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Electronic structure of hybrid halide perovskite photovoltaic absorbers[1]

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The performance of organometallic perovskite solar cells has rapidly surpassed those of both traditional dye-sensitized and organic photovoltaics, e.g. solar cells based on $CH_3NH_3PbI_3$ have recently reached 18% conversion efficiency. We analyze its electronic structure and optical properties within the quasiparticle self-consistent *GW* approximation (QS*GW*). Quasiparticle self-consistency is essential for an accurate description of the band structure: bandgaps are much larger than what is predicted by the local density approximation (LDA) or *GW* based on the LDA. Several characteristics combine to make the electronic structure of this material unusual. First, there is a strong driving force for ferroelectricity, as a consequence the polar organic moiety CH_3NH_3. The moiety is only weakly coupled to the PbI_3 cage; thus it can rotate give rise to ferroelectric domains. This in turn will result in internal junctions that may aid separation of photoexcited electron and hole pairs, and may contribute to the current-voltage hysteresis found in perovskite solar cells. Second, spin orbit modifies both valence band and conduction band dispersions in a very unusual manner: both get split at the R point into two extrema nearby. This can be interpreted in terms of a large Dresselhaus term, which vanishes at R but for small excursions about R varies linearly in k. Conduction bands (Pb 6p character) and valence bands (I 5p) are affected differently; moreover the splittings vary with the orientation of the moiety. We will show how the splittings, and their dependence on the orientation of the moiety through the ferroelectric effect, have important consequences for both electronic transport and the optical properties of this material.

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