Ab initio study of epitaxial anatase TiO_2 on Si for efficient solar water splitting

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Crystalline metal oxides on silicon have been studied for novel applications ranging from high-k gate devices to solar energy conversion due to the interesting properties at the abrupt oxide interface. In particular, in this work, we investigate the formation of a quasi-2D electron gas (2DEG) at the interface of an epitaxial anatase TiO_2 film on Si for efficient and sustainable solar water splitting. By forming a quasi-2DEG at the interface, electrostatic fields can be induced, thereby separating photo-excited charge carriers and increasing surface reactivity. TiO_2 is known to be a stable photoelectrocatalyst in aqueous solutions. Therefore, by enhancing the charge carrier separation with a quasi-2DEG, solar water splitting can be more efficient. We use ab initio density functional theory calculations to examine the electronic structure and thermodynamic stability of various interface configurations between TiO_2 and Si. We observe the formation of a quasi-2DEG based on the atom-projected density of states, the electronic band structure and charge density. We also observe the induced electrostatic fields and the polarization of TiO_2 monolayers. We further examine the formation energy and diffusion barrier of oxygen vacancies to verify the possibility of kinetically trapped metastable states that forms the quasi-2DEG. Sr-buffered TiO_2 is also evaluated as another possible set of interface configurations.