

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

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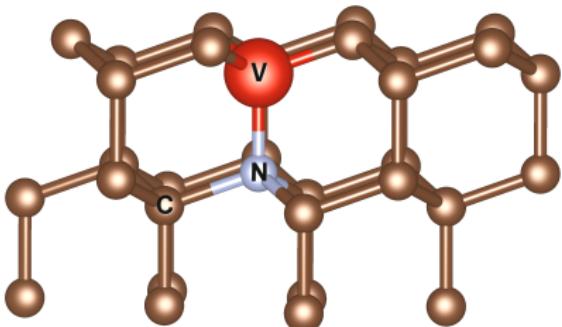


## **Aim and scope of the work**

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# Aims and motivation

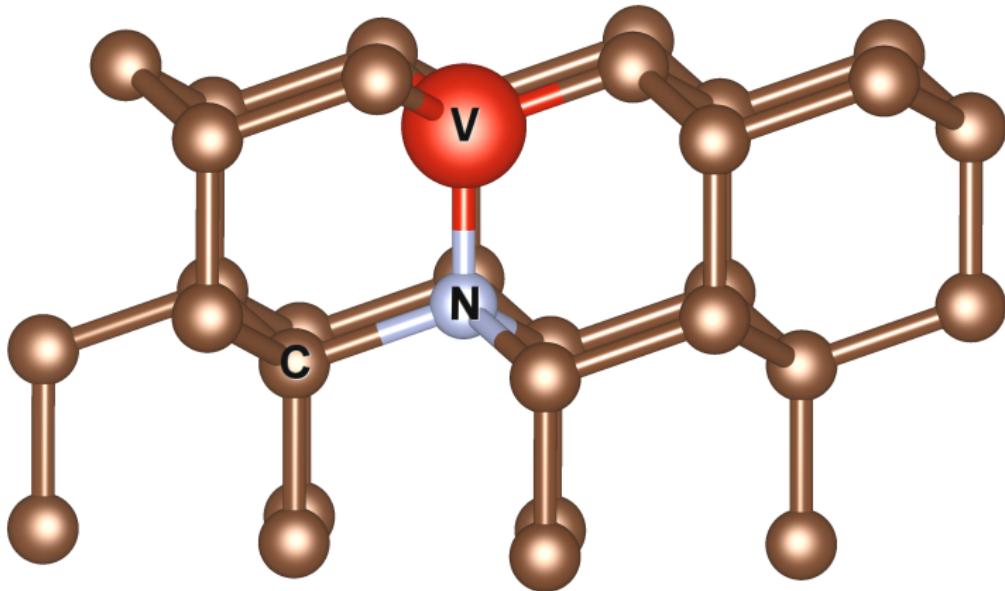
- Exploration of defect centres using state-of-the-art ab-initio theories.
- Benchmark ab initio theories with well-known experimental data.
- Systematic characterisation of defect fingerprints.
- Search for new defects with tailored properties which are useful for different applications.



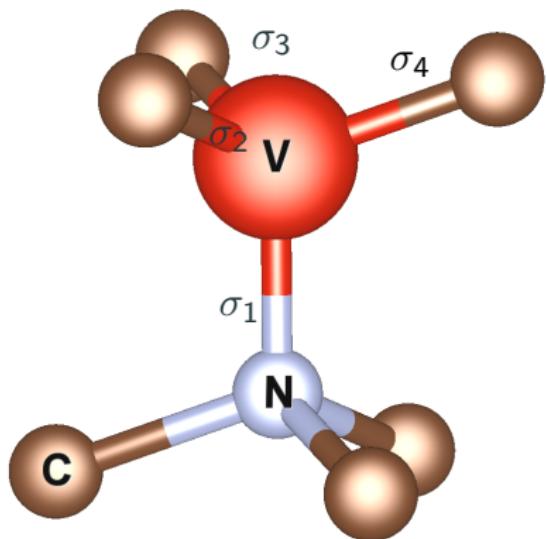
# Introduction

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# 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  $\sigma_i$  we obtain 4 levels  $a_1(1), a_1(2), e(1), e(2)$  classified according to their symmetry.

# Basic level overview

Conduction Band

$e(1)$  —————  $e(2)$

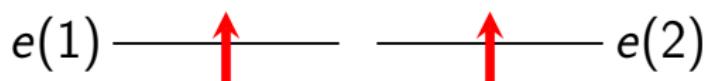
$a_1(2)$  —————

$a_1(1)$  —————



# Basic level overview: Ground State

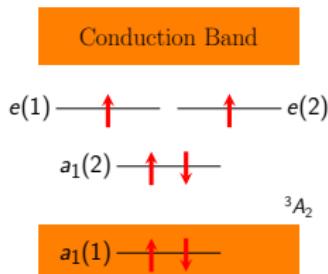
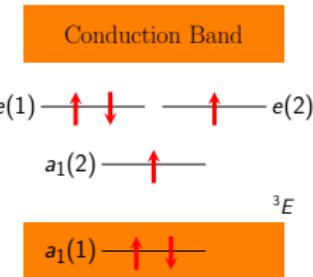
Conduction Band



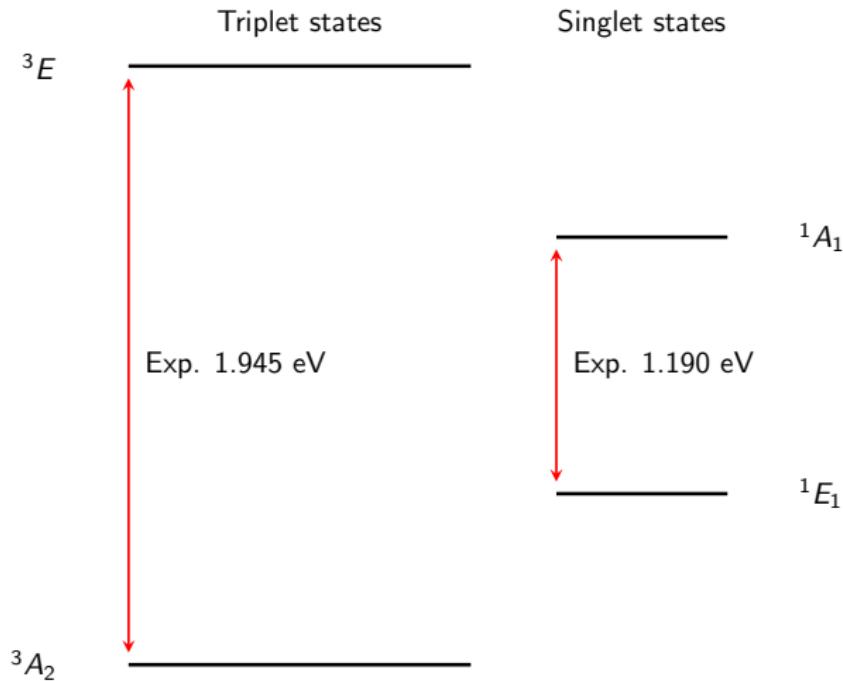
$^3A_2$



# Triplets overview: NV<sup>-</sup>

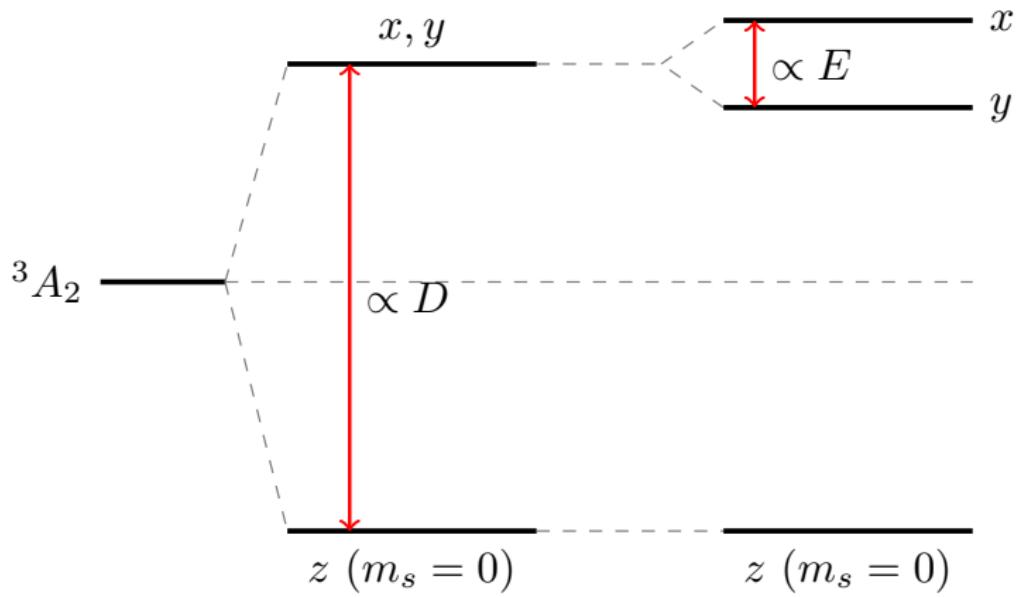


# Main level overview: NV<sup>-</sup>

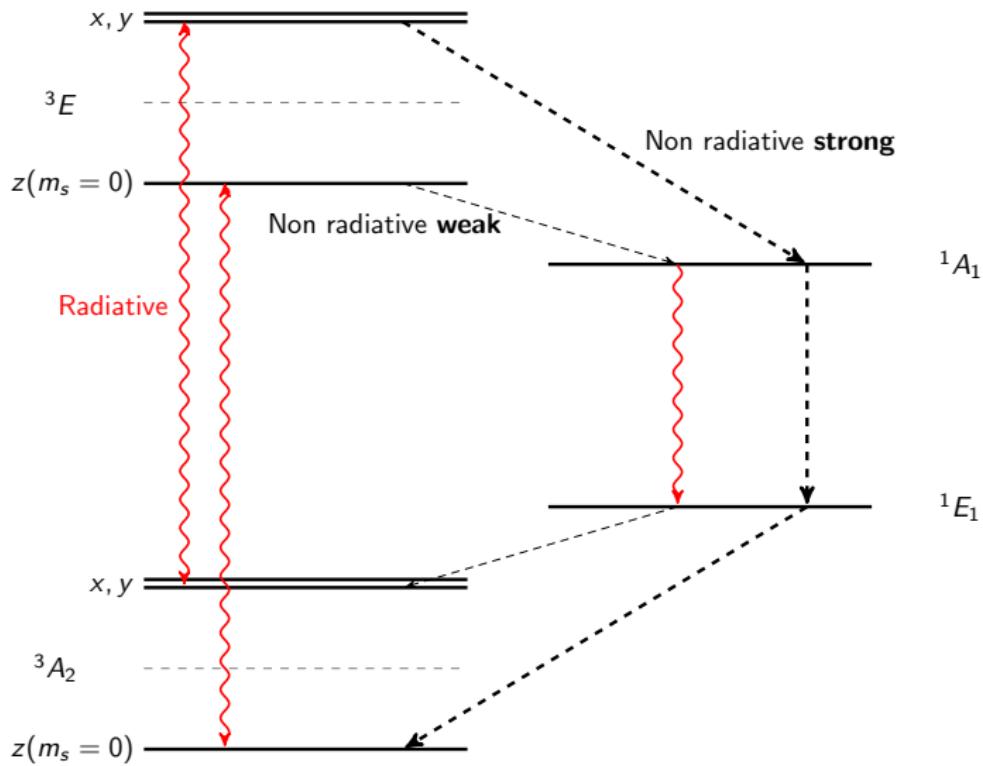


# Zero Field Splitting (ZFS)

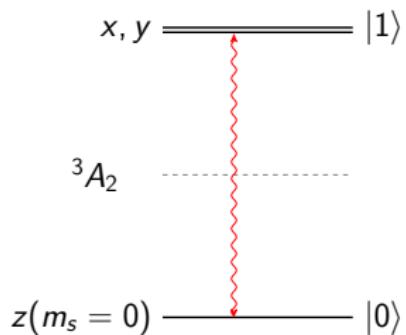
Spin-Spin/Zeeman/strain  
 $(\hat{\mathbf{S}}^t \mathbf{D} \hat{\mathbf{S}})$



# Transitions overview: NV<sup>-</sup>

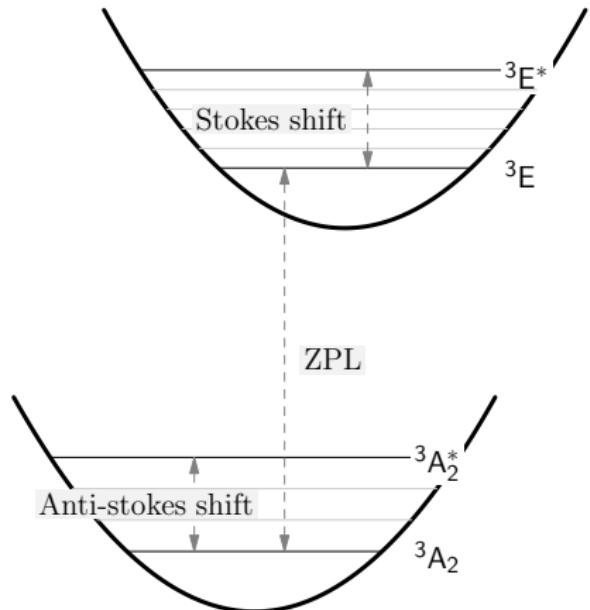
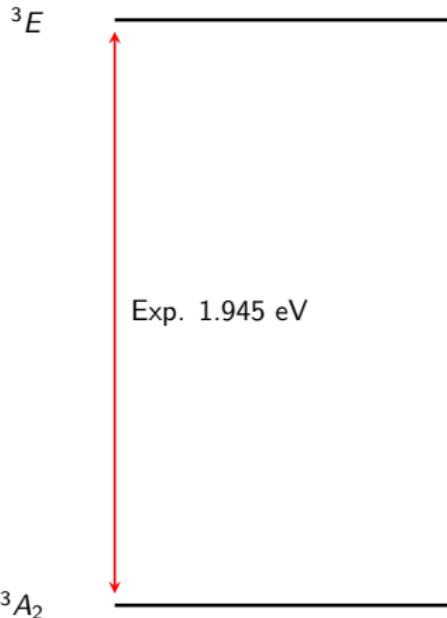


# Transitions overview: NV<sup>-</sup>



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

# Zero phonon line (ZPL)



# Density functional theory (DFT)

*" $\Psi$  contains too much information"*

- Popular saying

- In principle, DFT delivers the **exact ground state properties**.
- All quantities are written in terms of  $\rho$  (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.

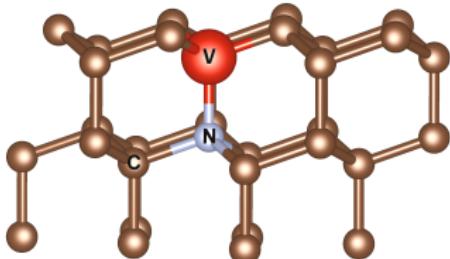


## **Hexagonal diamond and defects**

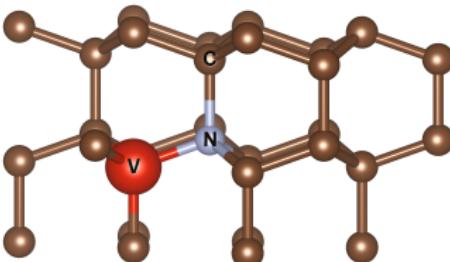
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# Defect hexagonal diamond

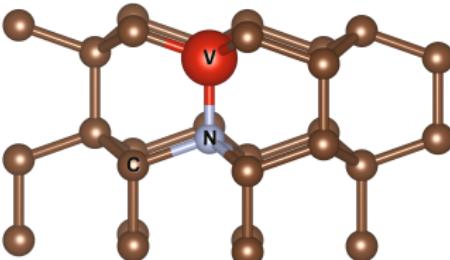
Cubic diamond



Hexagonal x-type



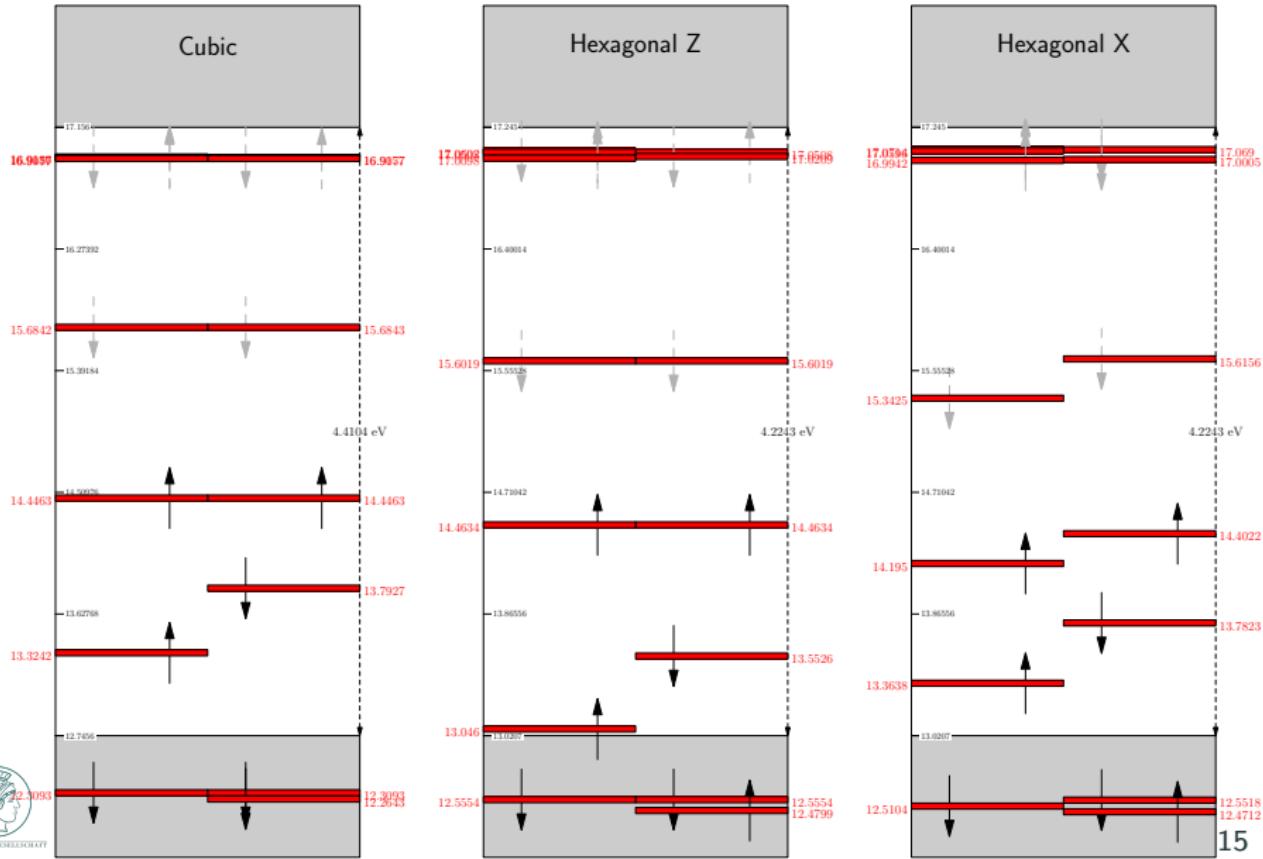
Hexagonal z-type



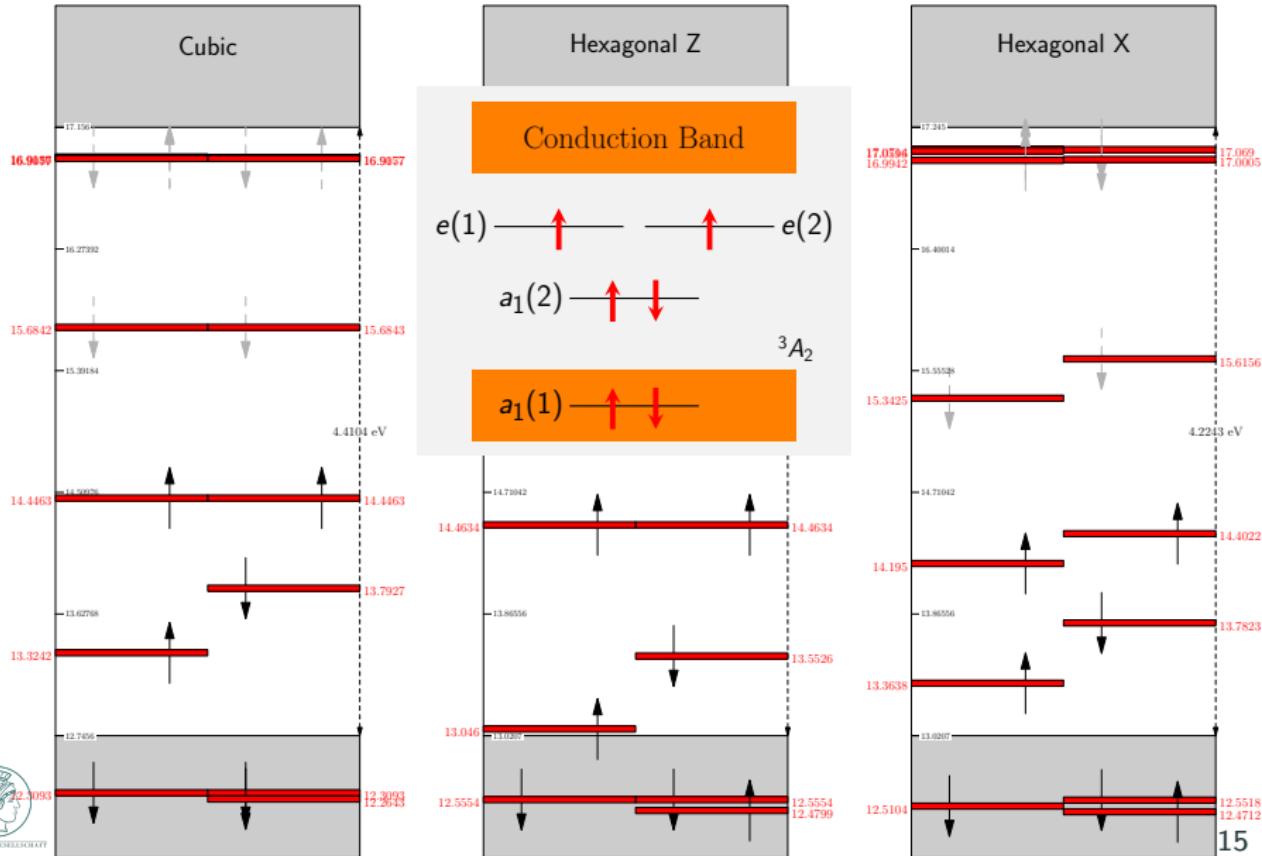
## Results

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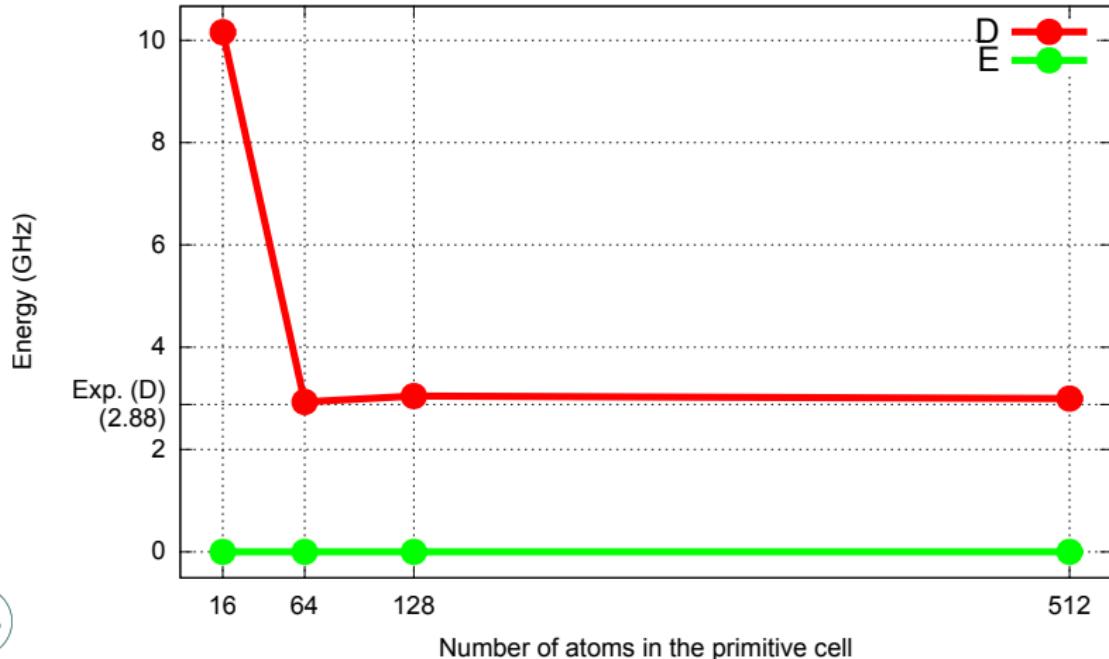
# NV<sup>-</sup>: Ground state $^3A_2$



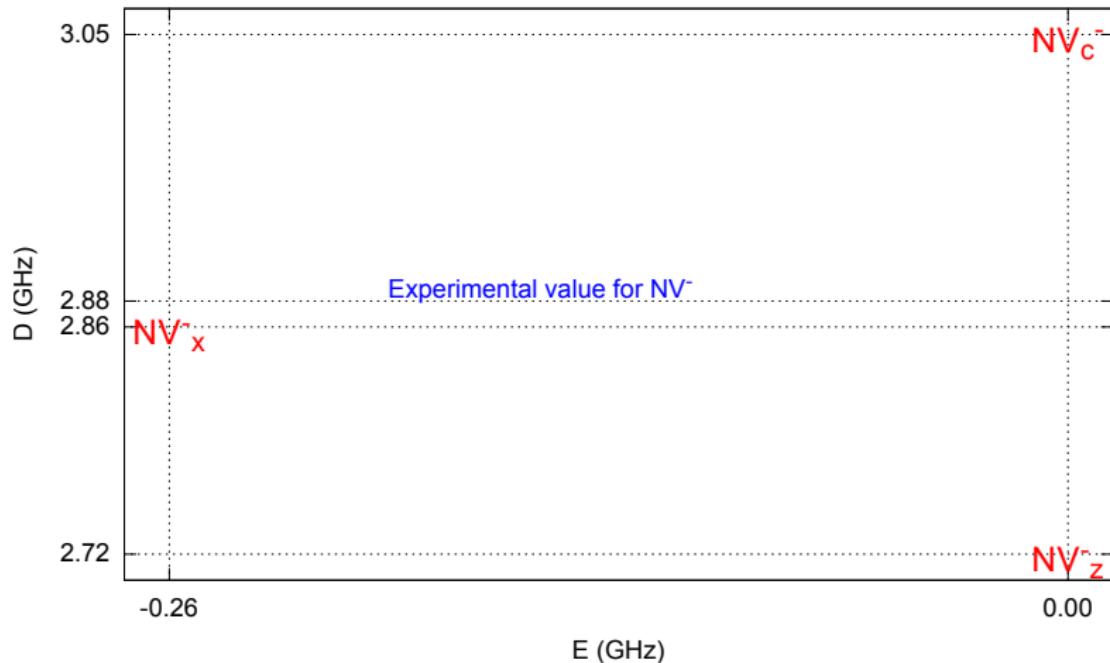
# NV<sup>-</sup>: Ground state $^3A_2$



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



# NV<sup>-</sup>: ZFS cubic ( $\text{NV}_c^-$ ), Hexagonal $x, z$ ( $\text{NV}_{x,z}^-$ )



# Expanding the defect: XV

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



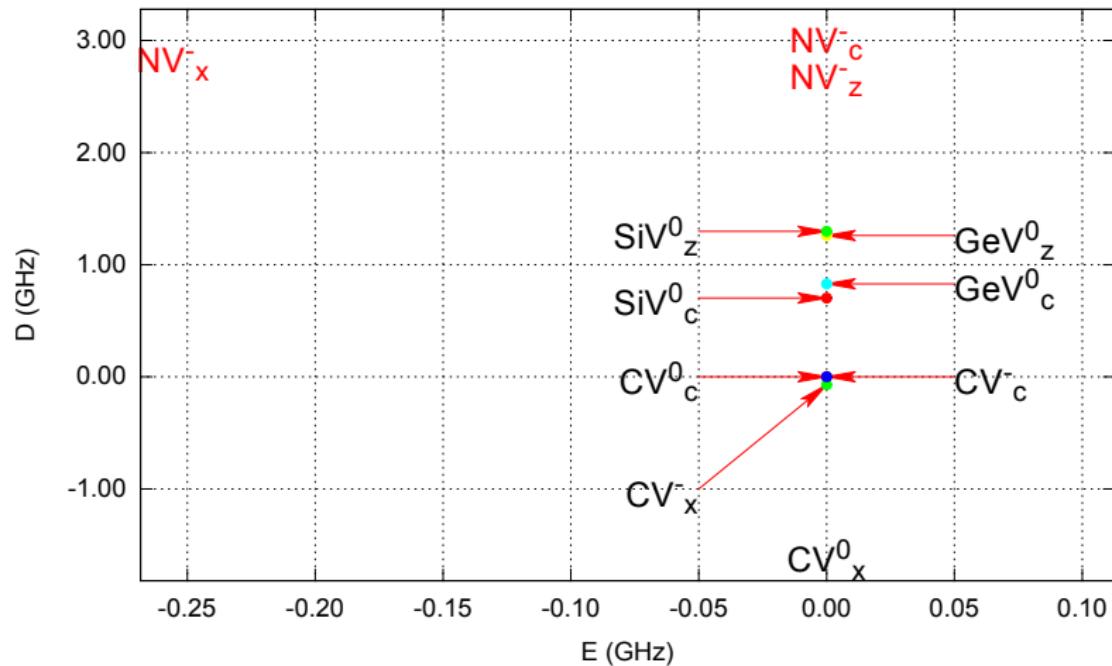
# Expanding the defect

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**

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# 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!**

