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Element-specific contributions to the electronic structures of inorganic cesium lead halide perovskites revealed by resonant X-ray photoelectron spectroscopy

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Inorganic cesium lead halide perovskites (such as CsPbIBr₂) constitute an emerging class of high performance semiconducting materials, delivering promising device performances in solar cells and light emitting devices. Despite the rapid development of device optimisation, many fundamental issues remain open. One of the questions attracting huge attention is: do the constitute elements contribute equally to the electronic structures of perovskites? To address this, synchrotron-based resonant photoelectron spectroscopy (resPES) was employed to resolve the low binding energy region from -5 eV to 100 eV (valence band and shallow core levels) of a model inorganic perovskite material -CsPbIBr₂, which delivers high air stability and satisfactory solar cell performance. Taking advantage of the energy tunability provided by synchrotron, the incident X-ray energy was tuned around the absorption edges of the component elements, including Cs⁺, Pb²⁺, I⁻, and Br⁻. With the high-resolution photoelectron spectroscopy at the low binding energy region from -5 eV to 100 eV, it was found that the photoemission cross-sections at the shallow core levels of the constitute elements and valence band region follow different trends when the incident X-ray energy was tuned toward the absorption edges of different elements. Intriguingly, additional photoemission bands emerged around the valence band region when Cs⁺ was selectively excited, while the X-ray excitations around other constitute elements do not show such extra photoelectron bands. With the aid of resPES, direct experimental evidence was obtained to support that different constitute elements contribute, in a dissimilar way, to the electronic structure of inorganic cesium lead halide perovskites. This exciting work is believed to be able to experimentally verify or disapprove the proposed electronic structure models of high performance perovskite materials.

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