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Structural Effects on the Bandstructure of Methylammonium Lead Iodide MARCO BERNARDI, California Institute of Technology, BRADFORD A BARKER, DEREK VIGIL-FOWLER, JEFFREY B NEATON, STEVEN G LOUIE, University of California at Berkeley, LOUIE TEAM — Metal-organic halide perovskites possess peculiar physical properties. The carrier diffusion length in methylammonium lead iodide (MAPbI) exceeds 1 μm, but this unusually high value for a solution-processed material is poorly understood. We developed first-principles calculations of carrier lifetimes and diffusion lengths in semiconductors, which require accurate knowledge of the bandstructure. In this talk, we show that in MAPbI the structure strongly affects the bandstructure and band edges, and that density functional theory (DFT) is unable to predict the room temperature tetragonal structure due to the polymorphism of MAPbI. The Rashba splitting induced by the spin-orbit interaction, and the DFT band gap and effective masses, all depend strongly on the chosen structure, a point that previous work failed to address. Working with multiple stochastic realizations of large unit cells with random methylammonium orientations, we compute average effective masses and show that the effective mass depends linearly on the band gap. The average Rashba coefficient we find is an order of magnitude smaller than previously reported, and the band edges are almost parabolic. Our structures possess the correct symmetry and are free of the spurious Pb off-centering assumed in previous work. We identify the correct starting point for GW bandstructure calculations and to compute the carrier lifetime and diffusion length.

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