Quasiparticle dispersions of the structural ground state CH₃NH₃PbI₃ observed by angle-resolved photoelectron spectroscopy

Jeehong Park^{1,2}, Soon Sang Huh^{3,4}, Young Woo Choi¹, Donghee Kang^{1,2}, Minsoo Kim^{3,4}, Donghan Kim^{3,4}, Soohyung Park⁵, Hyoung Joon Choi^{1,2},

Changyoung Kim^{3,*}, Yeonjin Yi^{1,2,*}

¹ Department of Physics, Yonsei University, Seoul, Republic of Korea
²Van der Waals Materials Research Center, Yonsei University, Seoul, Republic of Korea
³Center for Correlated Electron System, Institute for Basic Science, Seoul, Republic of Korea
⁴Department of Physics and Astronomy, Seoul National University (SNU), Seoul, Republic of

Korea

⁵Advanced Analysis Center, Korea Institute of Science and Technology, Seoul 02792, Republic of Korea Email: changyoung@snu.ac.kr, yeonjin@yonsei.ac.kr

Lead halide perovskites (LHP) are a novel class of semiconducting materials which have attracted a tremendous research interest in a wide range of optoelectronic applications due to their long charge carrier lifetimes and carrier diffusion lengths despite the low-cost fabrication method. Different hypotheses, including the Rashba effects and Fröhlich large polaron formation, have been proposed to elucidate the microscopic origin of their peculiar charge carrier dynamics. Although these effects are generally accompanied by the modifications of their underlying electronic structure, the key experimental evidence has not been precisely estimated so far. Here, we investigate the electronic structure of structural ground state of CH₃NH₃PbI₃, the archetypical LHP material, studied via angle-resolved photoelectron spectroscopy (ARPES) combined with *ab initio* density functional theory (DFT) calculations. By establishing careful optimizations of sample preparations and experimental protocols, we present the pristine quality of our measurements, we discuss the possibility of the Rashba effect and Fröhlich large polaron formation in the uppermost valence band of CH₃NH₃PbI₃.