Synthesis of large area single-crystalline freestanding oxide membranes

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Recently, a number of 2D materials have been the focus of intense study for the development of nanoelectronics [1]. One potential platform for study is the complex oxide perovskites. Complex oxides are known to exhibit a rich variety of properties such as superconductivity and ferroelectricity due to the local entanglement of the charge, spin and orbital degrees of freedom [2]. In addition, these properties have been shown to be highly tunable by strain, which is an advantage of the 2D geometry compared to bulk samples [3]. The synthesis of complex oxide nanomembranes, while highly challenging due to the 3D nature of the oxygen octahedral bonding, may offer pathways towards highly controllable flexible optical and electronic devices.

In order to take advantage of the unique properties of complex oxides, the final membrane or device must consist of high quality single-crystalline oxide materials. Recently, we have developed a new synthesis technique for crystalline complex oxide membranes and heterostructures compatible with state-of-the-art atomic-scale thin-film growth. The technique makes use of pH-neutral water to selectively etch the sacrificial layer ($Sr_3Al_2O_6$) to release the epitaxially grown thin-film from the substrate [4].

In contrast to van der Waals heterostructures, the lattice mismatch between the sacrificial layer and the thin-film plays a critical role in the growth process, membrane release, resulting nanocrystallite size, and material quality. A key question for further developing this highly promising route is to understand the scope of materials and lattice constants which can be accessed via the Sr₃Al₂O₆ family of materials. Here we take a representative transparent conducting oxide, La-doped BaSnO₃, as a model system. The large lattice mismatch of 3.9% with Sr₃Al₂O₆ leads to high crack density when released due to the large epitaxial strain at the sacrificial layer and thin-film interface. Employing a lattice matched sacrificial layer (Sr,Ba)₃Al₂O₆ (mismatch 0.4%) leads to successful fabrication of millimeter-scale large-area membranes (Fig. 1a) with crystallinity and electronic properties comparable to the epitaxial thin films (Fig. 1b), paving the way for the development of nanomembrane devices.

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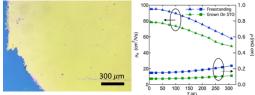


Figure 1. a) Freestanding BaSnO₃ membrane on silicon wafer, and b) mobility and resistivity of freestanding and epitaxial La-doped BaSnO₃ films.

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