Electronic structure of defects and band gap engineering in $RAlO_3$ (R = Y, La, Gd, Yb, Lu) perovskites

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Tuning of the charge trapping properties of oxide crystals via changing of their band-gap widths by cationic substitution is a promising approach in elaboration of efficient optical and scintillation materials [1, 2]. The main idea of this approach is that such substitution in oxide crystal may change its band gap and, consequently, the energy levels of defects states can change their depth with respect to the band edges. The band-gap engineering effect has been experimentally evidenced for several classes of oxide compounds. However specific electronic-level mechanisms which govern this effect in a particular crystal still remain unknown. The present work reveals such mechanisms in the case of $YAlO_3$ perovskite crystals in which Y cations are substituted with La.

The DFT-based theoretical calculations with use of the Plane-Wave Pseudopotential method were carried out in order to determine the influence of La doping on the energy positions defect levels in the crystals bandgap of $YAlO_3$. The geometry-optimized calculations were performed in the super-cell approach. The super-cells were constructed as 2x2x2 multiplication of the unit cell and comprised 160 atoms of $YAlO_3$ crystal. Several kinds of point defects and defect combinations were modeled in the super-cells: natural vacancies and vacancy complexes, interstitial oxygen defects and combinations of such interstitials with natural vacancies, cationic antisites, iso- and aliovalent cationic substitutions, as well as several other combinations of such defects in $Y_{0.75}La_{0.25}AlO_3$ mixed-cationic solid solution.

Results obtained in the calculations are compared with corresponding experimental data on the optical absorption in the VUV-UV range, luminescence spectroscopy under synchrotron radiation excitation, thermally stimulated luminescence (TSL) in 300 - 500 °C temperature range are carried out for the specially grown set of $RAlO_3$ (R = Y, La, Gd, Yb, Lu) single crystals and solid solutions.

The mechanisms of formation of TSL glow peaks of $(Y,Gd,La)AlO_3:Mn^{4+}$ crystals are discussed using the obtained computational results. The origin of traps, which form the TSL glow peaks of $YAlO_3$ crystals above room temperature, is revealed.

References

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