Supplementary Material for

Strong Optical Absorption in Cubic $RaMS_3$ Chalcogenide Perovskites for Optoelectronic Applications: A First-Principles DFT Study

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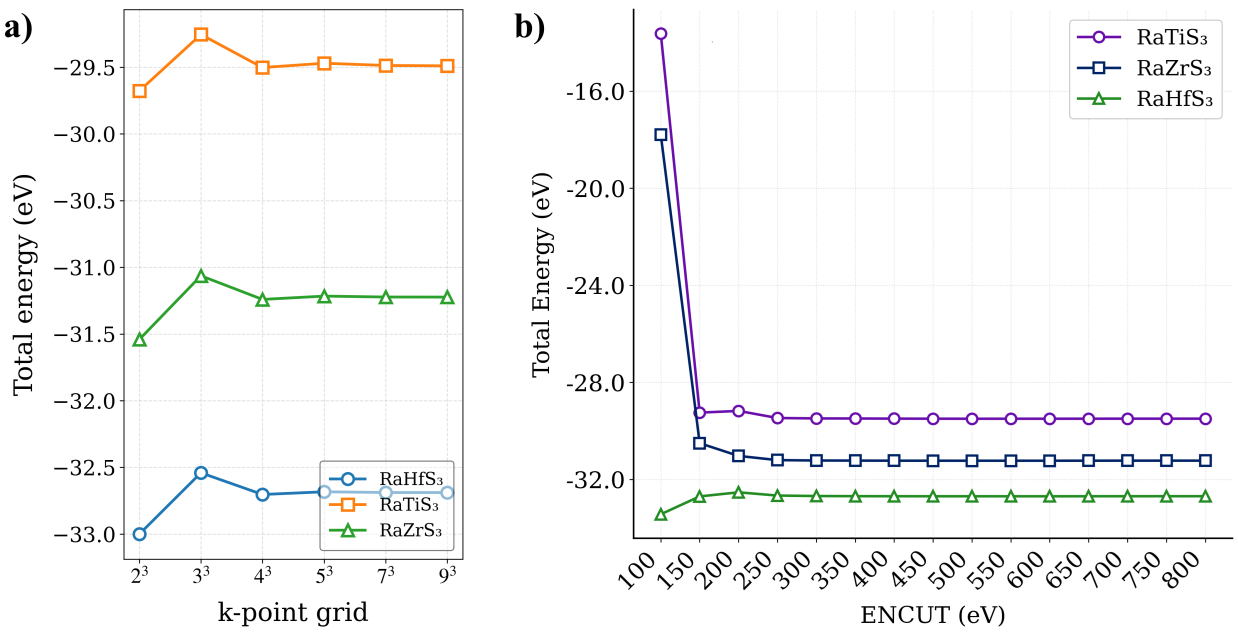

FIG. S1: Convergence tests for the ground-state calculations. (a) Total energy as a function of the k-point sampling for $RaHfS_3$, $RaTiS_3$, and $RaZrS_3$. The total energies become essentially stable for k-point meshes denser than $7 \times 7 \times 7$, indicating that the Brillouin-zone sampling is well converged. (b) Total energy convergence with respect to the plane-wave cutoff energy (ENCUT) for $RaHfS_3$. The total energy stabilizes for cutoff energies above 350 eV, which was therefore adopted for all subsequent calculations. These parameters were used consistently in both the electronic structure and optical property calculations.

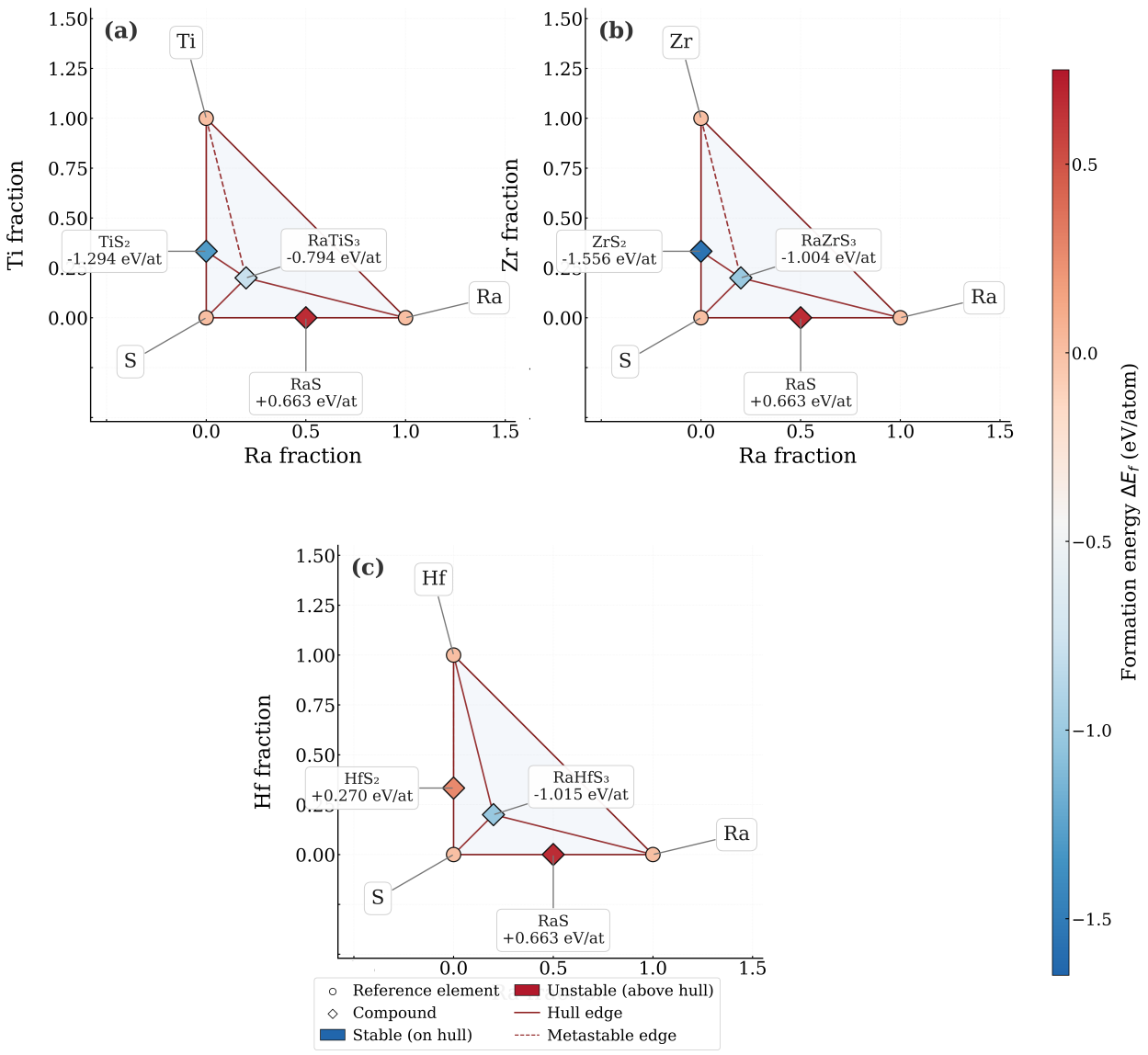

FIG. S2: Convex-hull diagrams for the Ra–M–S ternary systems (M = Ti, Zr, Hf) constructed from the calculated formation energies per atom (ΔE_f) obtained using the same elemental reference states and computational methodology. The formation energies of the ternary perovskites are $\Delta E_f = -0.794$, -1.004, and -1.015 eV/atom for $RaTiS_3$, $RaZrS_3$, and $RaHfS_3$, respectively. For comparison, the competing binary sulfides exhibit formation energies of -1.294 eV/atom (TiS_2), -1.556 eV/atom (ZrS_2), $+0.270$ eV/atom (HfS_2), and $+0.663$ eV/atom (RaS). Points lying on the convex hull correspond to thermodynamically stable phases, whereas points above the hull represent metastable configurations. The convex-hull analysis shows that $RaTiS_3$, $RaZrS_3$, and $RaHfS_3$ lie on the convex hull at 0 K, indicating thermodynamic stability with respect to decomposition into competing binary sulfide phases within the Ra–M–S chemical space.

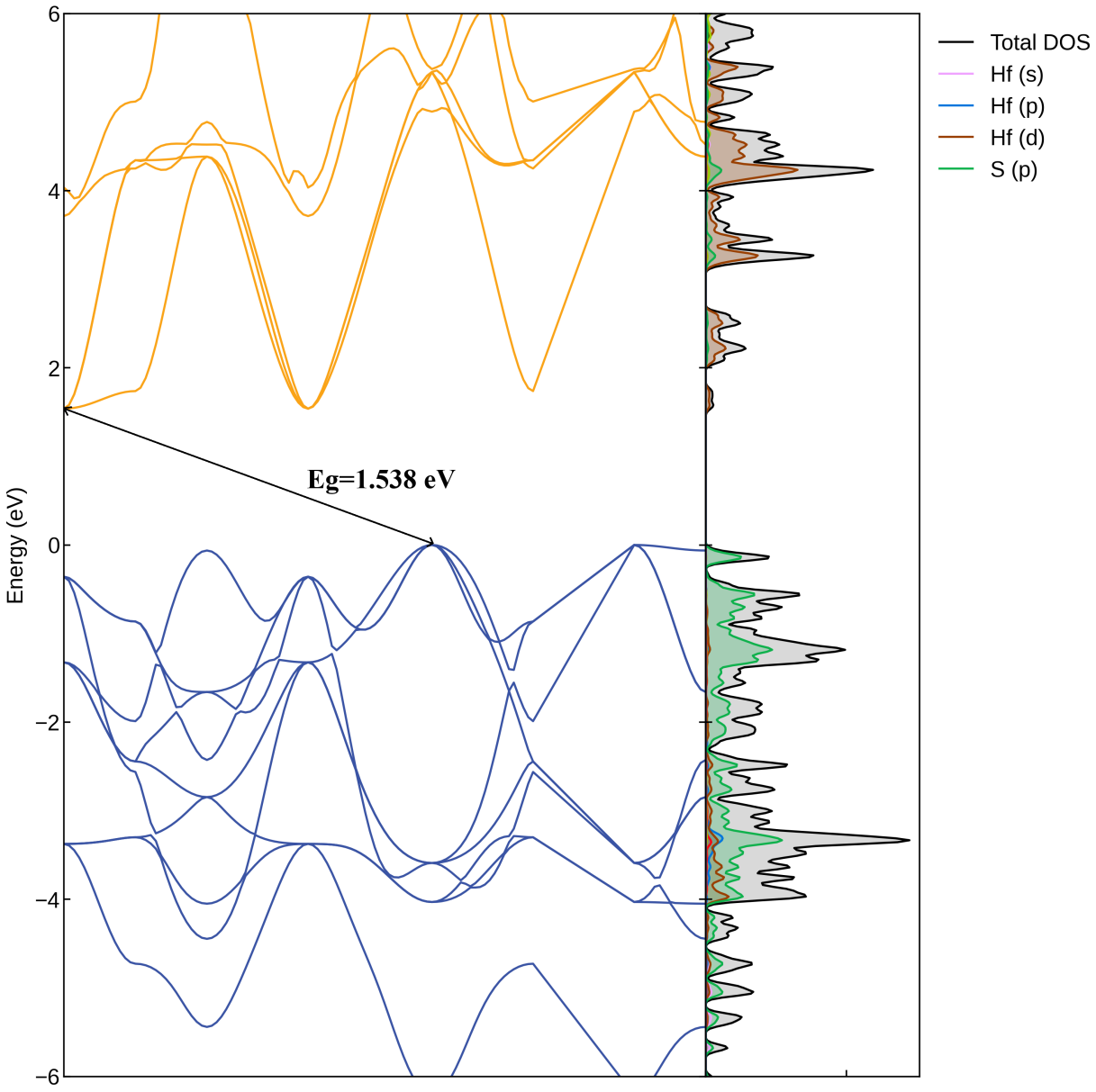

FIG. S3: Calculated electronic band structure and corresponding density of states (DOS) of $RaHfS_3$ obtained from HSE06 calculations incorporating spin–orbit coupling (SOC), with the Fermi level set to zero. The valence band is predominantly composed of S-derived states, whereas the conduction band is mainly contributed by Hf orbitals, while Ra atoms provide a negligible contribution to the electronic states near the Fermi level. Inclusion of SOC preserves the overall orbital character of the electronic states and does not significantly modify the semiconducting nature of $RaHfS_3$. The resulting band gap is 1.538 eV, which is very close to the non-SOC value of 1.546 eV, indicating a minimal SOC-induced effect on the band gap.

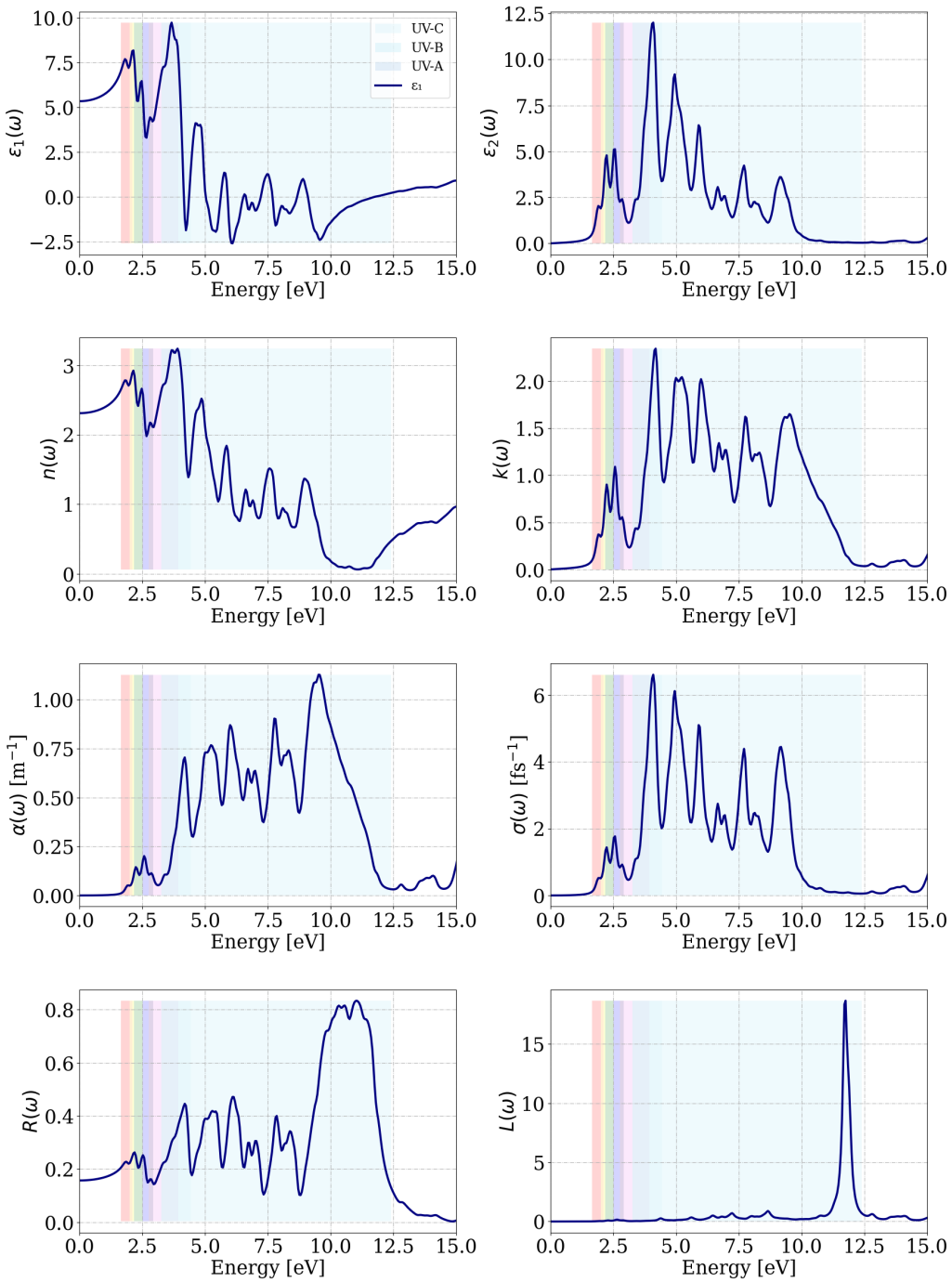

FIG. S4: Calculated optical properties of $RaHfS_3$ as a function of photon energy obtained with spin–orbit coupling (SOC) included. (a) Real part of the dielectric function, (b) imaginary part of the dielectric function, (c) refractive index, (d) extinction coefficient, (e) absorption coefficient, (f) optical conductivity, (g) reflectivity, and (h) electron energy loss function. The inclusion of SOC does not lead to noticeable changes in the spectral features or overall trends of the optical response, indicating that the optical properties of $RaHfS_3$ remain essentially the same as in the non-SOC case.