

5th International Conference on

Theoretical, Materials and Condensed Matter Physics

November 26-28, 2018 | Los Angeles, USA

Toward rational design of new photocatalytic materials for solar fuel generation using density functional theory

Moussab Harb and Luigi Cavallo

King Abdullah University of Science and Technology, Saudi Arabia

Solar hydrogen production through the challenging photocatalytic water splitting using powder semiconductor materials still remains a promising technology due to the low cost required. Designing new potential absorber semiconducting materials used in visible-light-driven photocatalytic water splitting cells requires the appropriate determination of different components that will be assembled in the final device. In addition to the required good chemical stability in aqueous solution, high crystallinity and adequate band gap energy of the prepared material (greater than 1.23eV and near 2.0eV) to absorb a wide range of photons in the visible region, which counts for 43% of the solar spectrum, other specific intrinsic parameters directly involved in the processes must be properly tuned. They include the solar light absorption intensity, exciton binding energy, the possibility of charge carrier diffusion throughout the crystal structure to the surface and their interaction with the solution. Experimentally, previous works on semiconductors widely used in photovoltaic devices revealed that high dielectric constant is needed to obtain a good ability for exciton dissociation into free holes and electrons at room temperature. A delocalization orbital character of photogenerated charge carriers is also required to give low effective masses and help for their good transport to the surface by minimizing the electron-hole pairs recombination. Moreover, suitable valence band (VB) and conduction band (CB) edge positions with respect to water redox potentials are also needed to give the driving force to the photogenerated holes and electrons to oxidize water and to reduce H⁺. To design and characterize new materials for solar energy conversion applications, the density functional theory (DFT) has emerged as a valuable computational tool to quantify these key intrinsic parameters because of the difficulties to their direct experimental measurement. Achieving accurate DFT computations is thus particularly relevant, and this is known to be in strong correlation with the type of the exchange-correlation functional used to describe the various electron-electron interactions. In previous theoretical studies on largely utilized semi-conducting materials in photocatalytic water splitting and photovoltaics, we have shown that the intrinsic parameters mentioned above can be predicted with good accuracy using DFT along with the screened Coulomb hybrid Heyd–Scuseria–Ernzerhof (HSE06) functional. Following this robust computational protocol, we have shown suitable band edge positions for visible-light-driven overall water splitting of (Ta_{3-x}Nb_{5-5x})O₅ (x≥0.16) compounds. Besides, we have predicted interesting dielectric, charge carrier transport and redox features of (Ta_{1-x}Nb_x)ON solid solution materials (0.25≤x≤0.5) for water splitting while they revealed almost UV light absorption features due to their large predicted direct bandgaps in the 2.8-3.0eV range. Recently, we have predicted Ta_{0.75}V_{0.25}ON as a promising photocatalyst for splitting of water driven by solar light, with an adequate band gap of 2.0eV, high absorption efficiency, a static dielectric constant greater than 10, smaller hole and electron effective masses than 0.5^{m₀} along the [001] and [010] crystallographic directions respectively, binding energy of the exciton lower than 25meV, and suitable energy levels of band edges for water splitting limits. The obtained solar energy absorption and redox features of Ta_{0.75}V_{0.25}ON were clearly better than those acquired for Ta₃N₅, which is the most common semiconductor photocatalyst used in visible-light-driven water splitting. In my talk, I will first show how DFT can greatly help the experimentalists for a rational design of new photocatalytic materials for solar energy conversion by giving relevant information on key and recent examples. Secondly, I will present a deep DFT-based computational study successfully achieved recently in combination with experiment aiming to understand the nature of the trap states that are significantly decreased upon hydrogen treatment and explain the electronic origin of the charge carrier lifetime enhancement in bismuth vanadate (BiVO₄) through mild hydrogen treatment. Overall, these findings provide further insights into the interplay between defect modulation and carrier transport in metal oxides, which benefit the development of low-cost and highly-efficient solar energy conversion devices.

Biography

Moussab Harb has completed his PhD in 2008 at the age of 25 years from Light-Matter Institute (ILM) at Claude Bernard University (UCBL-France). Until 2014, he has completed several Postdoctoral research studies from Multidisciplinary Research Institute on Environment and Materials (IPREM-France), French Petroleum Institute (IFPEN-France) and KAUST Catalysis Center (KCC-KSA). He is currently a Research Scientist in Computational Physical Chemistry for solar Energy Conversion at KAUST University working on the Design of new potential and efficient 3D and 2D materials for visible-light-driven photocatalytic water-splitting and photovoltaic devices using accurate first-principles quantum calculations. He has published more than 50 papers in peer-reviewed journals and has been serving as a member of ACS, APS, AIP and MRS societies and reviewer of many relevant scientific journals.

moussab.harb@kaust.edu.sa