Quantum machine learning for the prediction of two-dimensional photocathode growth on crystal substrates

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We present a high-throughput, material-agnostic strategy to study promising new photocathode materials for their epitaxial growth. Cs_3Sb films grown on 3C-SiC(100) substrates with molecular beam epitaxy (MBE) display lattice parameters intermediate to fully epitaxial layers and bulk structures, indicating partial relaxation at a thickness of only 38 Å. The surface phase diagrams and structures for monolayer thin films of epitaxial Cs-Sb photocathodes on SiC substrates have been predicted using a genetic algorithm (GA) coupled with density functional theory (DFT) calculations. This information helped identify the experimental growth conditions for uniform and stoichiometric films and possible surface reconstructions. However, the computational cost of DFT calculations limits the study to a single monolayer, and the nucleation and growth of these films cannot be studied with this method.

Hence, we employ machine learning accelerated GA simulations to predict complex surface structures with larger in-place unit cells and increasing film thickness to model the transition from monolayer to bulk. Following this, the growth of Cs-Sb layers on (100) and (001) 3C-SiC substrates is studied with exascale molecular dynamics (MD) simulations. Our atomistic modeling aims to elucidate the structural transition from potentially epitaxial monolayers to bulk structures and identify structures, nucleation mechanisms, and film growth processes on (100) and (001) 3C-SiC substrates. The GA and MD simulations are powered by machine-learned ultra-fast force fields (UF^3) trained on the data from the previous GA/DFT studies, greatly reducing the computational cost.[1] The resulting structures obtained from the simulations are compared with experimental XRD and RHEED patterns, and the structural energies provide the basis for a thin-film phase diagram.

While the work here is presented for Cs-Sb films on a SiC substrate, the framework can be applied to any material system to create similar phase diagrams. These phase diagrams, along with kinetic information gleaned from the molecular-dynamics simulations, can help identify optimal experimental processing conditions during MBE growth and guide the selection of other materials systems.

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[1] Stephen R. Xie, Matthias Rupp, and Richard G. Hennig, "Ultra-Fast Interpretable Machine-Learning Potentials," *Npj Computational Materials* 9, no. 1 (September 2, 2023): 1–9, https://doi.org/10.1038/s41524-023-01092-7.