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ABSTRACT

Accurate ab initio Modeling of Thermal Effects in High-Temperature Diffusion

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Accurately modeling thermal effects in high-temperature diffusion is essential for predicting material performance and stability in extreme environments. However, describing diffusion at elevated temperatures presents a significant challenge for density functional theory (DFT). At high temperatures, atomic vibrations, electronic and magnetic excitations, and their complex coupling effects play crucial roles in diffusion mechanisms. Neglecting or oversimplifying these thermal contributions can result in substantial errors, often spanning several orders of magnitude. To address these challenges, we propose an efficient ab initio framework that integrates DFT with molecular dynamics (MD) simulations and machine-learning interatomic potentials to compute the temperature-dependent Gibbs energy of vacancy formation and migration in vacancy-mediated diffusion. Our approach thoroughly accounts for relevant thermal effects, including anharmonic lattice vibrations and electronic excitations, ensuring a highly accurate description of diffusion processes.

Using body-centered cubic (BCC) tungsten as a prototype system, we reveal that anharmonic effects strongly influence the vacancy formation and migration Gibbs energies. This insight explains the physical origin of the experimentally observed non-Arrhenius behavior in tungsten self-diffusion. The resulting temperature-dependent self-diffusivity calculations show excellent agreement with experimental data, accurately capturing both magnitude and characteristic curvature. Furthermore, the robustness and transferability of the framework are demonstrated through initial applications to a multi-component high-entropy alloy. This comprehensive ab initio methodology lays the groundwork for developing accurate diffusion databases. It enhances the predictive power of first-principles modeling in the design of materials for high-temperature applications.

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[2] A. Dash, A. Paul, S. Sen, S. V. Divinski, J. Kundin, I. Steinbach, B. Grabowski, and X. Zhang, Annual Review of Materials Research 52 (2022) 383–409.