Macroscopic Tin Monochalcogenide Van der Waals Ferroelectrics: Growth, Alloys, Doping, Domain Structures, and Curie Temperatures

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2D and layered van der Waals crystals present opportunities for creating new families of ferroelectrics with switchable electric polarization, elastic strain, or magnetic order at thicknesses down to the single-layer limit. So far, synthesis was limited to small (few microns) crystals, where proximity to edges affects domain patterns and severely limits the ability to fabricate complex device architectures required for accessing functionalities in van der Waals ferroelectrics. Here, we report the realization of in-plane ferroelectric few-layer crystals of the monochalcogenides tin(II) sulfide, selenide, and sulfoselenide (SnS, SnSe, $SnSe_{1-x}S_x$) whose linear dimensions exceed the current state of the art by an order of magnitude [1-3]. Such large crystals allow the investigation of ferroelectric domain patterns that are unaffected by edges and finite size effects. Analysis by in-situ transmission electron microscopy and nanobeam diffraction determines the characteristics of the ferroelectric phase across the $SnSe_{1-x}S_x$ system [1-3], provides measurements of Curie temperatures [2,4], and sheds light on symmetry breaking and the transition between the polar and the symmetric (high-T) phase [4]. Finally, we demonstrate controlled n-type doping of the Sn monochalcogenide crystals, which in pure state show invariably p-type conductivity. The combined results highlight a new class of van der Waals ferroelectrics with promise for energy conversion, information storage, and novel computing paradigms.

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