Application of X-ray and optical spectroscopy methods for verification of DFT band-structure calculations

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During lastdecades, first-principles band-structure calculations carrying out within a density functional theory (DFT) framework is among the most important scientific challenges [1]. However, the accuracy of DFT calculations in many cases depend on the choice of approaches used for exchange correlation (XC) potential. As a result, during recent years X-ray photoelectron spectroscopy (XPS) and different kinds of optical spectroscopy were found very important tool for verification of the accuracy of DFT calculations in the case of semiconductors and insulators where the use of generally employed GGA and LDA approaches for XC potential bring far underestimated energy band gaps as compared to measurements of optical absorption edges [2]. Energy positions of the valence-band fine-structure peculiarities and those associated with semi-core electrons are essentially shifted towards the Fermi energy [3].

In the current work (NCN, 2024/55/B/ST5/01411), we demonstrate peculiarities of X-ray and optical spectroscopy techniques that are well suited for verification of DFT band-structure calculations of solids, in particular complexadvanced optical quaternary chalcogenides $A^{I}_2B^{II}D^{IV}X_4$ (A^{I}= Cu, Tl; B^{II}= Cd, Hg; D^{IV}= Si, Ge, Sn; X = S, Se) and halides $CsPb(Sn)X_3$ (X = Cl, Br, I). In particular, we demonstrate advantages of combination of XPS and optical spectroscopy techniques for probing theoretical DFT data on density of states (DOSs) and the main optical properties (absorption spectrum, complex dielectric function, reflectivity coefficient, refractiveindex, extinction coefficient, optical reflectivity, electron energy-loss spectrum) of the above semiconductors.

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