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Higher-order effects in the temperature dependence of the energy levels in Methylammonium Lead Iodide Perovskite WISSAM A. SAIDI, University of Pittsburgh, SAMUEL PONCE, University of Oxford, BARTOMEU MONSERRAT, Rutgers University — We determine temperature effects on the bandgap and band edges of $CH_3NH_3PbI_3$ from first principles by accounting for electron-phonon coupling and thermal expansion. From 290 to 380K, the computed bandgap change of 40 meV coincides with the experimental change of 30-40meV. We show that the calculation of electron-phonon coupling in $CH_3NH_3PbI_3$ is particularly intricate, as the commonly used Allen-Heine-Cardona (AHC) theory significantly overestimates the bandgap change, and excellent agreement with experiment is only obtained when high-order terms in the electron-phonon coupling are included in conjunction with spin-orbit corrections. In contrast, we find that the inclusion of nonlocal correlations using hybrid functionals has little effect. We show that the low-energy phonon modes contribute the most to the energy levels renormalization due to temperature, which suggests that the AHC failure is likely common to metal-halide perovskites and not only to hybrid ones. We have verified this on $CsPbI_3$. Our results unambiguously confirm for the first time the importance of higher-order terms in the electron-phonon coupling by directly comparing with experimental results

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