Effect of Gd doping on the structural and electronic properties of Cu3N from first principles

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Resumen

Cu_3N is an indirect band gap material, with a cubic crystal structure similar to the ABX_3 antiperovskite having a vacant body-center position, allowing for the introduction of a wide variety of impurities in the crystal lattice. In this way Cu_3N is attractive for energy conversion and storage applications, i.e., for photovoltaics as well as photoelectrochemical (PEC) CO_2 and hydrogen reduction reactions, and batteries. As a functional photocathode material [1], p-Cu_3N is of particular interest as an emerging material as it possesses a 1.8 eV bandgap, high optical absorption coefficient, and high carrier mobility. However, current growth methods result in high-density of intrinsic defects, e.g. copper interstitial defects Cu_I, causing comparably low PEC performances. In this work, we use first principles calculations to investigate the defect properties of Cu_3N, and to understand the effect of Gd doping to help overcome the defect-mediated photocarrier recombination losses. We use density functional theory (DFT) techniques and the supercell method to study point defects in the cubic Pm-3m ground state structure of Cu_3N. Formation energies and defect transition levels for Gd and K doping are computed with well-converged supercells containing up to 256 atoms. Our results are in line with previous DFT results [2, 3] showing that the lower energy maxima at 1.53 and 1.79 eV correspond to the M and R direct band gaps of bulk Cu_3N. Our computational results are supported by experimental measurements from collaborators and provide insights to enhance the PEC performance of Cu_3N films [4].

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