Nanoscale charge localization induced by random orientations of organic molecules in hybrid perovskite CH₃NH₃PbI₃ JIE MA, LIN- WANG WANG, Lawrence Berkeley National Lab — Perovskite-based solar cells have achieved high solar-energy conversion efficiencies and attracted wide attentions nowadays. Despite the rapid progress in solar-cell devices, many fundamental issues of the hybrid perovskites have not been fully understood. Experimentally, it is well known that in CH₃NH₃PbI₃, the organic molecules CH₃NH₃ are randomly orientated at the room temperature, but the impact of the random molecular orientation has not been investigated. Using linear-scaling ab-initio methods, we have calculated the electronic structures of the tetragonal phase of CH₃NH₃PbI₃ with randomly orientated organic molecules in large supercells up to ~ 20,000 atoms. Due to the dipole moment of the organic molecule, the random orientation creates a novel system with long-range potential fluctuations unlike alloys or other conventional disordered systems. We find that the charge densities of the conduction-band minimum and the valence-band maximum are localized separately in nanoscales due to the potential fluctuations. The charge localization causes electron-hole separation and reduces carrier recombination rates, which may contribute to the long carrier lifetime observed in experiments. We have also proposed a model to explain the charge localization.

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