

Electronic and Optical Properties of Lanthanide-Doped MoS₂: Impact of Ionic Size and Orbital Configuration Mismatch

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Single-photon emitters (SPEs) are crucial for quantum technologies such as quantum simulation [1], secure quantum communication [2], and precision measurements [3]. Two-dimensional transition-metal dichalcogenides (TMDCs) provide an attractive platform for SPEs due to their atomically thin structure, high extraction efficiency, and compatibility with chip-based photonic devices [4]. However, conventional TMDC SPEs emit mainly in the visible range, which limits their use for telecommunication applications that require infrared wavelengths. Lanthanide doping in TMDCs, such as MoS₂, offers a potential solution by introducing sharp, *f* orbital-derived emissions in the infrared range. Yet, the feasibility and impacts of introducing these dopants remain uncertain due to the large ionic radii of the lanthanides.

In this context, we employ density functional theory calculations to investigate the structural, electronic, and optical impact of lanthanide-doped MoS₂ monolayers (Ln=Ce, Er). By evaluating formation energies with adjacent S vacancies, we assess that sulfur vacancies adjacent to Ln sites play a key role in mitigating lattice strain, enabling thermodynamically stable lanthanide incorporation. Charge-state and band structure analysis reveal that *f* orbital-derived defect states and additional host-related states emerge near the band gap, originating from the mismatch of the orbital configuration between the dopant and the host lattice (Fig. 1(a)). Furthermore, optical absorption analysis reveals multiple defect- and *f* orbital-related transitions within the band gap range of the host material (Fig. 1(b)). Notably, ErMo₂ exhibits sharp, weak *f-f* optical transitions (0.9-1.1 eV), suggesting the feasibility of defect engineering for SPE. In contrast, CeMo₂ shows only defect-related absorption due to its empty *f* shell.

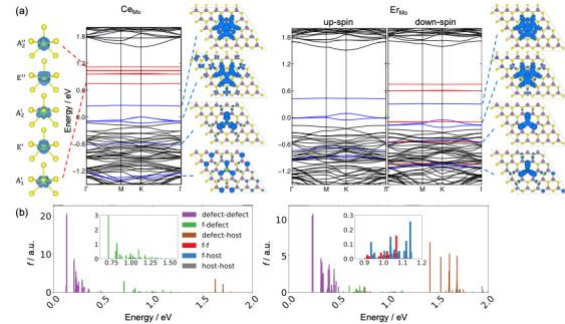


Figure 1. (a) Band Structures and charge distributions of *f* orbitals (red bands) and defect states (blue bands) of CeMo₂ and ErMo₂. (b) Optical absorption coefficients of CeMo₂ and ErMo₂ projected along the in-plane (xy) direction. Each peak shows the oscillator strengths of individual transitions computed using TDDFT.

- [1] A. Aspuru-Guzik and P. Walther, Nat. Phys. **8**, (2012).
- [2] H.-K. Lo, M. Curty, and K. Tamaki, Nat. Photon. **8**, (2014).
- [3] V. Giovannetti, S. Lloyd, and L. Maccone, Nat. Photon. **5**, (2011).
- [4] C. Palacios-Berraquero, D. Kara, A. Montblanch, M. Barbone, P. Latawiec, D. Yoon, A. Ott, M. Loncar, A. Ferrari, and M. Atatüre, Nat. Commun. **8**, (2017).