A machine learning force field for albite and the diffusion mechanisms of its defects

A. Gorfer[1,2], R. Abart[2], C. Dellago[1]

[1]*Faculty of Physics, University of Vienna, Boltzmanngasse 5, 1090, Vienna, Austria*
[2]*Department of Lithospheric Research, University of Vienna, Josef-Holaubuek-Platz 2, 1090, Vienna, Austria*
e-mail: alexander.gorfer@univie.ac.at

Feldspar is the most abundant mineral in the Earth's crust and the nature of its diffusive phase transformations are essential for reconstructing the thermal histories of magmatic and metamorphic rocks. Due to the large timescales over which these transformations proceed, the mechanism responsible for sodium diffusion and its possible anisotropy has remained a topic of debate. To elucidate the process, we have developed a machine learning force field (MLFF) (reviewed in Unke et al. 2022) trained on first-principle calculations of Albite (Na-feldspar) and its charged defects.

The MLFF has been trained to accurately predict a range of experimental macroscopic properties as well as defect formation energies, incorporating electrostatic corrections of the underlying first-principle calculation. Notably, we have discovered a new type of dumbbell interstitial defect, which is found to be the most favorable interstitial, and its formation free energy at finite temperature has been computed using thermodynamic integration.

Using the force field to drive molecular dynamics (MD) simulations allowed us to gain unprecedented insight into the diffusion mechanisms, as depicted in Figure 1. Through the analysis of jump rates, diffusion coefficients and tracer correlation factors we have determined that correlation plays a significant role, particularly in the ⊥(010) direction due to a distinct dumbbell/interstitialcy mechanism. Moreover, we have observed a high degree of anisotropy in diffusion, with variations up to 27-fold at 1000K, as illustrated in Figure 2. The strong agreement between our results and experimental diffusion coefficients leads us to conclude that MLFFs represent a mature and powerful methodology for investigating the dynamical properties of feldspar and other silicates.

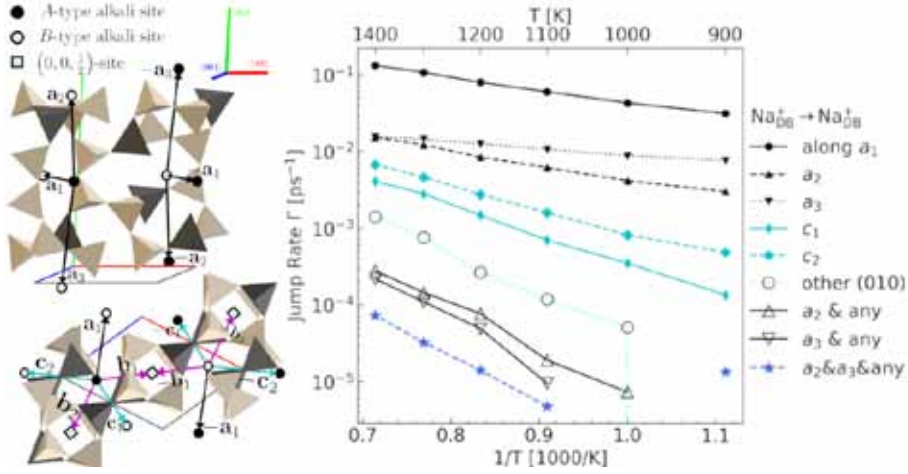

Figure 1. Left: diffusion pathways connecting the different sites. Right: jump rates for paths between alkali sites measured during an MD simulation using the MLFF. The dominance of diffusion along $\mathbf{a}_{1,2,3}$ leads to the high anisotropy.

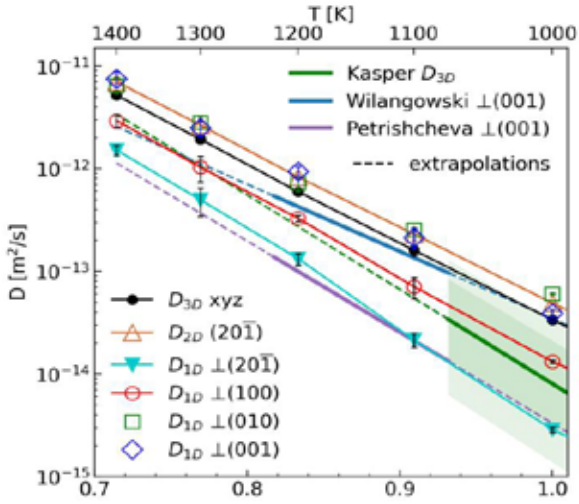

Figure 2. Diffusion coefficients for an interstitial defect measured over all dimensions D_{3D} and projected onto two D_{2D} or one dimension D_{1D} including experimental data of Kasper (1975), Wilangowski et al. (2015), and Petrishcheva et al. (2020)

Unke O, Chmiela S, Sauceda E, Gastegger M, Poltavsky I, Schütt K, Tkatchenko A, Müller KR (2021): Machine Learning Force Fields. - Chem Rev 121 (16), 10142-10186

Kasper RB (1975): Cation and Oxygen Diffusion in Albite - Ph.D. Thesis, Brown University.

Wilangowski F, Divinski S, Abart R, Stolwijk NA (2015) (1962): Radiotracer experiments and Monte Carlo simulation of sodium diffusion in alkali feldspar: evidence against the vacancy mechanism. - Defects Diffus Forum 363, 79–84

Petrishcheva E, Tiede L, Heuser D, Hutter H, Giester G, Abart R (2020): Multicomponent diffusion in ionic crystals: theoretical model and application to combined tracer- and interdiffusion in alkali feldspar. - Phys Chem Minerals 47, 35