Stoichiometric control and optical properties of $BaTiO_3$ thin films grown by hybrid MBE

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$BaTiO_3$ is a technologically tremendously relevant material in the perovskite oxide class with above room temperature ferroelectricity and a very large electro-optical coefficient, making it highly suitable for emerging electronic and photonic devices. [1,2] An easy, robust, straightforward, and scalable growth method is required to synthesize epitaxial $BaTiO_3$ thin films with reproducible thin film properties allowing sufficient control over the film's stoichiometry. Here, we report on the growth of $BaTiO_3$ thin films by hybrid molecular beam epitaxy. A self-regulated growth window was identified using complementing information (Fig. 1a, b), the evolution of reflection high energy electron diffraction images during film growth, the deviation of the intrinsic lattice parameter, and film surface morphology. Subsequent optical characterization of the $BaTiO_3$ films by spectroscopic ellipsometry revealed refractive index and extinction coefficient values closely resembling stoichiometric bulk $BaTiO_3$ (Fig. 1c, 1d). In addition to the expected degradation of optical properties with increasing deviation from the self-regulated growth window, an optical absorption peak emerged at 700 nm even in the absence of a detectable lattice parameter expansion of $BaTiO_3$ thin films. This feature was identified by first-principles calculation as Ba vacant lattice site. In addition, optical second harmonic generation analysis was performed to determine the polar symmetry of the films which revealed bulk-like SHG coefficients. In this talk, we will discuss how the optical properties of $BaTiO_3$ can be utilized as a much finer probe than intrinsic lattice parameter expansion to determine the stoichiometry level present in $BaTiO_3$ films. This work is supported by the US Department of Energy, Basic Energy Sciences, under Award Number DE-SC0020145 as part of the Computational Materials Sciences.

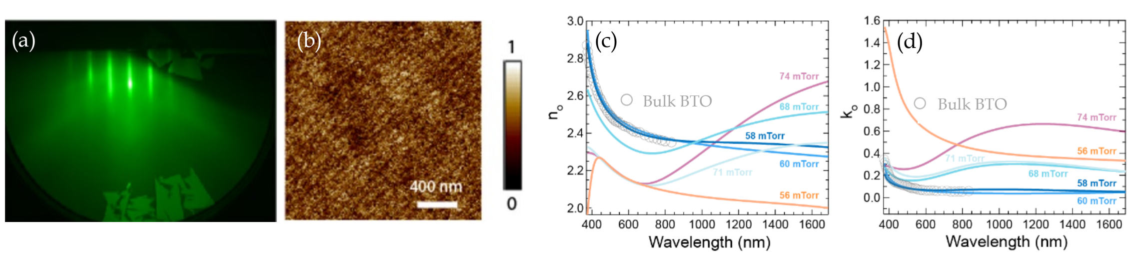

Figure 1. (a) a RHEED image taken along the [100] direction (b) an AFM micrograph of a $BaTiO_3$ film grown at 58 mTorr (c) Ordinary refractive indices, n_o, and (d) ordinary extinction coefficients, k_o, of $BaTiO_3$ thin films plotted together with bulk $BaTiO_3$

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[2] L. Mazet, *et al*., Sci. Technol. Adv. Mater. **16**, 36005 (2015)

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