Increasing our use of renewable but intermittent energy sources, such as wind and solar, requires grid-scale energy storage, such as via the electrocatalytic production of hydrogen from water. While high performance water-splitting devices and fuel cells have been developed, their high cost limits widespread use. Significant reductions in cost are expected if we can switch from using today's acid electrolytes to an alkaline electrolyte, however the activity of most electrocatalysts used to produce hydrogen is 2-3 orders of magnitude lower in an alkaline electrolyte. In this talk I will discuss how we have used a combination of density functional theory (DFT) modeling and experimental measurements on single crystals in both the electrochemical and ultra-high vacuum (UHV) environments to determine the mechanism of the alkaline hydrogen evolution and oxidation reactions. We then use this mechanism to produce a three-dimensional hydrogen activity volcano, providing clear design goals (in terms of hydrogen and hydroxide binding energy) to construct a high-performance alkaline hydrogen evolution/oxidation catalyst.