

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

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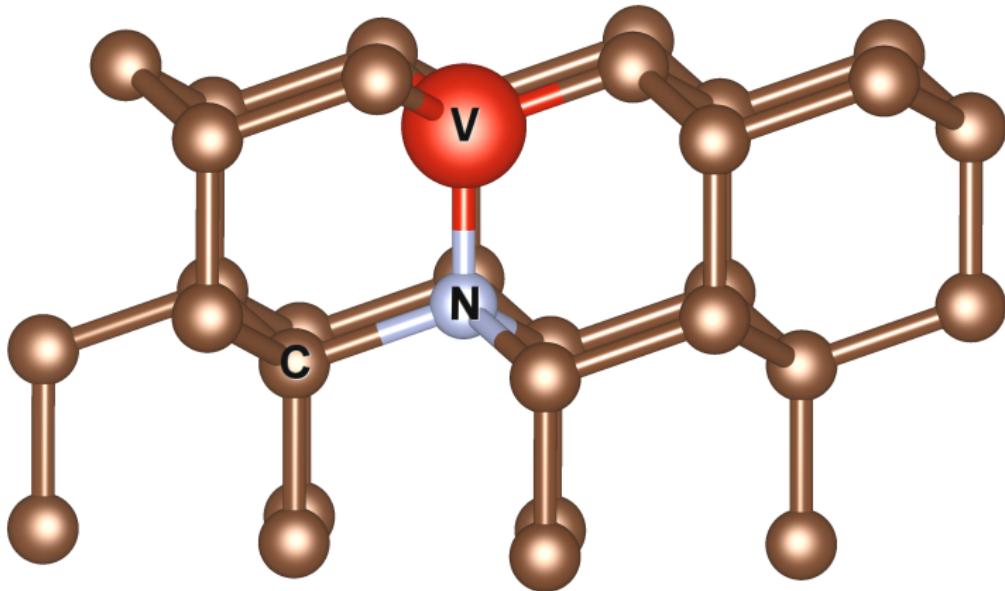
Aim and scope of the work

Aims

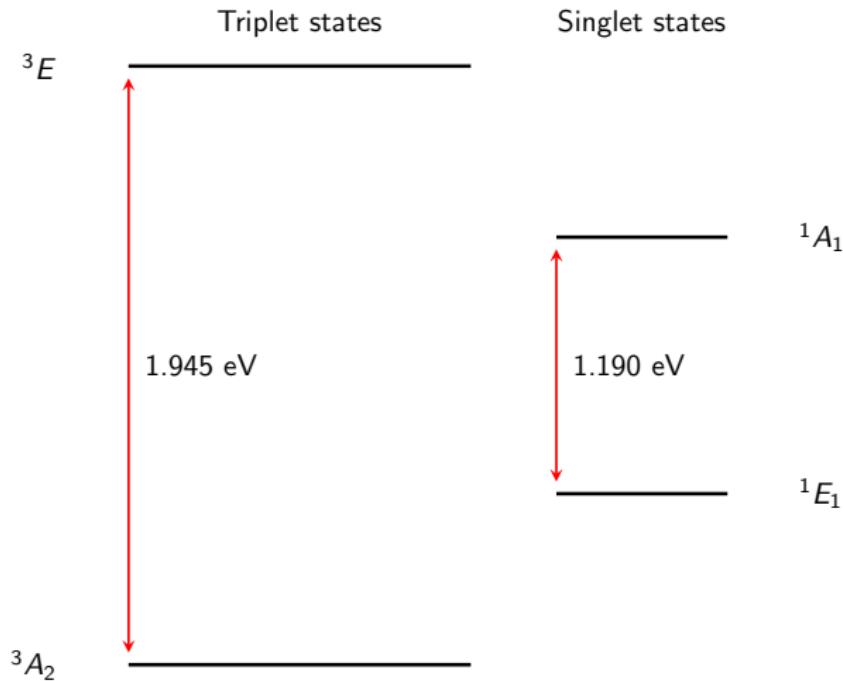
- Exploration of defect centres using state-of-the-art ab-initio theories.
- Reproduce and obtain classical results for nitrogen vacancy impurity complexes in diamond.

Introduction

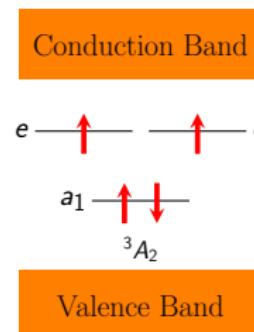
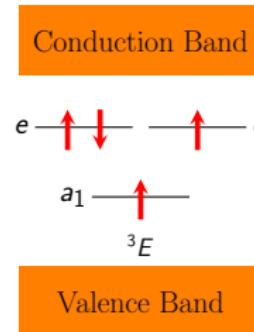
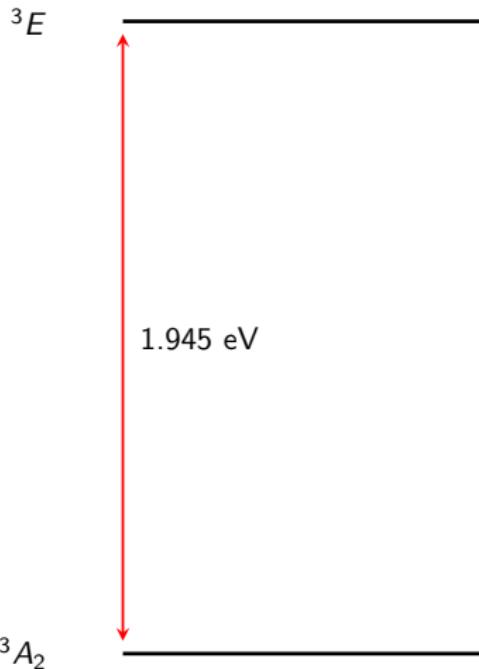
Nitrogen Vacancy Centre in diamond (NV center)



Main level overview: NV⁻



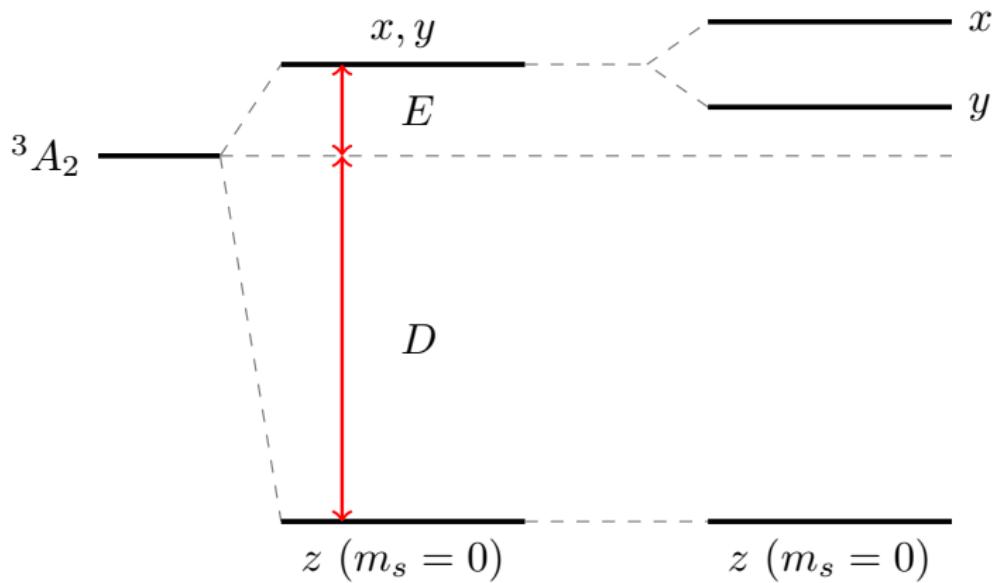
Triplets overview: NV⁻



Zero Field Splitting (ZFS)

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

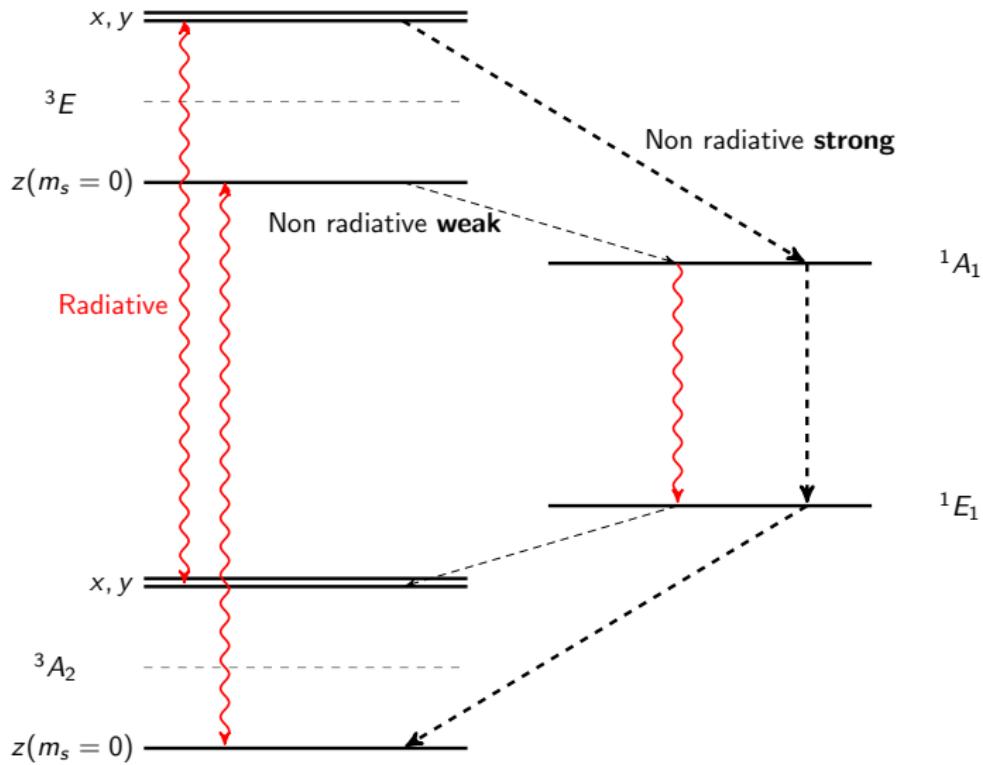
Zeeman/strain



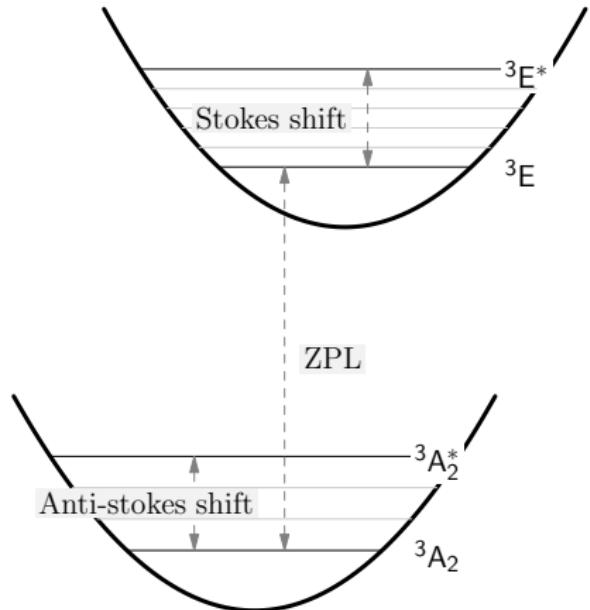
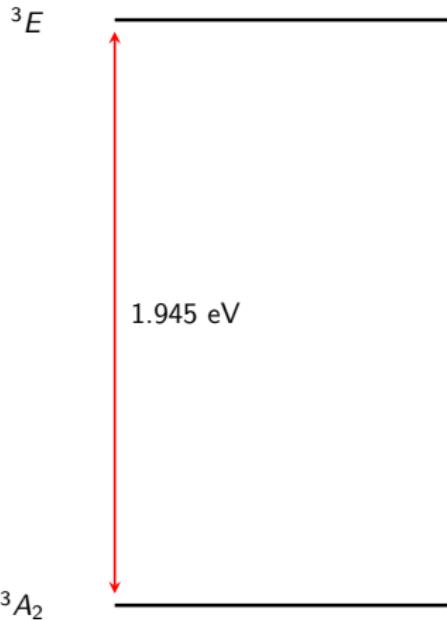
REVISE E AND D



Transitions overview: NV⁻



Zero phonon line (ZPL)



Density functional theory (DFT)

“ Ψ contains too much information”

- Popular saying

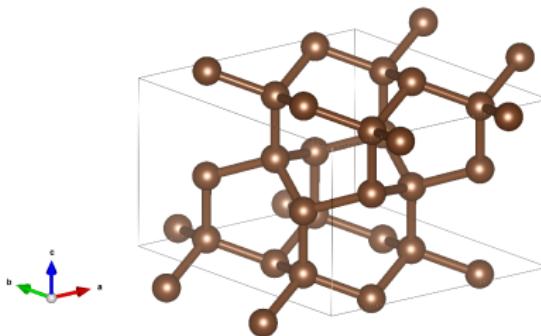
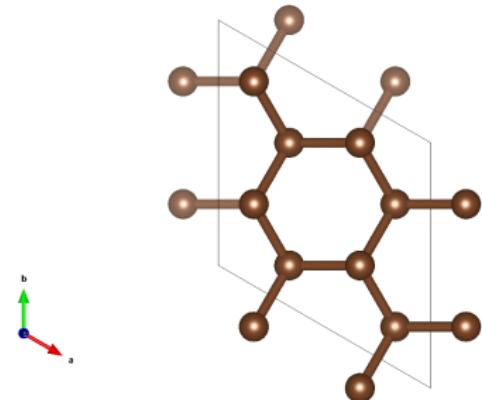
- The exact electronic ground state of a system is only dependent on the electronic density ρ .
- In principle, DFT delivers the **exact ground state**.
- All quantities written in terms of ρ (functional formalism).
- E.g.:

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

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.

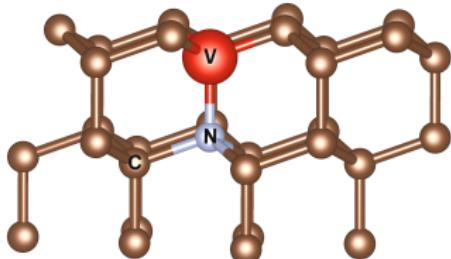
Hexagonal diamond and defects

Hexagonal diamond

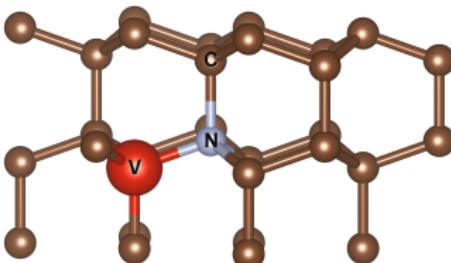


Defected hexagonal diamond

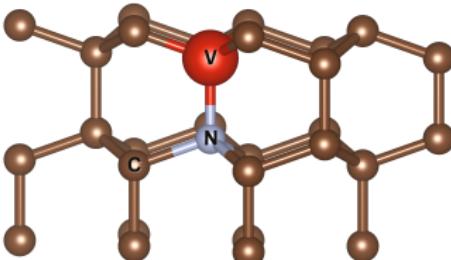
Cubic diamond



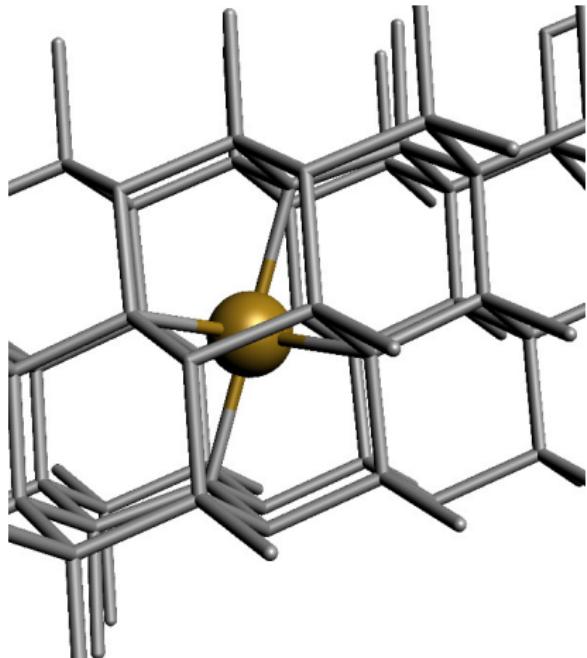
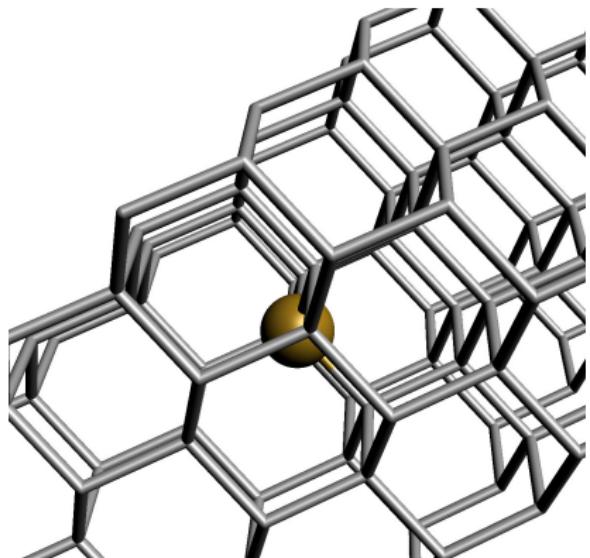
Hexagonal *x*-type



Hexagonal *z*-type

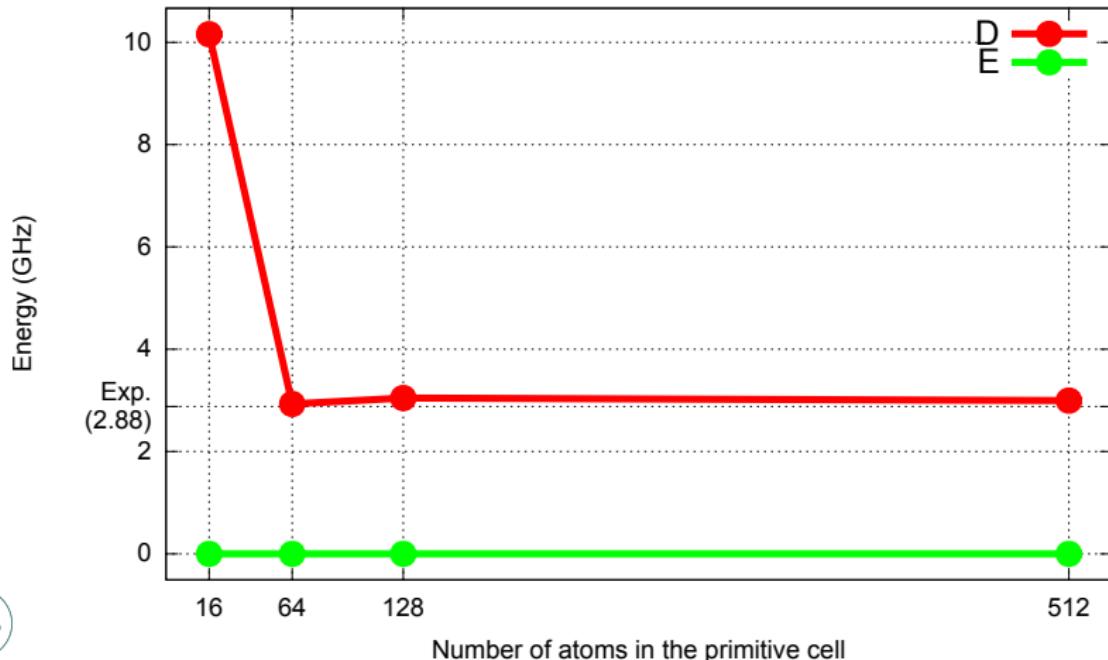


Split vacancies: SiV⁻

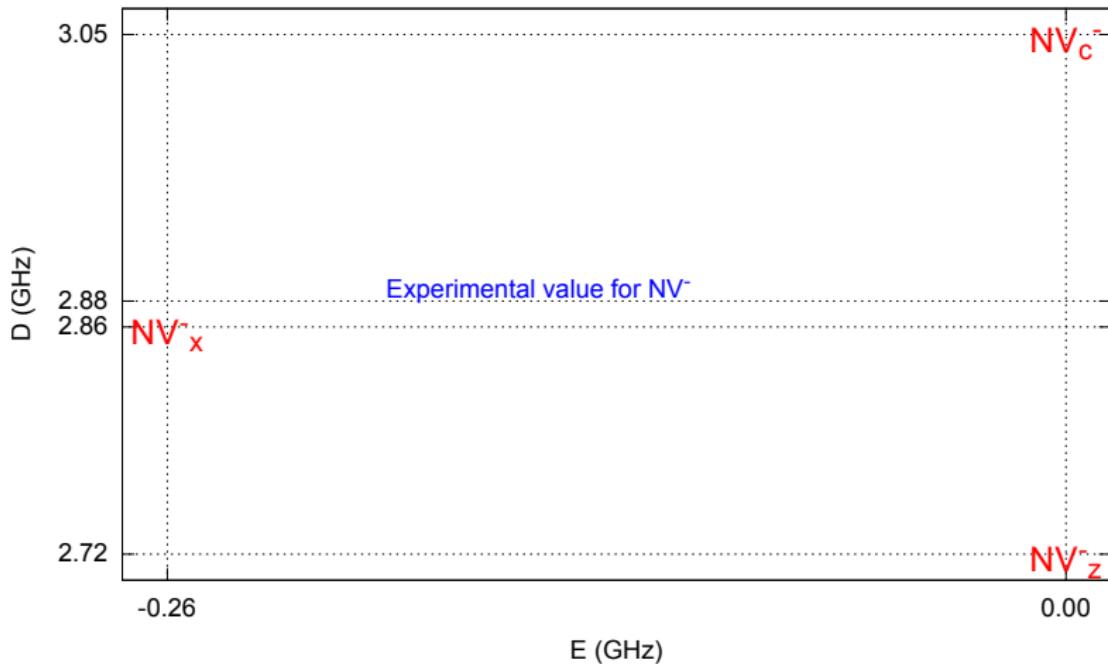


Results

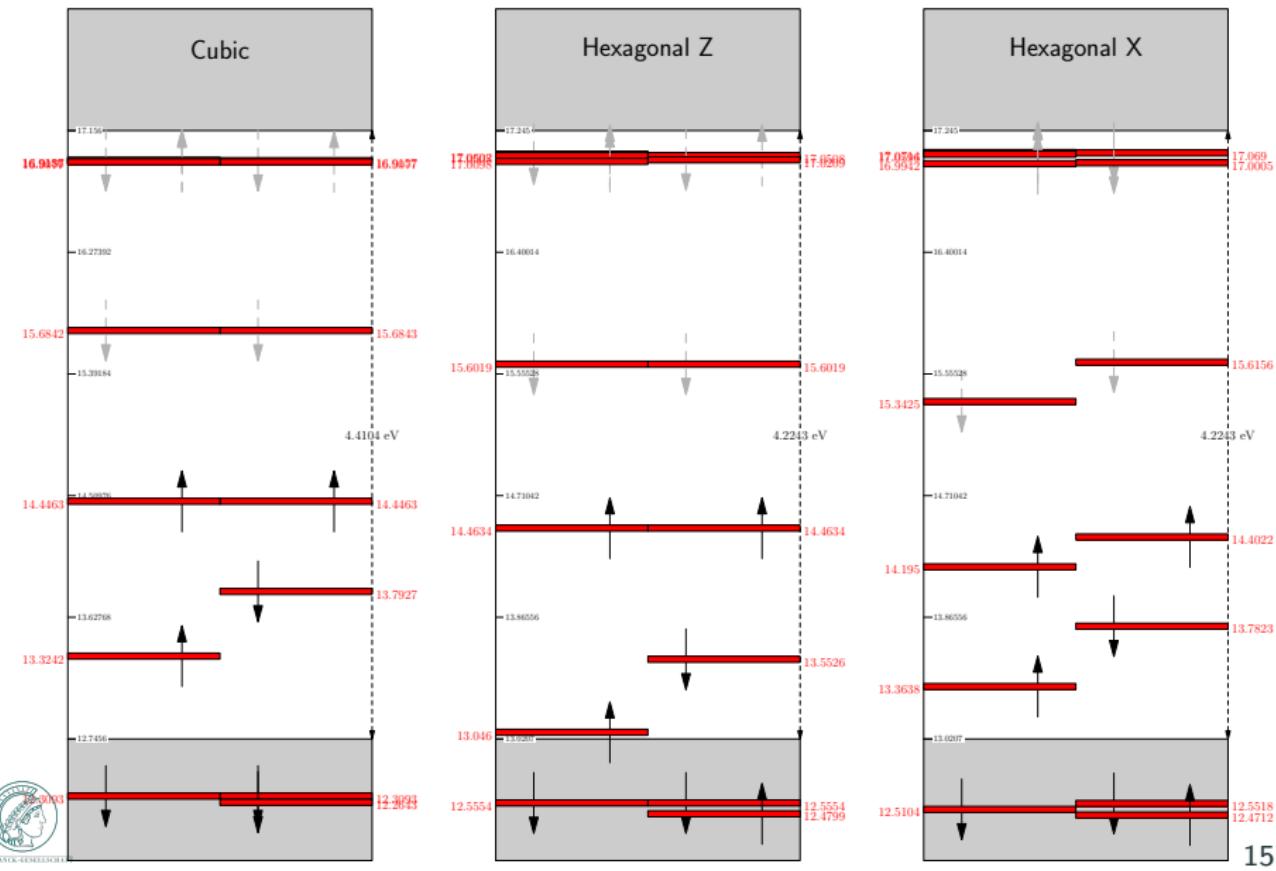
- Cubic diamond, convergence and comparison with the experimental result.



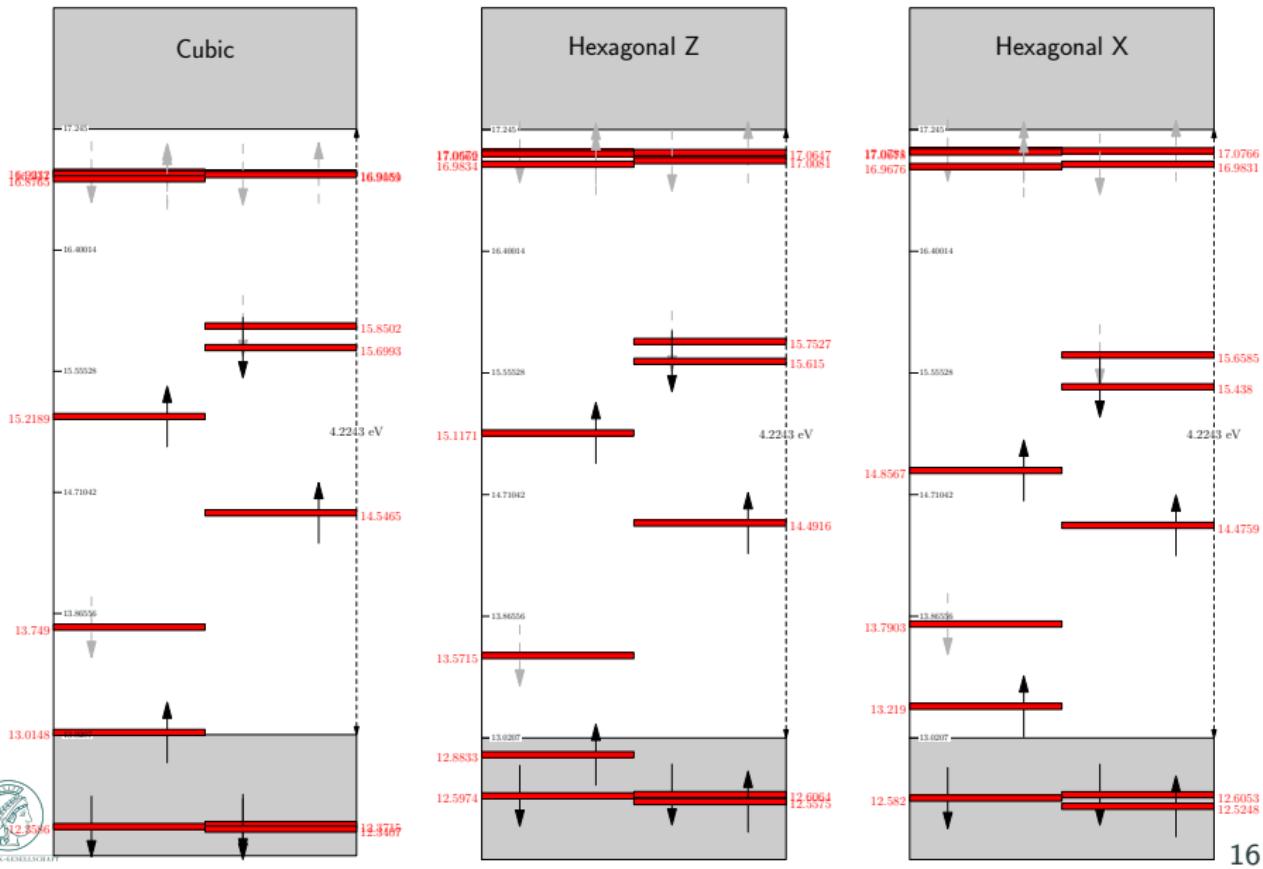
NV*: ZFS cubic (NV_c^*), Hexagonal x, z ($\text{NV}_{x,z}^*$)



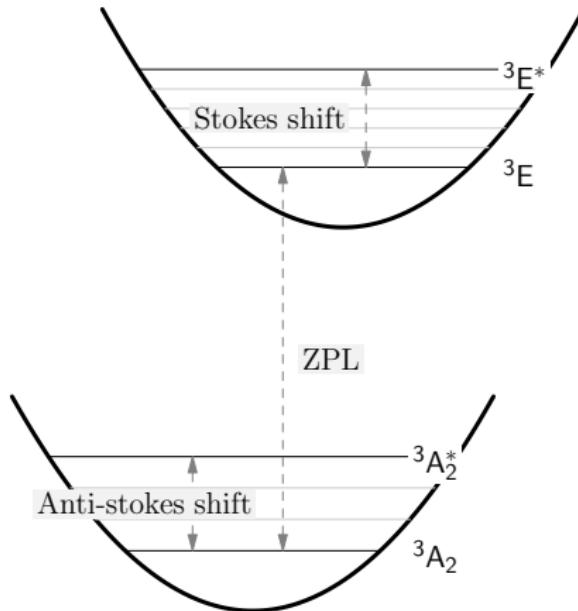
NV⁻: Ground state 3A_2



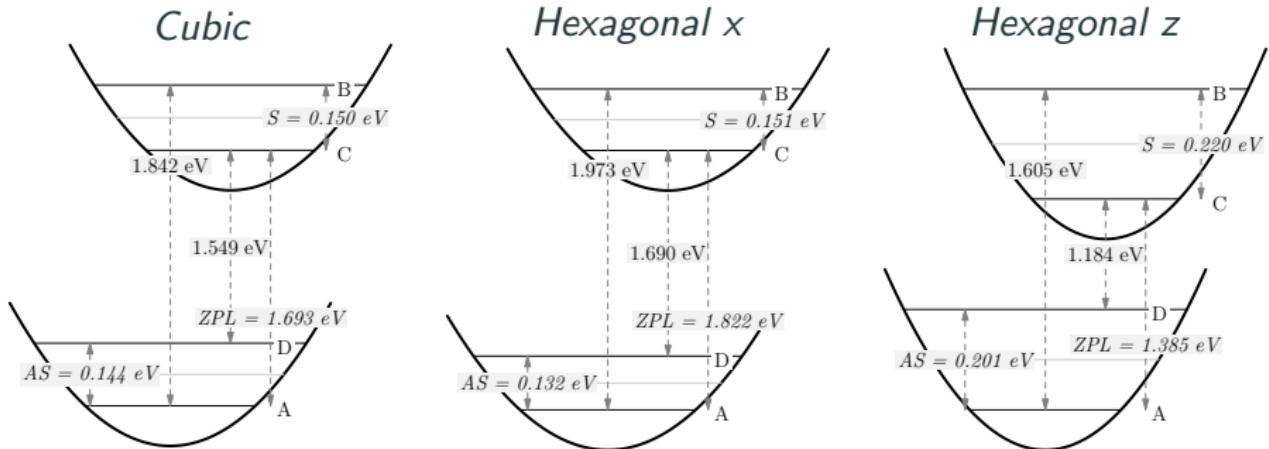
NV⁻: Excited state 3E



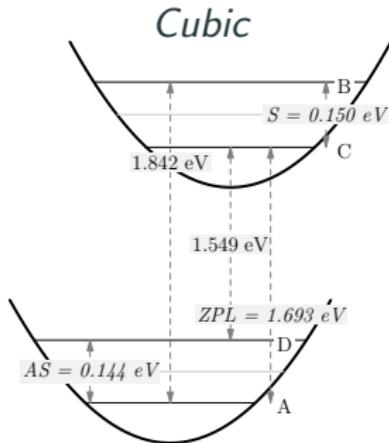
NV⁻: Vibronic scheme (PBE + 128 atomic cell)



NV⁻: Vibronic scheme (PBE + 128 atomic cell)



NV⁻: Vibronic scheme (PBE + 128 atomic cell)



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 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		



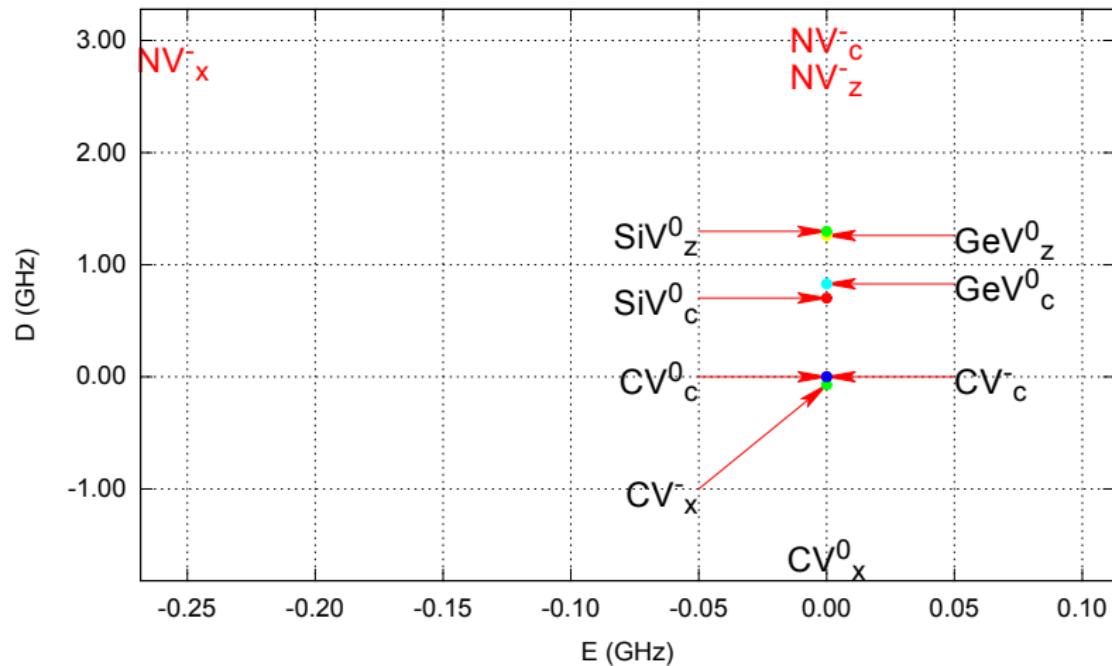
Expanding the defect

14 IVA 15 VA

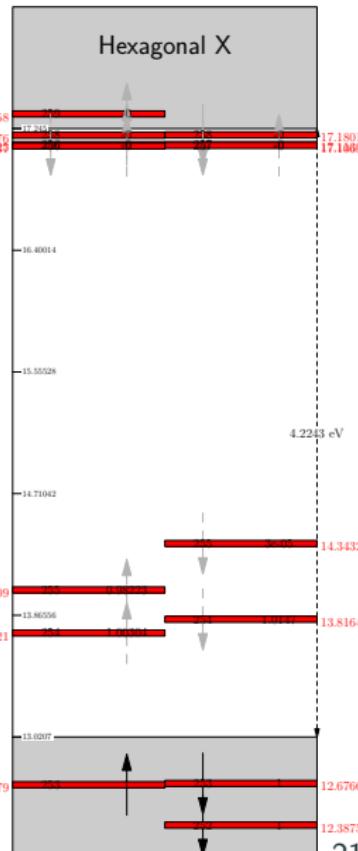
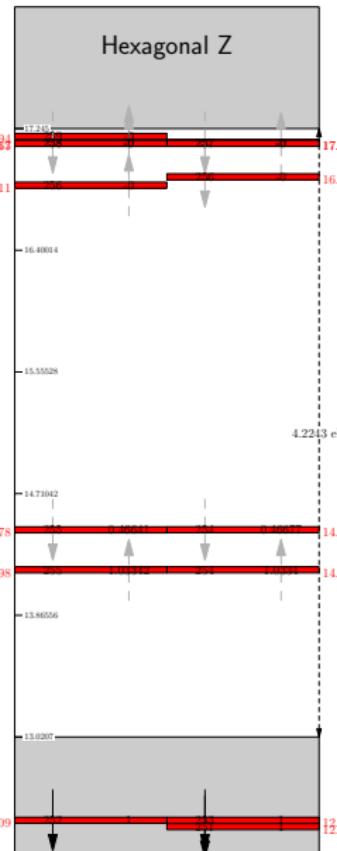
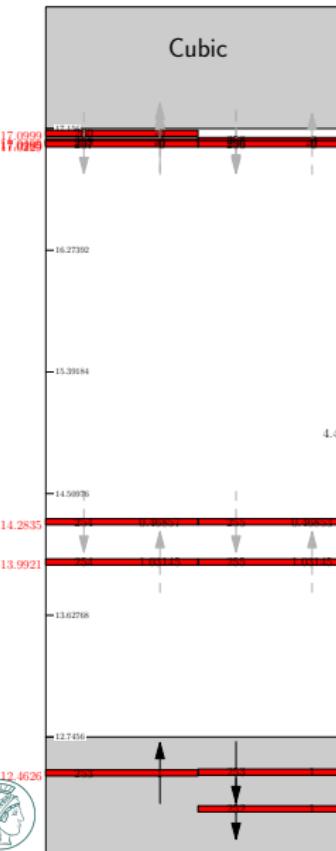
6	12.011	7	14.007
$\text{CV}_{\textcolor{red}{a}}^q$		$\text{NV}_{\textcolor{red}{a}}^q$	
Carbon		Nitrogen	
14	28.086	15	30.974
$\text{SiV}_{\textcolor{red}{a}}^q$		$\text{PV}_{\textcolor{red}{a}}^q$	
Silicon		Phosphorus	
32	72.64		
$\text{GeV}_{\textcolor{red}{a}}^q$		$a = \{c, x, z\}$ $q = \{+, -,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



ZFS map



Ground state example: GeV^-



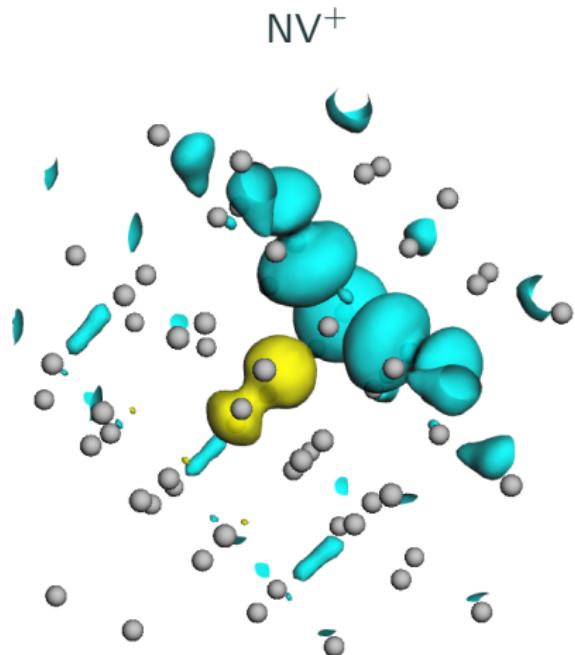
Summary and outlook

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

Using DMRG for excited state calculations

- XV^q as molecules.
- Localized states as isolated.
- Issues like charge transfer, charge corrections.



Thank you!

