Ab initio study of the polarization dependence of the optoelectronic properties of hybrid halide perovskites

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With efficiencies as high as 20%, hybrid organic-inorganic halide perovskites have garnered much of the photovoltaic community’s attention. In light of recent experimental results [1], we investigate the coupling mechanism between polarization and optoelectronic properties of methylammonium (MA) lead iodide, (CH$_3$NH$_3$)$_2$PbI$_3$, and related halide perovskites. In particular, we study the conditions that promote a combined effect of strong spin-orbit coupling and inversion symmetry breaking and that lead to a sizable Rashba/Dresselhaus effect. Using density functional theory calculations, we elucidate the emergence of Rashba/Dresselhaus splitting associated with local distortions and long-range coherent alignment of MA moieties in the material. We examine the extent to which the magnitude of the splitting, as well as other important electronic and optical properties [1], can be altered by increasing the macroscopic polarization. This opens avenues for manipulation of optoelectronic properties by an external electric field and/or chemical substitution of the MA molecule.