Quasiparticle and optical band gaps of $\text{Sr}_{n+1}\text{Ti}_n\text{O}_{3n+1}$ from \textit{ab initio} many-body perturbation theory

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The Ruddlesden-Popper (RP) homologous series $\text{Sr}_{n+1}\text{Ti}_n\text{O}_{3n+1}$ provides a unique opportunity to study the effect of dimensionality and confinement on the band gap and band edges states of the complex oxide $\text{SrTiO}_3$ \footnote{Lee \textit{et al.}, APL \textbf{102}, 122901 (2013)}. The structure of the $n$-th member of the RP series consists of a periodic stacking of $n$ perovskite $\text{SrTiO}_3$ monolayers separated by single SrO planes along one axis. As $n$ becomes large, the structure converges toward bulk $\text{SrTiO}_3$. 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 \footnote{Lee \textit{et al.}, APL \textbf{102}, 122901 (2013)} 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 $\infty$) 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 \textit{ab initio} optical gaps and spectra, and compare with experiment.

\footnote{Lee \textit{et al.}, APL \textbf{102}, 122901 (2013)}