Oxygen Vacancy Diffusion in $BaTiO_3$ via Machine Learning Potential

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Computer simulations have garnered significant attention for their role in designing new materials at the atomic level. We have proposed a method to achieve highly accurate meso-scale ion dynamics [1]. This approach comprises three key components: (1) Optimizing the parameters of empirical interatomic potentials using ab-initio calculations. (2) Calculating free energies for specific events through multicanonical Monte Carlo (MUCA-MC) simulations [2,3], leveraging these optimized potentials. (3) Simulating ion dynamics with the parallelized kinetic Monte Carlo (kMC) simulation [4], which incorporates the temperature-dependent activation energies and attempt frequencies obtained from Step 2. This multi-scale simulation approach is a novel method poised to make substantial contributions to next-generation materials development in the energy and sustainability fields. In this study, we are taking Step 1 in a new direction: we will use a machine learning potential instead of traditional empirical interatomic potentials and apply it to oxygen vacancy defects in perovskite oxide $BaTiO_3$.

The calculation process begins by performing ab-initio molecular dynamics (AIMD) simulations of $BaTiO_3$ with an oxygen vacancy at 1000 K, 1500 K and 2000 K using the VASP software [5,6]. From these simulations, a total of 18,000 data points (6,000 from each temperature) are extracted to train the high-dimensional neural network potential (HD-NNP) using the n2p2 software [7,8]. This machine learning potential is then used to perform MUCA-MC simulations to estimate the kinetic parameters. Finally, these parameters are used in kMC simulations to model the diffusion of oxygen vacancies around 1100 K.

Figure 1 shows the energy profile along the minimum energy path (MEP) of an oxygen vacancy defect. The MEP is the lowest energy pathway for the diffusion event shown in the inset. The blue squares represent the results obtained from the ab-initio calculation using the Wu-Cohen functional [9], and the red circles show the results from the optimized HD-NNP. Although there is a noticeable discrepancy near the energy barrier maximum, the HD-NNP generally reproduces the behavior of the ab-initio calculation well. It should be noted that the ab-initio calculation are at 0 K, whereas the HD-NNP was constructed from the high-temperature AIMD simulations. Therefore, they are not expected to be in perfect agreement. The important point is that the optimized HD-NNP accurately reproduces the behavior of the ab initio calculation, including its symmetry. We plan to present the results of the MUCA-MC and kMC simulations during the presentation.

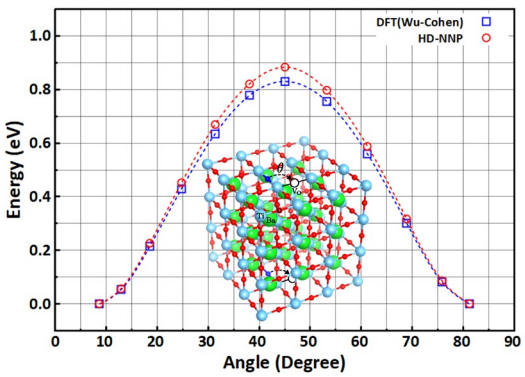

Figure 1 Energy profile along minimum energy path of oxygen vacancy diffusion at 0 K.

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