Defects in materials

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Outline

- Motivation.
- Computational methods.
- Defects in oxides.
  - Why are defects challenging?
  - Defects in high-κ dielectric – HfO_2.
  - Defects in technologically important material – TiO_2.
  - Defects for quantum computing.

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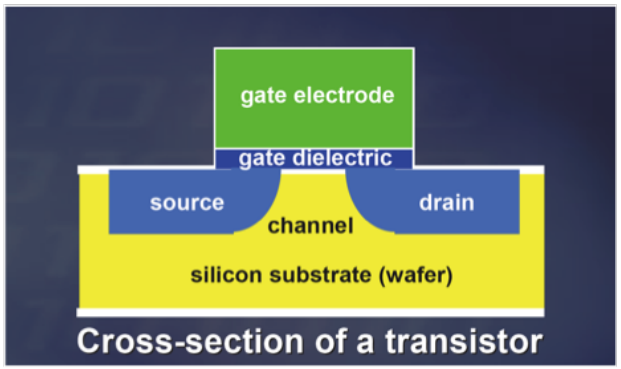
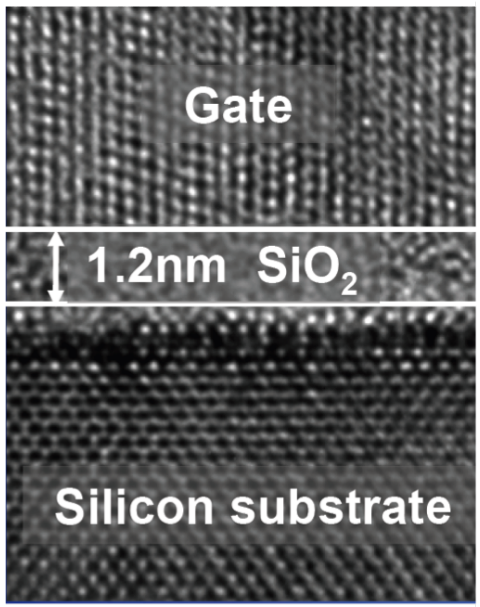
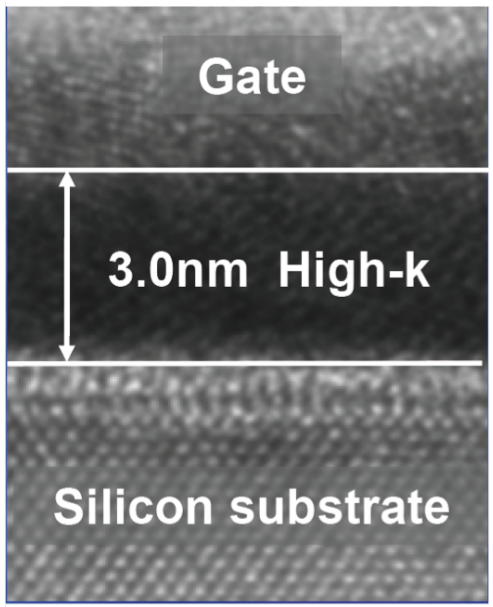
  - Defects for quantum computing.

Motivation – High-κ dielectric material HfO_2

- Moore's law about size of transistor.
- HfO_2 has replaced SiO_2 in todays MOSFET devices.







$$\kappa_{SiO_2} \approx 3.9$$

$$\kappa_{HfO_2} \approx 16 - 17$$

[1] http://www.intel.com/technology/45nm/index.htm

High-κ dielectric materials – Motivation

- High defect density.
- Charge trapping by defects in the interfacial layer or oxide – threshold voltage instability[1].
- Oxygen-related defects – vacancy[2] (V_O) or interstitials (I_O) major cause.
- Study the stability of point defects – V_O.

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[1] G. Ribes et. al., IEEE Trans. Device Mater. Reliab. **5**, 5 (2005).
[2] K. Shiraishi et. al., Jpn. J. Appl. Phys., Part 2 **43** L1413 (2004); H. Park et. al., IEEE Electron Device Lett. **29**, 54 (2008).

Motivation – TiO_2

- Photovoltaic material in solar cells[1].
- Photocatalytic material – hydrolysis of water[2].
- Memresistive switches for non-volatile memory[3].
- Oxygen vacancies are believed to play a very important role in all these applications.

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[1] B. O'Regan and M. Gr̈atzel, Nature (London) **353**, 737 (1991).
[2] A. Fujishima and K. Honda, Nature (London) **238**, 37 (1972).
[3] J. J. Yang, M. D. Pickett, X. Li, D. A. A. Ohlberg, D. R. Stewart, and R. S. Williams, Nature Nanotechnology **3**, 429 (2008).

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  - Defects for quantum computing.

Methods – Ground-state properties

Properties that are intrinsic to a system with all its electrons in equilibrium.

- Density functional theory is the "standard model" for understanding ground-state properties.
- Total energy is a functional of the charge density.
- Kohn-Sham formulation: Map the interacting many-electron problem to non-interacting electrons moving in a self-consistent field.

$$\left(-\frac{\nabla^2}{2} + \mathrm{V}_{\mathrm{ionic}}(\mathbf{r}) + \mathrm{V}_{\mathrm{Hartree}}(\mathbf{r}) + \mathrm{V}_{\mathrm{xc}}(\mathbf{r}) \right) \psi(\mathbf{r}) = \epsilon\, \psi(\mathbf{r})$$

Local density approximation
Generalized gradient approximation

Methods – Excited-state properties

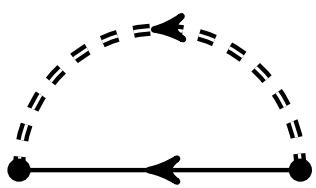
Spectroscopic properties that involve experiments creating an excited particle above the ground state.

- Concept and formalism of the interacting particle Green's function (G).
- Many-body perturbation theory is the "standard model" for understanding excited-state properties.

$$\mathrm{G}^{-1} = \mathrm{G}_0^{-1} + \Sigma$$

- GW approximation to the self-energy (Σ).

$$\Sigma = \quad = i\mathrm{GW}$$



$$\mathrm{W} = \varepsilon^{-1} v$$

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Why are defects challenging?

- Potentially strong electron-electron correlations.
  - Can be open-shell systems.
  - Multiple localized, interacting electrons.
- Lattice relaxation effects.
- Screening from the host.
  - Mimicking the system by isolated cluster may be incorrect.
- Experiments often involve excited-state properties (deep level transient spectroscopy or optical absorption etc.)
- Computational difficulty – isolated defect.

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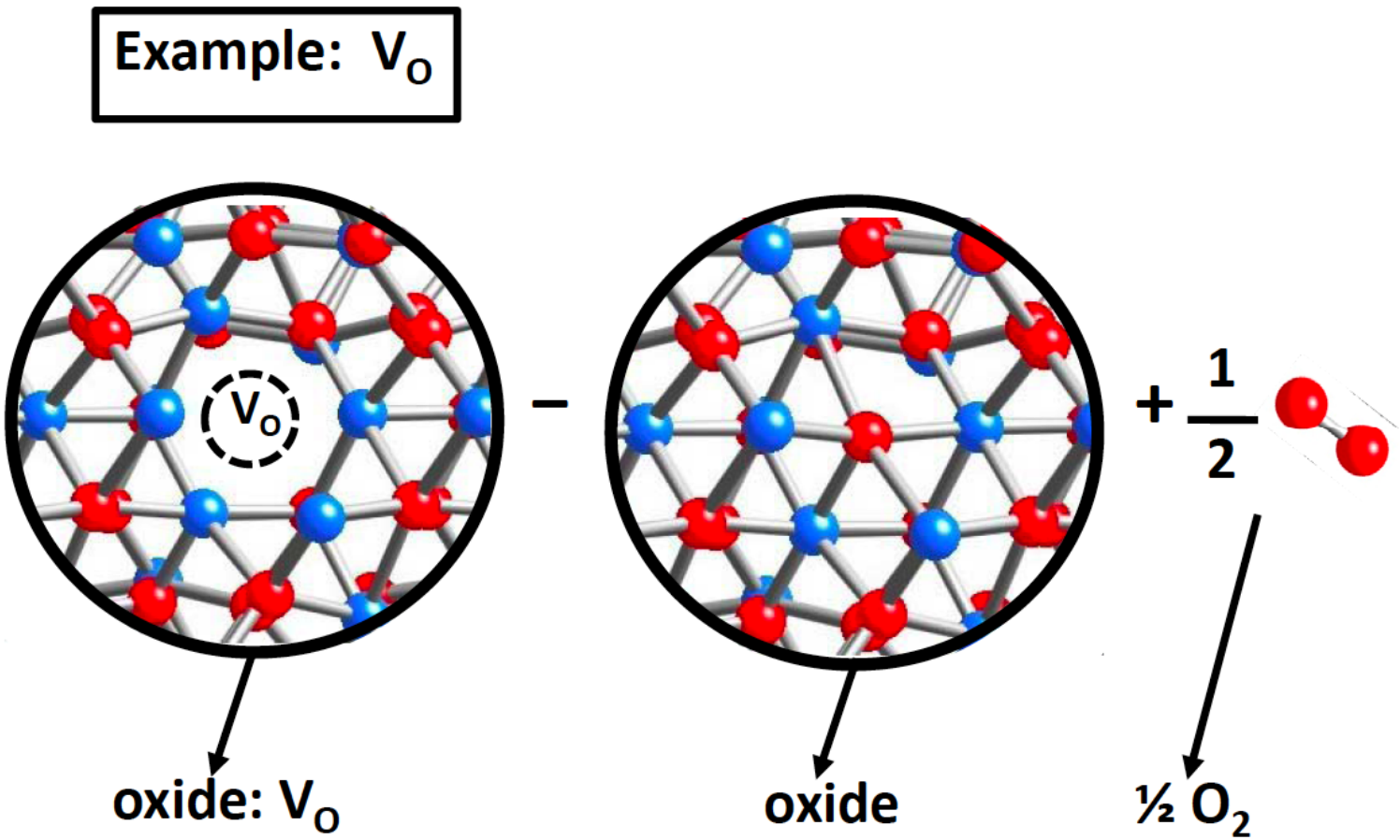
High-κ dielectric materials – Definitions

Formation energy of an oxygen-vacancy:

$$\mathrm{E}^f[\vec{\mathrm{R}}] = \mathrm{E}[\vec{\mathrm{R}}] - \mathrm{E}_{\mathrm{ref}} + \frac{1}{2}\mathrm{E}_{\mathrm{O_2}}$$

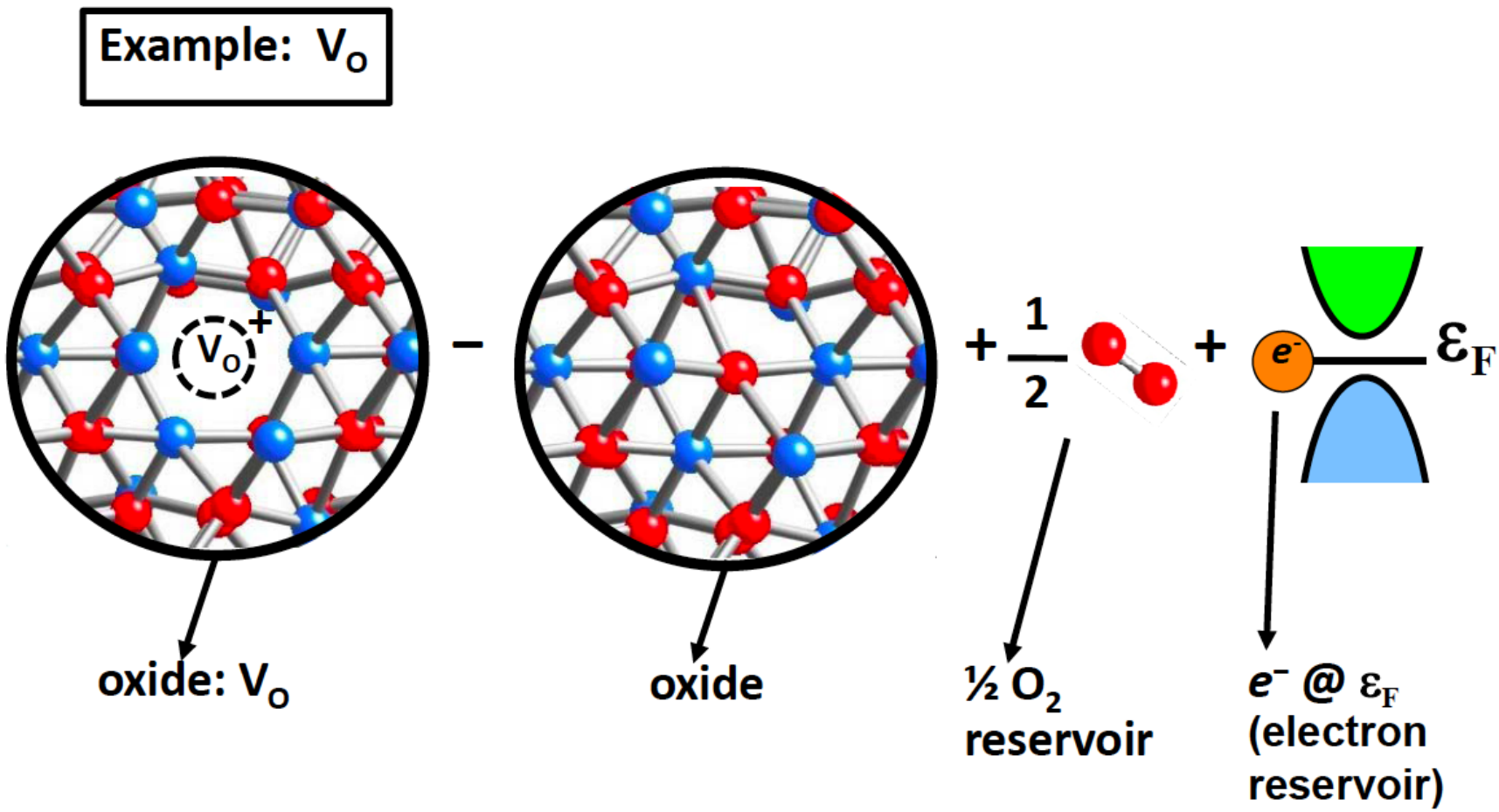


From slides by C. G. Van de Walle

High-κ dielectric materials – Definitions

Formation energy of an oxygen-vacancy:

$$\mathrm{E}_{\mathrm{q}}^{f}[\vec{\mathrm{R}}_{\mathrm{q}}](\mathrm{E}_{\mathrm{F}}) = \mathrm{E}_{\mathrm{q}}[\vec{\mathrm{R}}_{\mathrm{q}}] - \mathrm{E}_{\mathrm{ref}} + \frac{1}{2}\mathrm{E}_{\mathrm{O_2}} + \mathrm{q}(\mathrm{E}_{\mathrm{F}} + \mathrm{E}_{\mathrm{v}})$$



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High-κ dielectric materials – Definitions

Formation energy of an oxygen-related defect in hafnia :

$$\mathrm{E}_\mathrm{q}^f[\vec{\mathrm{R}}_\mathrm{q}](\mathrm{E}_\mathrm{F}) = \mathrm{E}_\mathrm{q}[\vec{\mathrm{R}}_\mathrm{q}] - \mathrm{E}_{\mathrm{HfO_2}} - \mathrm{n}_\mathrm{O}\mu_\mathrm{O} + \mathrm{q}(\mathrm{E}_\mathrm{F} + \mathrm{E}_\mathrm{v})$$

where μ_O is the oxygen chemical potential.

High-κ dielectric materials – Definitions

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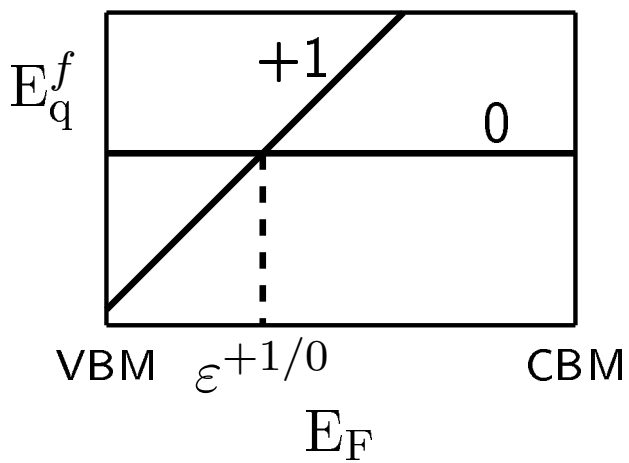
where μ_O is the oxygen chemical potential.

Charge transition level : $\varepsilon^{\mathrm{q/q\text{-}1}}$ = Fermi energy where defect $\mathrm{q} \to \mathrm{q} - 1$.



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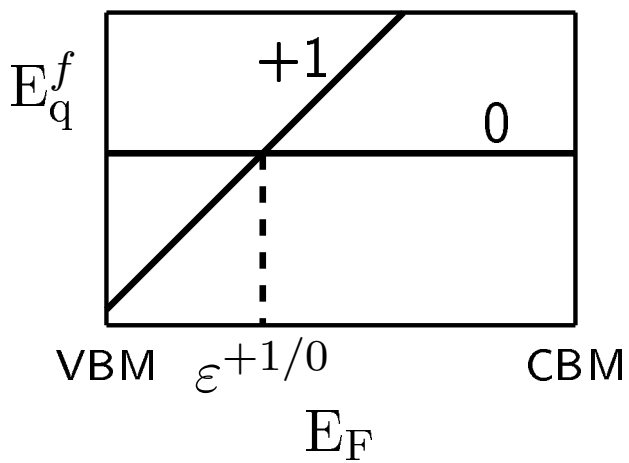
where μ_{O} is the oxygen chemical potential.

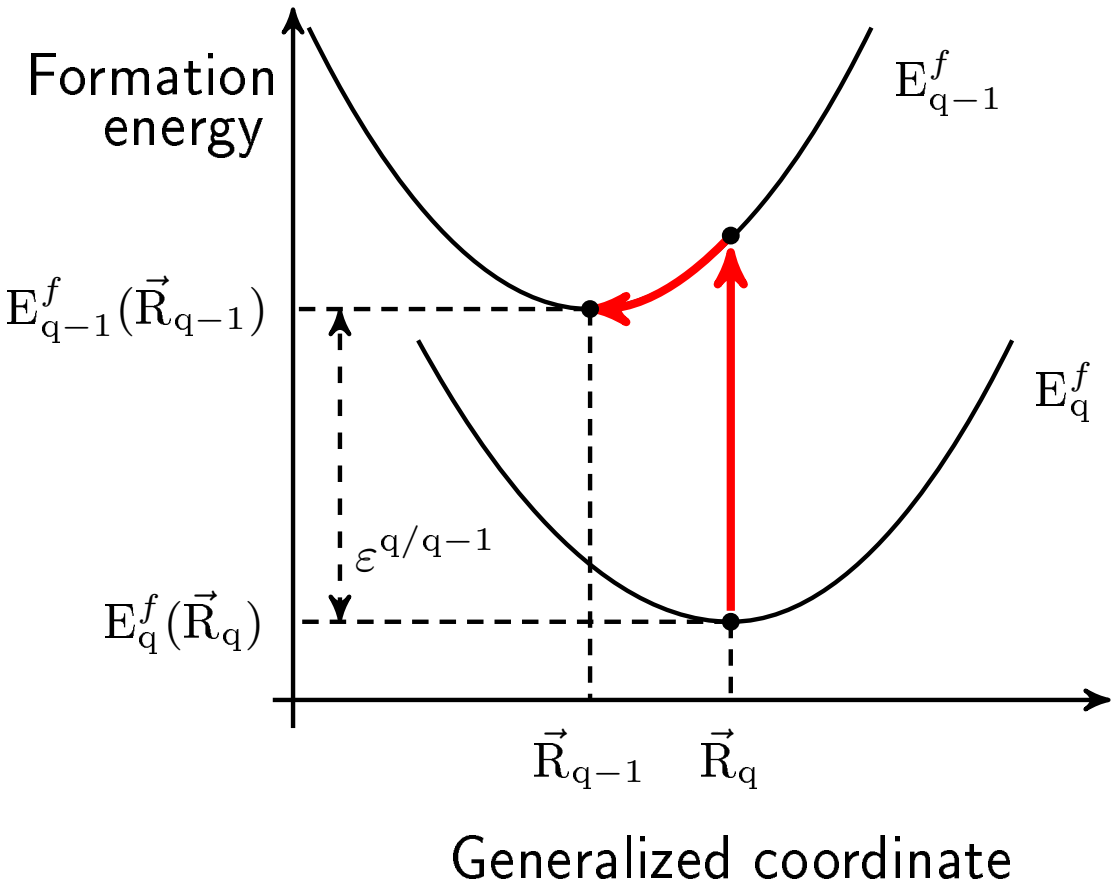
Charge transition level : $\varepsilon^{\mathrm{q/q\text{-}1}}$ = Fermi energy where defect $\mathrm{q} \rightarrow \mathrm{q}-1$.

$$= \mathrm{E}_{\mathrm{q\text{-}1}}^{f}[\vec{\mathrm{R}}_{\mathrm{q\text{-}1}}](\mathrm{E}_{\mathrm{F}} = 0) - \mathrm{E}_{\mathrm{q}}^{f}[\vec{\mathrm{R}}_{\mathrm{q}}](\mathrm{E}_{\mathrm{F}} = 0)$$



High-κ dielectric materials – Methodology

DFT + GW methodology.



M. Hedström, A. Schindlmayr, G. Schwarz, and M. Scheffler, Phys. Rev. Lett. **97**, 226401 (2006); P. Rinke, A. Janotti, M. Scheffler, and C. G. Van de Walle, Phys. Rev. Lett. **102**, 026402 (2009); M. Jain, J. R. Chelikowsky and S. G. Louie, Phys. Rev. Lett. **107**, 216803 (2011).

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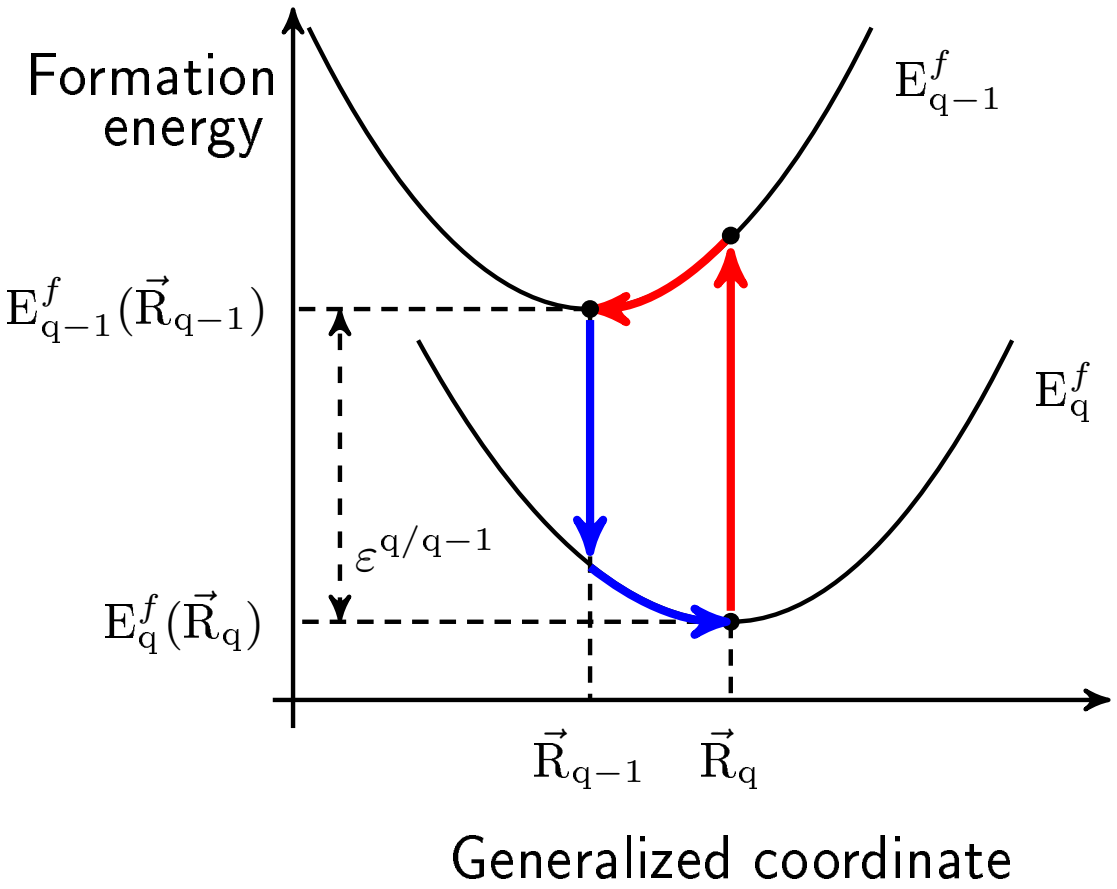

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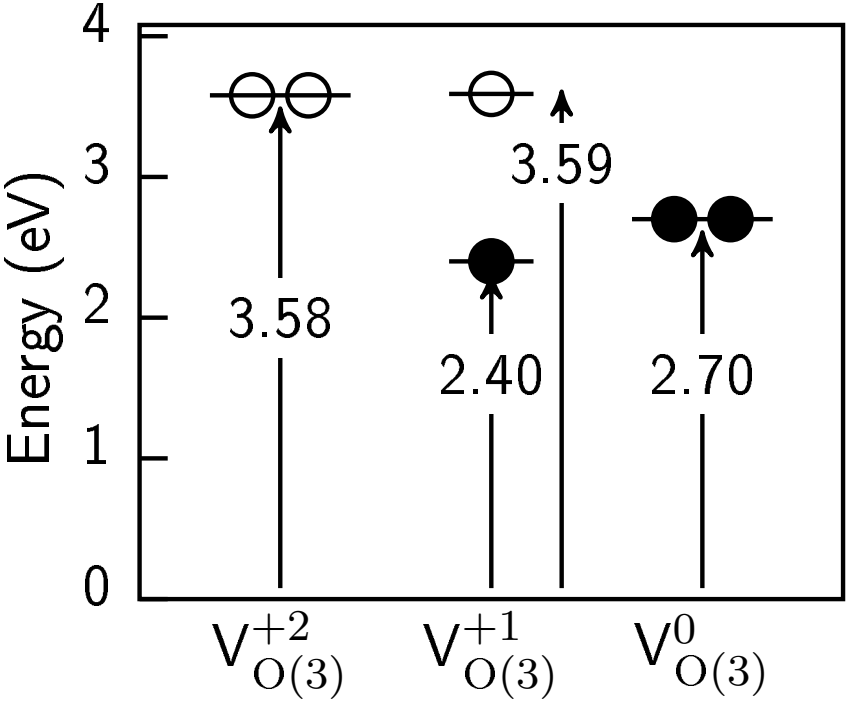
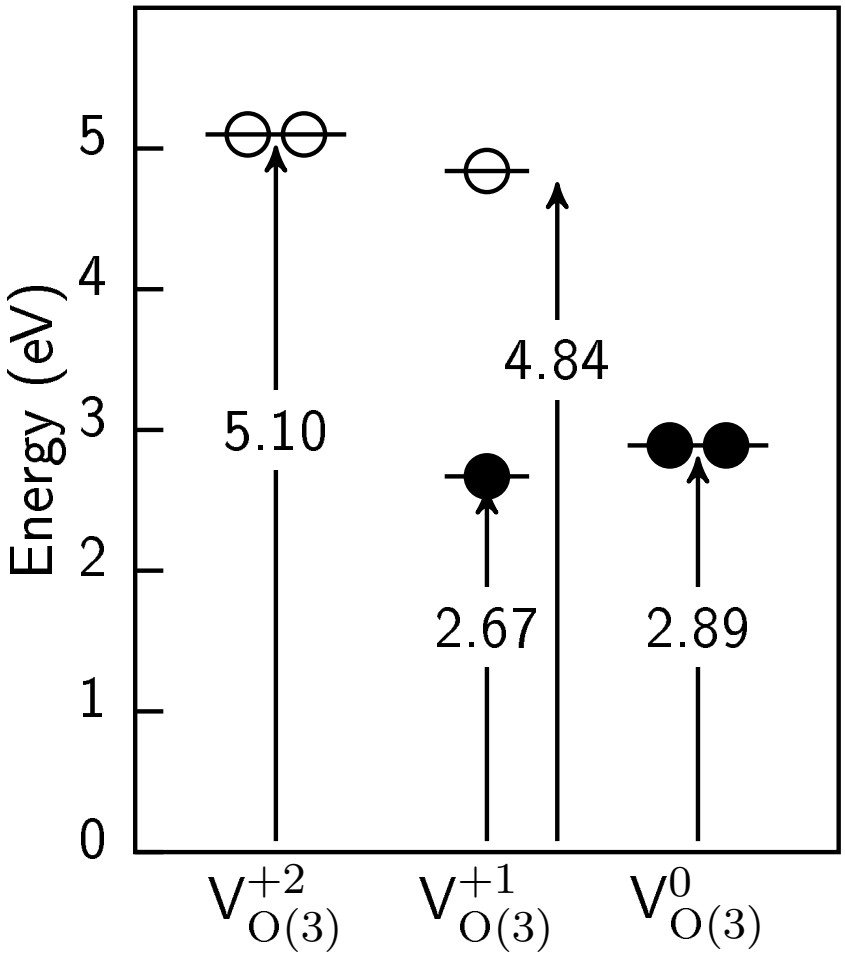
High-κ dielectric materials – Computational details

- Three fold coordinated ($V_{O(3)}$) and four fold coordinated ($V_{O(4)}$) vacancies in charge states {0, +1, +2}
- 96 atom super cells.
- Norm conserving pseudopotentials - Hf $5s^2 5p^6 5d^2 6s^2$ and O $2s^2 2p^4$.
- 250 Ry energy cutoff for wavefunctions.
- PBE functional for structural energies.
- 'One shot' GW done within generalized plasmon pole model done with `BerkeleyGW`.
- Bulk structural parameters and band gap (6.00 eV) in good agreement with experiment.
- `SIESTA` for large supercells to get electrostatic corrections.

PBE and quasiparticle level diagram - $V_{O(3)}$



PBE

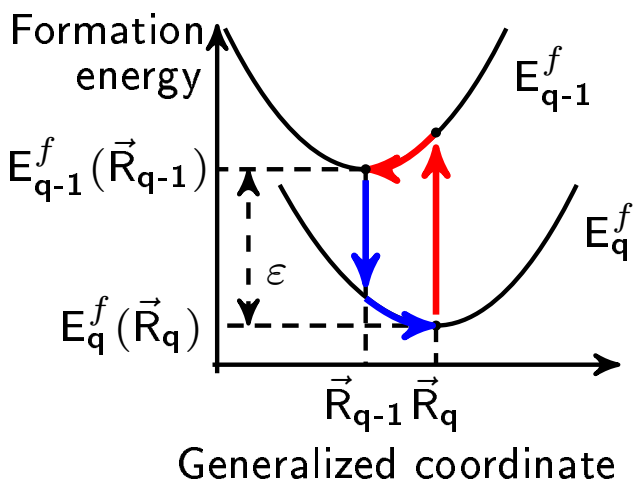


GW

Electrostatic corrections - $V_{O(3)}$

GW charge transition levels (eV)

$\varepsilon^{+2/+1}$	4.50
$\varepsilon^{+2/+1}$	3.42
$\varepsilon^{+1/0}$	3.55
$\varepsilon^{+1/0}$	4.33



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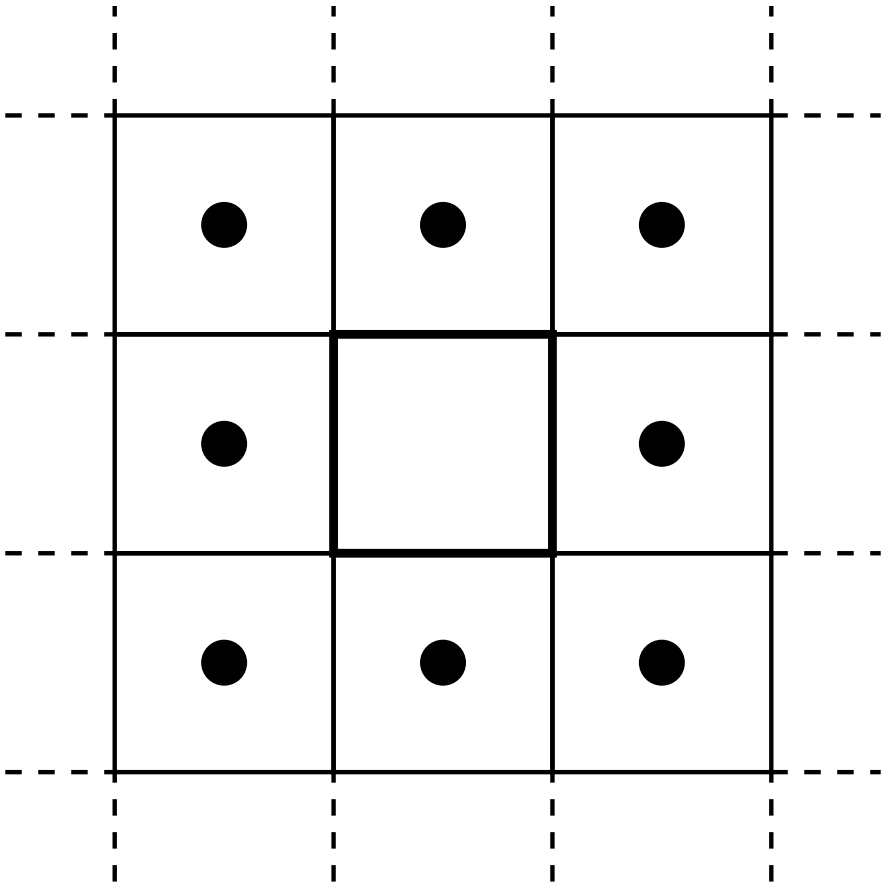

S. Lany and A. Zunger, Phys. Rev. B **81** 113201 (2010)

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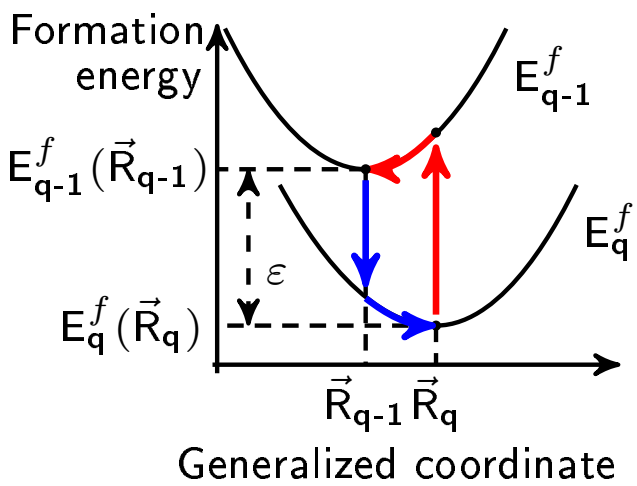

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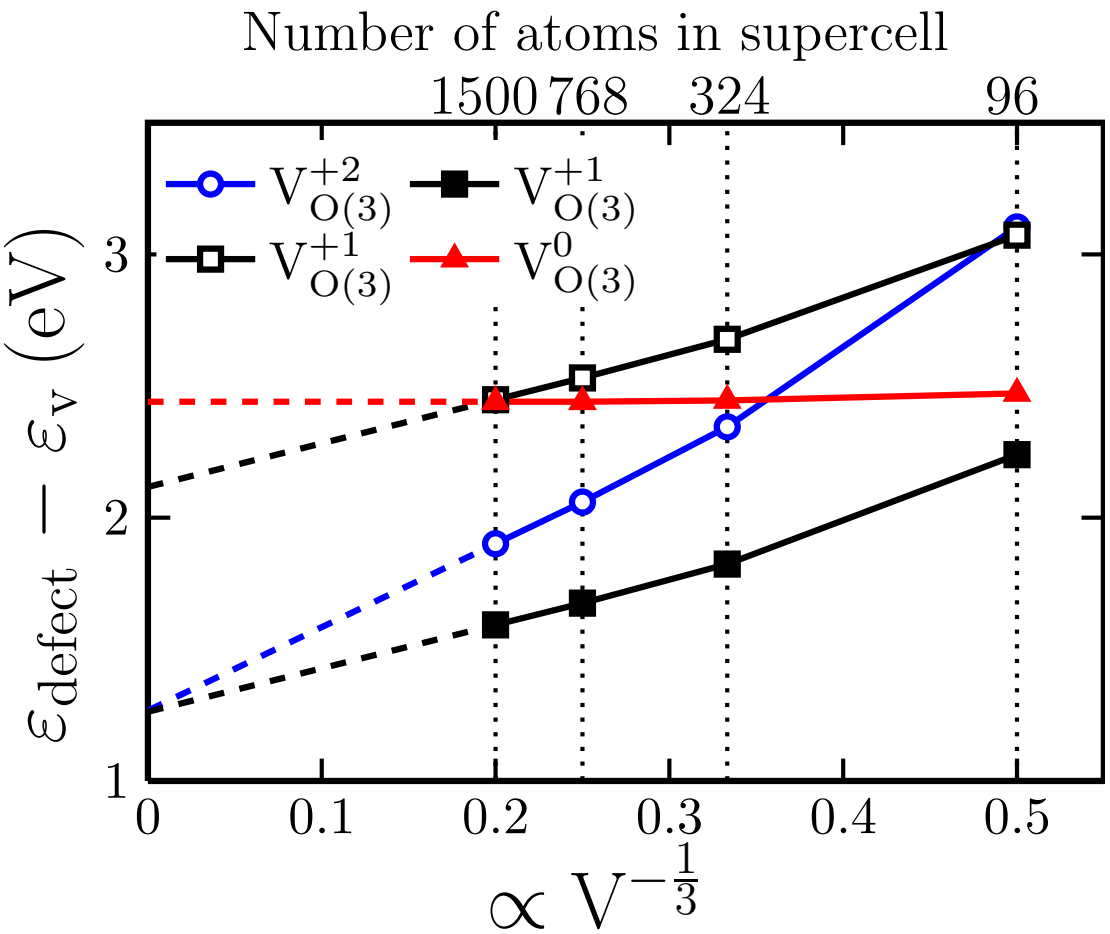

PBE defect levels.

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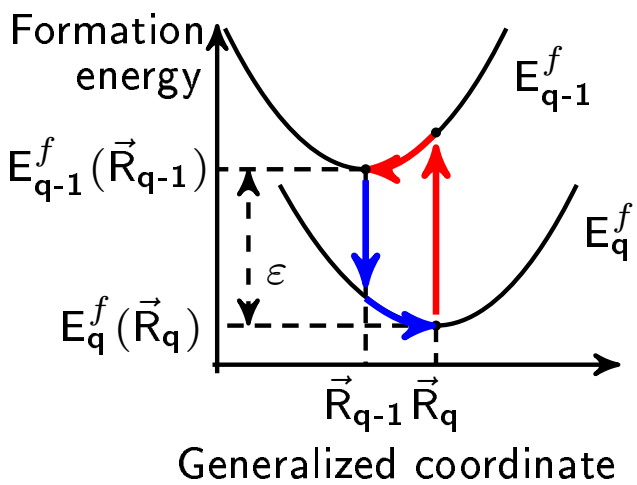

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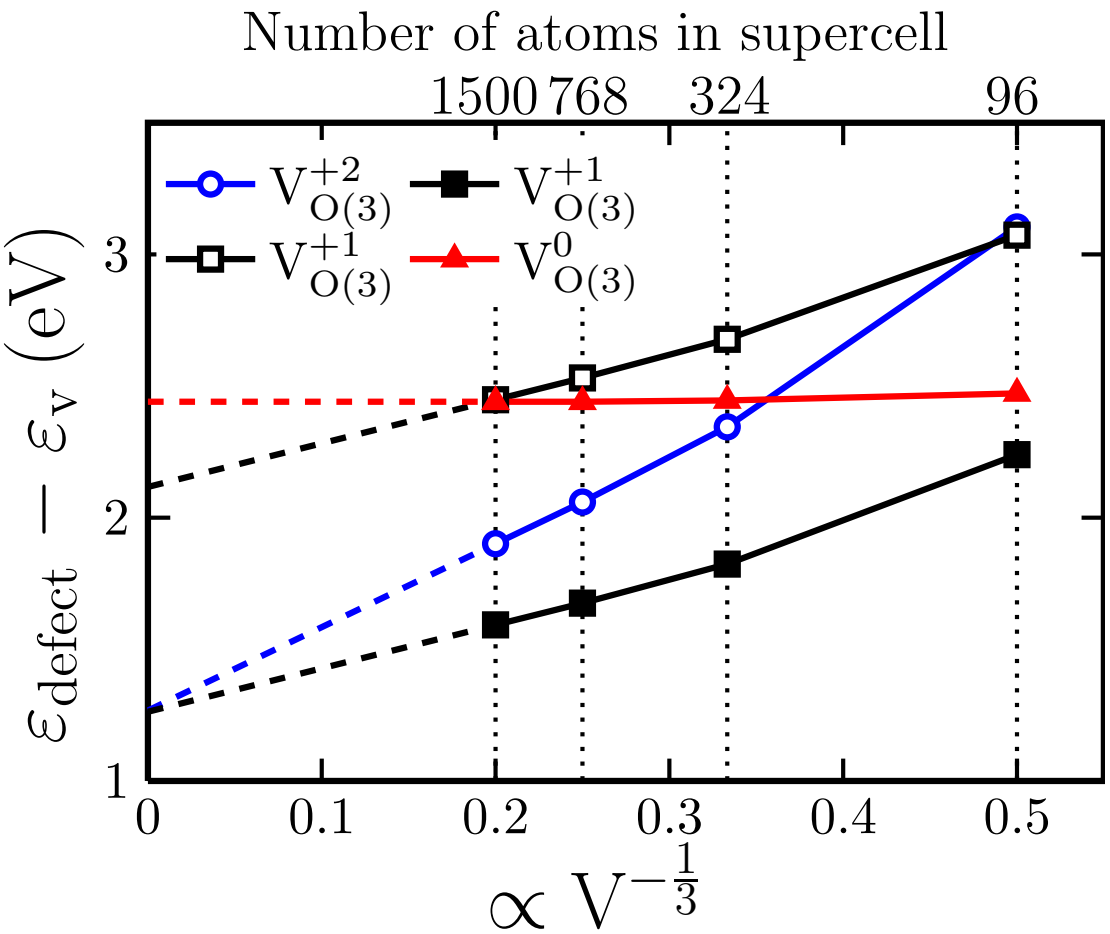
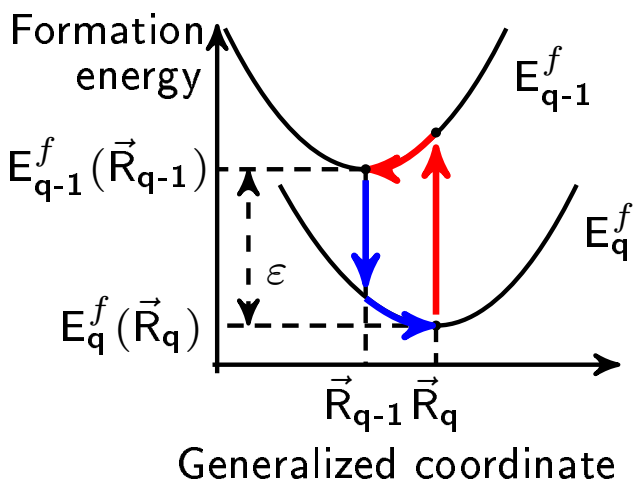


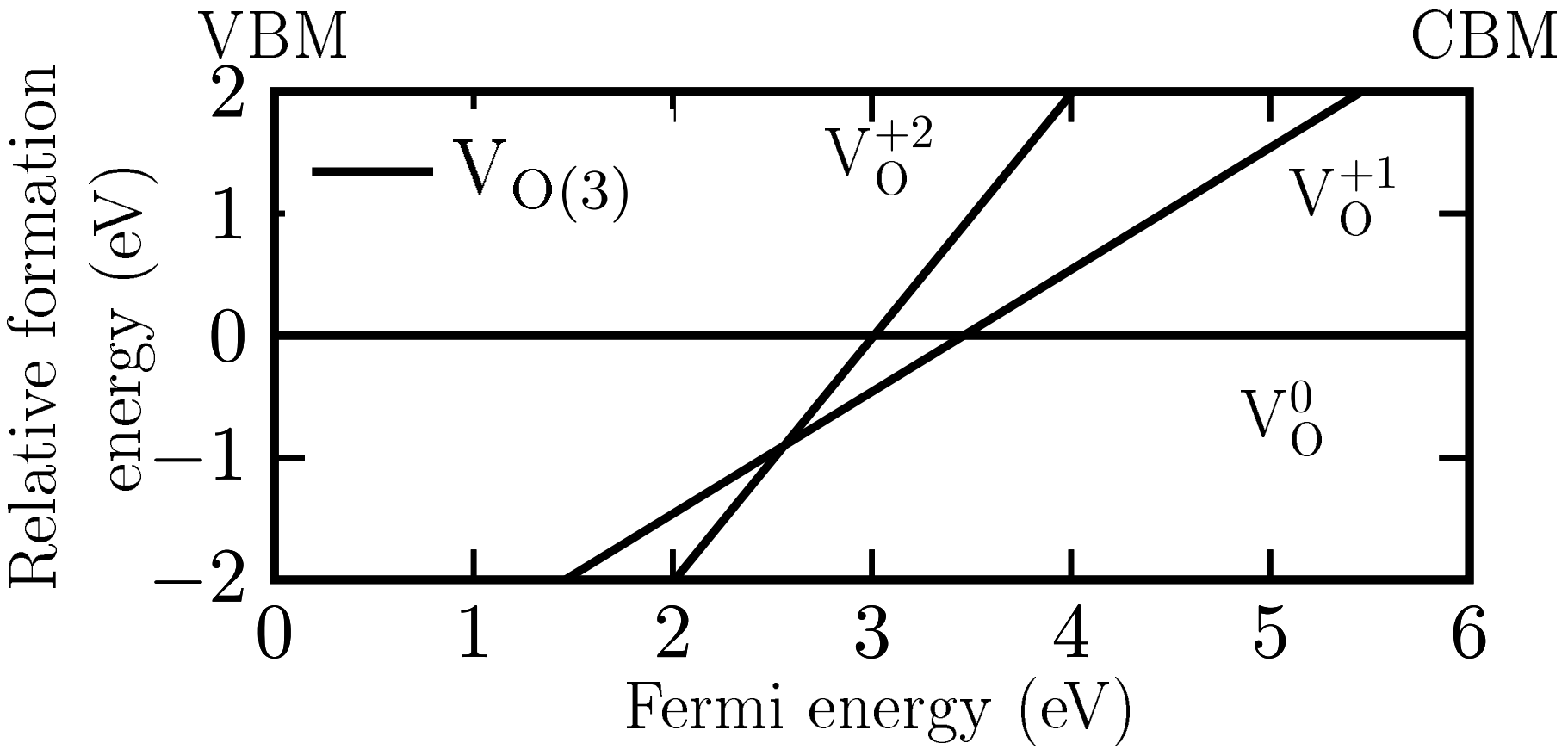
PBE defect levels.

GW charge transition levels (eV)

	w/o cor.	with cor.
$\varepsilon^{+2/+1}$	4.50	2.66
$\varepsilon^{+2/+1}$	3.42	2.45
$\varepsilon^{+1/0}$	3.55	3.55
$\varepsilon^{+1/0}$	4.33	3.36



High-κ dielectric materials – Formation energy of V_O



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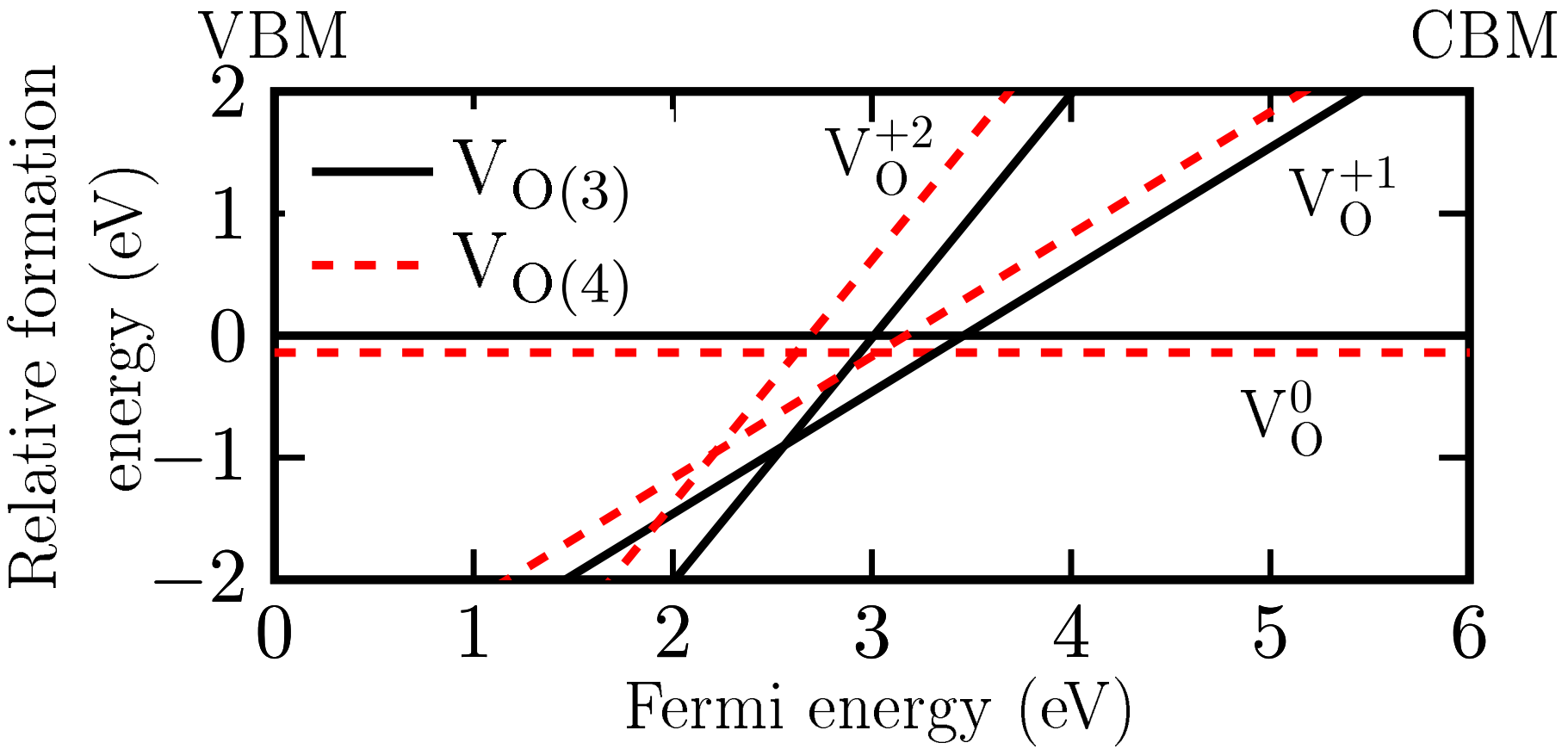

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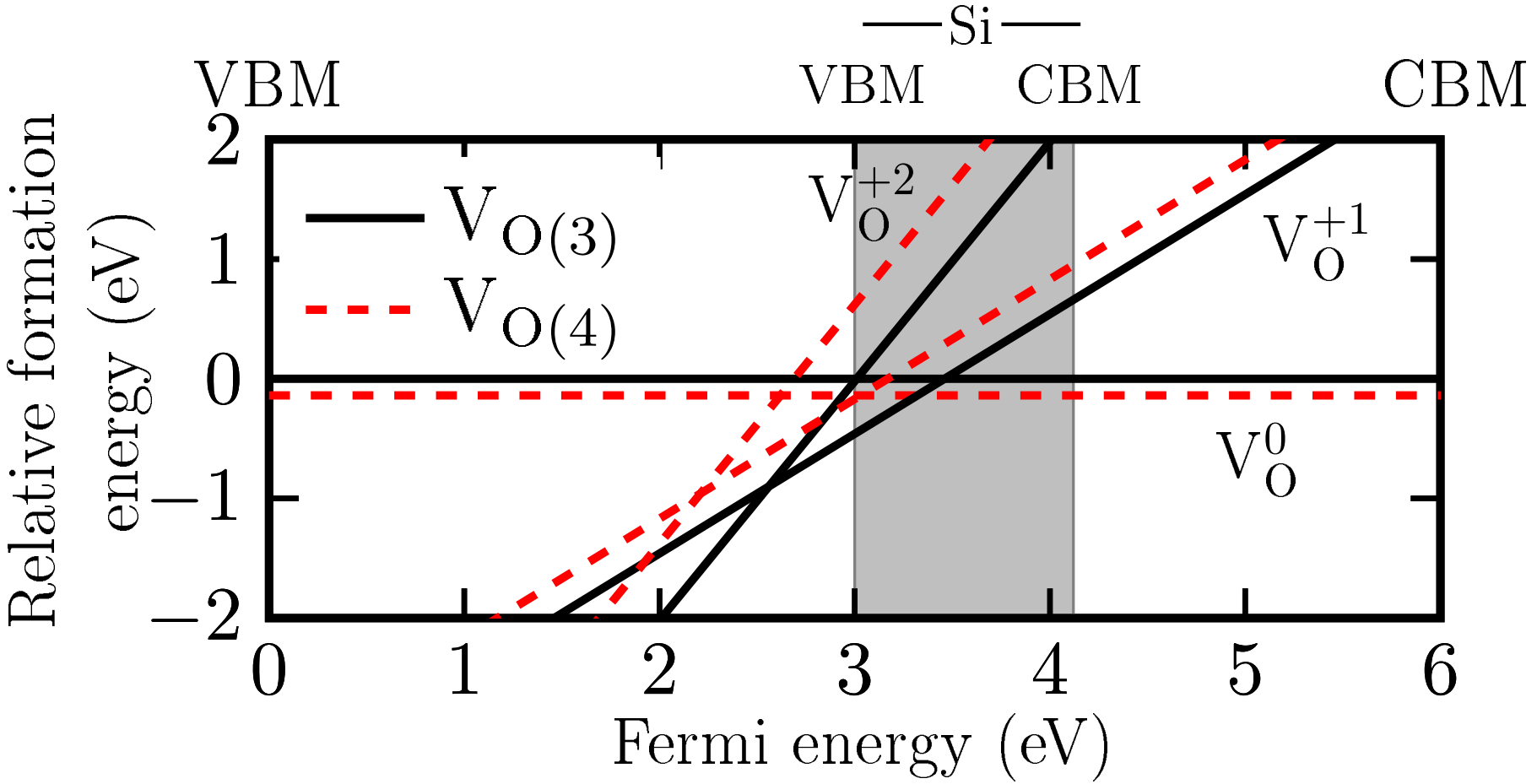
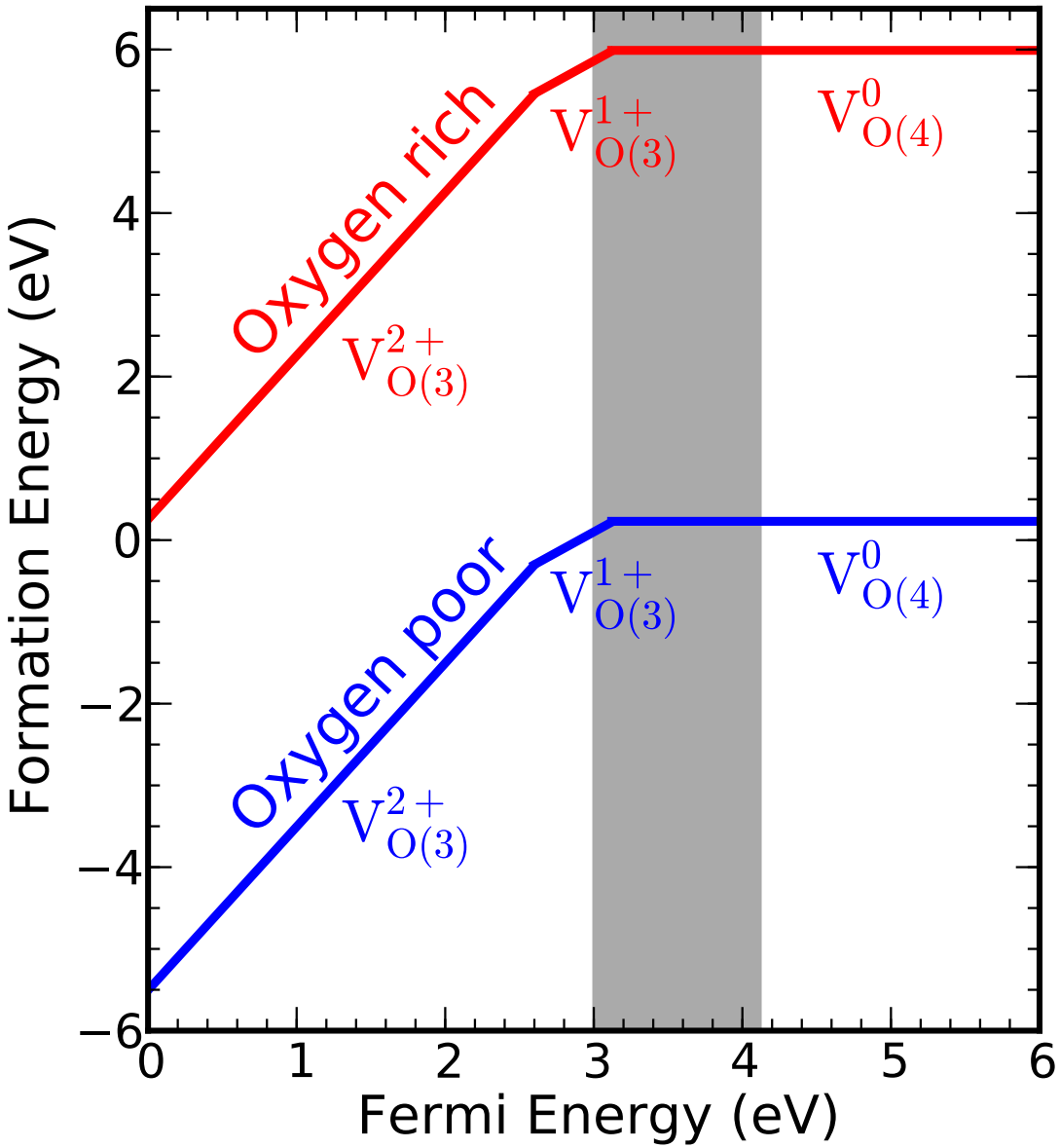


[1] M. Jain, J. R. Chelikowsky and S. G. Louie, Phys. Rev. Lett. **107**, 216803 (2011).

High-κ dielectric materials – Formation energy of vacancies



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Defect charging energy – U

U, defect charging energy, is defined as the energy of the reaction:

$$2\mathrm{V}^{+} \rightarrow \mathrm{V}^{2+} + \mathrm{V}^{0}$$

$$\begin{aligned} \mathrm{U} &= \mathrm{E}^{f}_{+2}(\vec{\mathrm{R}}_{+2}) + \mathrm{E}^{f}_{0}(\vec{\mathrm{R}}_{0}) - 2\mathrm{E}^{f}_{+1}(\vec{\mathrm{R}}_{+1}) \\ &= -\varepsilon^{+2/+1} + \varepsilon^{+1/0} \end{aligned}$$

Comparison with previous studies

All values are in eV.

	$V_{O(3)}$			$V_{O(4)}$		
	$\varepsilon^{+2/+1}$	$\varepsilon^{+1/0}$	U	$\varepsilon^{+2/+1}$	$\varepsilon^{+1/0}$	U
GGA	2.74	2.69	-0.05	2.38	2.41	-0.03
HSE[1]	3.93	4.42	0.49	–	–	–
PBE0[2]	3.7	4.1	0.4	–	–	–
GW[3] (24 atoms)	4.00	3.10	-0.90	3.22	2.43	-0.79
GW[4] (96 atoms)	2.56	3.46	0.90	2.21	3.03	0.82

[1] J.L. Lyons, A. Janotti and C.G. Van de Walle, Microelectronic Engineering **88**, 1452 (2011).
[2] P. Broqvist and A. Pasquarello, Appl. Phys. Lett. **89**, 262904 (2006).
[3] E.-A. Choi, and K. J. Chang, Appl. Phys. Lett., **94**, 122901 (2009).
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&= -\varepsilon^{+2/+1} + \varepsilon^{+1/0} \\
&\equiv \mathrm{U}_{\mathrm{elec}} + \mathrm{U}_{\mathrm{relax}} \\
&= \{\mathrm{E}^{f}_{+2}(\vec{\mathrm{R}}_{+1}) + \mathrm{E}^{f}_{0}(\vec{\mathrm{R}}_{+1}) - 2\mathrm{E}^{f}_{+1}(\vec{\mathrm{R}}_{+1})\} + \\
&\{[\mathrm{E}^{f}_{+2}(\vec{\mathrm{R}}_{+2}) - \mathrm{E}^{f}_{+2}(\vec{\mathrm{R}}_{+1})] + [\mathrm{E}^{f}_{0}(\vec{\mathrm{R}}_{0}) - \mathrm{E}^{f}_{0}(\vec{\mathrm{R}}_{+1})]\}
\end{aligned}$$

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\end{aligned}$$

$$\mathrm{U}_{\mathrm{elec}} \geq 0 \text{ and } \mathrm{U}_{\mathrm{relax}} \leq 0$$

Defect charging energy – U

$$2V^{+} \rightarrow V^{2+} + V^{0}$$

	U_{elec}	U_{relax}	U
$V_{O(3)}$	2.24	-1.33	0.90
$V_{O(4)}$	2.13	-1.35	0.81

High-κ dielectric materials – Conclusions

- DFT+GW method for calculating the stability of oxygen vacancies.
- *Qualitative* agreement with previous hybrid functional calculations on the vacancies.
- *Quantitative* disagreement with previous hybrid functional calculations - vacancies near a Si/HfO_2 interface.

	DFT+GW	HSE
V_O	+1,0	+2,+1

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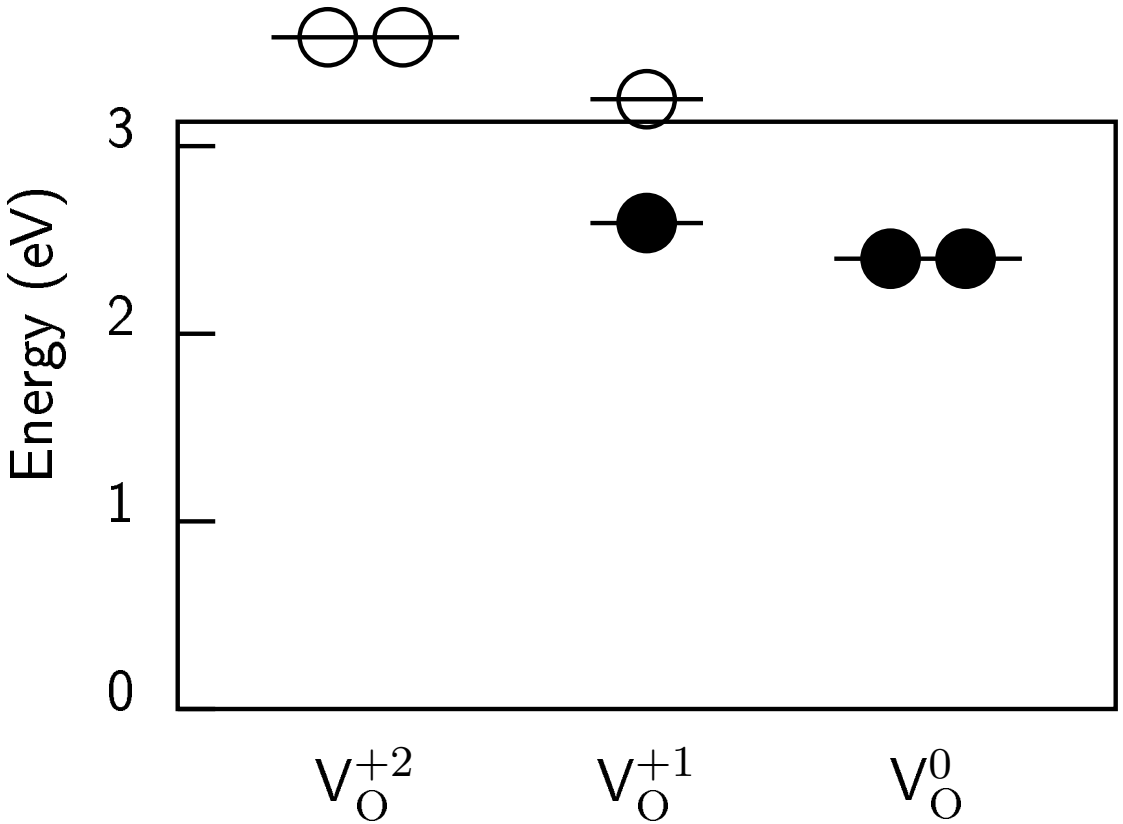
  - Defects for quantum computing.

Vacancies in TiO_2 – Computational details

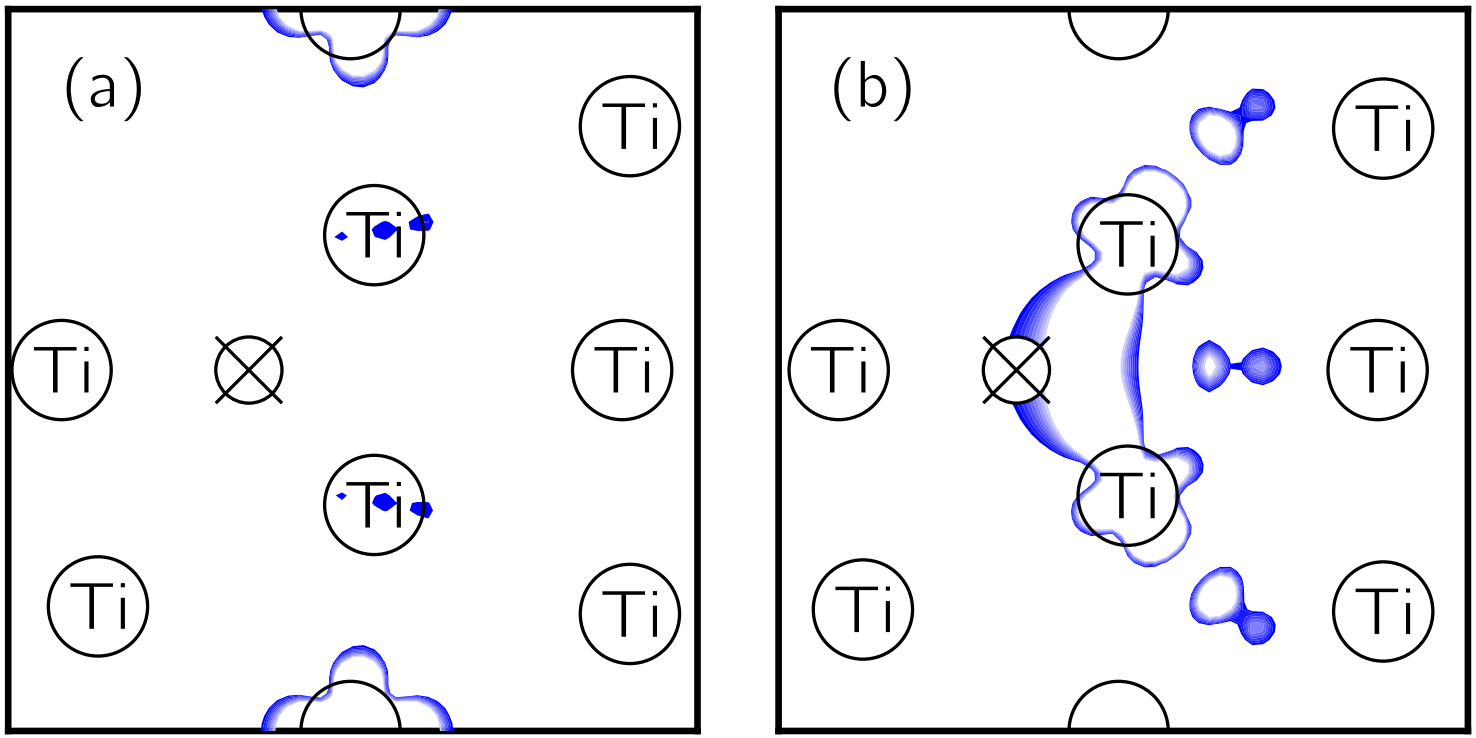
- Oxygen vacancies (V_O) in rutile in charge states $\{0, +1, +2\}$
- 72 atom super cells.
- 200 Ry energy cutoff for wavefunctions.
- HSE functional for structural energies.
- 'One shot' GW done within generalized plasmon pole model done with `BerkeleyGW`.
- Bulk structural parameters and band gap (3.13 eV) in good agreement with experiment.

Schematic quasiparticle level diagram - V_O



Polaron formation – V_O^{+1}

Relaxation of the atoms : Two minima



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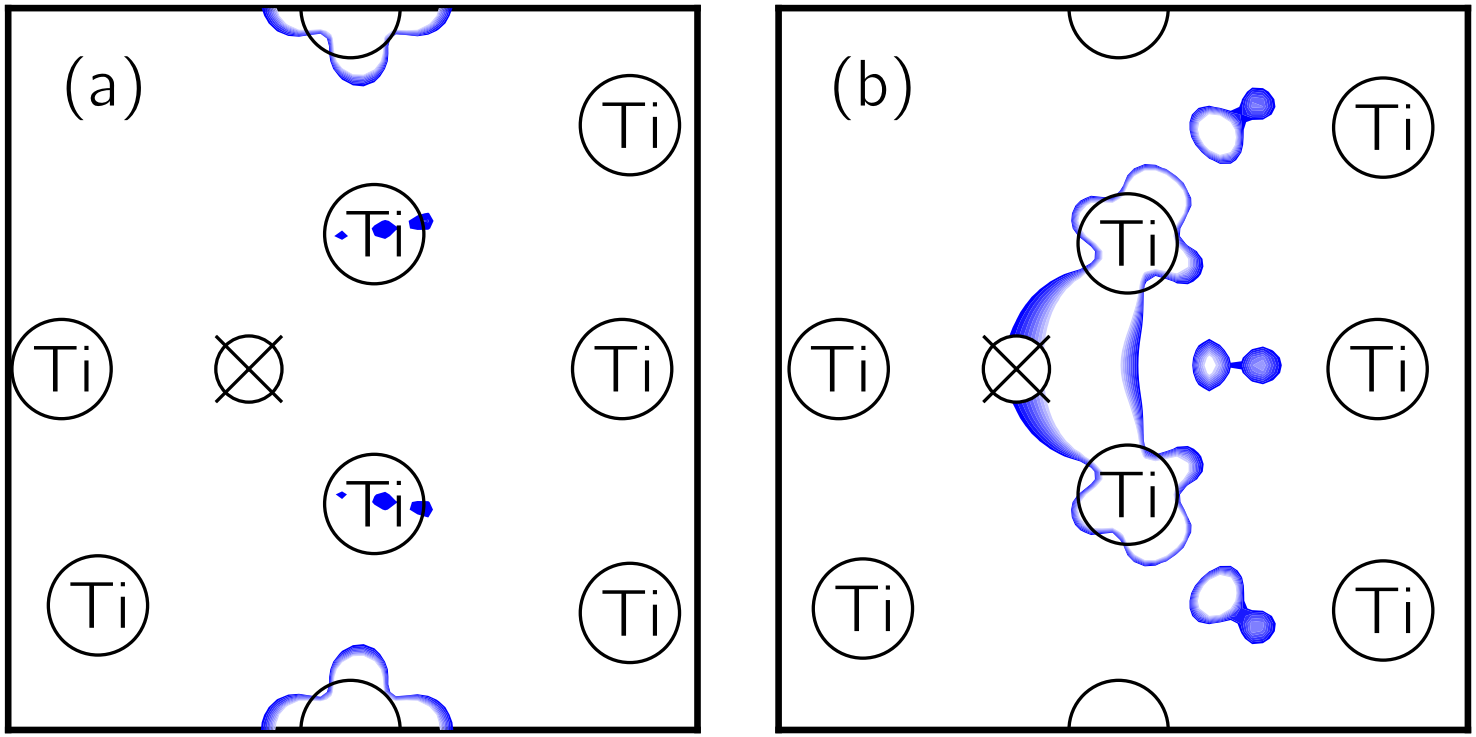
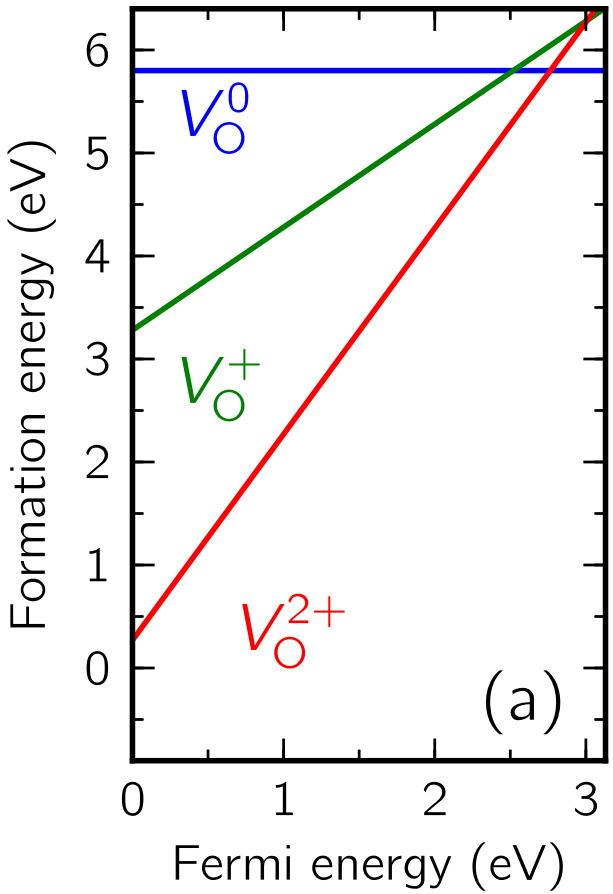


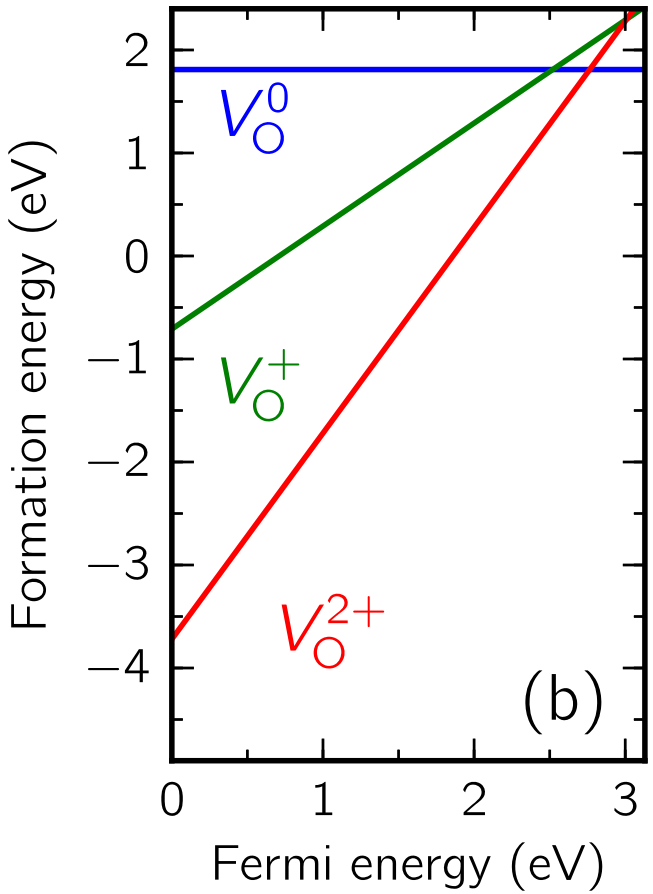
- A state in the gap – polaron.
- $V_O^{+1} \rightarrow V_O^{+2}$ + polaron.
- Lower in energy by 1.2 eV.
- Not relevant for charge transition level

- A state in the gap – defect state.
- Charge density on the defect site.
- Local minima.
- Relevant for charge transition level

TiO_2 – Formation energy of vacancies



Oxygen rich



Titanium rich

Comparison with previous studies

All values are in eV.

	V_O		
	$\varepsilon^{+2/+1}$	$\varepsilon^{+1/0}$	U
GGA[1]	2.0	2.0	0
HSE[2]	3.8	3.1	–0.7
GW[3]	3.01	2.56	–0.45

Negative U defect.

---

[1] H. Iddir, S. Ogut, P. Zapol, and N. D. Browning, Phys. Rev. B **75**, 073203 (2007). (value from plot)

[2] A. Janotti, J. B. Varley, P. Rinke, N. Umezawa, G. Kresse, and C. G. Van de Walle, Phys. Rev. B **81** 085212 (2010). (value from plot)

[3] A. Malashevich, M. Jain and S. G. Louie, Phys. Rev. B **89**, 075205, (2014).

TiO_2 – Conclusions

- DFT+GW method for calculating the stability of oxygen vacancies.
- Negative U defect.
- *Qualitative* agreement with previous hybrid functional calculations.
- *Quantitative* disagreement with previous hybrid functional calculations.

	DFT+GW	HSE
V_O	+2,0	+2

Outline

- Motivation.
- Computational methods.
- Defects in oxides.
  - Why are defects challenging?
  - Defects in high-κ dielectric – HfO_2.
  - Defects in technologically important material – TiO_2.
  - Defects for quantum computing.

Defects for spin-qubit applications

- Room temperature individually addressable spin systems in the solid-state for quantum computing.

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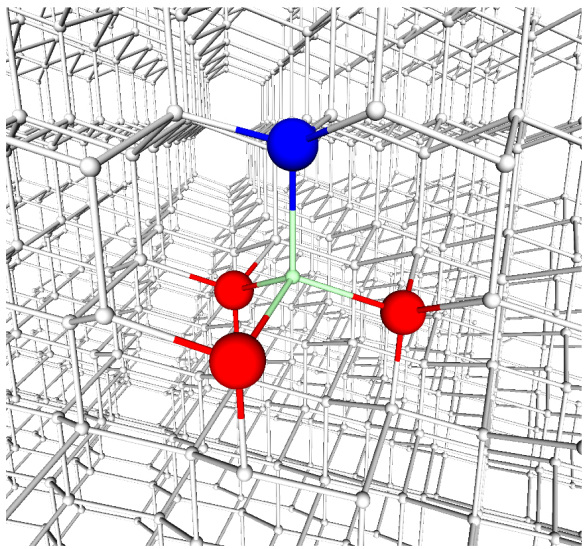
- NV^- center in diamond is the leading candidate.



Defects for spin-qubit applications

- Room temperature individually addressable spin systems in the solid-state for quantum computing.
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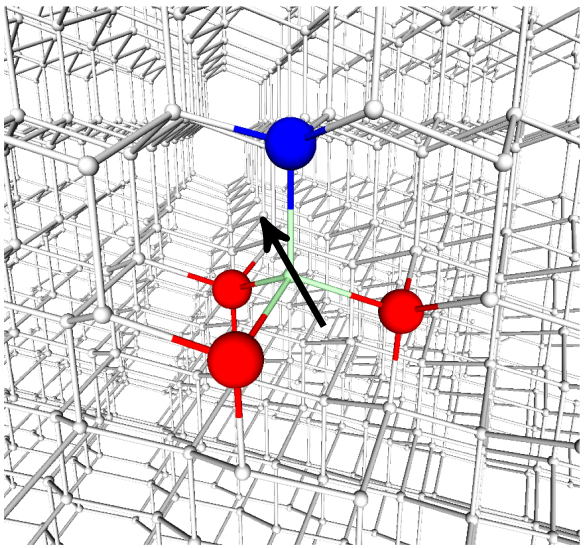

- Possible room-temperature qubit with long coherence time ($\sim$1ms) for quantum computing.
- High sensitivity, high-spatial resolution magnetometry.

NV^- center in diamond – Motivation

Optical initialization at room-temperature provides initial spin pure state for spin-qubit operation.

Notation : ${}^{2S+1}\Lambda$

Λ : Irreducible representation of the orbital symmetry

S : Total Spin

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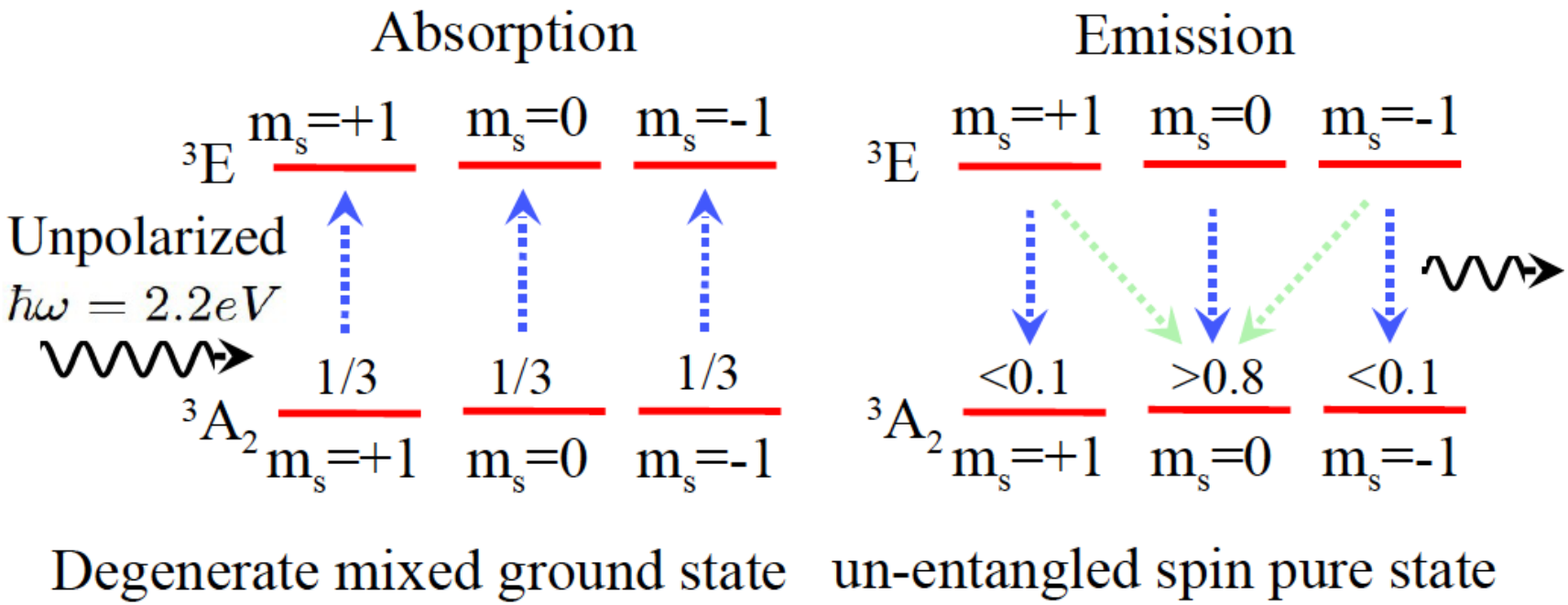
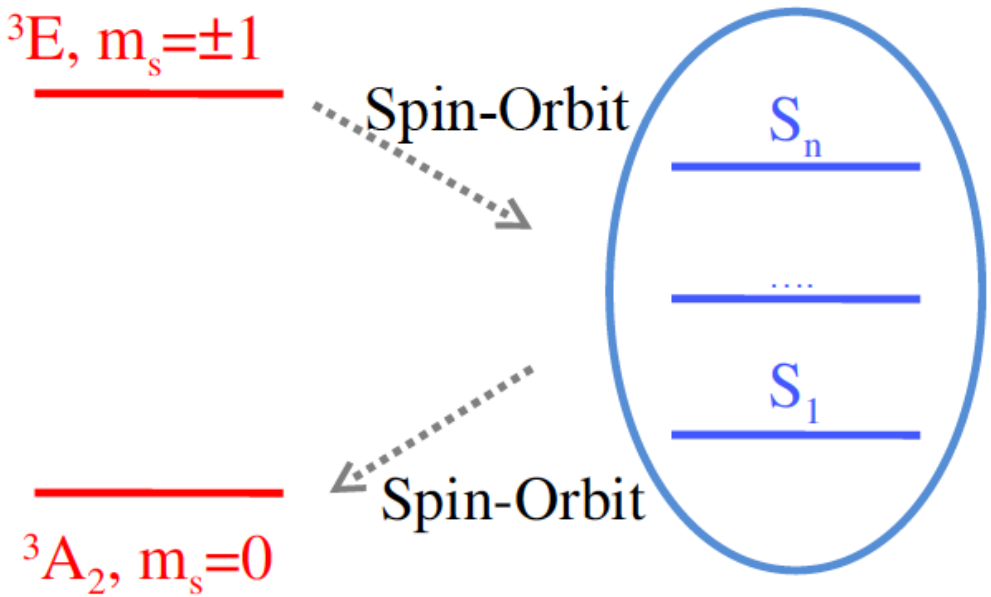
At Room Temperature



Degenerate mixed ground state un-entangled spin pure state

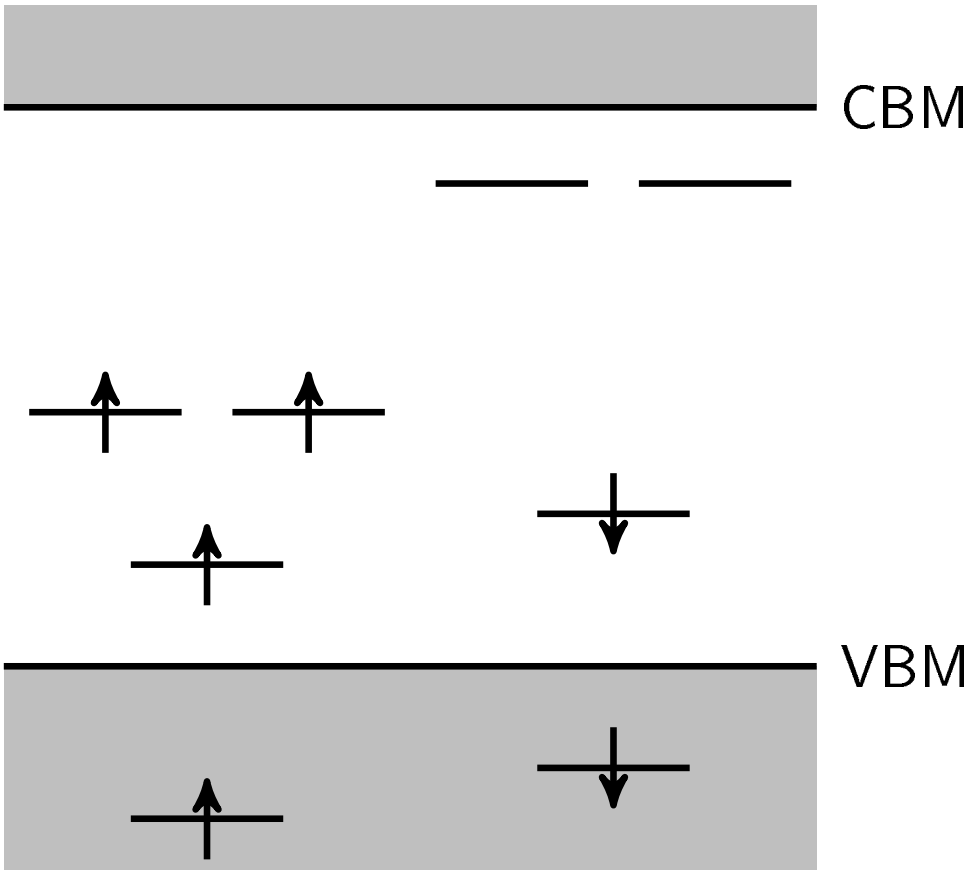
NV^- center in diamond – Unsolved problem



- Identification of singlet-level structure.
- Effective optical initialization path between the two triplet levels.

NV^- center in diamond – Single particle levels

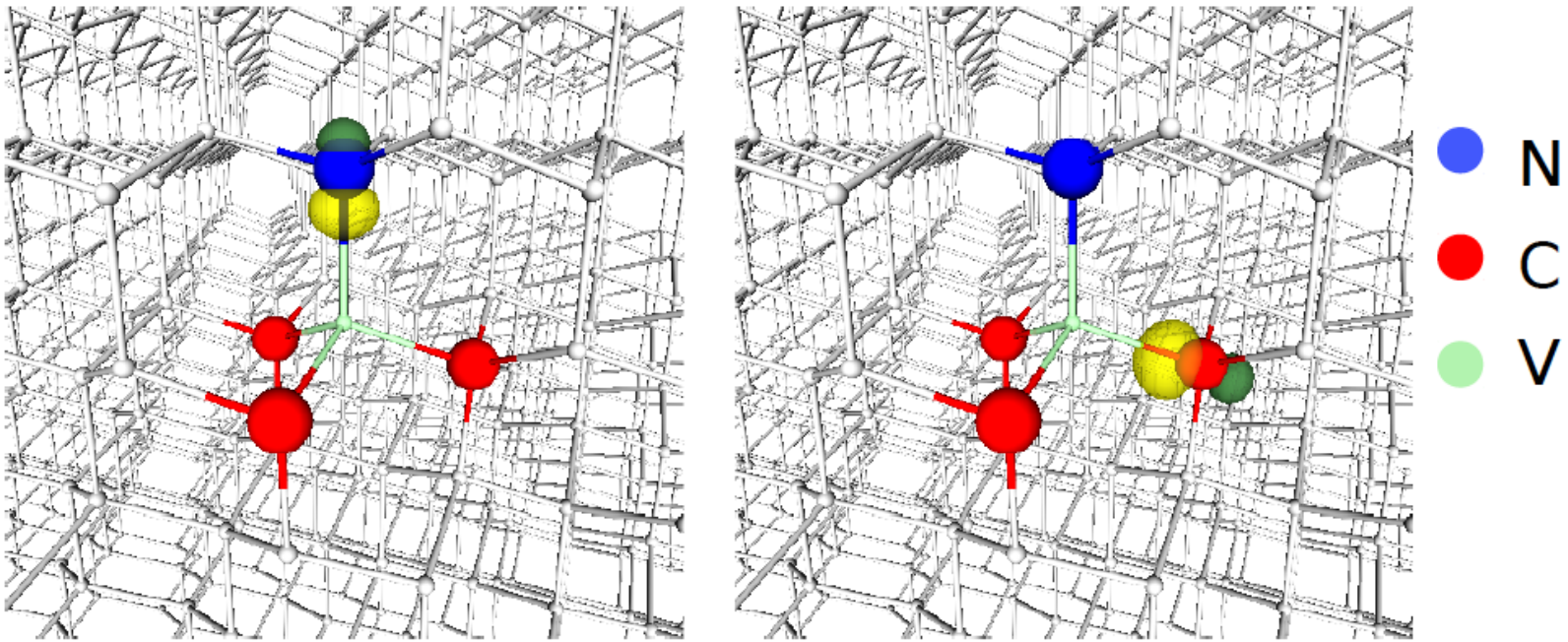
NV^- is a deep level center in a band gap of diamond with multiple localized, interacting electrons.



- Open-shell problem[1].
- Correlation due to degenerate ground-states.

[1] J. Lischner, J. Deslippe, M. Jain and S. G. Louie, Phys. Rev. Lett. **109**, 036406 (2012)

NV^- center in diamond – Dangling orbitals



8 dangling spin-orbitals in and near band-gap of diamond → Good basis for low energy excitations.

NV^- center in diamond – Extended Hubbard model

$$\hat{\mathrm{H}} = \sum_{i,\sigma} \mathrm{E}_i n_{i\sigma} + \sum_{i \neq j,\sigma} t_{i,j} c^{\dagger}_{i,\sigma} c_{j,\sigma} + \sum_{i} \mathrm{U} n_{i,\uparrow} n_{j,\downarrow} + \sum_{i \neq j,\sigma,\sigma'} \mathrm{V} n_{i,\sigma} n_{j,\sigma'}$$

i, j :Atomic sites (C or N)
σ, σ' :Spin direction
E_i :On-site energy
$t_{i,j}$:Hopping integral
U :On-site Coulomb repulsion
V :Nearest neighbor Coulomb repulsion

Effective Coulomb interaction parameters → Strong electron-electron correlation

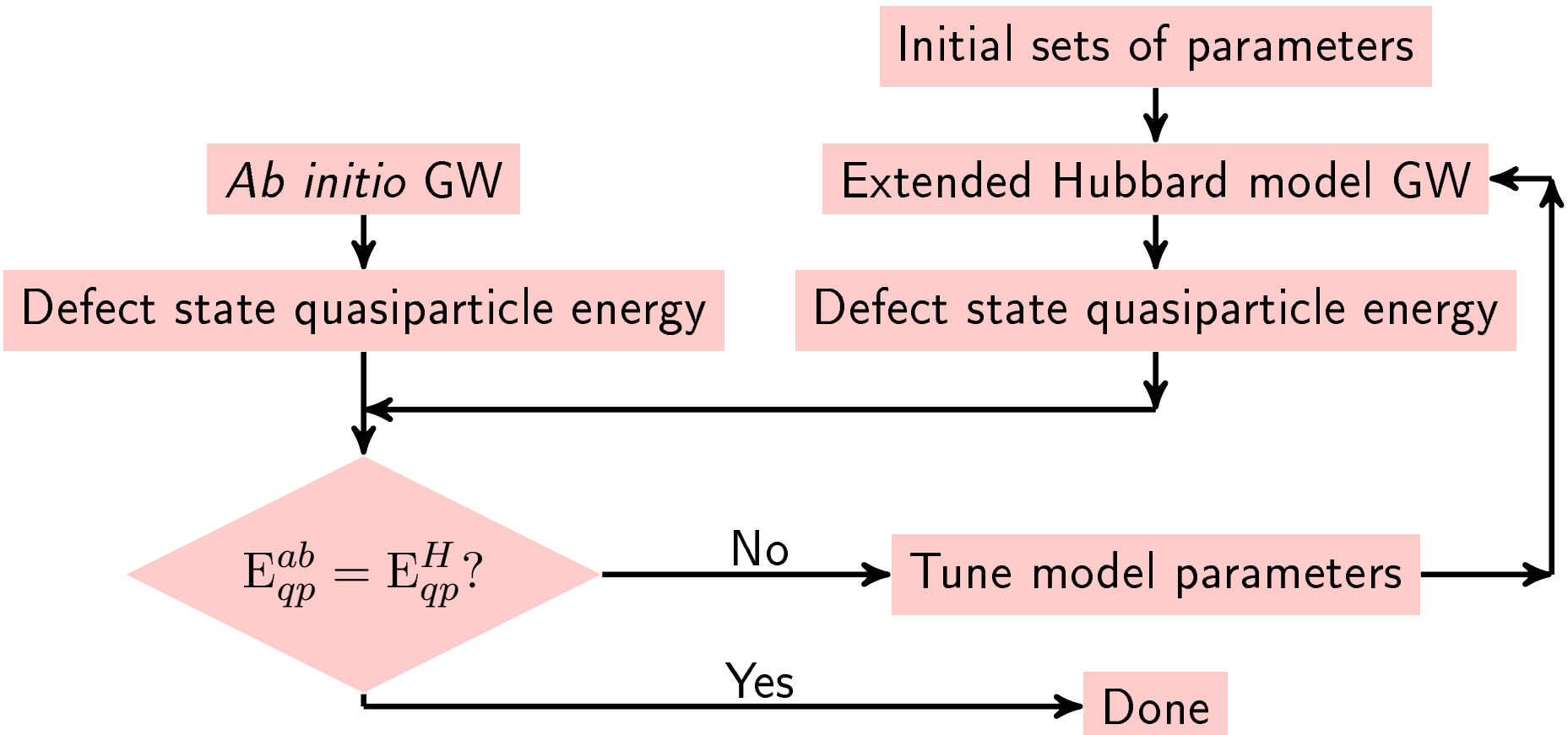
Effective Coulomb interaction parameters → Screening from diamond

Geometry dependent parameter sets → Large structural relaxation

Hindered by the difficulty in getting physically grounded model parameters.

NV^- center in diamond – Model parameters from GW

Use *ab initio* GW to get model parameters, incorporating realistic electron-electron interactions.



NV^- center in diamond – Model parameters from GW

- $U/t > 3 \rightarrow$ strongly correlated system.
- Reasonable values of parameters.
- Geometry dependent parameters.

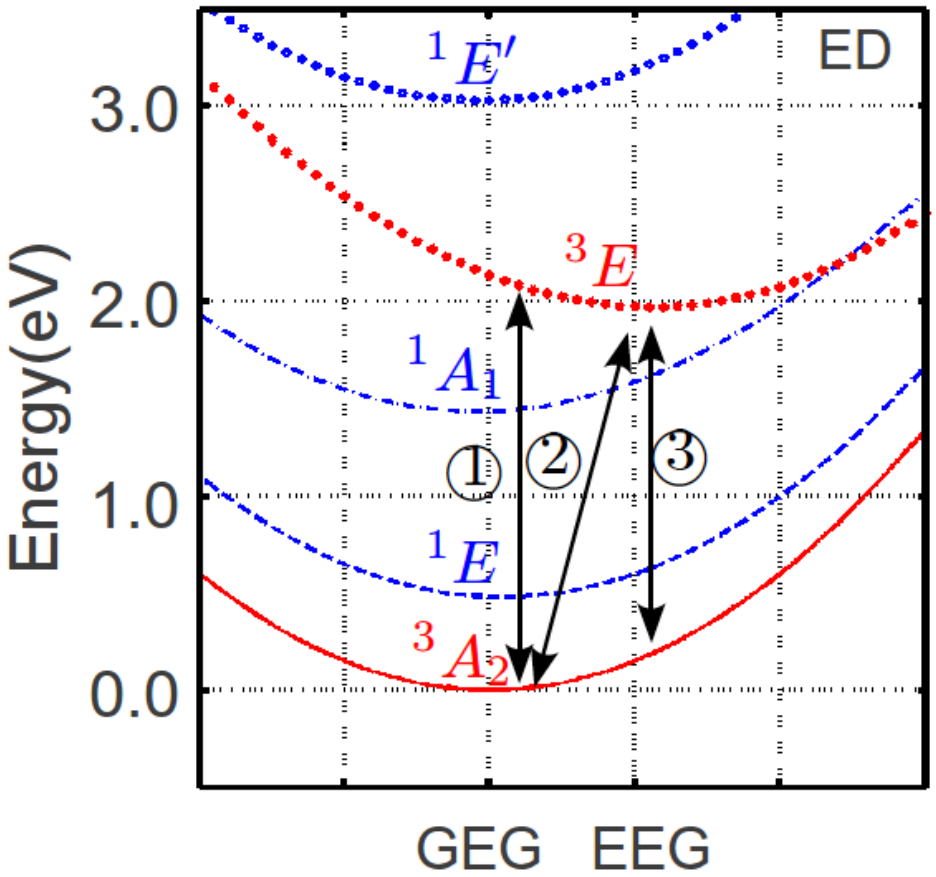
All parameters are in eV.

	E_C-E_N	t_{NC}	t_{CC}	U	V
Ground-state	2.56	–0.68	–1.03	3.43	0.83
Excited-state	2.86	–0.75	–0.90	3.45	0.67

NV^- center in diamond – Level diagram

Level diagram[1] from exact diagonalization of model Hamiltonian – all many-electron correlation effects within our Hilbert space.

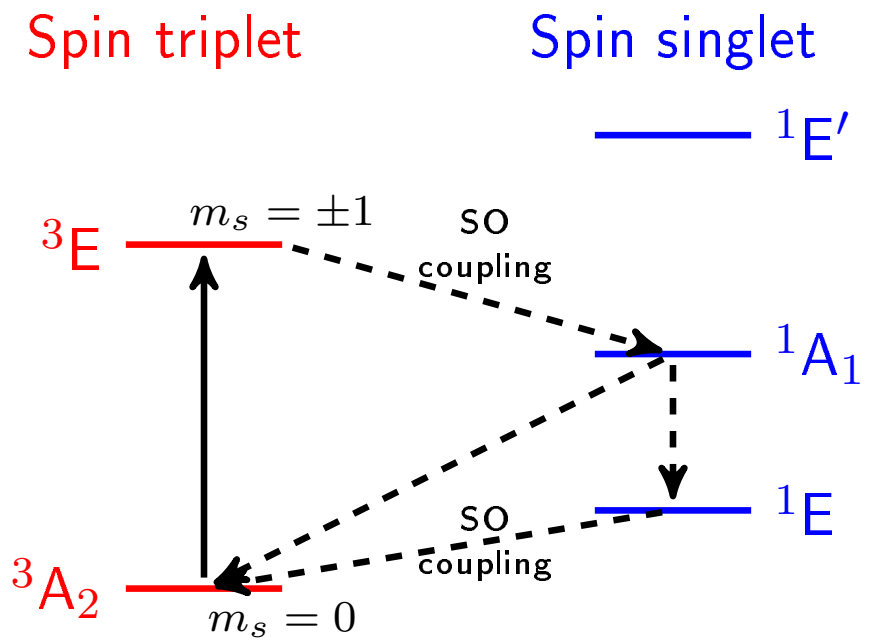
All energies in eV.



	Theory	Exp
①	2.1	2.2
②	2.0	1.945
③	1.8	1.8
$\tau_{rad}^{^3E}$	20ns	13ns
$\tau_{nonrad}^{^3E}$	30ns	50ns

[1] S. Choi, M. Jain and S. G. Louie, Phys. Rev. B **86**, 041202 (2012)

NV^- center in diamond – Proposed optical pathway[1]



Qualitatively and quantitatively consistent with available experimental data.

[1] S. Choi, M. Jain and S. G. Louie, Phys. Rev. B **86**, 041202 (2012)

NV^- center in diamond – Conclusion

- Constructed extended Hubbard Hamiltonian from *ab initio* GW calculations.
- Through exact diagonalization, many-electron effects strongly affect the energy level diagram qualitatively and quantitatively.
- Computed ground- and excited-state energy surfaces and transition rates between them provided a consistent picture with experiments.
- Proposed an optical initialization pathway in which inter-sytstem crossing plays a crucial role.

S. Choi, M. Jain and S. G. Louie, Phys. Rev. B **86**, 041202 (2012)

Conclusion

- *Ab initio* methods to understand and predict properties of defects in materials.
- Oxygen vacancies in high-κ material – HfO_2 and photocatalytic material – TiO_2.
- NV^- center in diamond for quantum computing application.

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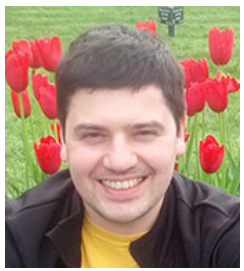

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