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**Total Energy Calculations using DFT+DMFT: Application to the Pressure-composition Phase Diagram of Rare-earth Element Nickelates**[1]

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Ab-initio total energy calculations have been implemented within the fully self-consistent density functional theory plus dynamical mean field theory (DFT+DMFT) method, using a Wannier orbital basis. The method is used to calculate the structural and metal-insulator transition phase diagrams of the rare-earth element nickelate $R\mathrm{NiO}_3$ perovskites as a function of rare-earth ion, pressure and temperature. This phase diagram is of interest because the insulating phase arises from a remarkable site-selective Mott state, in which unusual electronic physics is strongly coupled to a breathing-mode Ni-O bond disproportionation. Conventional DFT fails to stabilize the breathing distortion and thus does not reproduce the insulating phase. DFT+U overpredicts order, in particular finding that $La\mathrm{NiO}_3$ is disproportionated, in disagreement with experiment. In contrast to these theories, the DFT+DMFT method can quantitatively reproduce the metal-insulator and structural phase diagram of all $R\mathrm{NiO}_3$ perovskites in the plane of pressure and rare-earth elements. The calculated temperature dependence of the energetics of the phase transformation indicates that the thermal transition is driven by phonon entropy effects. This present method can be generally applied to nano-structured or artificially structured strongly correlated materials including heterostructures and thin films, whose electronic phases are strongly coupled to their lattice degrees of freedom.

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