further enhance transition strength. From these considerations, we identified $CuTaS_3$ as a potentially strong absorber and set out to assess its optical and electronic properties computationally and then to confirm them experimentally.

$CuTaS_3$ also brings advantages of low toxicity,[19] material abundance,[20] and low cost.[21] Prior reports have focused only on its preparation and crystal structure.[1,2]

. All calculations were performed via density functional theory (DFT) and the plane-wave projector-augmented wave (PAW) method,[22,23] as implemented in the VASP code.[24] The atomic positions were relaxed by the Heyd–Scuseria–Ernzerh hybrid functional (HSE06)[25] until all atomic forces were less than 0.02 eV $Å^{-1}$, with lattice parameters fixed to experimental values. The band structure and optical absorption coefficients were calculated with the many-body GW approximation[26] perturbatively on the top of wave functions and energy eigenvalues

R–Γ–Y represents dispersion along [010], i.e., the direction of the one-dimensional Cu- and Ta-centered chains where orbital overlap is significant.

Figure 3 shows total and projected densities of states. A mixture of Cu 3 and S 3 orbitals characterizes the valence band, while Ta 5 dominates the conduction band. Hence, the electronic structure fulfills the design principle for states from two different elements, each prevailing near VBM and CBM. The spikes in the densities of states reflect the flat-band portions of the band diagram and one-dimensional character of the crystal structure. Based on the total number of electrons available, the valence band is filled, the conduction band empty. Consequently, atomic charges are formally Cu^{1+}, Ta^{5+}, and S^{2-}, consistent with a Cu ^{10} valence band and a Ta ^{0} conduction band. In contrast, Sunshine and Ibers[2] proposed that the valence states were likely to be $Cu^{2+}Ta^{4+}S_3$, similar to $Cu_{0.8}Ta_2S_6$. They accounted for the low conductivity by assuming the unpaired electrons of Cu and Ta are sequestered in a strong Cu–Ta bond. Recent X-ray photoelectron spectroscopy (XPS)[1] and magnetic susceptibility[31] results, however, are consistent with our DFT calculations and assignment of formal oxidation states: $Cu^{1+}Ta^{5+}S_3$.

3. We evaluated optical properties by analysis of ground single crystals and thin films. Figure 4 shows a diffuse reflection spectrum of the ground crystals, revealing a band gap near 1 eV.

XRD analysis confirmed deposition of a crystalline $CuTaS_3$ film (Figure S1). The band gap and absorption coefficients were assessed from UV–vis–NIR transmittance (T) and reflectance (R) spectra. Absorption coefficients were calculated with eq 1,[32] researchers did not explicitly describe their contacts.[2] We did not measure the resistivity, ρ_{ac}, in the ac plane. The resistivity of pellets, ρ_p = 0.4 MΩ cm, prepared from ground crystals, however, is higher than ρ_b. ρ_p represents and average of ρ_b and ρ_{ac}. ρ_{ac} is anticipated to be higher than ρ_b based on the anisotropic nature of the crystal structure (Figure 1) and the band dispersion (Figure 3). Significant dispersion along [010] contributes to low ρ in the single crystal, while in pressed pellets, contributions from the flat, off-axis dispersion in the ac plane raises ρ.

The negative Seebeck coefficient, −1650 μV K^{-1}, of the pellets indicates electrons are majority carriers. This n-type behavior likely arises from sulfur vacancies or Ta-on-Cu antisites (Ta_{Cu}) defects, which place excess electrons in the Ta cry4-672m7.1ri3(s1Tf21.63390T0891650)Tjp1Tf.45920TD3