Title: First-principles study of high-performance scintillators: Tl-doping in $CsSrI_3$ perovskite

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Abstract: Understanding the fundamental physics of electronic behavior and emission mechanisms in scintillators is essential for optimizing their performance in practical applications including medical imaging and other fields. In this study, DFT is performed to decipher scintillating properties of pristine $CsSrI_3$ and Tl-doped $CsSrI_3$ by means of investigating the electronic and optical properties. The electronic band structure's investigation reveals that Tl doping modifies the band gap with transitioning from a direct band gap of 3.8 eV in pristine $CsSrI_3$ to indirect band gap of 3.03 eV. In addition, electronic band structures endorse that Tl act as a single-level activator like Tl-doped CsI scintillators. Besides, core-level transitions involving Tl-s states further influence the electronic and optical features by offering Auger free cross-luminescence. Moreover, the highest (theoretical limit) light yield (LY) is estimated at 132,013 photons/MeV (or 132 photons/keV) for 3 % Tl-doped $CsSrI_3$ compounds, which is much higher than the value of 52,000 to 56,000 photons/MeV (or 52–56 photons/keV) for well-established CsI:Tl scintillator. Furthermore, optical property analysis confirms that an electronic transition from the Tl-s state to the Tl-p state is becoming more pronounced with respect to doping concentration. These findings demonstrate that Tl doping may enhance the scintillator's performance by introducing additional electronic transition and radiative recombination pathways. In addition, this study will provide deeper insight into the underlying physics and guide the design of more efficient scintillation materials with Tl-activator.

Keywords: DFT; Scintillation; Tl-activator; CsSrI3; Cross-luminescence

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