Abstract: Solid Oxide Fuel Cells (SOFCs) extract energy from fuels electrochemically, and offer a clean, low-emission, quiet, reliable, fuel adaptable, and highly efficient way to obtain power. High-performing SOFC cathodes catalyze the oxygen reduction reaction, $\text{O}_2(\text{gas}) + 2\text{e}^- \rightarrow 2\text{O}^2-(\text{solid})$, and rapidly transport oxygen through their bulk to the electrolyte. This reaction can presently only be done practically at high temperature, limiting the durability and applicability of the fuel cells. There is therefore a strong interest in developing more active catalysts for SOFCs. However, the cathodic reaction mechanism is still poorly understood, inhibiting the development of improved SOFC technologies. In this talk I will discuss how my group and collaborators are using ab initio quantum mechanical methods to better understand and design improved cathode materials. I will briefly discuss the use of ab initio methods for relevant bulk properties, like defect chemistry and diffusion, as well as predicting surface characteristics, like Sr segregation. I will then focus on the challenge of predicting and optimizing SOFC cathode activity. I consider the surface exchange rate, $k^*$, which determines how quickly oxygen can enter the surface and is closely related to the perovskite’s catalytic ability. First, I will briefly mention some past successes using the O 2p band descriptor, which we have demonstrated correlates well with $k^*$ (see Figure 1) [1]. Then I will discuss some recent progress on a fully atomistic model of $k^*$ and show that using ab initio energies we can determine the rate limiting step in the oxygen reduction reaction and predict $k^*$ with reasonable accuracy for select materials.