

Ab initio studies of vacancy-impurity complexes in cubic and hexagonal diamond

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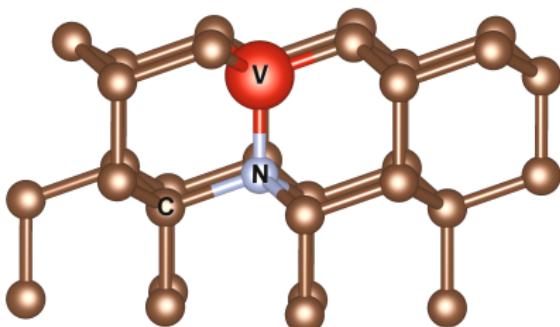
Summary and outlook



Aim and scope of the work

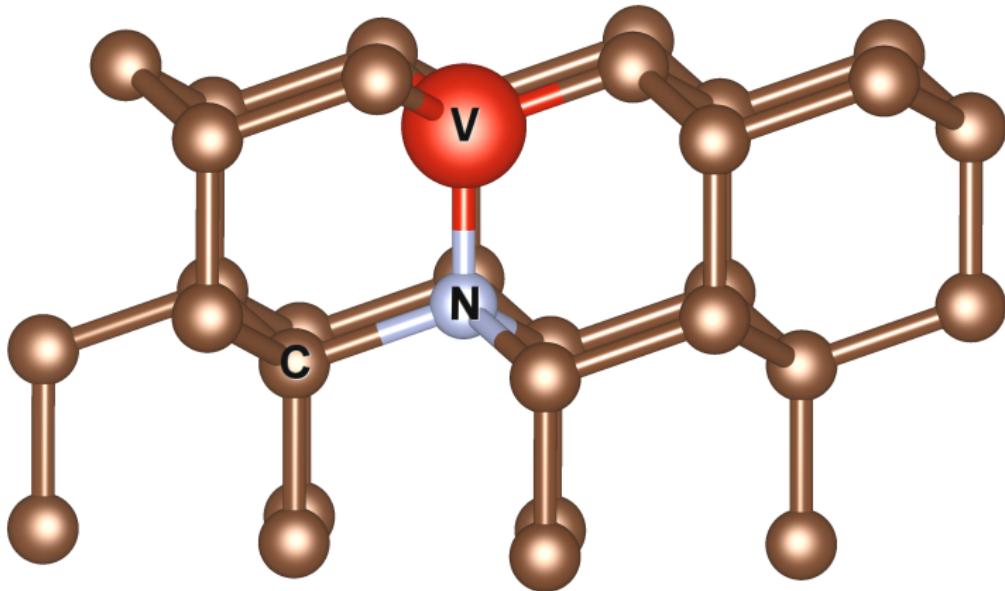
Aims and motivation

- Exploration of defect centres using state-of-the-art ab-initio theories.
- Benchmark ab initio theories with well-known experimental data.
- Systematic characterisation of defect fingerprints: ZFS, ZPL.
- Search for new defects with tailored properties which are useful for different applications.

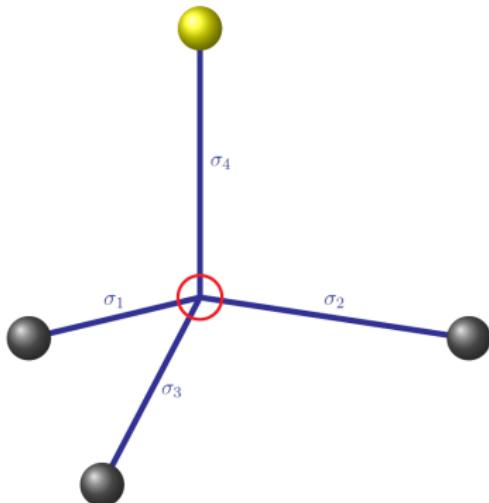


Introduction

Nitrogen Vacancy Centre in diamond (NV center)



Nitrogen Vacancy Center (Dangling bonds)



- Model for the defect $4\ sp^3$ dangling bonds $\{\sigma_1, \dots, \sigma_4\}$
- Linear Combination of Atomic Orbitals (LCAO) to account for C_{3v} symmetry.
- In the case of NV^- we have $1\#C + 2 \times \#N + 1e = 6$ electrons.
- From σ_i we obtain 4 levels $a_1(1), a_1(2), e(1), e(2)$ classified according to their symmetry.

Basic level overview

Conduction Band

$e(1)$ ————— $e(2)$

$a_1(2)$ —————

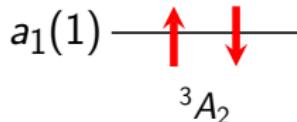
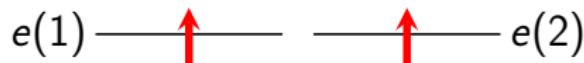
$a_1(1)$ —————

Valence Band



Basic level overview: Ground State

Conduction Band

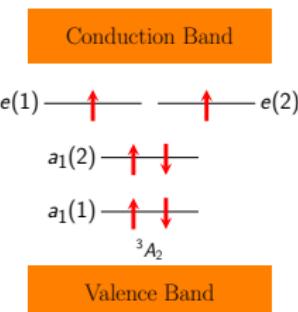
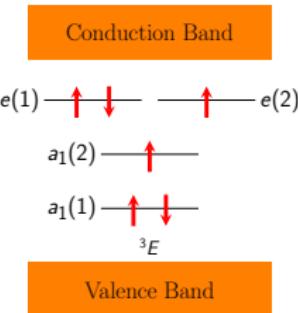
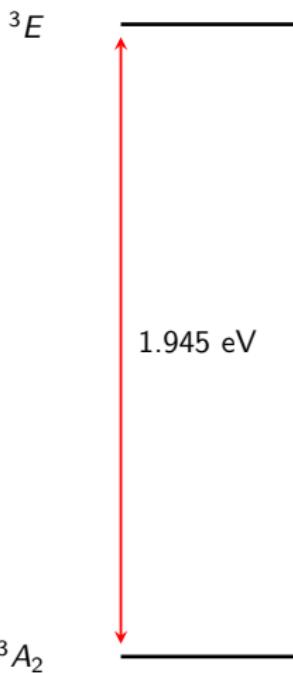


3A_2

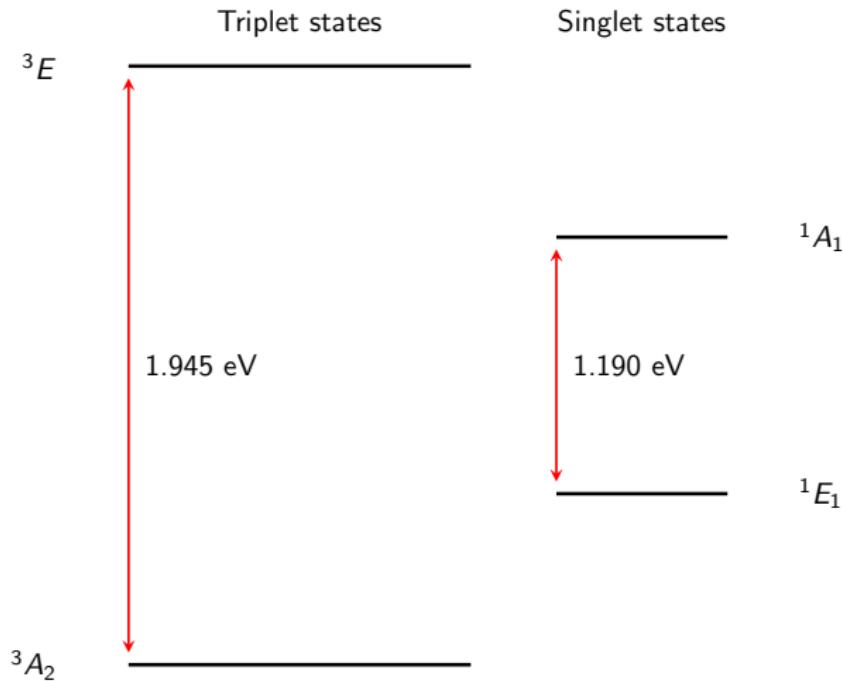
Valence Band



Triplets overview: NV⁻



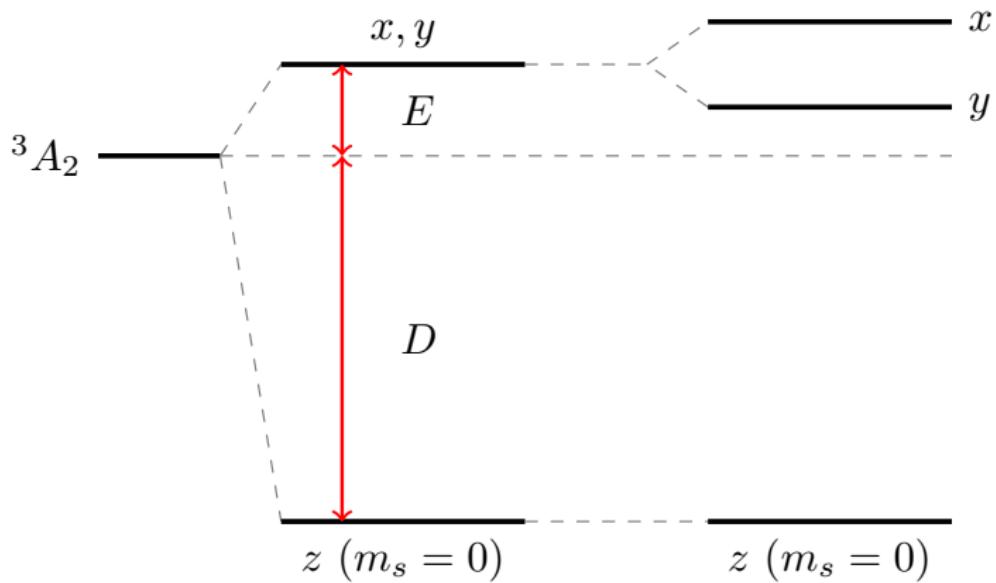
Main level overview: NV⁻



Zero Field Splitting (ZFS)

$\hat{\mathbf{S}}^t \mathbf{D} \hat{\mathbf{S}}$

Zeeman/strain

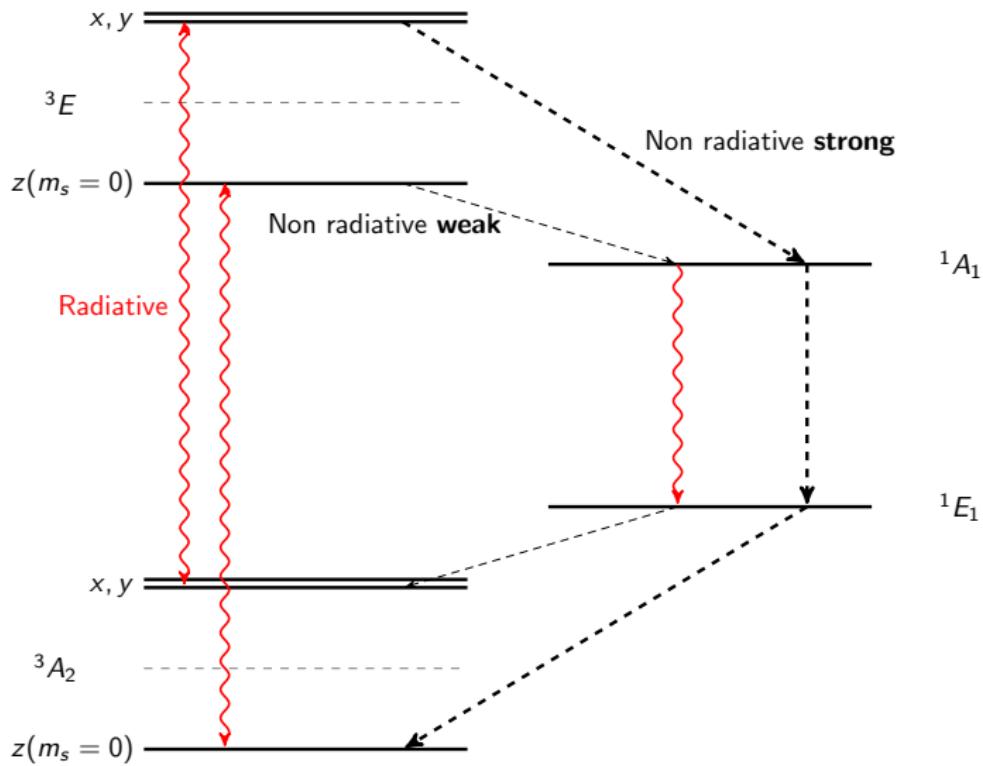


REVISE E AND D

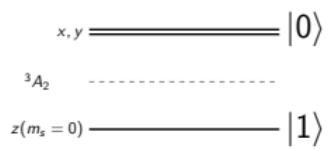


MAX-PLANCK-GESSELLSCHAFT

Transitions overview: NV⁻

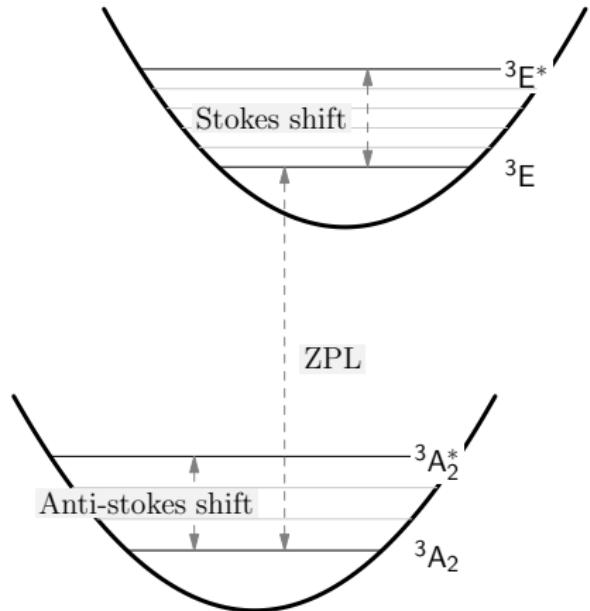
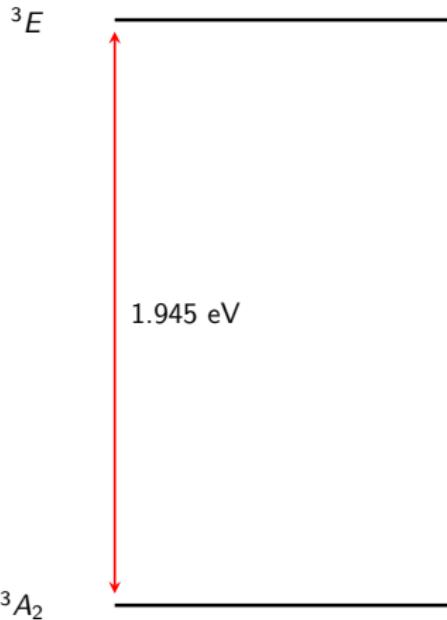


Transitions overview: NV⁻



- Detection of spin state.
- Realization of a *qubit*.
- Initialization of the state.

Zero phonon line (ZPL)



Density functional theory (DFT)

" Ψ contains too much information"

- Popular saying

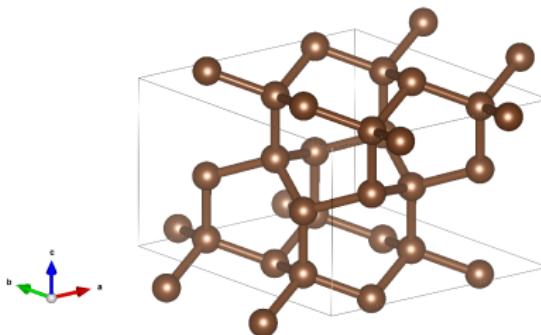
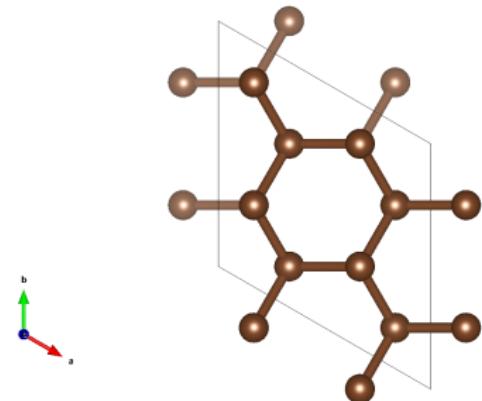
- In principle, DFT delivers the **exact ground state**.
- All quantities are written in terms of ρ (functional formalism).
- E.g.:

$$E[\rho] = T_s[\rho] + \int V(\mathbf{r})\rho(\mathbf{r}) d\mathbf{r} + \frac{1}{4\pi\epsilon_0} \int \frac{\rho(\mathbf{r})\rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r} d\mathbf{r}' + E_{\text{xc}}^{\text{exact}}[\rho]$$

The exchange correlation potential $E_{\text{xc}}^{\text{exact}}[\rho]$ determines the DFT flavor. In many calculations we use the so-called **PBE** (*Perdew-Burke-Ernzerhof*) functional.

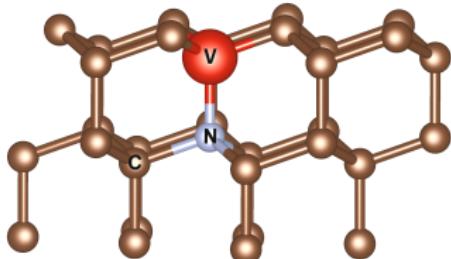
Hexagonal diamond and defects

Hexagonal diamond

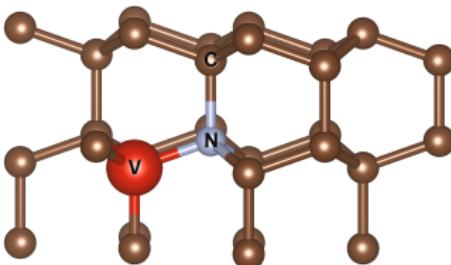


Defected hexagonal diamond

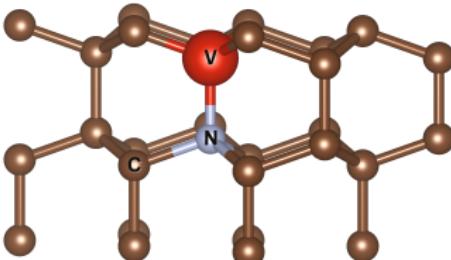
Cubic diamond



Hexagonal *x*-type

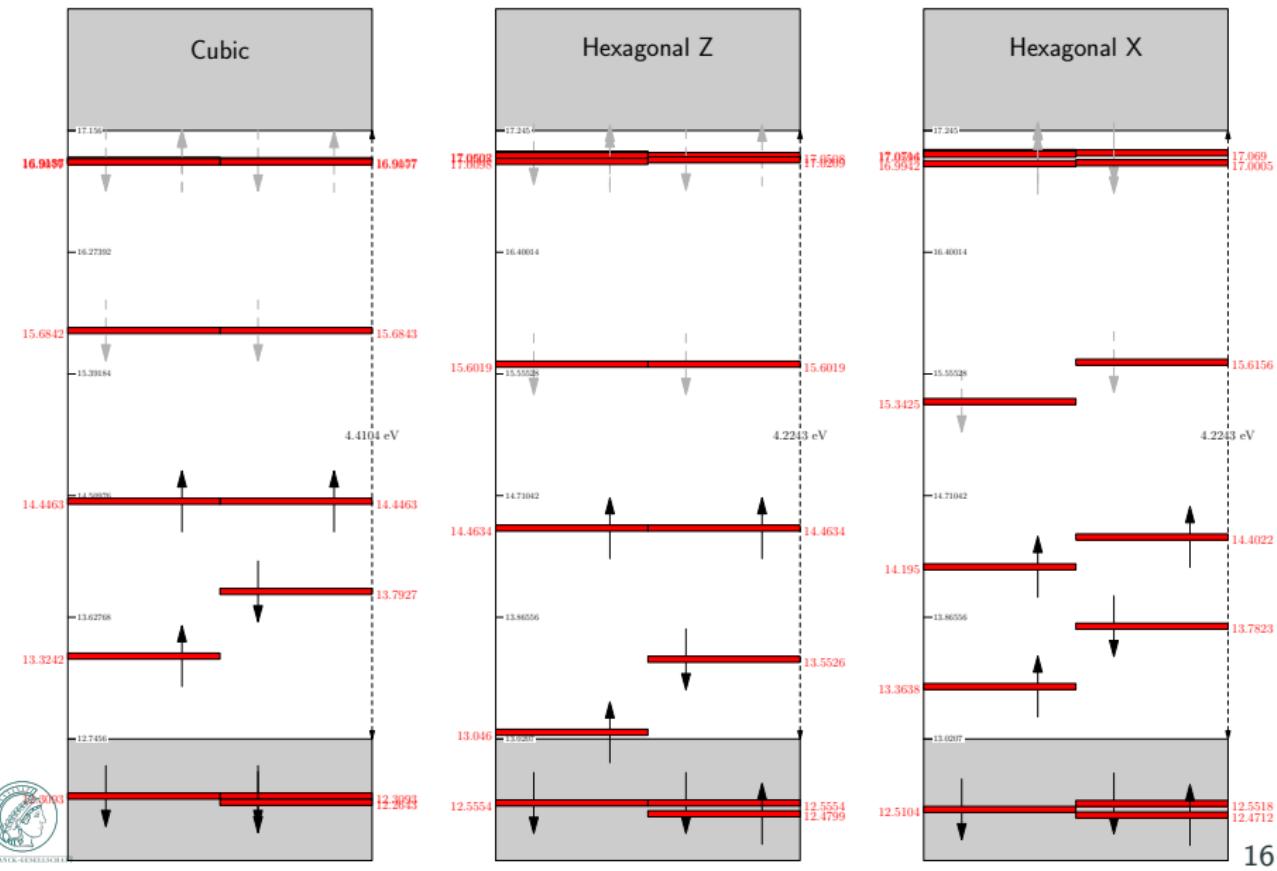


Hexagonal *z*-type

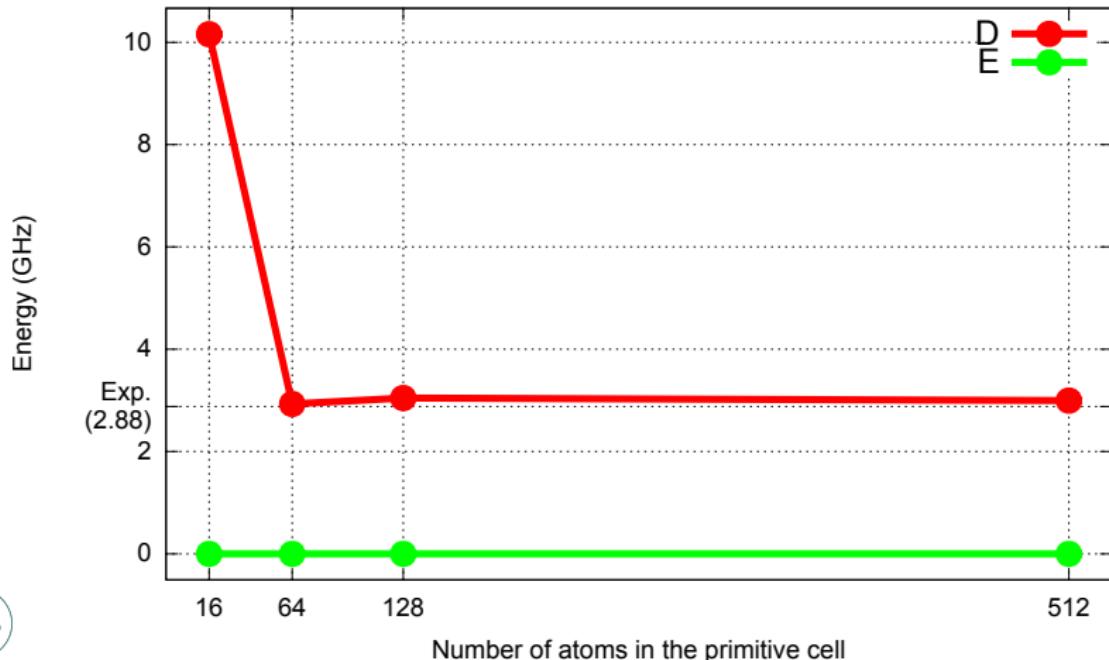


Results

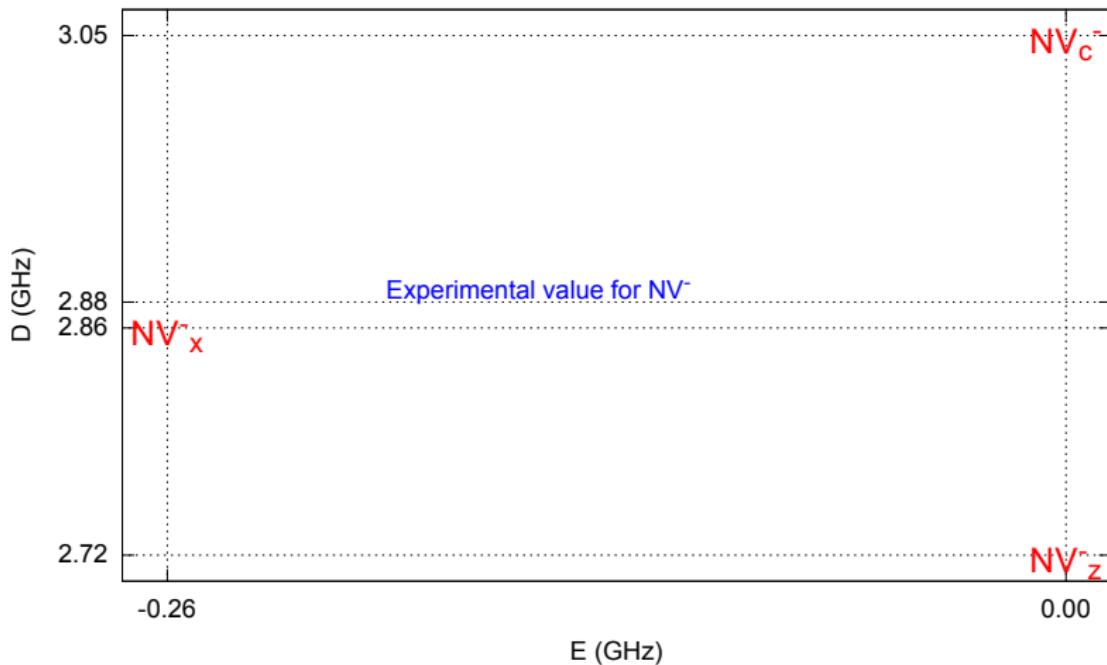
NV⁻: Ground state 3A_2



- Cubic diamond, convergence and comparison with the experimental result.



NV*: ZFS cubic (NV_c^*), Hexagonal x, z ($\text{NV}_{x,z}^*$)



Expanding the defect

18 VIIA							
5	10.811	6	12.011	7	14.007	8	15.999
B	C	N	O	F	Ne		
Boron	Carbon	Nitrogen	Oxygen	Flourine	Neon		
13	26.982	14	28.086	15	30.974	16	32.065
Al	Si	P	S	Cl	Ar		
Aluminium	Silicon	Phosphorus	Sulphur	Chlorine	Argon		
31	69.723	32	72.64	33	74.922	34	78.96
Ga	Ge	As	Se	Br	Kr		
Gallium	Germanium	Arsenic	Selenium	Bromine	Krypton		
49	114.82	50	118.71	51	121.76	52	127.6
In	Sn	Sb	Te	I	Xe		
Indium	Tin	Antimony	Tellurium	Iodine	Xenon		
81	204.38	82	207.2	83	208.98	84	209
Tl	Pb	Bi	Po	At	Rn		
Thallium	Lead	Bismuth	Polonium	Astatine	Radon		
113	284	114	289	115	288	116	293
Uut	Uug	Uup	Uuh	Uus	Uuo		
Ununtrium	Ununquadium	Ununpentium	Ununhexium	Ununseptium	Ununoctium		



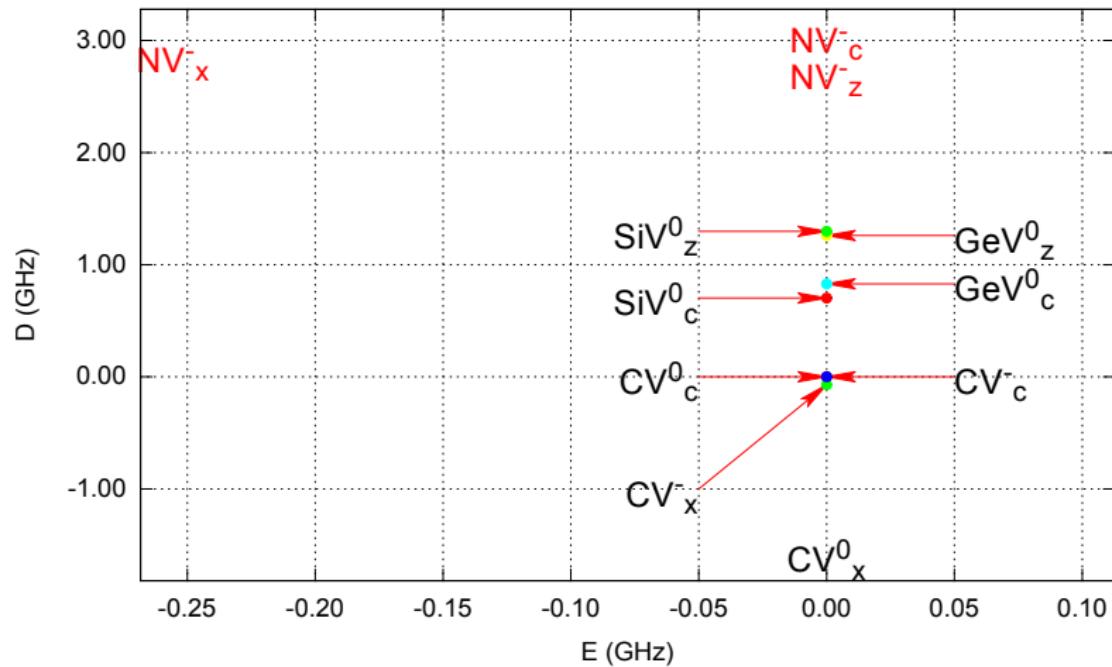
Expanding the defect

14 IVA 15 VA

6 12.011 $\text{CV}_{\textcolor{red}{a}}^q$ Carbon	7 14.007 $\text{NV}_{\textcolor{red}{a}}^q$ Nitrogen
14 28.086 $\text{SiV}_{\textcolor{red}{a}}^q$ Silicon	15 30.974 $\text{PV}_{\textcolor{red}{a}}^q$ Phosphorus
32 72.64 $\text{GeV}_{\textcolor{red}{a}}^q$ Germanium	$a = \{c, x, z\}$ $q = \{+, -, 0\}$



ZFS map



Summary and outlook

Where we are, and where to go next...

- Structural properties
- ZPL calculation.
- ZFS tensor calculation.
- **Beyond the Ground state:**
Using DMRG (*Density Matrix Renormalization Group*) for excited state calculations

Thank you!

