Examination of the Photo-Physical Properties of Single and Multiple Perovskite Layer Two-Dimensional Hybrid Lead Halide Perovskites

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Abstract

Lead halide perovskites are being studied due to their favorable properties for light emitting and photovoltaic devices[1]. The main challenge currently limiting the application of perovskite solar cells is the long-term stability, which has thermal, photo, and moisture stability weaknesses. Two-dimensional inorganic-organic hybrid perovskites offer increased stability and higher tunability of physical properties[2-3]. However, these materials show decreased efficiency for photovoltaic devices compared to their three-dimensional versions[4]. Here we use density functional theory and excited state dynamics treated by reduced density matrix method to examine the effects that varying the thickness of the perovskite layer has on the ground state and excited state photo-physical properties of the materials. Nonadiabatic couplings were computed based on the on-the-fly approach along a molecular dynamics trajectory at ambient temperatures. Density matrix-based equation of motion for electronic degrees of freedom is used to calculate the dynamics of electronic degrees of freedom. We find that single perovskite layer models offer tunability of transition energy, where the thinner perovskite layers have a larger band gap, with band gap decreasing with thickness. Perovskite layers of all considered thickness values of perovskite layers show similar trends for non-radiative relaxation. There is an open challenged of the decreased photovoltaic efficiency for the two-dimensional hybrid perovskites, which we offer to address as follows. A combination of layers absorbing in different spectral ranges is expected to help in catching photons of a broader range of wavelength. Specifically, we take our single perovskite layer models and combine them into heterostructures where we have two perovskite layers of different thickness separated by the organic layer of our model. This heterostructure demonstrates promising performance, with the heterostructure models showing photo-physical properties that have been compared to those of the independent single perovskite layers.

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