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Optical gaps of F-center defects in LiF using many-body methods

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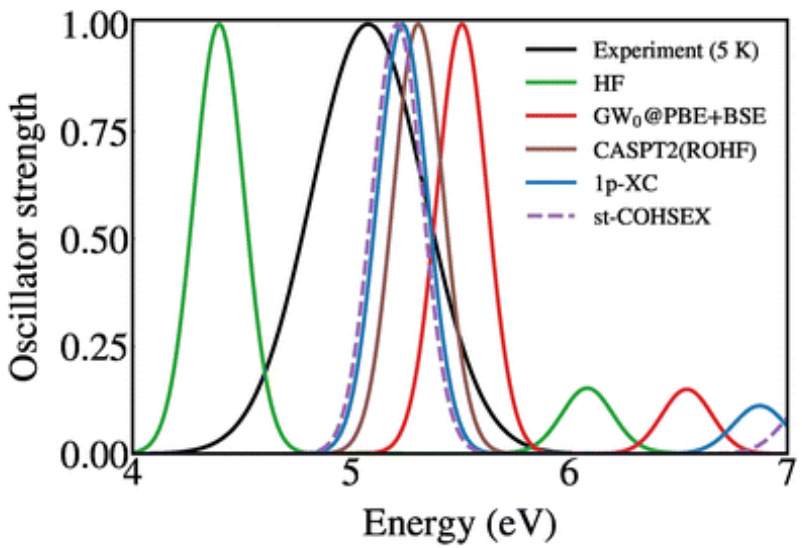

Abstract

F-center defects, occurring in technologically important wide-bandgap solids, influence the properties of the parent material. Their experimental characterization can be challenging due to low concentrations and spectral broadening. Modeling of these defects poses computational challenges due to their aperiodic nature, lattice distortion effects, and the need to describe many-body polarization effects induced by the surrounding environment. Here, we study bulk and surface F-center defects in LiF by combining random phase approximation (RPA) based potentials with periodic electrostatic embedding. We evaluate the accuracy of computationally efficient potentials such as single-pole exchange–correlation (1p-XC) and static-Coulomb hole screened exchange (st-COHSEX), and find the former to be suitable for bulk and surface F-center defects. For the commonly studied case of bulk F-centers, the 1p-XC approach predicts an optical gap (Δ_{og}) of 5.24 eV, closely matching high-level quantum chemistry methods like CASPT2 (Δ_{og} = 5.31 eV) and within 0.16 eV of the experimental absorption maximum. For surface F-centers, which are largely unexplored, we find a significantly lower optical gap of 1.85 eV. The smaller gap arises from reduced confinement effects and significantly larger lattice relaxation induced by surface F-center defects compared to the bulk. On the other hand, the polarization corrections to the optical gaps are similar for both cases.