Electronic states in electron-doped rare-earth nickelates from first principles

Michele Kotiuga & Karin Rabe

Department of Physics and Astronomy, Rutgers University

Correlation effects in transition metal based materials give rise to many interesting and exotic properties. The rare-earth nickelates, with a rich composition-phase diagram, are no exception. Doping rare-earth nickelates can lead to electron localization, introducing defect states that are unlike typical shallow or deep donor states familiar in conventional semiconductors. We present first-principles density-functional-theory-based calculations of rare-earth nickelates, with a focus on samarium nickelate, in which we add electrons to the material. Here, we investigate doping concentrations on the order of one electron per formula unit with the goal of changing the orbital occupation and triggering a phase transition, akin to the phase control seen with strain modulation. We carry out calculations where a uniform compensating background charge (‘jellium”) has been added to maintain charge neutrality when electrons are added, as well as supercell configurations with defects that electron dope the system. In particular, we explore the effects of intercalated hydrogen and lithium as well as oxygen vacancies in samarium nickelate. In comparing these calculations, we find the jellium-background calculations capture the changes to the electronic structure seen with the explicit inclusion of defect. The resulting changes to the electronic structure, intimately linked to structural changes, cannot be understood with a rigid shift of the states: the bands are reorganized and the character of the gap is fundamentally altered. This class of doping effects introduces a new knob to turn in the field of materials design.