Stoichiometric growth of a wide-bandgap semiconductor $SrTiO_3$ film via Bayesian optimization with adaptive prior mean

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In recent years, the rapid advance in materials informatics has enabled efficient and speedy development of materials through the application of information science. As for one such development/application, we have developed a technique called "machine-learning-assisted molecular beam epitaxy" (ML-MBE), which combines molecular beam epitaxy with Bayesian optimization (BO) of statistical machine learning. Aiming to explore new materials and enhance crystallinity, we have focused on perovskite oxides [1,2]. Notably, our ML-MBE successfully produced ultra-high-quality thin films of $SrRuO_3$, a 4*d* magnetic metal, on which we observed quantum transport of Weyl fermions [2]. While BO is a sample-efficient method for optimizing a black-box function by machine learning to find the maximum (or minimum) value of the function, further enhancing the efficiency of ML-MBE requires improving the BO method itself to better suit crystal growth.

To satisfy that requirement, we developed a new BO method for efficiently and automatically searching for growth parameters in the ML-MBE framework. This BO method has two strengths: first, the algorithm directly searches a wide parameter space while compensating for missing growth data caused by growth failure when growth conditions deviate significantly from optima [3]. Second, the method adaptively modifies the prior mean function of the prediction model so that the function can escape from a locally optimal region that exhibits mediocre evaluation values. As a result, the search for unseen regions is expanded so that the function reaches an improved global optimum [4]. In addition to validating the effectiveness of the method by using simulation functions, we successfully applied the method to ML-MBE and thereby enabled efficient fabrication of high-quality $SrTiO_3$ (STO) thin films.

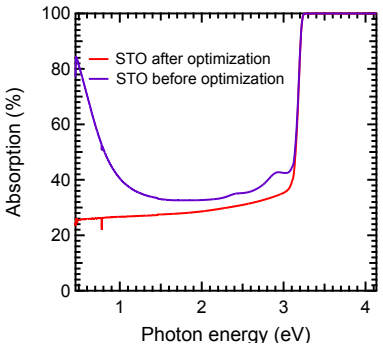

Fig. 1. Optical absorptions of STO films before and after BO.

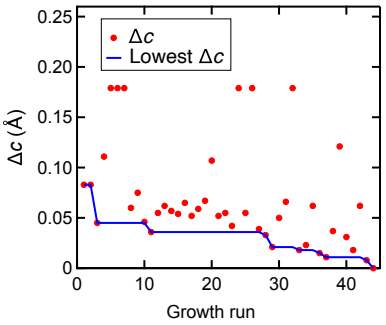

Fig. 2. Actual Δc values and the lowest experimental Δc plotted as a function of growth run.

Wide-bandgap semiconductor STO, a material extensively studied for its high dielectric constant (100-200), high electron mobility over 53,000 cm^2/Vs, and chemical stability, is promising for next-generation high-*k* capacitors and photocatalytic materials. To harness STO as a functional material, it is essential to grow a highly insulating stoichiometric STO film with no absorption in the bandgap. However, reproducibly growing such a film has been challenging. To overcome that challenge, we used our new BO method as follows. The evaluation value to be minimized by BO is the difference in lattice constants [Δc (Å)] of grown STO films and bulk STO. The BO method predicts and explores Δc (Å) of STO films grown under various growth conditions (substrate temperature, Ti/Sr atomic flux ratio, and ozone supply). As a result, we developed highly insulating stoichiometric STO films with lattice constants identical to those of bulk STO. Observation of the STO films by visible-to-UV light spectroscopy showed no optical absorption other than that from the substrate in the bandgap (3.2 eV) (Fig. 1), and the films were grown in only 44 MBE growth runs (Fig. 2). The proposed BO algorithm provides an efficient experimental design platform that is not as dependent on the experience and skills of individual researchers. Since ML-MBE has also grown ultra-high-quality $SrRuO_3$ films that exhibit quantum transport of Weyl fermions, this approach is generally promising for growing various kinds of semiconductors with enhanced efficiency.

References [1] Y. K. Wakabayashi *et al*., APL Mater. **7**, 101114 (2019). [2] K. Takiguchi *et al*., Nat. commun. **11**, 4969 (2020). [3] Y. K. Wakabayashi *et al*., npj Comput. Mater. **8**, 180 (2022). [4] Y. K. Wakabayashi *et al*., APL Mach. Learn. **1**, 026104 (2023).