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Phase inversion and frequency doubling of reflection high-energy electron diffraction intensity oscillations in the layer-by-layer growth of complex oxides¹ ZHANGWEN MAO, WEI GUO, DIANXIANG JI, TIANWEI ZHANG, CHENYI GU, CHAO TANG, ZHENGBIN GU, YUEFENG NIE*, XIAOQING PAN, National Laboratory of Solid State Microstructures, College of Engineering and Applied Sciences, Nanjing University, Nanjing 210093, China — *In situ* reflection high-energy electron diffraction (RHEED) and its intensity oscillations are extremely important for the growth of epitaxial thin films with atomic precision. The RHEED intensity oscillations of complex oxides are, however, rather complicated and a general model is still lacking. Here, we report the unusual phase inversion and frequency doubling of RHEED intensity oscillations observed in the layer-by-layer growth of SrTiO₃ using oxide molecular beam epitaxy. In contrast to the common understanding that the maximum(minimum) intensity occurs at SrO(TiO₂) termination, respectively, we found that both maximum or minimum intensities can occur at SrO, TiO₂, or even incomplete terminations depending on the incident angle of the electron beam, which raises a fundamental question if one can rely on the RHEED intensity oscillations to precisely control the growth of thin films. A general model including surface roughness and termination dependent mean inner potential qualitatively explains the observed phenomena, and provides the answer to the question how to prepare atomically and chemically precise surface/interfaces using RHEED oscillations for complex oxides.

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