Density functional theory and quantum embedding studies of Er^{3+} in WS_2

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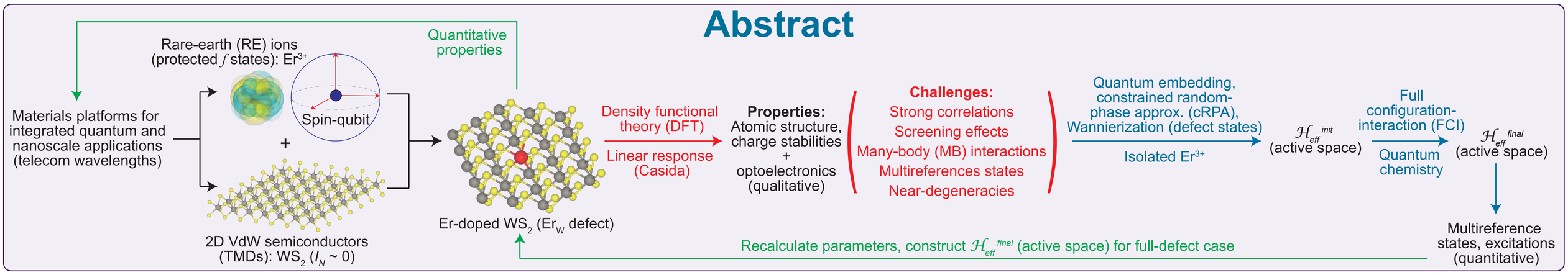
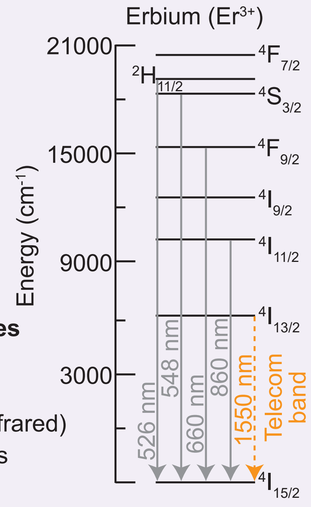
Abstract

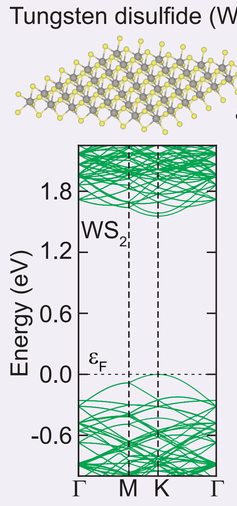


Motivation

RE ions:
- ► Protected 4f states
- ► Spin-selective transitions
- ► Long coherences, narrow linewidths
- ► Transitions insensitive to host
- ► Access to nuclear memories

Transition metal dichalcogenides (TMDs):
- ► Accessible as 2D (monolayer)
- ► Wide bandgap (visible, near-infrared)
- ► Easily integrated with photonics
- ► Large in-plane lattice constant



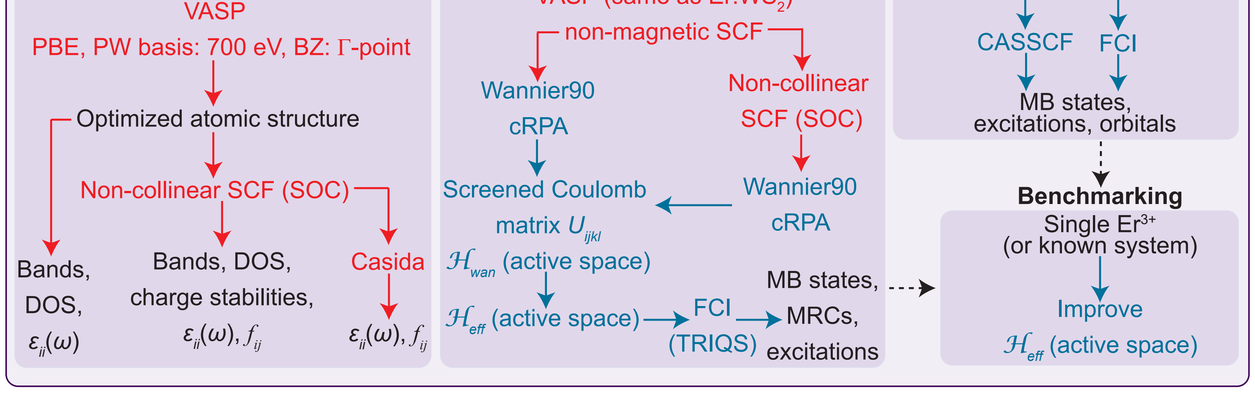


Er^{3+}:
- ► Transition within telecom band
- ► ms-long electron-spin coherence

WS_2:
- ► Bandgap ~2.1 eV
- ► $I_N \sim 0 \rightarrow \mathcal{H}_{HF} \sim 0$

Hypothesis ($Er:WS_2$):
$^4I_{13/2} \leftrightarrow {}^4I_{15/2}$ within WS_2

Methods



Results (ongoing work)

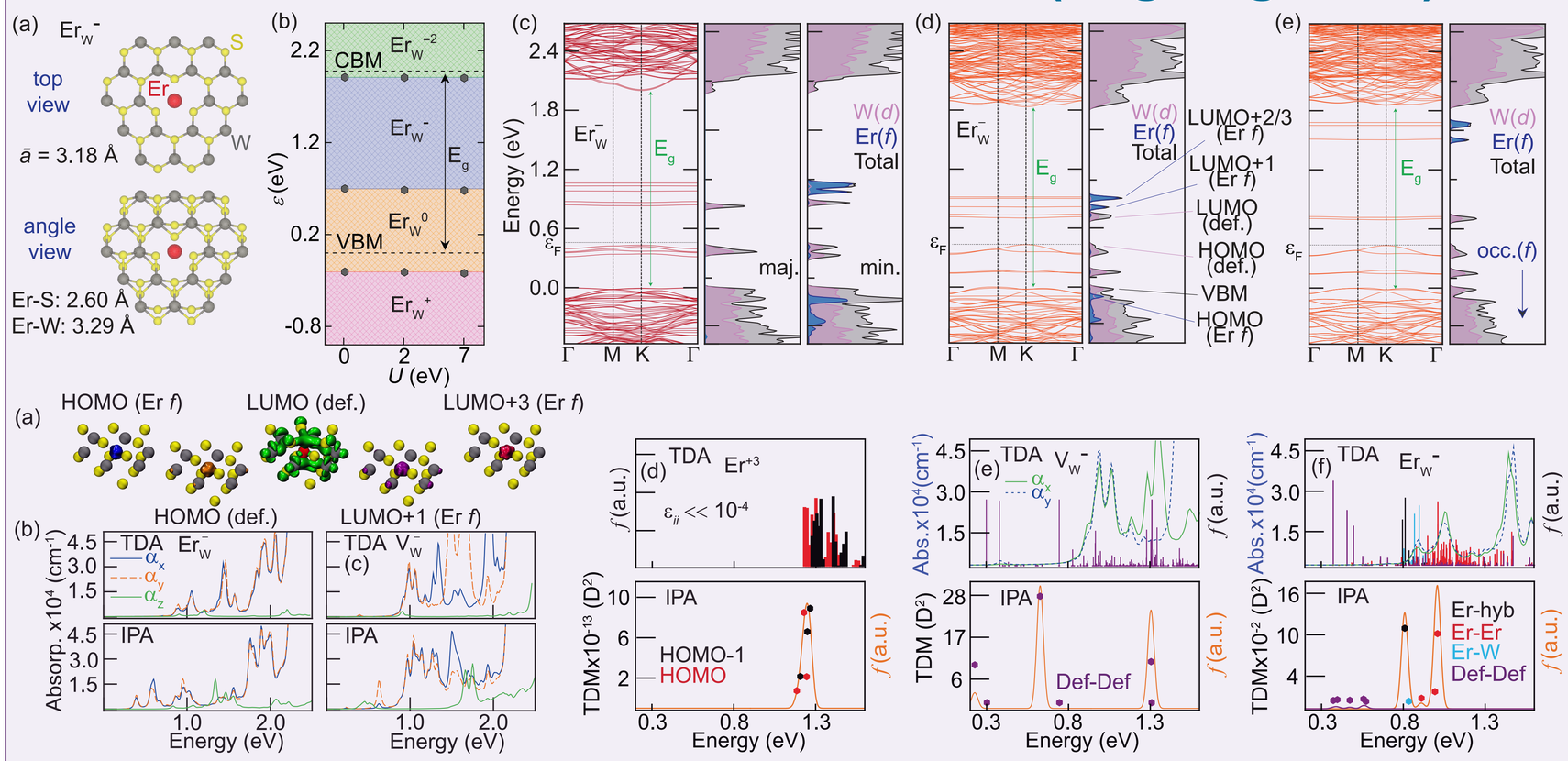


Figure 1. Summarized DFT results for Er^{3+}, Er_W and V_W in WS_2. **TOP**: (a) atomic structure for Er_W^-, (b) charge-state transition energies for Er_W, band structure (c) without SOC, (d) including SOC, (e) including SOC within DFT+U (U = 2.5 eV). **BOTTOM**: (a) charge density of relevant defect states (as labeled), absorption spectra at two levels of approximation for (b) Er_W^-, (c) V_W^-, transition dipole moment and oscillator strengths for (d) isolated Er^{3+}, (e) V_W^-, (f) Er_W^-.

Embedding (VASP via W90) vs BAGEL

Non-magnetic ΔE (FCI)	Non-magnetic MRC	After SOC ΔE (FCI)	After SOC MRC	CASSCF (ROHF) MB Occup.	CASSCF (ROHF) ΔE (FCI)
1.046	2.102	1.602	1.421	1.341	1.184
0.993	1.380	1.423	1.543	1.436	0.844
0.904	1.155	1.301	1.395	1.478	0.793
0.819	1.355	0.989	1.284	1.590	0.769
0.789	1.625	0.800	0.986	1.630	0.506
0.436	1.066	0.253	1.128	1.676	0.338
0.000	1.181	0.000	0.531	1.847	0.000

Figure 2. Embedding/quantum chemistry results for Er^{3+}. **LEFT**: (spinless) orbitals contained within active space, as obtained from W90 within VASP. Excitation energies and multireference character of MB states obatained from $\mathcal{H}_{eff}$ through FCI. **RIGHT**: orbitals and MB occupations in the active space from constrained active space self-consistent field (CASSCF) and excitation energies via FCI, both within BAGEL.

References

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Summary / Outlook

- ► Excitation energies for Er^{3+} in good agreement with experiment from FCI via $\mathcal{H}_{eff}$
- ► $\mathcal{H}_{eff}$ could be further improve by explicit inclusion of SOC terms (not from U_{ijkl})
- ► Methods to be extended for the full $Er:WS_2$ case to calculate excitation energies; MB states
- ► Quantum embedding may offer a promising description of RE impurities in semiconductors

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