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Machine learning prediction of 2D perovskite photovoltaics and interaction with energetic ion implantation

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Hong-Jian Feng[a] and Ping Ma

AFFILIATIONS

School of Physics, Northwest University, Xi'an 710127, China

[a]Author to whom correspondence should be addressed: hjfeng@nwu.edu.cn and fenghongjian@126.com

ABSTRACT

Atomic-level prediction combined with machine learning (ML) and density functional theory (DFT) is carried out to accelerate the fast discovery of potential photovoltaics from the 2D perovskites. Based on the ML prediction, stability test, optical absorption, and the theoretical power conversion efficiency (PCE) evaluation, two promising photovoltaics, i.e., Sr_2VON_3 and Ba_2VON_3, are discovered with PCE as high as 30.35% and 26.03%, respectively. Cu, Ag, C, N, H, and He ion implantation are adopted to improve the photovoltaic performance of the high-efficiency and best stable perovskite Sr_2VON_3. The time-dependent DFT electronic stopping calculations for energetic ion implanted Sr_2VON_3 indicate that the excited electrons from the valence band contribute to the electron–phonon coupling, the evolution and formation of the defects, and the photovoltaic performance. This work opens the way to the high-accuracy fast discovery of the high-efficiency and environmentally stable 2D perovskites solar cells and the further engineering improvement in photovoltaic performance by ion implantation.

Organic–inorganic hybrid perovskites solar cells have reached the record power conversion efficiency (PCE) of 25.5% that has never been achieved by the traditional solar cells in such a short period since the first endeavor of the perovskite cells.[1–3] The excellent photovoltaic performance of the perovskites cells is ascribed to the optimal bandgap,[4,5] the large optical absorption associated with its strong *p-p* transition across the band edge,[6,7] the defect tolerance,[8,9] and the higher carrier mobility and diffusion length associated with the strong band dispersion near the band edge.[10] However, the large lattice constants and the vibration of the organic moieties render the decomposition of perovskites solar cells, the so-called the instability issue, which severely hinders further industrial application.[11] Thus, finding perovskite-type photovoltaic materials becomes the alternative way to solve the instability issue and the Pb-related toxic problem. For example, 32 A-site organic cations, 43 B-site divalent cations, and 4 X-site halogen ions are selected to build the 5504 perovskites prediction dataset,[12] the machine learning (ML) and density functional theory (DFT) have been combined to screen the potential perovskites solar materials. Six perovskites have been predicted with optimal bandgap and good dynamic stability. Except for the three-dimensional (3D) perovskites, the double perovskites, i.e., $A_2B^{1+}B^{3+}X_6$, have been another alternative solution to the instability and toxic problem. Im *et al.* selected 540 double perovskites materials to predict the heat of formation (ΔH_F) and the bandgap (E_g).[13] 3686 ABX_3 perovskites and 1509 $A_2B^{1+}B^{3+}X_6$ double perovskites materials in the Materials Projects (MP) are considered in a ML procedure to evaluate the formability of the 3D perovskites and double perovskites.[14]

Ruddlesden–Popper (RP) two-dimensional (2D) perovskites like $(BA)_2(MA)_2Pb_3I_{10}$ (n = 3) and $(BA)_2(MA)_3Pb_4I_{13}$ (n = 4) exhibit PCE of 12.5% with improved stability subject to light, humidity, and heat stress.[15] The 2D $(CH_3(CH_2)_3NH_3)_2(CH_3NH_3)_{n-1}Pb_nI_{3n+1}$ (n = 1, 2, 3, and 4) perovskite light absorbing layer has also been synthesized, and the corresponding solar cells show good light and humidity stability after long exposure in the environment.[16] Dion–Jacobson (DJ) 2D perovskite $(3AMP)(MA)_3Pb_4I_{13}$ has achieved the PCE of 7.32% and improved stability performance.[17] RP 2D perovskites have the general formula as $A'_2A_{n-1}B_nX_{3n+1}$, while the DJ 2D perovskites have the form $A'A_{n-1}B_nX_{3n+1}$. A′ ions in the DJ phase are divalent and those in the RP phase are monovalent; therefore, the inorganic layer in the latter case is displaced along in the plane.[18–20] The stability improvement in the 2D perovskites is attributed to the organic layer, which acts as a capping layer and suppresses the decomposition of the inorganic