Machine learning force fields for accurate defect calculations

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Abstract

Point defects control the properties of most functional materials, ranging from ionic conductivity to solar cell performance. Their dilute concentrations render experimental characterisation challenging, thus requiring computational methods to understand their impact on macroscopic properties.

The standard approach to modelling the thermodynamics of defects relies on a static description, where the change in Gibbs free energy is approximated by the internal energy at 0 K. This approach has a low computational cost, but ignores contributions from atomic vibrations and structural configurations that can be accessed at finite temperatures [1-3]. To assess these limitations, we train a machine learning force field (MLFF) to explore the dynamic defect behaviour at the device operating temperature, using Te_i^{+1} and V_{Te}^{+2} in CdTe and V_{Cl}^{+1} in $CsPbCl_3$ as exemplars [4]. We consider the different entropic contributions (e.g., electronic, spin, vibrational, orientational, and configurational) and compare methods to compute the defect free energies, ranging from a harmonic treatment to a fully anharmonic approach based on non-equilibrium thermodynamic integration. We find that metastable configurations are populated at the device operating temperature and thermal effects increase the predicted concentration of Te_i^{+1} by two orders of magnitude — and can thus significantly affect the predicted properties. Overall, our study underscores the importance of finite-temperature effects and the potential of MLFFs to accurately model complex defect processes.

References

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Figures

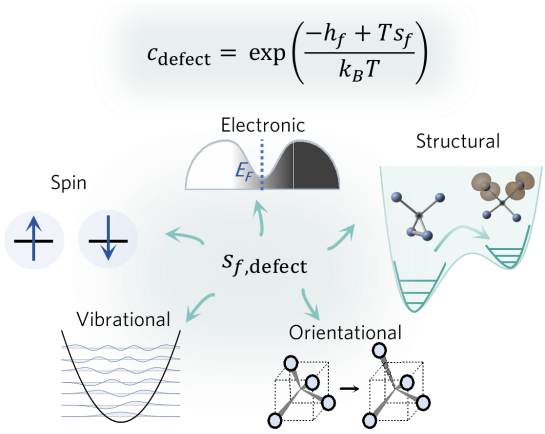

Figure 1. Different degrees of freedom that contribute to the defect formation entropy and predicted defect concentration.