Strong optical anisotropy in epitaxial $SrO(SrTiO_3)_n$ Ruddlesden–Popper thin layers

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Optical anisotropy is at the hearth of many essential optical devices raging from phase-matching elements or modulators[1] to more exotic devices exploiting unconventional optical modes,[2] with potential applications ranging from sensors, displays and medicine to non-linear and quantum optics.[3] Device fabrication requires anisotropic materials to be integrated compactly, in the form of thin films, with silicon based photonic circuits. Flexibility in controlling anisotropy is also required for device design. So-called optical metamaterials based on planar/multilayer nanostructuration enable strong birefringence and significant flexibility for permittivity engineering, but generally their morphology complicates their integration into devices.[4] Intrinsically anisotropic homogeneous materials are quite few in number, present a fixed anisotropy that cannot be engineered, are barely available in the form of thin films, and their elaboration processes are not compatible with the constraints of the photonics industry.[5] A homogeneous material exhibiting strong and controllable optical anisotropy, compactedly integrable as thin-film in silicon photonic platforms, is therefore still lacking.

In this contribution we will show that $SrTiO_3$ (STO) based Ruddlesden-Popper thin layers (noted STO-RP_N, general formula $Sr_{N+1}Ti_NO_{3N+1}$), grown by molecular beam epitaxy by inserting an extra SrO plane every N STO unit cells in the STO lattice, present a strong optical anisotropy, higher than the highest values reported in the literature in the UV and visible ranges. In contrast to known highly anisotropic materials, STO-RP_N anisotropy can be controlled by changing the STO-RP_N order N, which leverages flexibility to optimize the material properties depending on the targeted functionality/device design. STO-RP_N thin layers combines the advantages of optical metamaterials in terms of engineering flexibility with the integrability of homogeneous anisotropic materials, bridging the gap between both approaches. They can be epitaxially grown on Si and GaAs platforms thanks STO templates.[6] by using industry-standard growth processes. This collection of unique properties confers on these materials considerable interest for the design of novel integrated optical devices.

[1] L.H. Nicholls *et al.*, Nat. Photonics **11** 628, (2017)

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[3] X. Xu *et al.*, Nat. Photonics **16**, 698 (2022)

[4] J.D. Caldwell *et al.,* Nanophotonics **4**, 44 (2015)

[5] H. Mei *et al.*, Adv. Mater. **35**, 2303588 (2023)

[6] G. Saint-Girons *et al.,* Chem. Mater. **28**, 5347 (2016), L. Louahadj *et al.*, Appl. Phys. Lett. **103**, 212901 (2013)

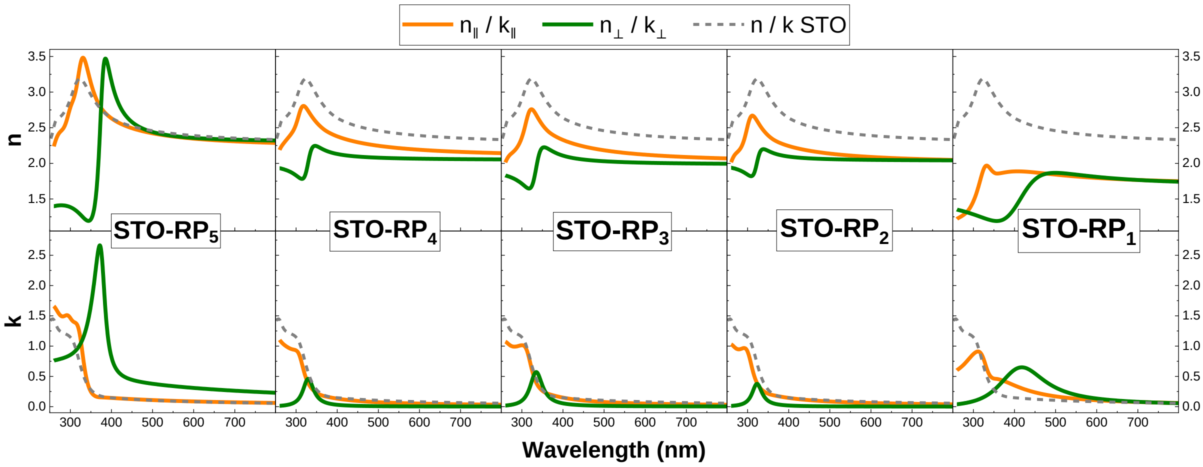

Fig. 1: Refractive indices and extinction coefficients for the five STO-RP_N samples, compared to that of STO.

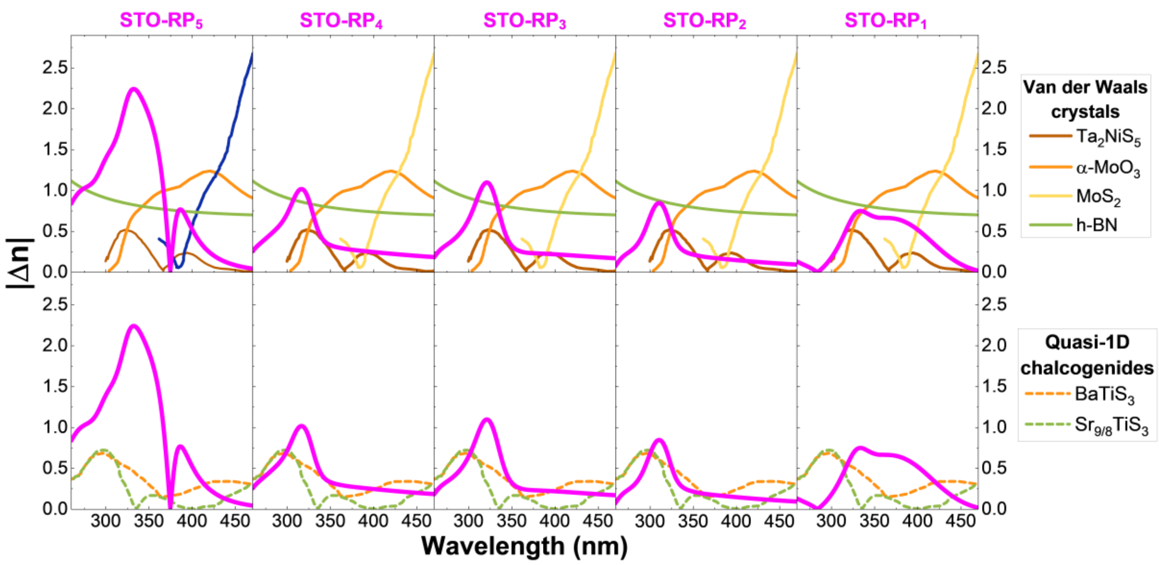

Fig. 2: Birefringence of the STO-RP_N layers (pink) compared to that of materials with the highest anistropy in the UV-visible spectral range : van der Waals crystals Ta2NiS5, α-MoO_3 , MoS_2 and h-BN and quasi-1D chalcogenides $BaTiS_3$ and $Sr_{9/8}TiS_3$.