Quasiparticle and optical band gaps of $Sr_{n+1}Ti_nO_{3n+1}$ from *ab initio* many-body perturbation theory

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The Ruddlesden-Popper (RP) homologous series $Sr_{n+1}Ti_nO_{3n+1}$ provides a unique opportunity to study the effect of dimensionality and confinement on the band gap and band edges states of the complex oxide $SrTiO_3$ [1]. The structure of the n-th member of the RP series consists of a periodic stacking of n perovskite $SrTiO_3$ monolayers separated by single SrO planes along one axis. As n becomes large, the structure converges toward bulk $SrTiO_3$. Experimental measurements have shown a decrease in the direct and indirect optical gaps, composed primarily of transitions between occupied O 2p and unoccupied Ti 3d states, as a function of n. Previous theoretical work [1] focused on the indirect gap and used density functional theory to reproduce the qualitative decrease of band gap as a function of n. In this work, we study the electronic and optical properties of selected members of the series (n=1-5 and ∞) quantitatively, calculating their direct and indirect quasiparticle band gaps using hybrid functionals and many-body perturbation theory within the GW approximation. Our GW calculations are in good agreement with measured direct optical gaps ($\Gamma \rightarrow \Gamma$), suggesting that excitonic effects are modest in these materials. Our computed indirect GW gaps ($M \rightarrow \Gamma$) overestimate reported optical gaps for small values of n but converge to experimental values at large n. Accounting for the electron-hole interaction via the Bethe-Salpeter equation approach for selected n, we compute *ab initio* optical gaps and spectra, and compare with experiment.

[1] Lee *et al.*, APL **102**, 122901 (2013).