Understanding the surface reconstruction during chemical switching of ultrathin PbTiO$_3$ films from density functional theory$^1$ JUN HE, BRIAN STEPHENSON, SERGE NAKHMANSOHN, Argonne National Laboratory — First-principles calculations are used to understand the structure and energetics of the newly discovered 4×1 surface reconstruction that forms under reducing conditions during chemical switching of the ferroelectric polarization in ultrathin films of PbTiO$_3$ with SrRuO$_3$ bottom electrodes coherently strained to SrTiO$_3$ (001). Relaxed surface structures are obtained for polar films with various oxygen stoichiometries in the outermost PbO layer. To model the behavior of many-unit-cell thick films, which are observed to have polarizations near the bulk value, the lowest unit cell(s) of the PbTiO$_3$ film are forced to be polar. The observed surface reconstructions are compared with experimental synchrotron x-ray measurements.

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