

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

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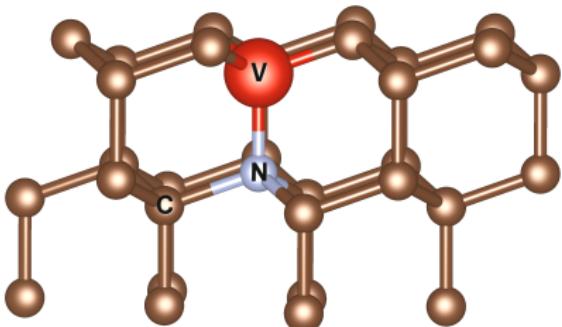
Summary and outlook



Aim and scope of the work

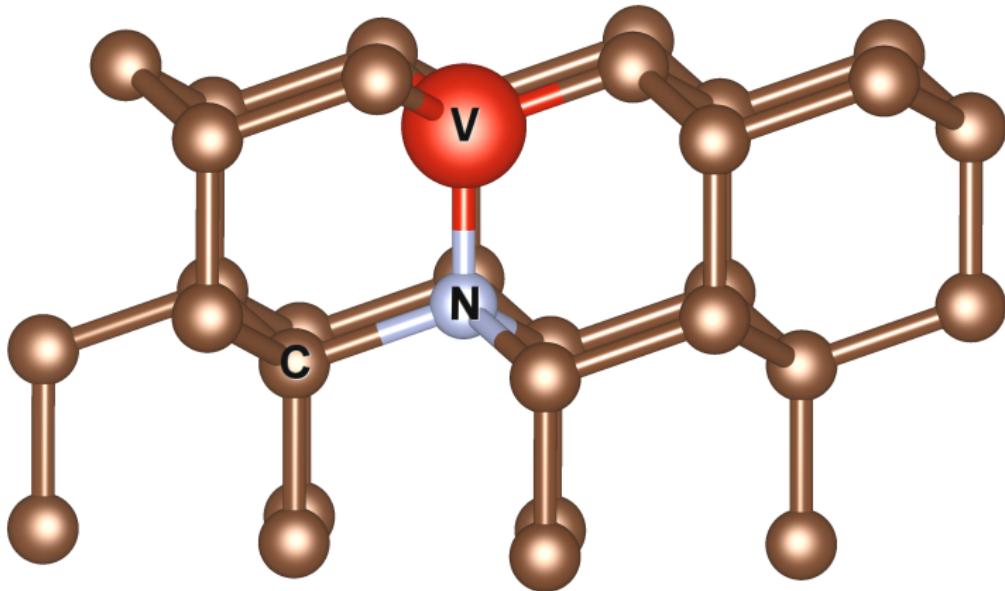
Aims and motivation

- 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.
- Search for new defects with tailored properties which are useful for different applications.

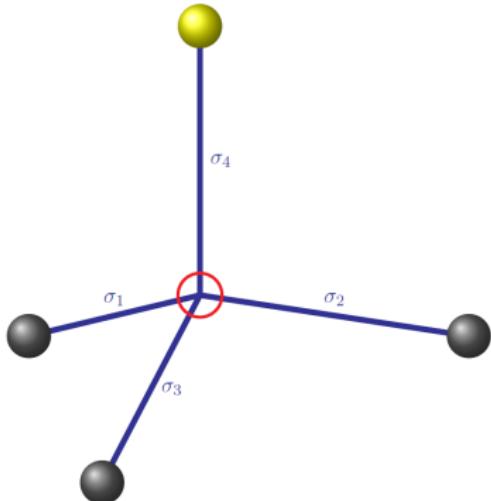


Introduction

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 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

$e(1)$ ————— $e(2)$

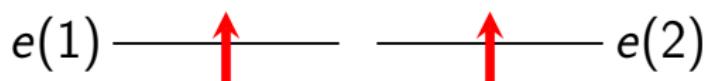
$a_1(2)$ —————

$a_1(1)$ —————



Basic level overview: Ground State

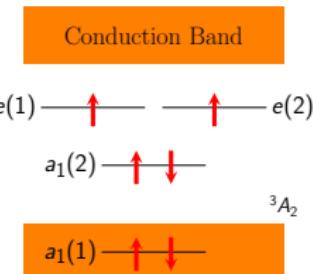
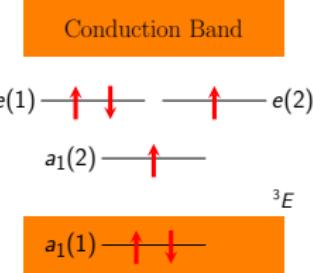
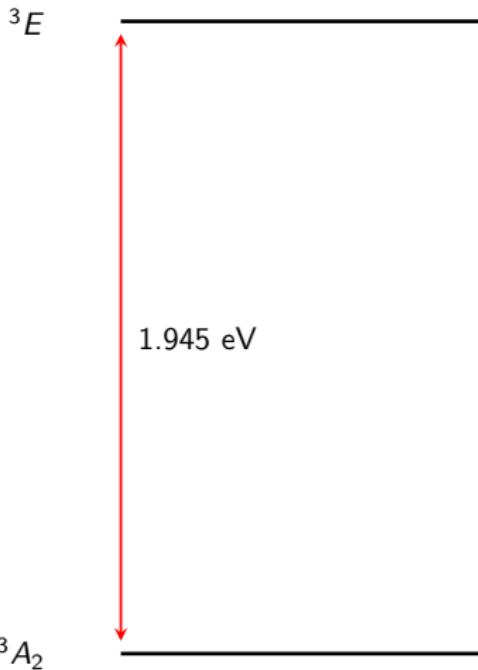
Conduction Band



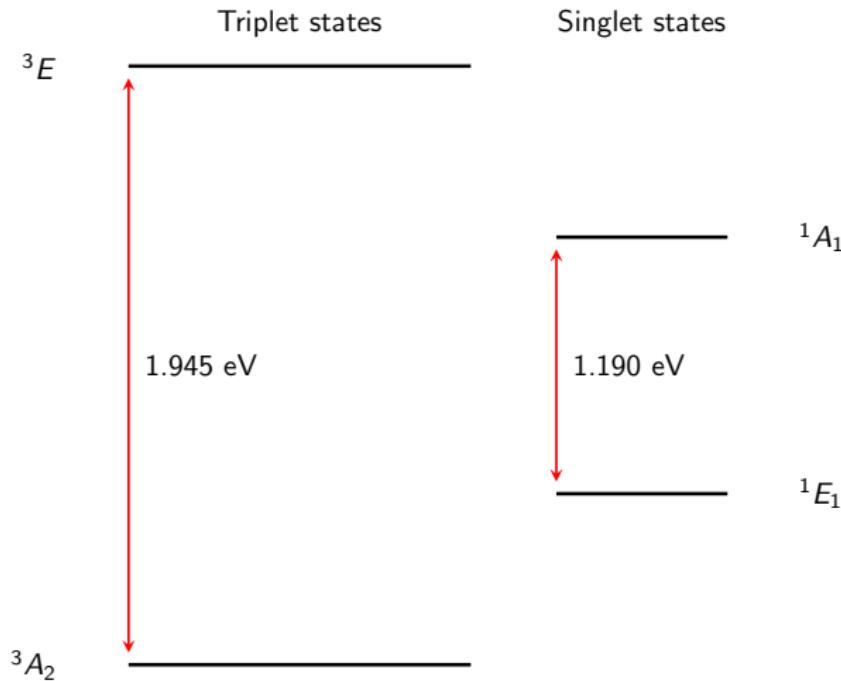
3A_2



Triplets overview: NV⁻

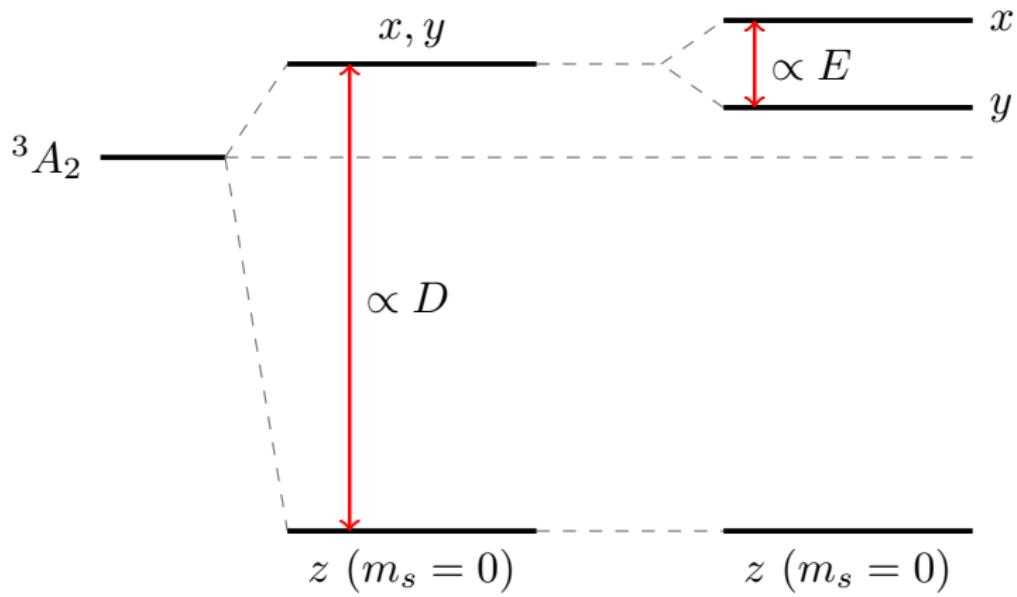


Main level overview: NV⁻

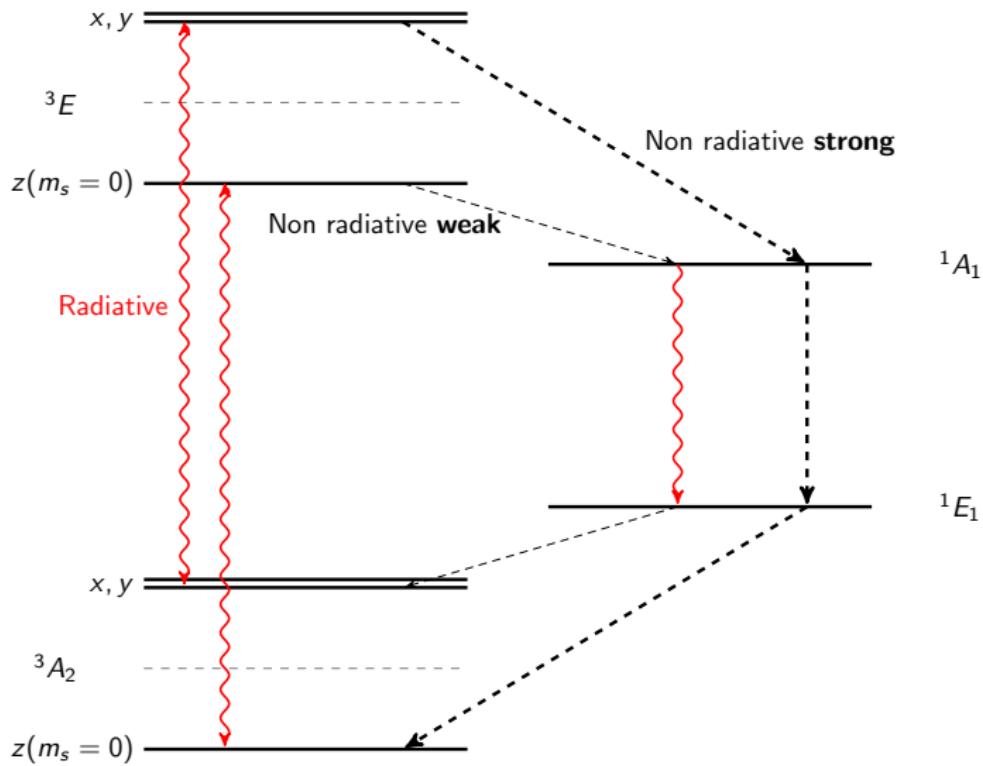


Zero Field Splitting (ZFS)

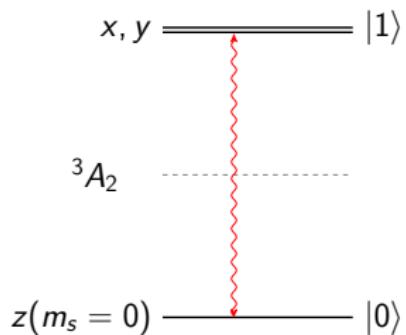
Spin-Spin/Zeeman/strain
 $(\hat{\mathbf{S}}^t \mathbf{D} \hat{\mathbf{S}})$



Transitions overview: NV⁻

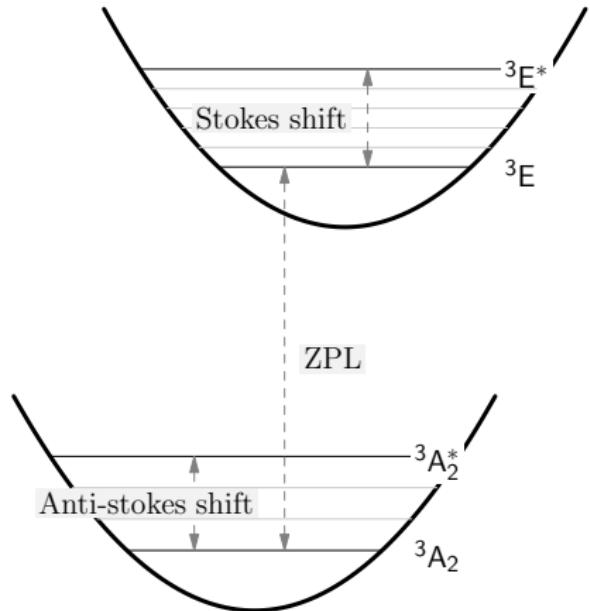
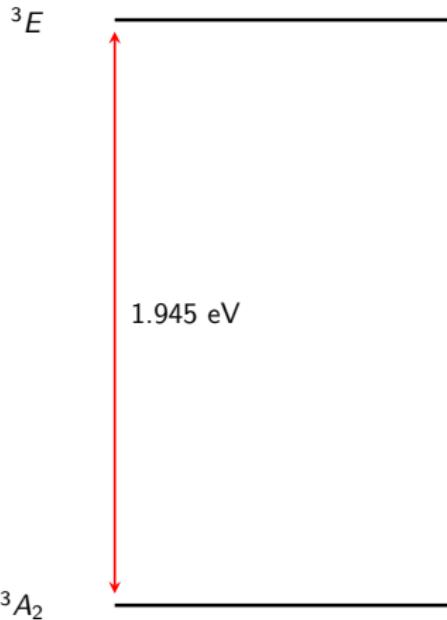


Transitions overview: NV⁻



- 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 properties**.
- All quantities are written in terms of ρ (functional formalism).
- E.g.:

$$E[\rho] =$$

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

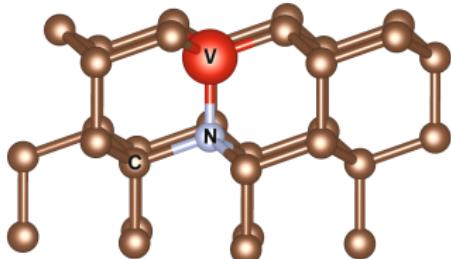
The exchange correlation potential $E_{\text{xc}}^{\text{exact}}[\rho, \nabla\rho]$ determines the DFT *flavor*. In many calculations we use the so-called **PBE** (*Perdew-Burke-Ernzerhof*) functional.



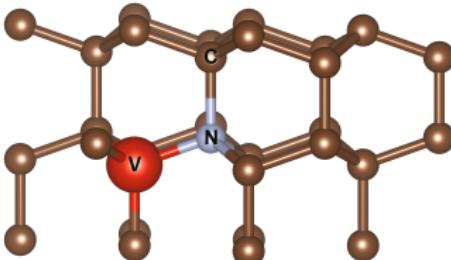
Hexagonal diamond and defects

Defected hexagonal diamond

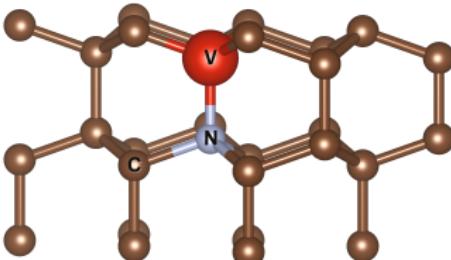
Cubic diamond



Hexagonal x-type

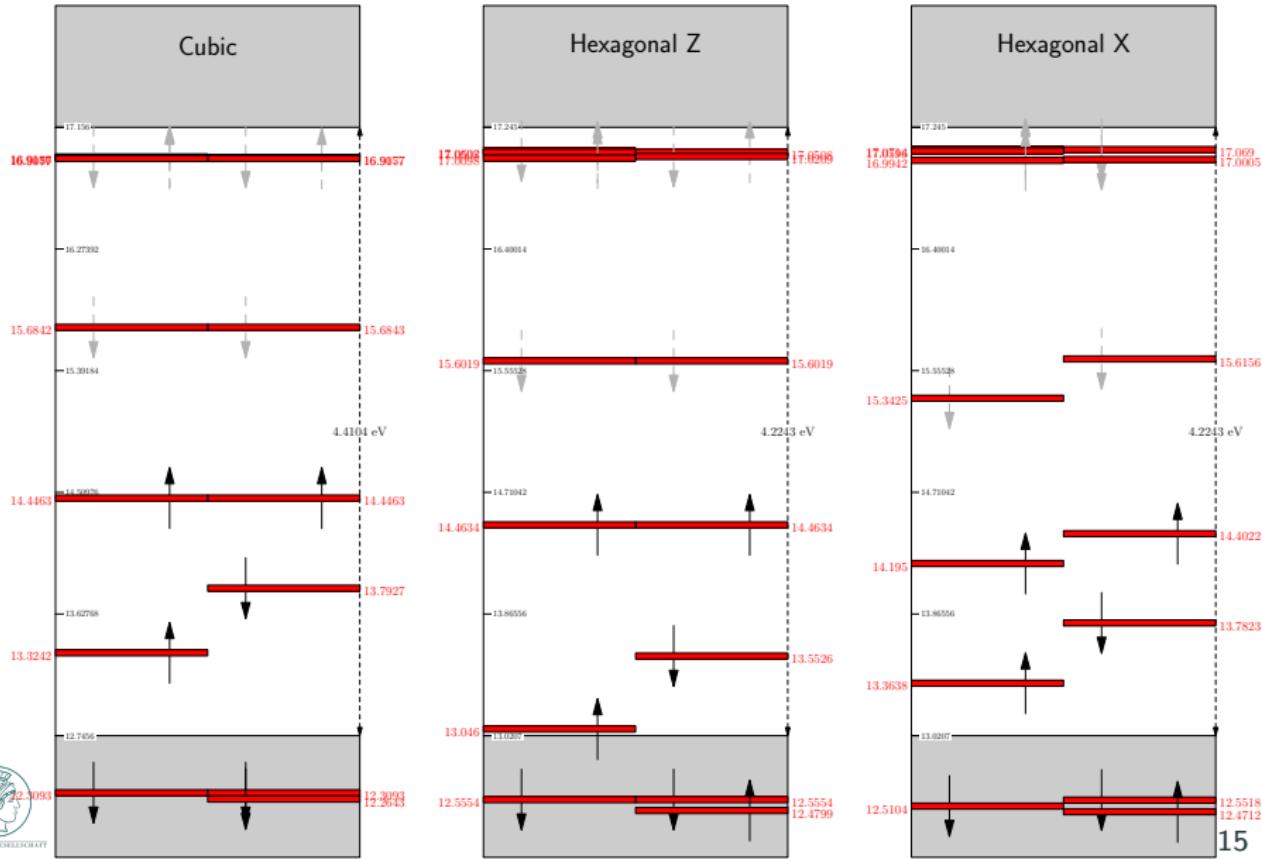


Hexagonal z-type

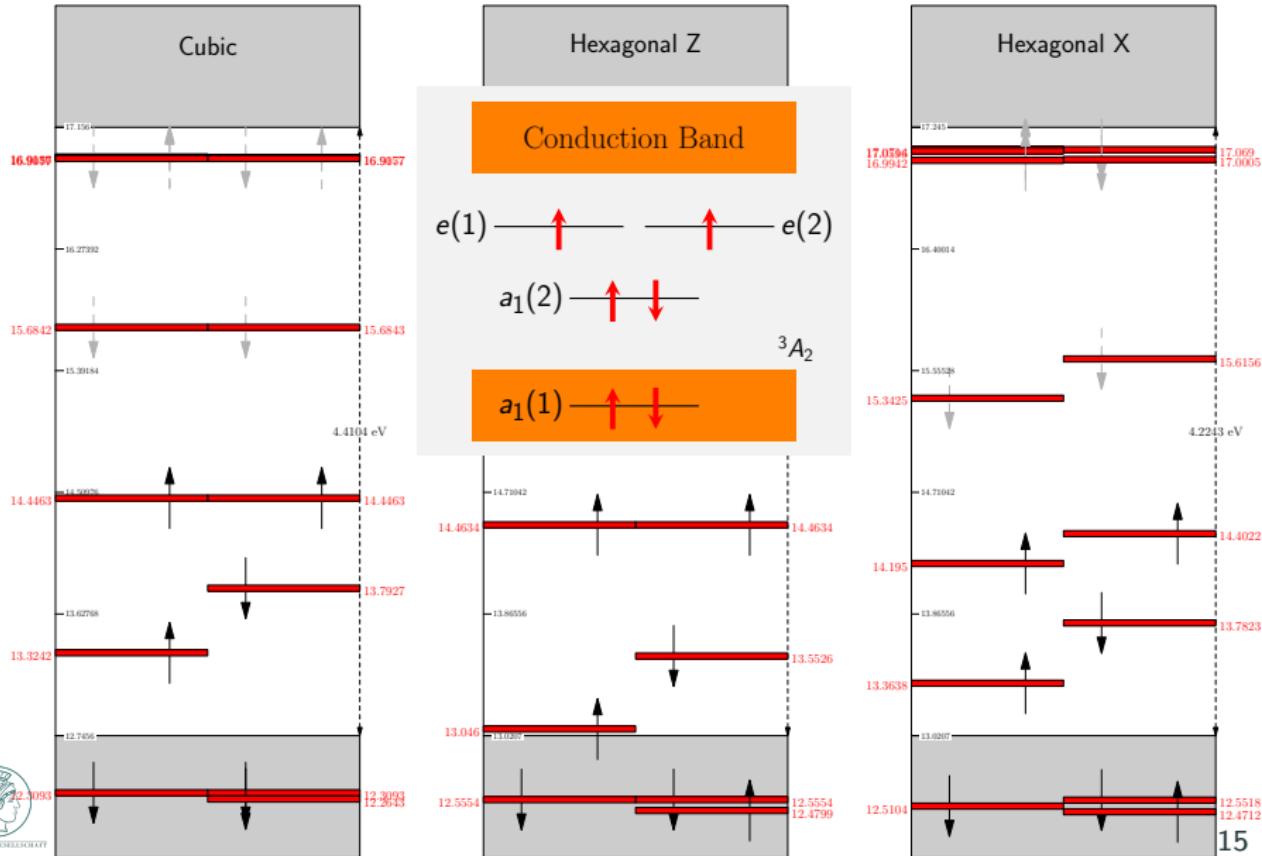


Results

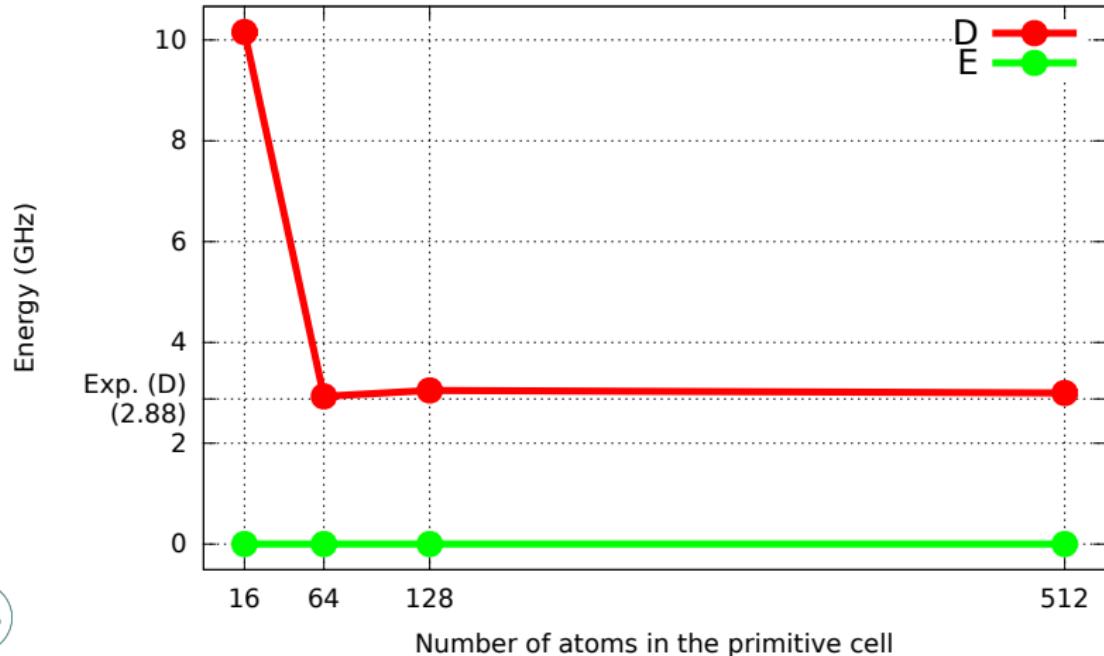
NV⁻: Ground state 3A_2



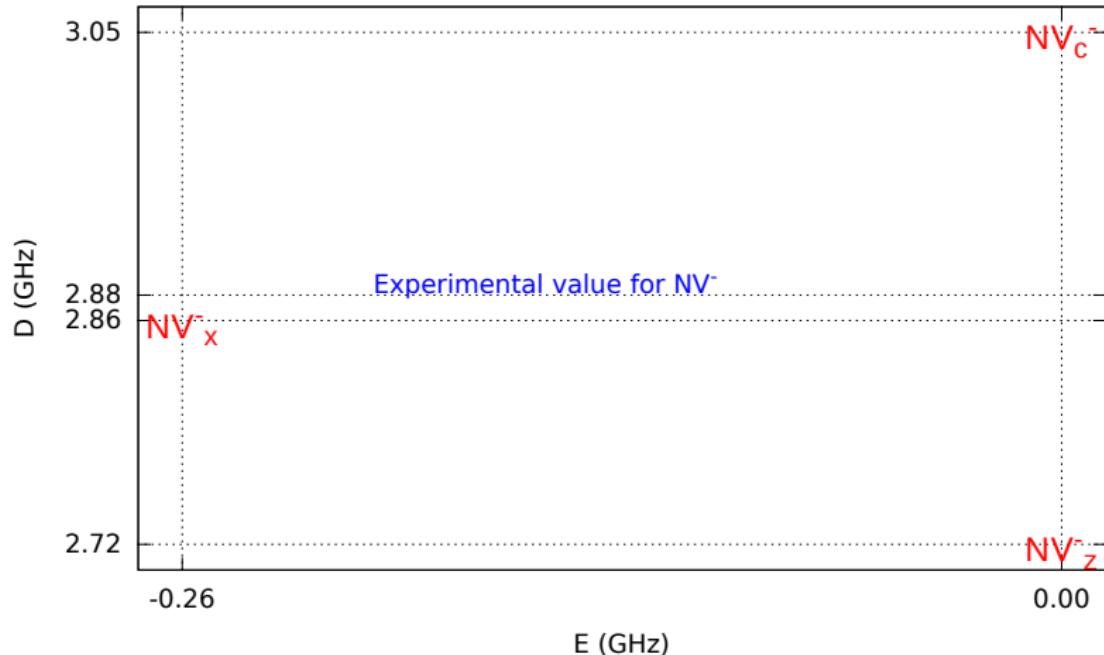
NV⁻: Ground state 3A_2



- 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							
							2 4.0025 He Helium
13 IIIA	14 IVA	15 VA	16 VIA	17 VIIA			
5 10.811 B Boron	6 12.011 C Carbon	7 14.007 N Nitrogen	8 15.999 O Oxygen	9 18.998 F Flourine	10 20.180 Ne Neon		
13 26.982 Al Aluminium	14 28.086 Si Silicon	15 30.974 P Phosphorus	16 32.065 S Sulphur	17 35.453 Cl Chlorine	18 39.948 Ar Argon		
31 69.723 Ga Gallium	32 72.64 Ge Germanium	33 74.922 As Arsenic	34 78.96 Se Selenium	35 79.904 Br Bromine	36 83.8 Kr Krypton		
49 114.82 In Indium	50 118.71 Sn Tin	51 121.76 Sb Antimony	52 127.6 Te Tellurium	53 126.9 I Iodine	54 131.29 Xe Xenon		
81 204.38 Tl Thallium	82 207.2 Pb Lead	83 208.98 Bi Bismuth	84 209 Po Polonium	85 210 At Astatine	86 222 Rn Radon		
113 284 Uut Ununtrium	114 289 Uug Ununquadium	115 288 Uup Ununpentium	116 293 Uuh Ununhexium	117 292 Uus Ununseptium	118 294 Uuo Ununoctium		



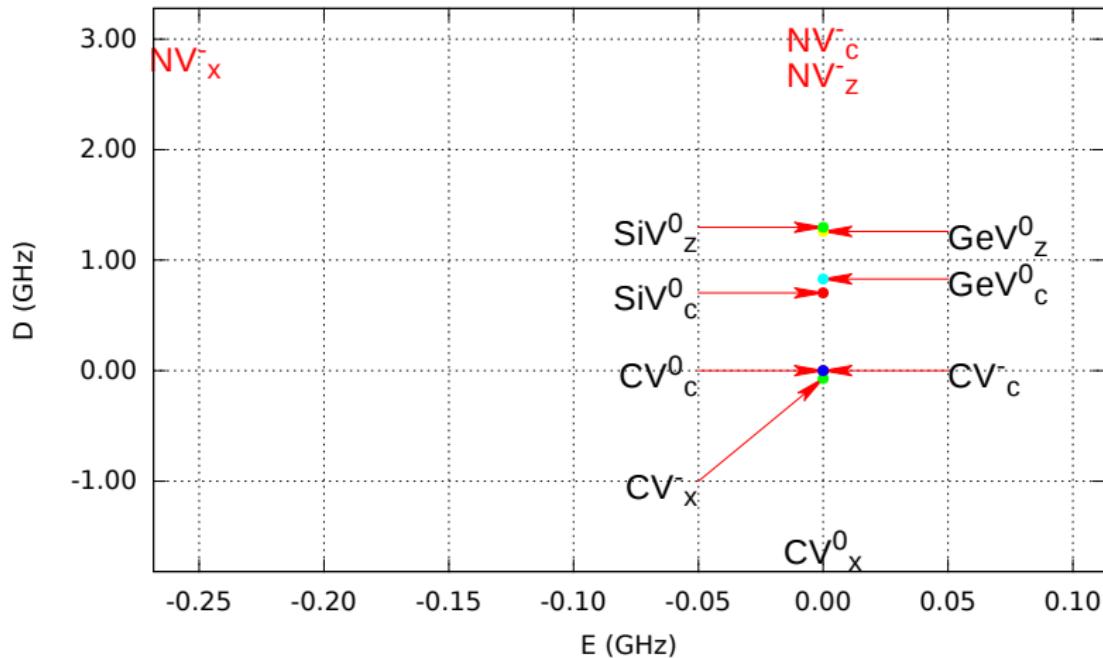
Expanding the defect

14 IVA 15 VA

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



ZFS map



Summary and outlook

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

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



Thank you!

