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Ab-initio calculations of the temperature-dependent band gap of inorganic halide perovskites

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Inorganic metal-halide perovskites have drawn much attention as candidates for producing low cost high-performance solar cells having efficiency reported as high as 20% in single-junction architecture. Their electronic and structural properties have been the case of intense study in both experimental setups and computational simulations. Since solar cells are expected to perform outside of such laboratory conditions, it is important to have an understanding of how their performance will be affected by their surroundings. Temperature is the main parameter which governs both the crystal and the electronic structure of inorganic metal-halide perovskites. Theoretical approaches that rely on either Monte-Carlo sampling of vibrational states [1] or classical molecular dynamics [2] have been successful to some extent. However, quantum mechanical ab-initio studies that yield accurate temperature dependence of band gaps of inorganic halide perovskites are still lacking.

In this work, we perform such studies for $CsPbC_3$, $CsPbBr_3$ and $CsPbI_3$. First, we use the density functional theory (DFT) with PBEsol exchange-correlation functional to obtain a decent starting estimate at zero temperature. Such an approach has a well-known error of underestimating the band gap. To overcome this, we employ the PBE0 hybrid functional whose parameters are chosen to enforce Koopman's condition for localized defect states [3]. Second, we include the effects of electron-phonon interaction that depend on the temperature of the material using the Allen-Heine-Cardona (AHC) theory [4]. Phonon modes are obtained from density functional perturbation theory, that in the present case produces unphysical imaginary frequencies in the phonon spectrum due to the harmonic approximation. We overcome this issue by calculating the phonon spectrum using the self-consistent phonon method that accounts for the effects of anharmonicity [5]. We obtain the electronic band gaps for the cubic phase of $CsPbCl_3$, $CsPbBr_3$, and $CsPbI_3$, at temperatures of 320, 403, and 300 K, respectively, of 3.07, 2.30, and 1.70 eV. These results differ by no more than 0.25 eV from the experimental ones found in the literature.

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