Ordinary and Extraordinary Complex Refractive Index of Barium Titanate by using Mueller-Matrix Spectroscopic Ellipsometry

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Barium titanate (BTO) is one of the earliest reported lead-free ferroelectric polydomain crystal materials, first documented in the 1940s [1]. Numerous studies on BTO have been conducted to analyze and engineer its exotic physical characteristics—including ferroelectricity, pyroelectricity, piezoelectricity, and biocompatibility—which primarily originate from differences in the bonding energy of ionic bonds (Ba^{+2}/TiO^3) and covalent bonds (Ti/O) [2]. Additionally, the high dielectric constant (ε_r > 2,000) of a-axis BTO facilitates various applications, such as multilayer ceramic capacitors, ferroelectric tunnel junctions, and positive temperature coefficient thermistors. BTO exhibits a tetragonal structure (symmetry group *P4mm*) at room temperature with anisotropy between the crystallographic c-axis and the other axes. From an optical perspective, BTO shows low optical losses from the near-ultraviolet to near-infrared range, along with transparency, and the Pockels effect, an ultrafast second-order linear electro-optic effect, enables the development of diverse optical components and nonlinear optical systems. However, the absence of accurate optical constants of anisotropic BTO along ordinary and extraordinary axes due to the ferroelectric multidomain makes it difficult to utilize BTO as an active matrix of nanophotonic devices.

Standard spectroscopic ellipsometry (SE) is a widely used, efficient, and non-destructive optical measurement technique for determining materials' optical thickness and refractive index. The measurement quantities are based on the complex ratio of p- to s-polarized Fresnel reflection coefficients, corresponding to the two on-diagonal elements of the 2×2 complex-valued Jones matrix. While SE effectively quantifies the amplitude and phase ratio of completely polarized light, it struggles to describe the non-ideal polarization states of light, which can frequently arise in anisotropic materials. To overcome these limitations, Mueller-matrix (MM) SE extends the analysis by employing 4×4 real-valued Mueller matrices and 4×1 Stokes vectors, offering a comprehensive representation of polarization effects, including depolarization and cross-polarization [3]. This capability enables precise optical characterization of complex optical systems and intricate materials. In this work, we optically characterize the refractive indices, extinction coefficients, and birefringence of BTO across a broad spectral range, from the visible to the mid-infrared range, using transmission and reflection measurement modes. Due to the multidomain in *a*-axis BTO, where the c-axis lies in the sample plane but lacks macroscopic long-range order, we introduce a smearing effect to the Euler angle associated with the optic axis in our ellipsometry model. This modeling approach allows us to extract a single set of ordinary and extraordinary refractive indices consistent for both a-axis and c-axis BTO samples. Furthermore, we utilize strong interference fringes observed in the ellipsometry data, caused by the differing phase velocities of the *p*- and *s*-polarized waves propagating through the sample. These fringes enhance sensitivity to birefringence in the transparent region. We envision that our work will contribute to flourishing BTO-based nanophotonic devices such as photonics integrated circuits and active metasurfaces in various operating ranges, leveraging the physically fitted and practically verified optical characteristics of BTO.

References

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