Toward Cost-Effective Configuration Coordinate Diagrams in Nonradiative Capture Calculations

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Nonradiative carrier capture at deep-level defects limits the performance of optoelectronic and power semiconductor devices, and its first-principles evaluation is now routinely carried out within the one-dimensional configuration-coordinate (1D-CC) framework [1–4]. In this approach, the potential-energy curves of the initial and final defect states are obtained by fitting density functional theory (DFT) total energies computed at discrete points along the configuration coordinate. Because each point requires an expensive supercell calculation, often with a hybrid functional, the number and placement of sampling points largely determine the computational cost. This becomes a bottleneck for high-throughput and database-scale studies. Yet in current practice, sampling points and fitting functions are typically chosen by visual inspection or experience.

In this work, we explore an objective, data-driven approach to configuration-coordinate diagram construction. We use leave-one-out cross-validation (LOOCV) to estimate the predictive accuracy of fitted potential-energy curves without additional DFT calculations. Several sampling orderings and fitting functions, ranging from harmonic and polynomial forms to Morse-type and spline representations, are compared on a dataset of potential-energy curves combining our own hybrid-functional calculations with data from published studies [5]. We further examine how fitting accuracy propagates to the calculated capture coefficients for representative defects with nearly harmonic and strongly anharmonic lattice relaxation.

Our analysis suggests that the order in which points are sampled and the choice of fitting function both strongly affect convergence, and that monitoring the LOOCV error offers a practical stopping criterion. We will present guidelines that can reduce unnecessary DFT calculations in large-scale nonradiative capture studies.

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