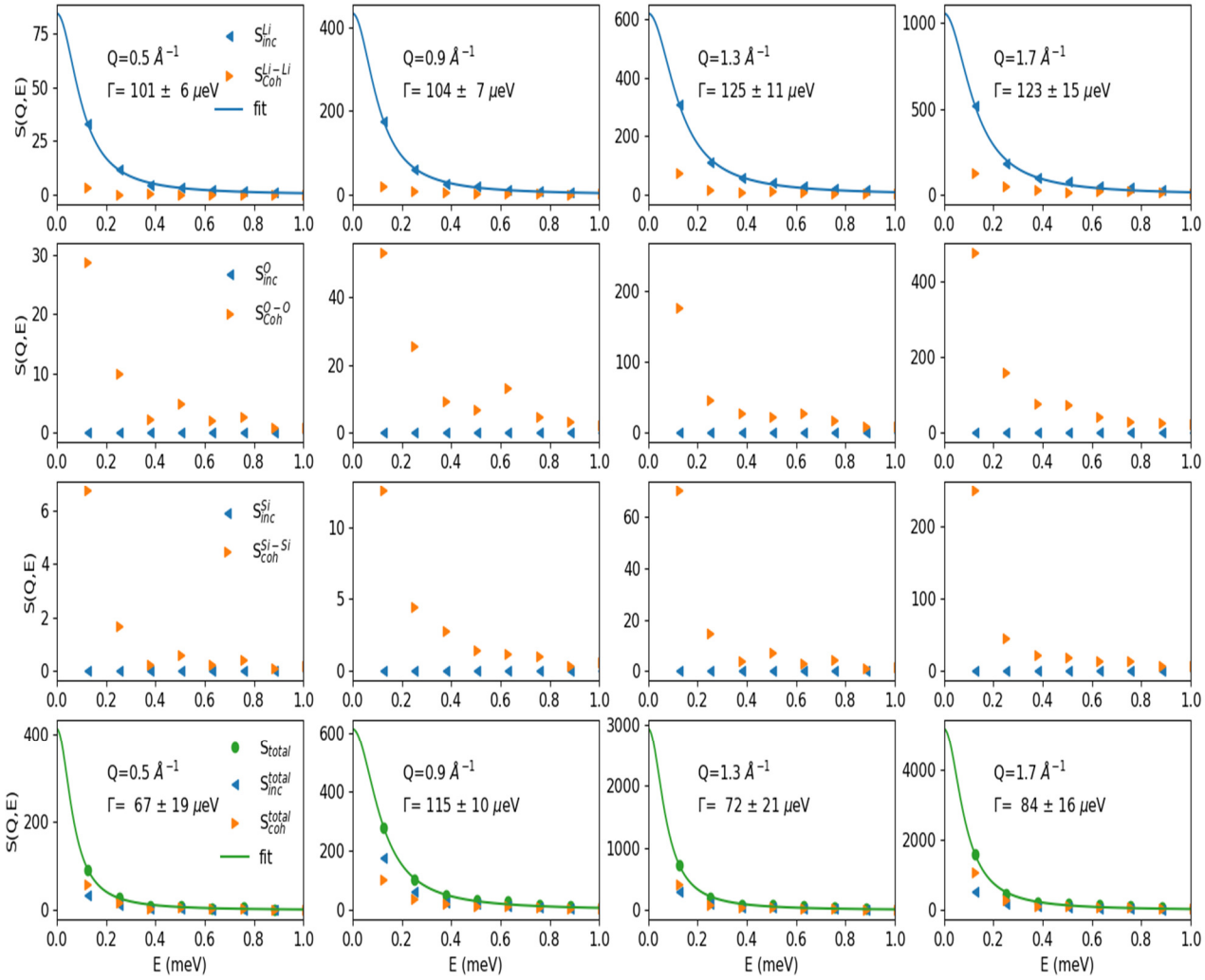


Figure S11. The calculated time averaged pair distribution function among all possible pairs in crystalline phase of $Li_2Si_2O_5$ at 300 K and 1000 K using AIMD.

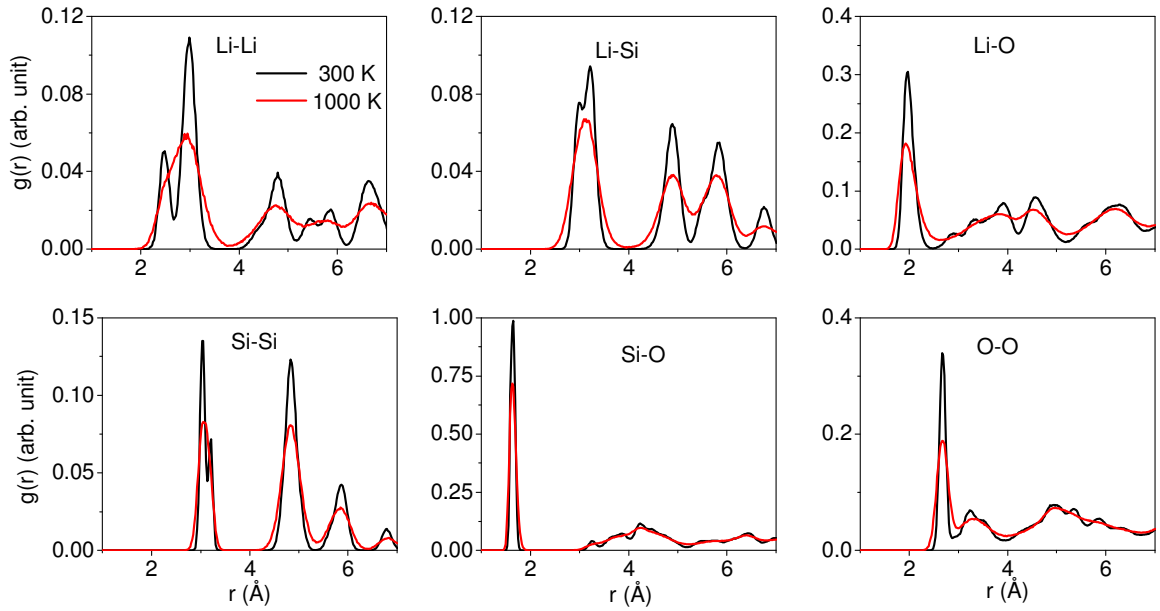


Figure S12. The calculated time averaged pair distribution function among all possible pairs in amorphous phase of $Li_2Si_2O_5$ at 300 K to 1000 K using AIMD.

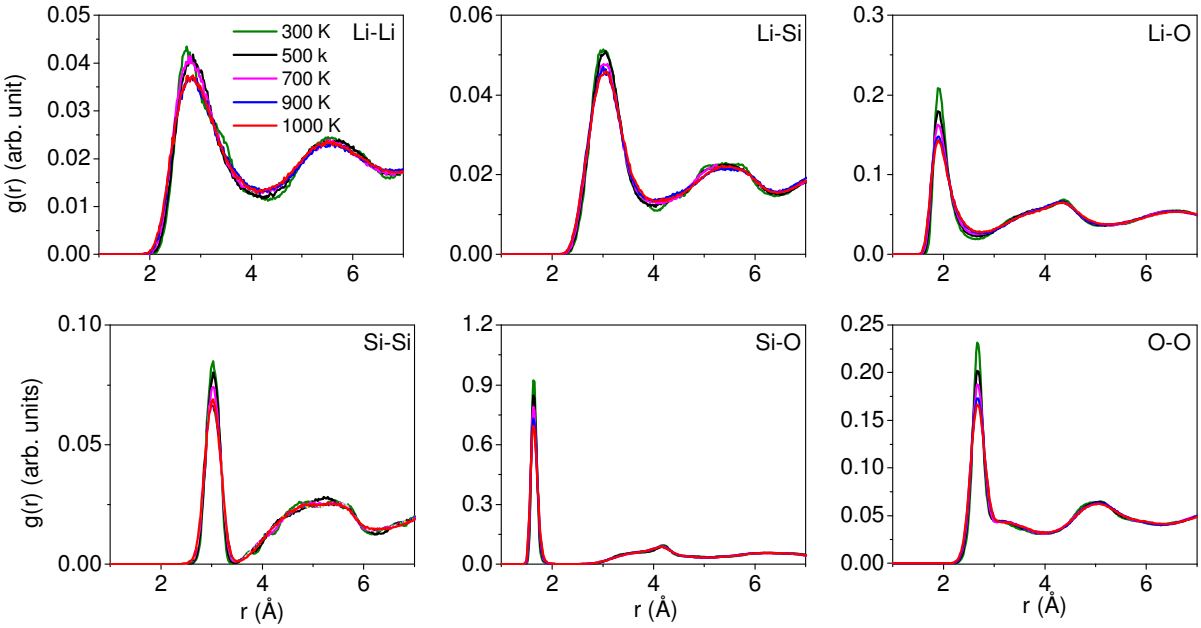


Figure S13. The calculated MSD of Individual species in crystalline phase of $Li_2Si_2O_5$ at 300 K and 1000 K using AIMD

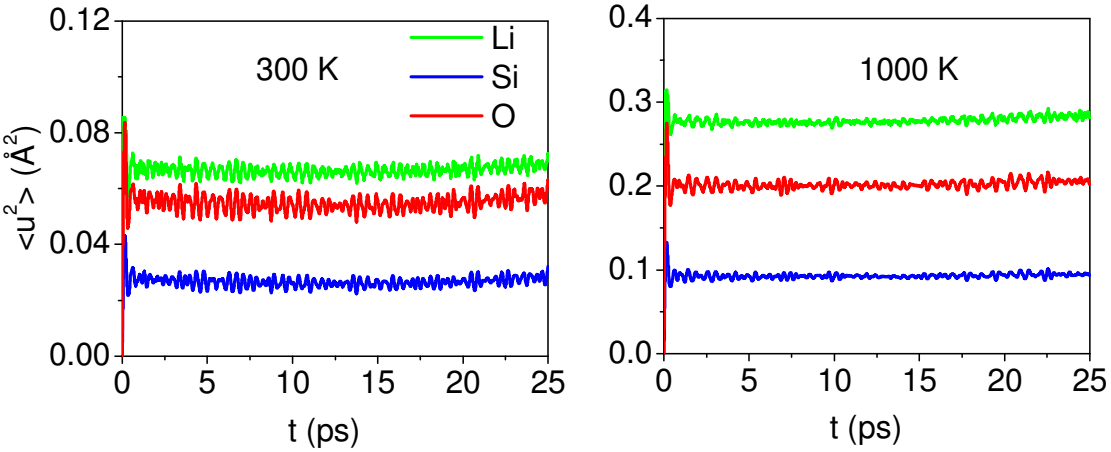


Figure S14. The calculated MSD of individual species in amorphous structure of $Li_2Si_2O_5$ at various temperature using AIMD. Bottom panel gives the calculated MSD of Si and O at an expanded scale.

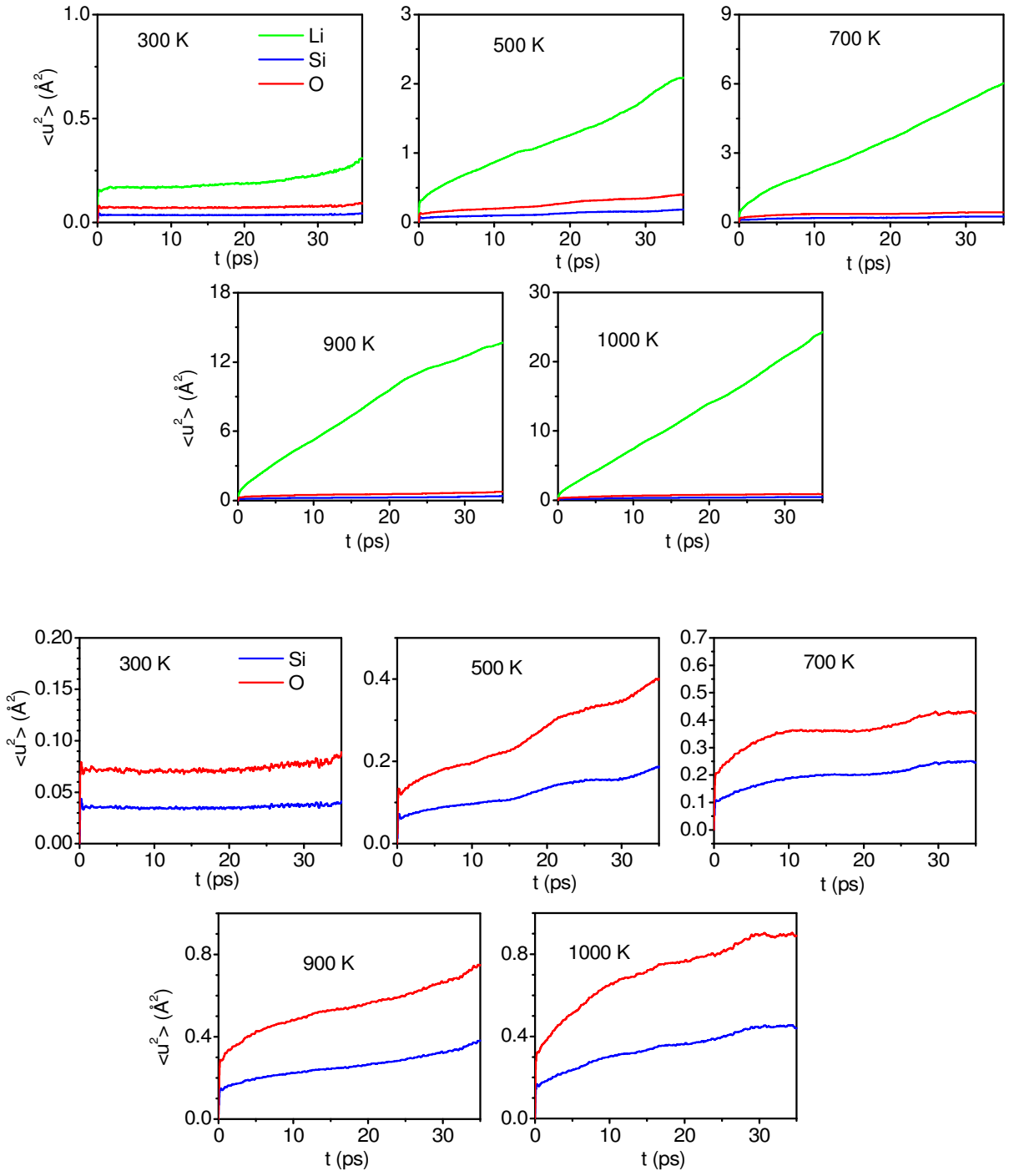


Figure S15. The calculated trajectory of Li-ion in *amr*-LSO for 40 ps in the inerval of 8 fs step at 700 K, 900K and 1000 K. SiO_4 polyhedra repersented by blue colour and O atom hidden for better visiual.

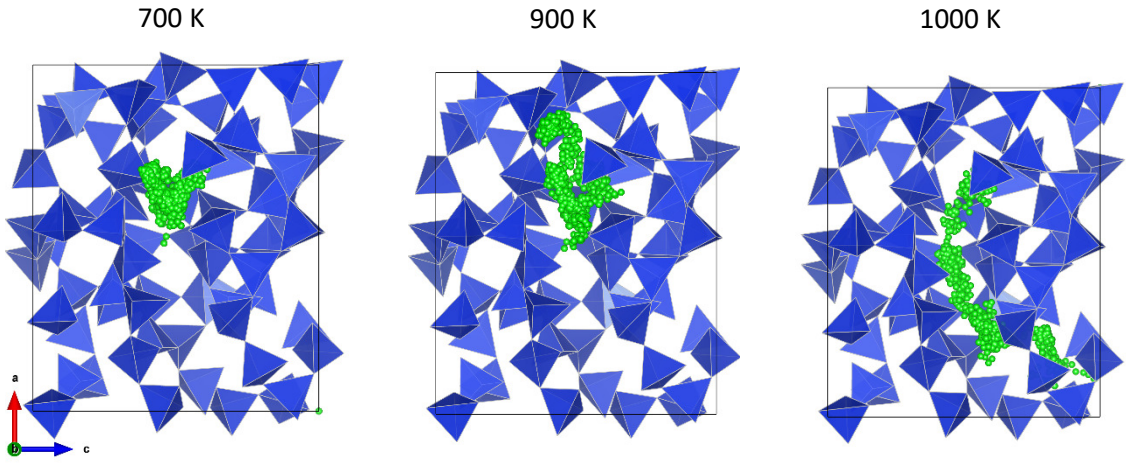
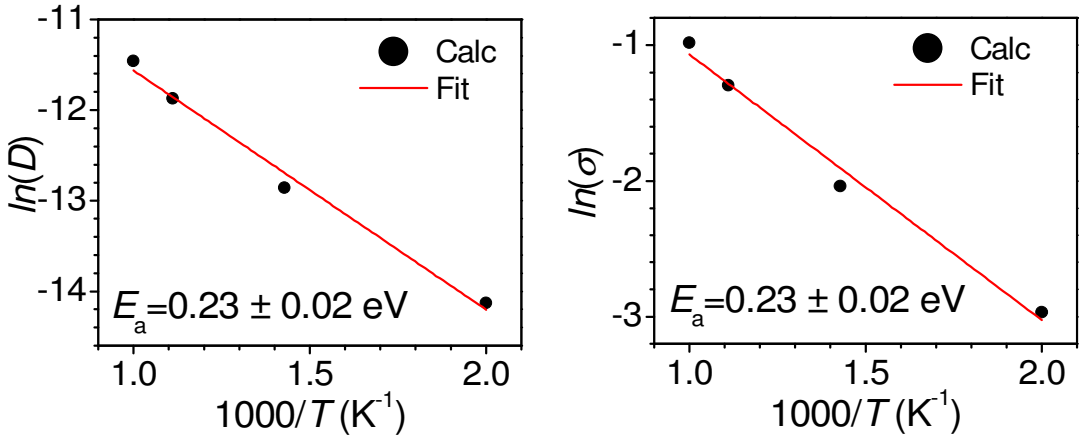


Figure S16. The calculated diffusion coefficient, D (cm^2sec^{-1}) and Li-ionic conductivity, σ (Scm^{-1}) with temperature in *amr*-LSO. The temperature dependence is fitted with the Arrhenius model and obtained a barrier energy for Li diffision ~ 0.23 ± 0.02 eV. $\sigma_{Li} = \frac{Nq^2 D_{Li}}{k_B T}$ where, N is the number density of mobile ion in unit cell, q be the charge of the mobile ion, T be the temperature and k_B is the Boltzmann constant.



1. M. K. Gupta, S. K. Mishra, R. Mittal, B. Singh, P. Goel, S. Mukhopadhyay, R. Shukla, S. N. Achary, A. K. Tyagi and S. L. Chaplot, *Physical Review Materials*, 2020, **4**, 045802.
2. W. Paszkowicza, A. Wolskaa, M. Klepkaa, S. abd el Allb and F. Ezz-Eldinb, *Proceedings of the 8th National Meeting of Synchrotron Radiation Users, Podlesice, September 24–26,* , 2009.
3. M. K. Gupta, J. Ding, N. C. Osti, D. L. Abernathy, W. Arnold, H. Wang, Z. Hood and O. Delaire, *Energy & Environmental Science*, 2021, **14**, 6554-6563.
4. M. K. Gupta, J. Ding, D. Bansal, D. L. Abernathy, G. Ehlers, N. C. Osti, W. G. Zeier and O. Delaire, *Advanced Energy Materials*, 2022, **12**, 2200596.