

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

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# Contents

Aim and scope of the work

Introduction

Hexagonal diamond and defects

Results

Summary



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

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# Aims

- Exploration of defect centres using state-of-the-art ab-initio theories.

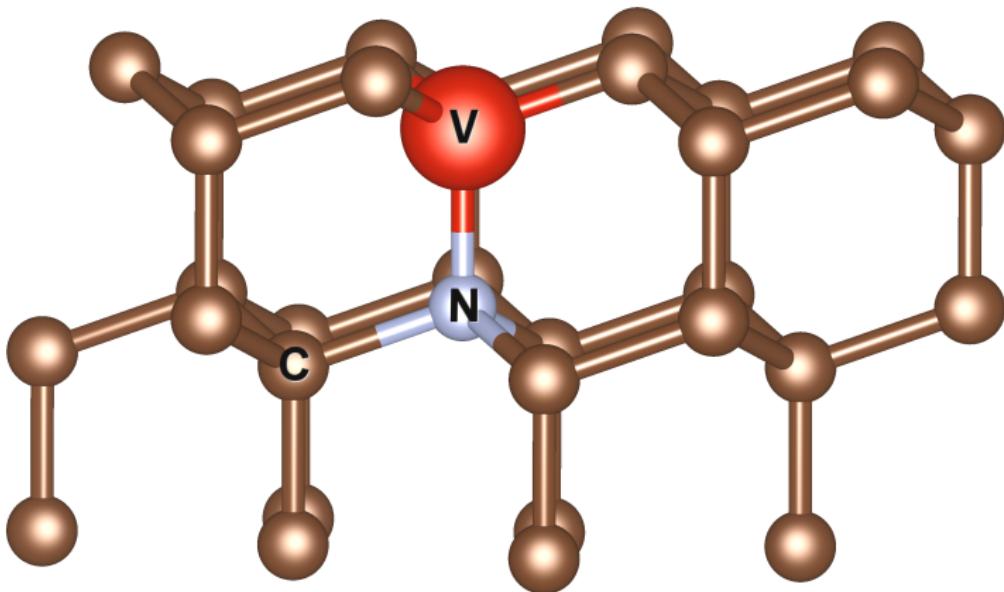
# Aims

- Exploration of defect centres using state-of-the-art ab-initio theories.
- Reproduce and obtain classical results for nitrogen vacancy impurity complexes in diamond.

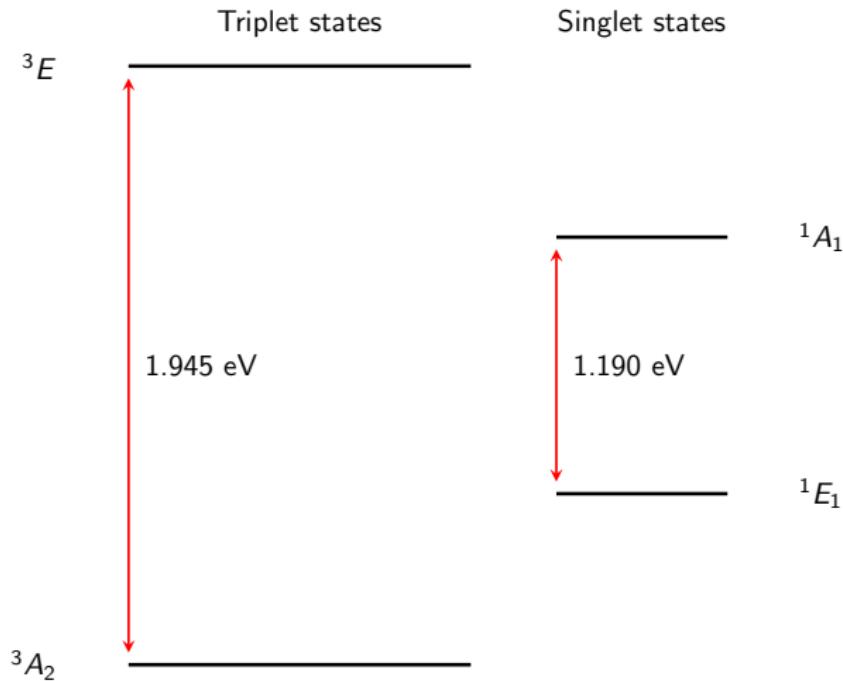
# Introduction

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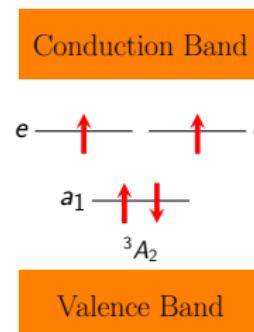
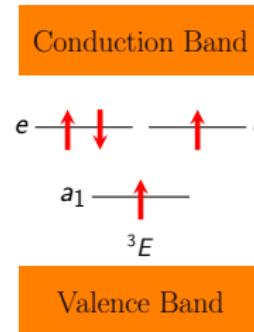
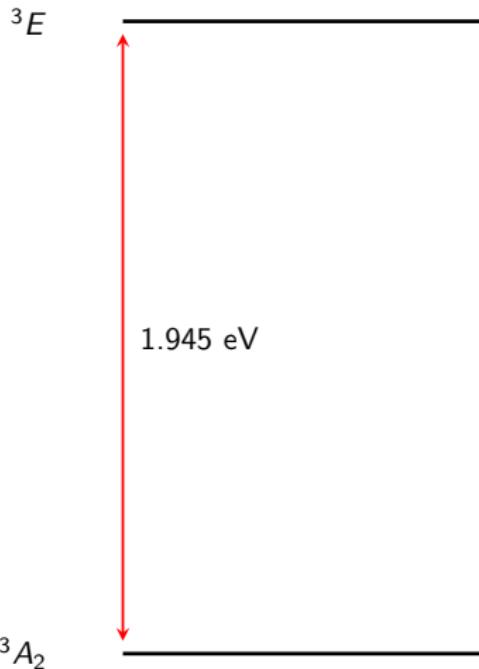
# Nitrogen Vacancy Centre in diamond



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



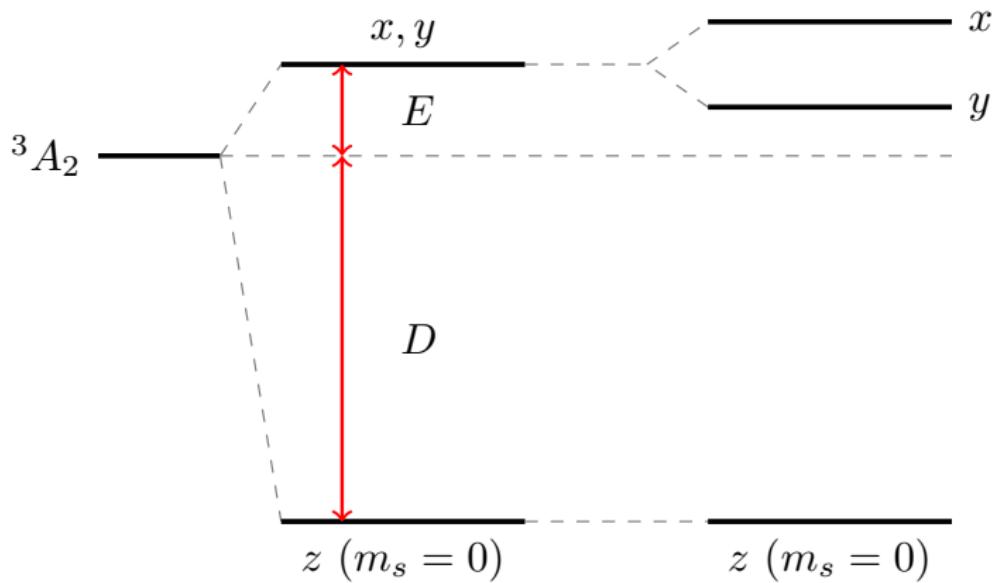
# Triplets overview: NV<sup>-</sup>



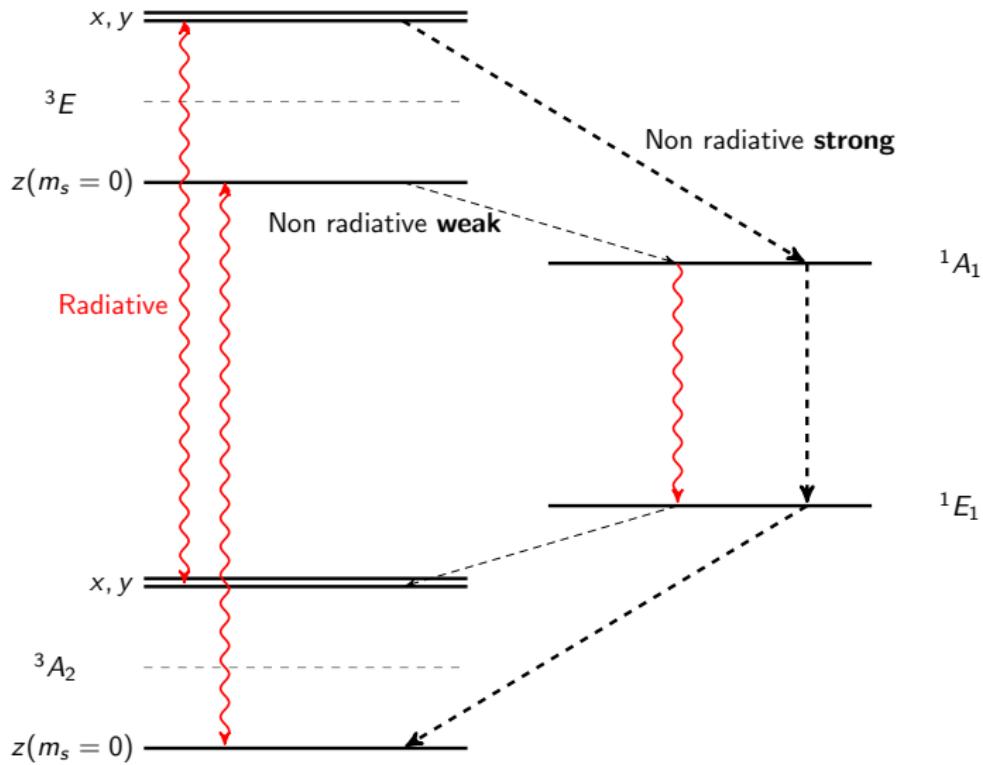
# Transitions

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

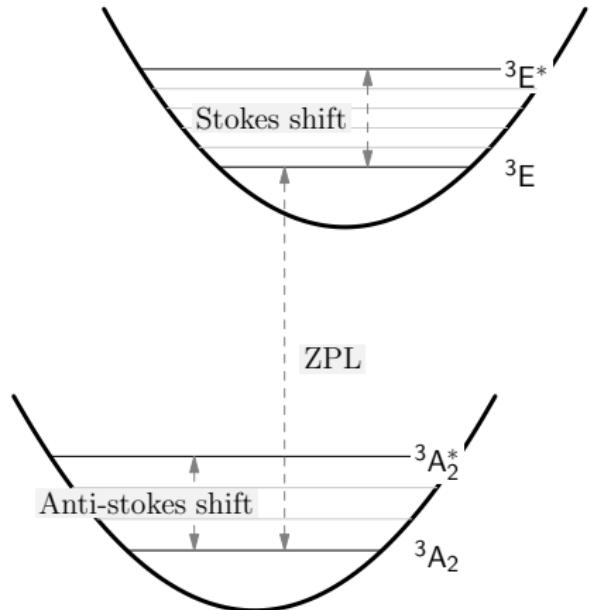
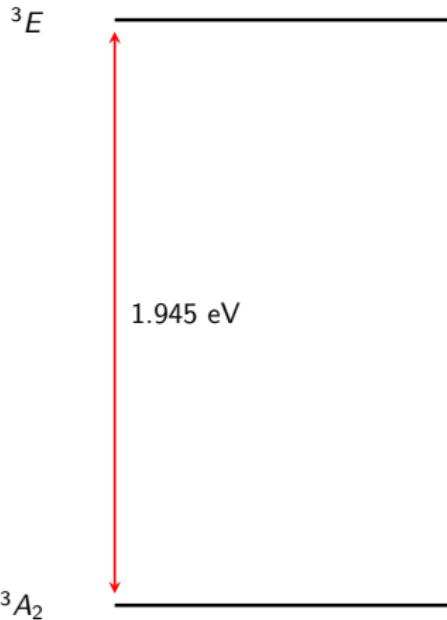
Zeeman/strain



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



# Zero phonon line (ZPL)



# Density functional theory (DFT)

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

- Popular saying

- The exact electronic ground state of a system is only dependent on the electronic density  $\rho$ .
- In principle, DFT delivers the **exact ground state**.
- All quantities written in terms of  $\rho$  (functional formalism).
- E.g.:

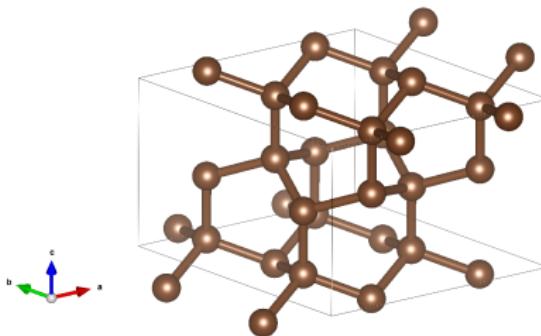
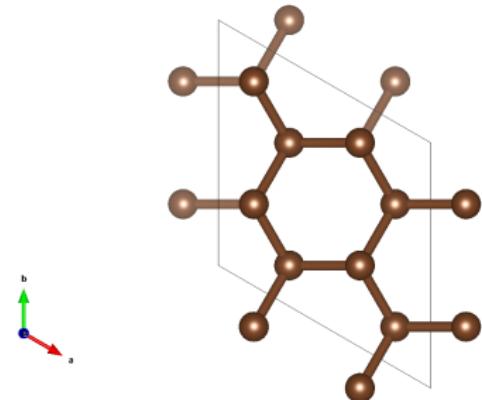
$$E[\rho] = T_s[\rho] + \int V(\mathbf{r})\rho(\mathbf{r}) \, d\mathbf{r} + \frac{1}{4\pi\epsilon_0} \int \frac{\rho(\mathbf{r})\rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} \, d\mathbf{r} d\mathbf{r}' + E_{xc}^{\text{exact}}[\rho]$$

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

## **Hexagonal diamond and defects**

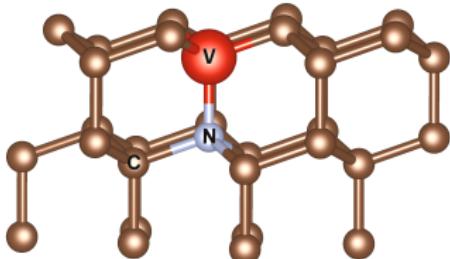
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# Hexagonal diamond

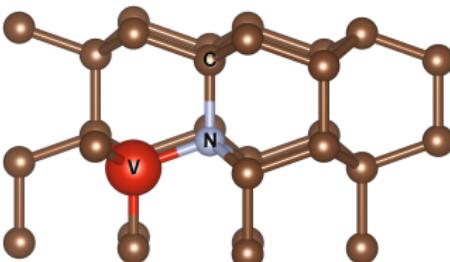


# Defected hexagonal diamond

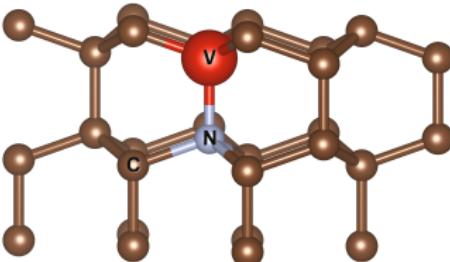
Cubic diamond



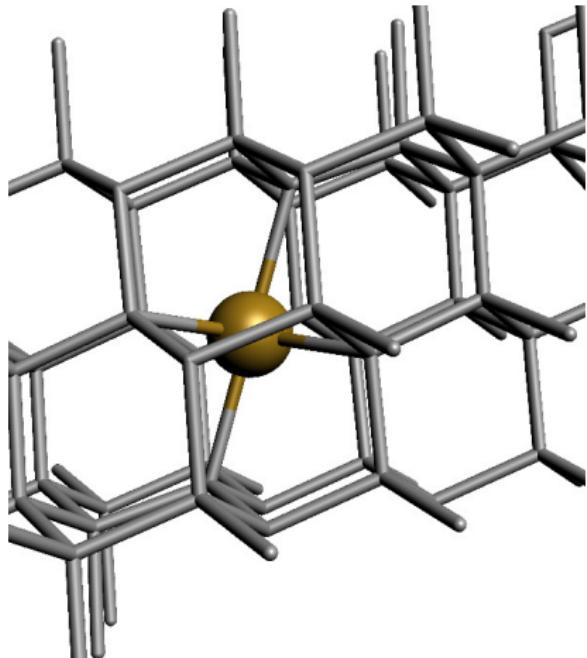
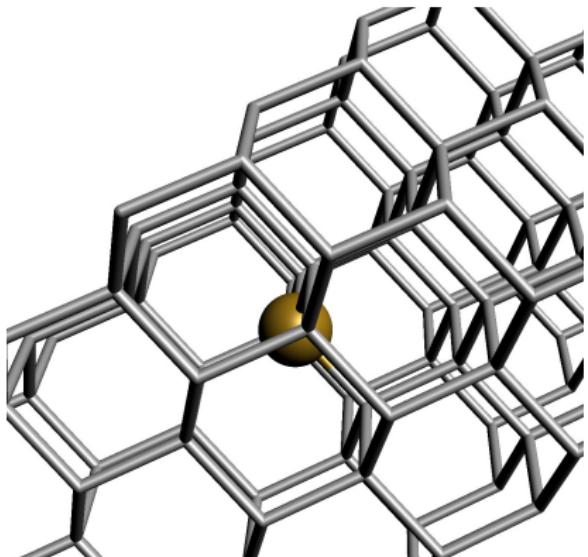
Hexagonal *x*-type



Hexagonal *z*-type



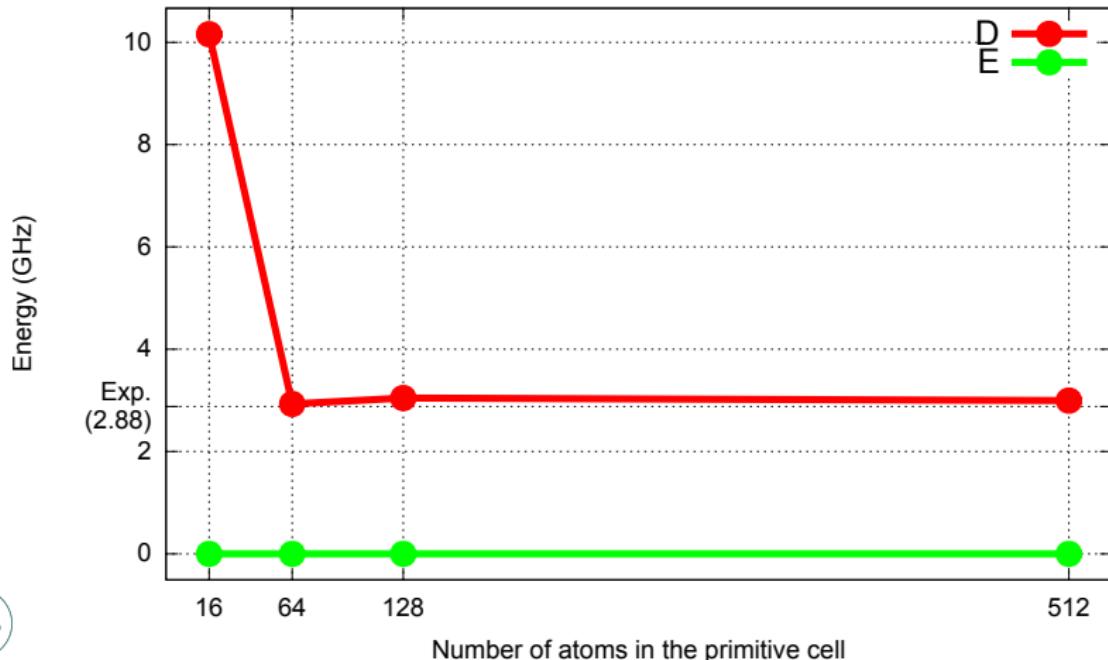
## Split vacancies: SiV<sup>-</sup>



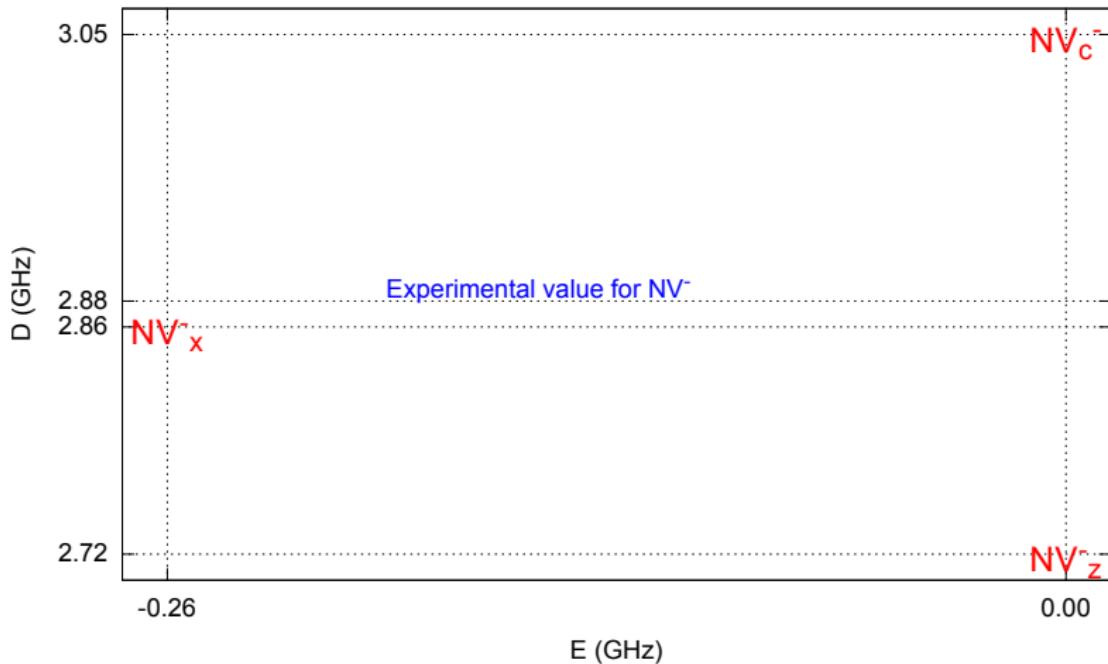
## Results

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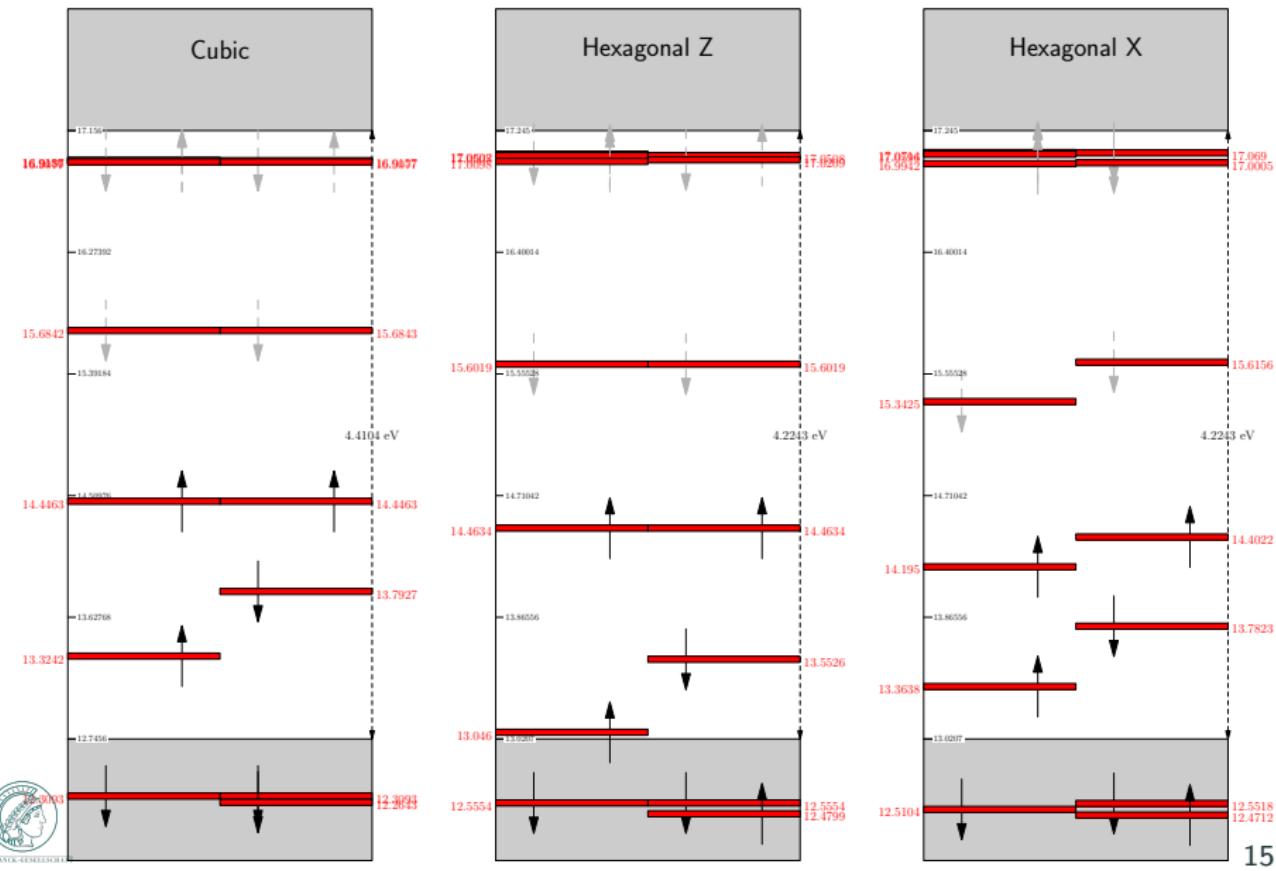
- Cubic diamond, convergence and comparison with the experimental result.



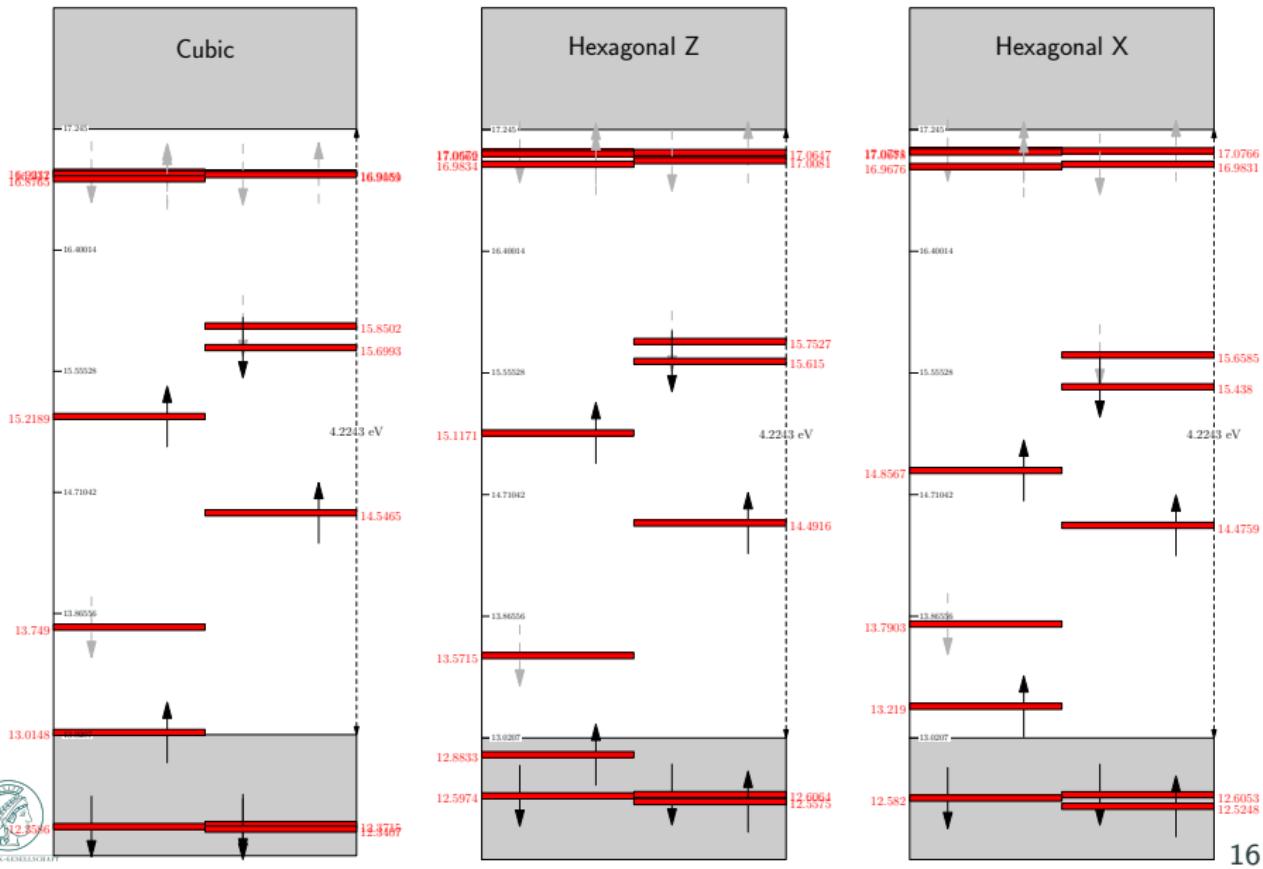
# NV\*: ZFS cubic ( $\text{NV}_c^*$ ), Hexagonal $x, z$ ( $\text{NV}_{x,z}^*$ )



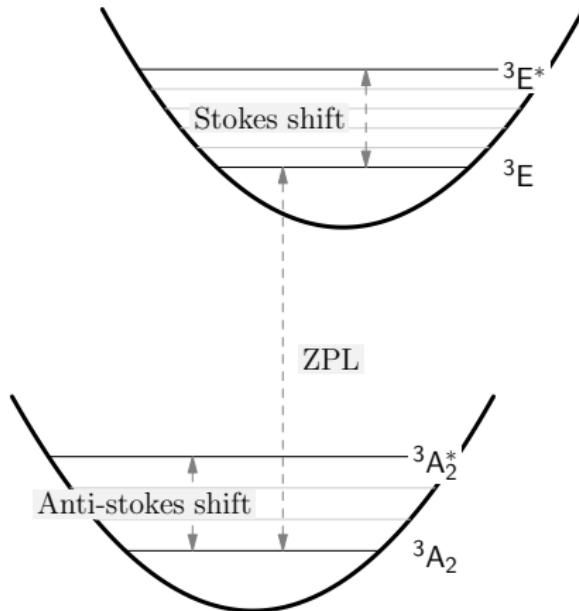
# NV<sup>-</sup>: Ground state $^3A_2$



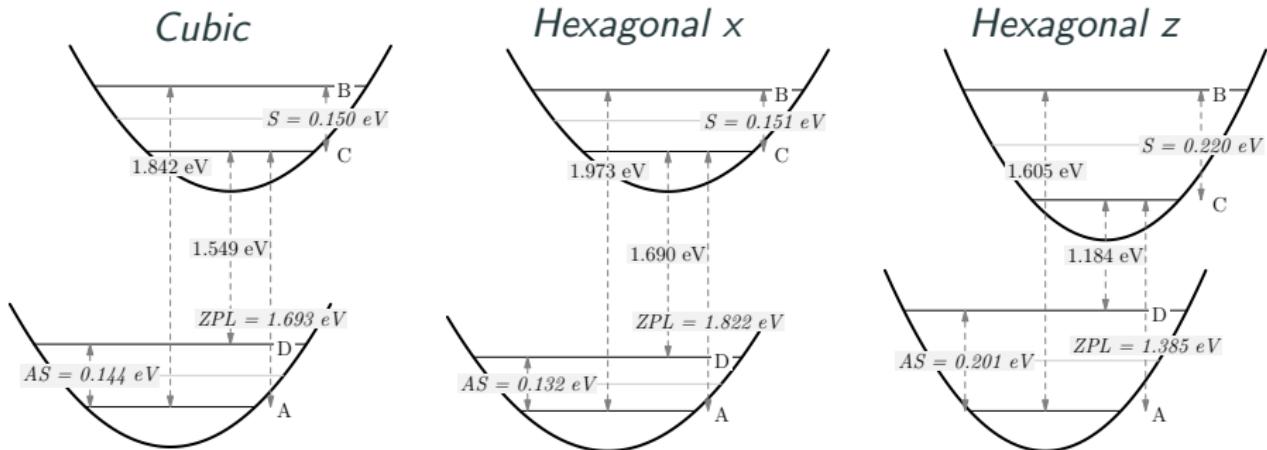
# NV<sup>-</sup>: Excited state $^3E$



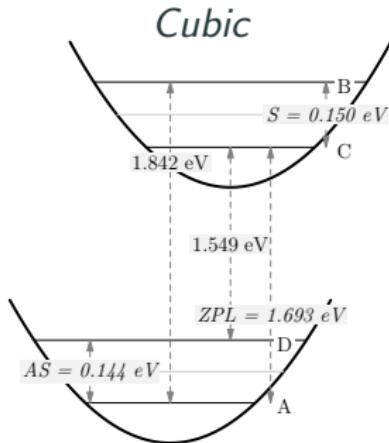
# NV<sup>-</sup>: Vibronic scheme (PBE + 128 atomic cell)



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Experimental data	
ZPL	1.945
${}^3A_2 \rightarrow {}^3E^*$	2.180
$S$	0.235
${}^3E \rightarrow {}^3A_2^*$	1.760
AS	0.185

Calculations	
ZPL	1.693
${}^3A_2 \rightarrow {}^3E^*$	1.842
$S$	0.150
${}^3E \rightarrow {}^3A_2^*$	1.549
AS	0.144

# Expanding the defect

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



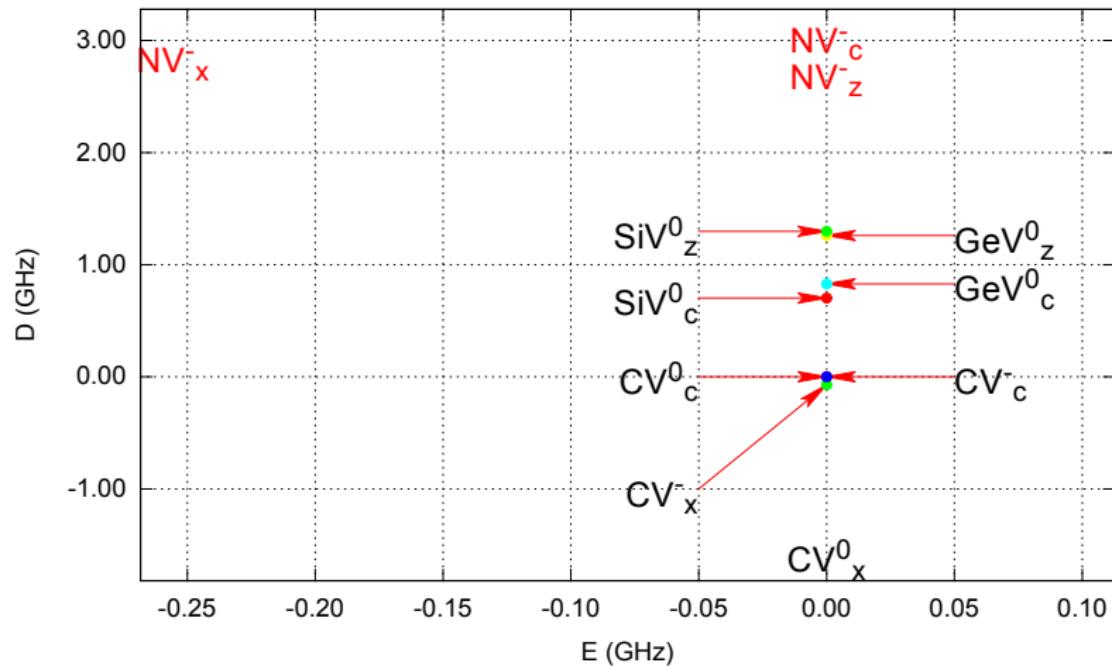
# Expanding the defect

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

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

- Structural properties
- ZPL calculation.
- ZFS tensor calculation.
- **Beyond the Ground state:**  
Using DMRG for excited state calculations



**Thank you!**

