

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

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Aim and scope of the work

Aims

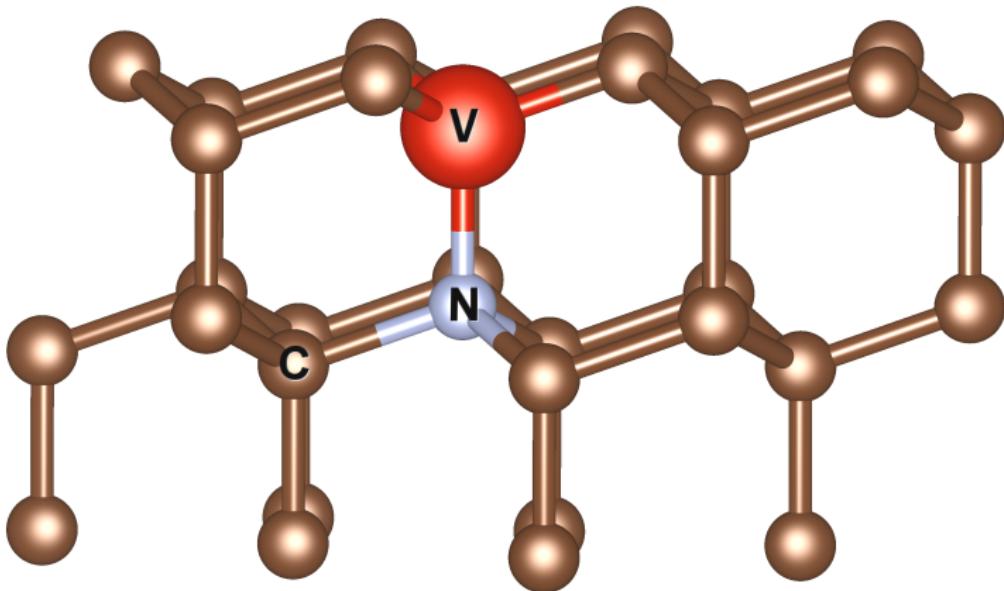
- Exploration of defect centres using state-of-the-art ab-initio theories.

Aims

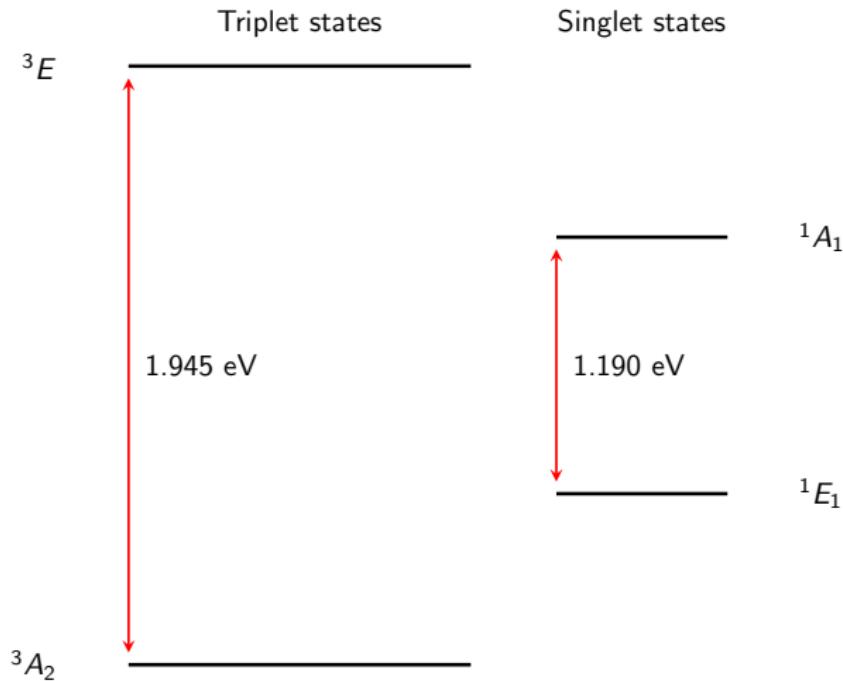
- Exploration of defect centres using state-of-the-art ab-initio theories.
- Reproduce and obtain classical results for nitrogen vacancy impurity complexes in diamond.

Introduction

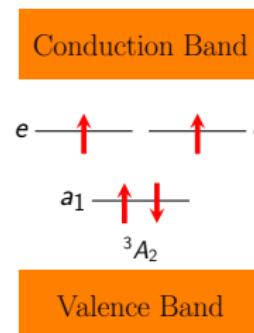
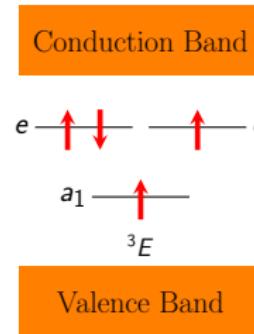
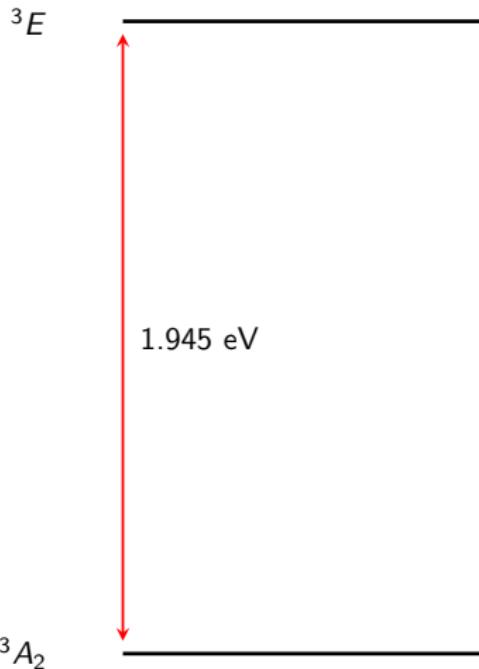
Nitrogen Vacancy Centre in diamond



Main level overview: NV⁻



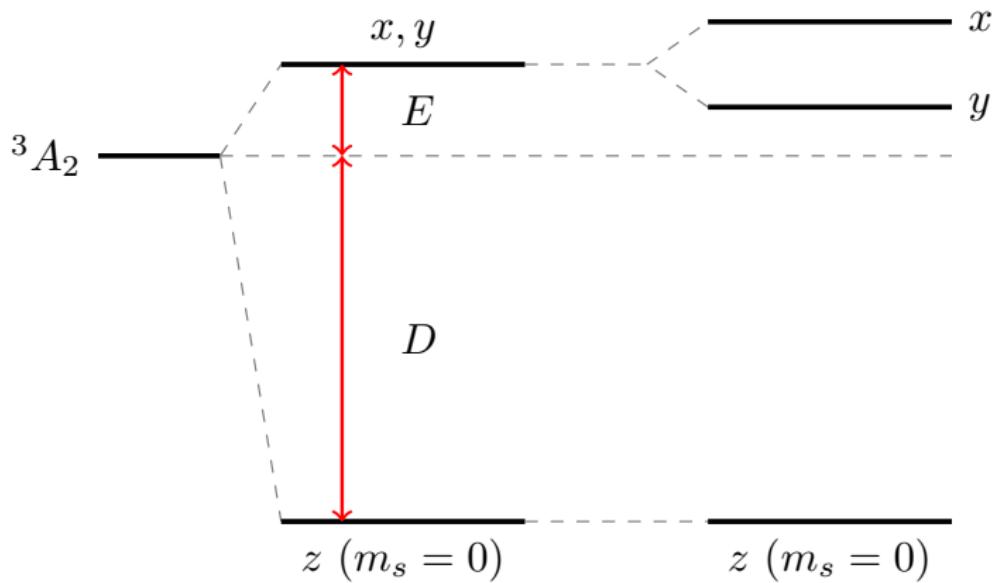
Triplets overview: NV⁻



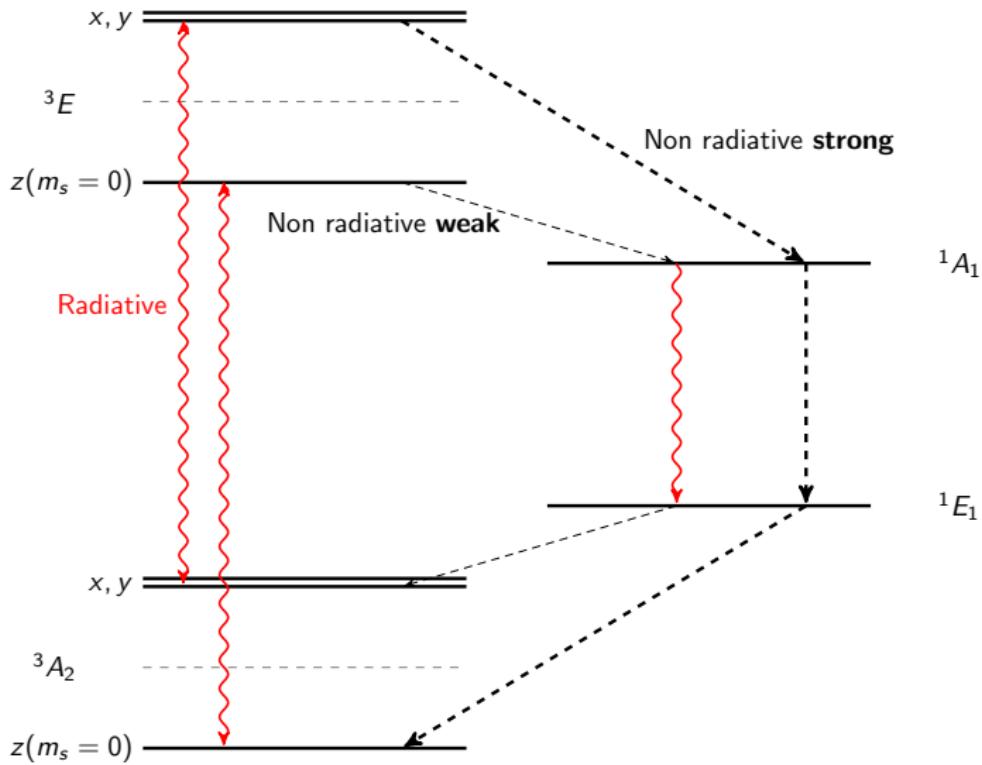
Transitions

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

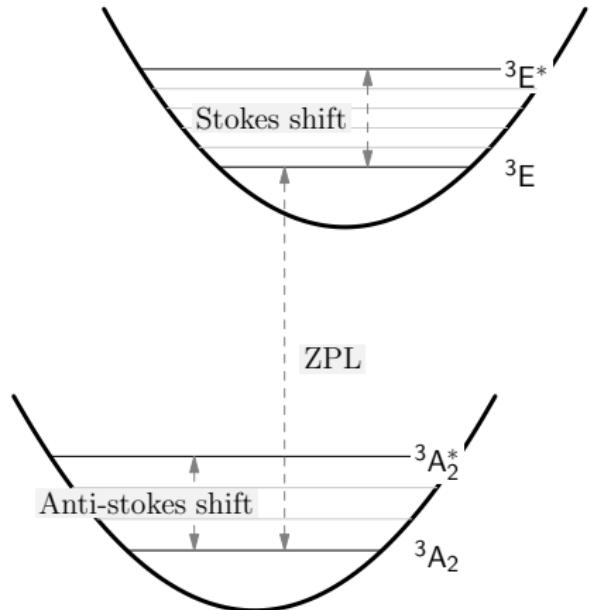
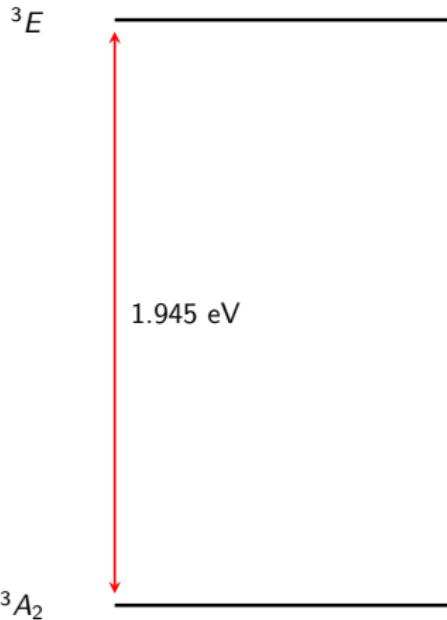
Zeeman/strain



Transitions overview: NV⁻



Zero phonon line (ZPL)



Density functional theory (DFT)

" Ψ contains too much information"

- Popular saying

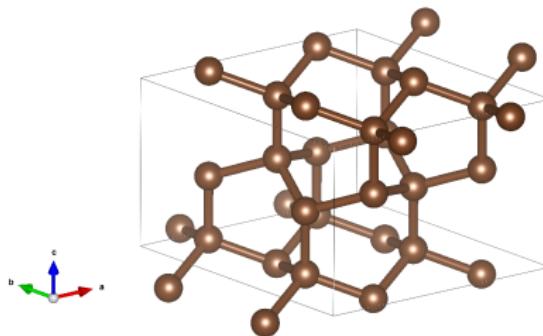
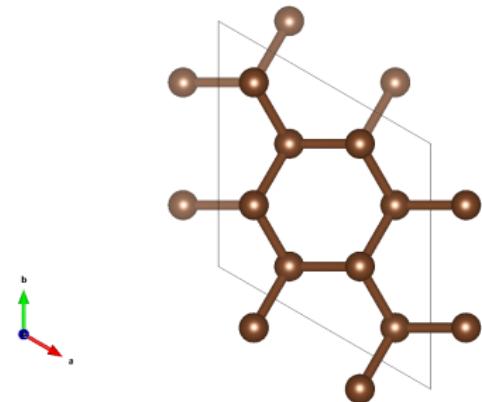
- The exact electronic ground state of a system is only dependent on the electronic density ρ .
- In principle, DFT delivers the **exact ground state**.
- All quantities written in terms of ρ (functional formalism).
- E.g.:

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

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.

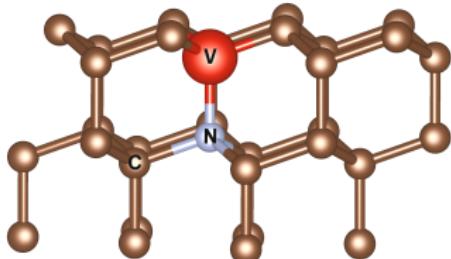
Hexagonal diamond and defects

Hexagonal diamond

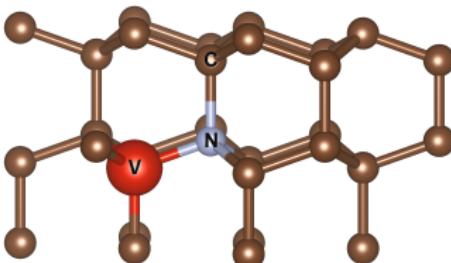


Defected hexagonal diamond

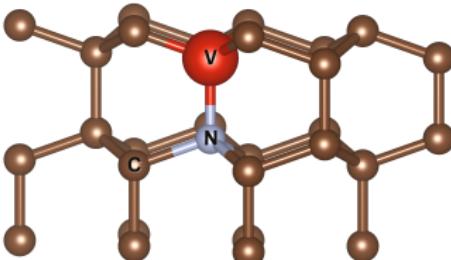
Cubic diamond



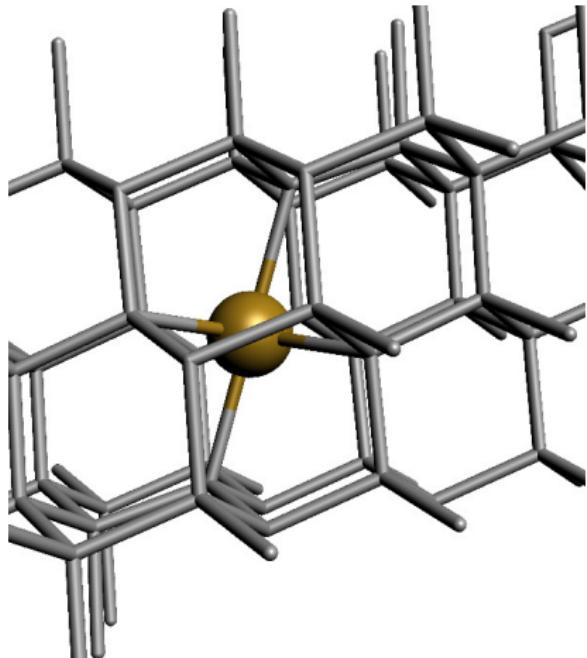
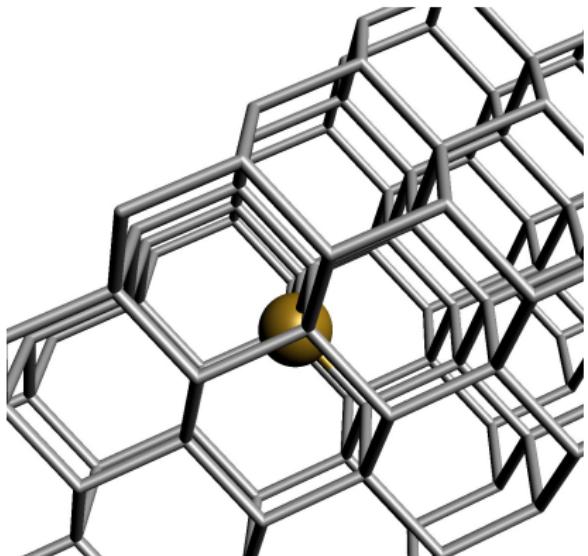
Hexagonal x-type



Hexagonal z-type

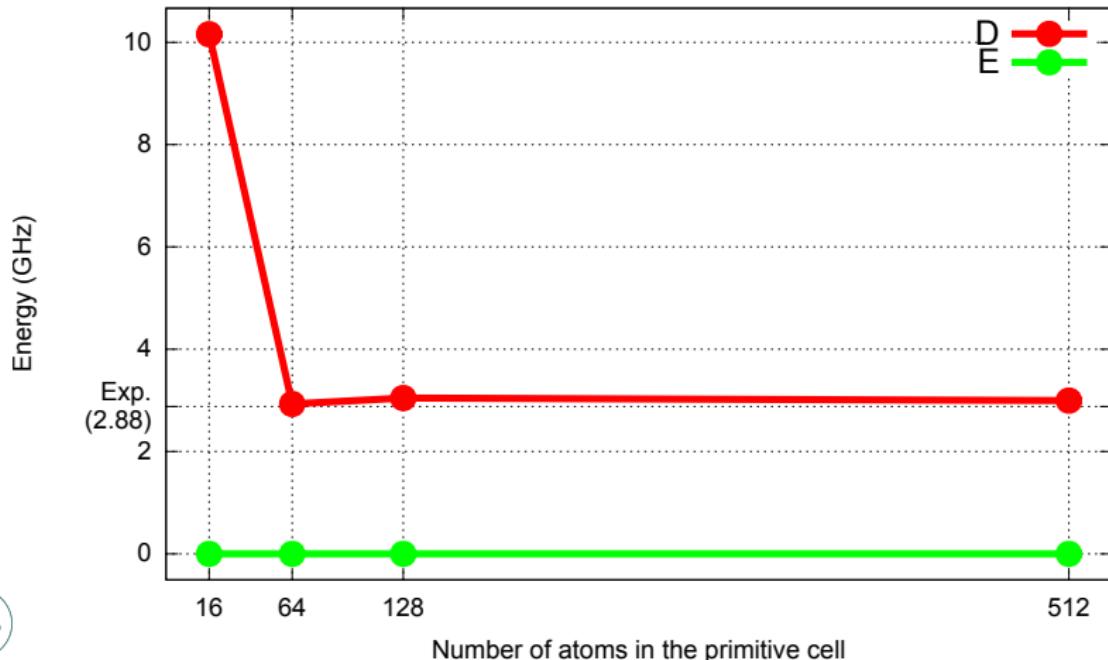


Split vacancies: SiV⁻

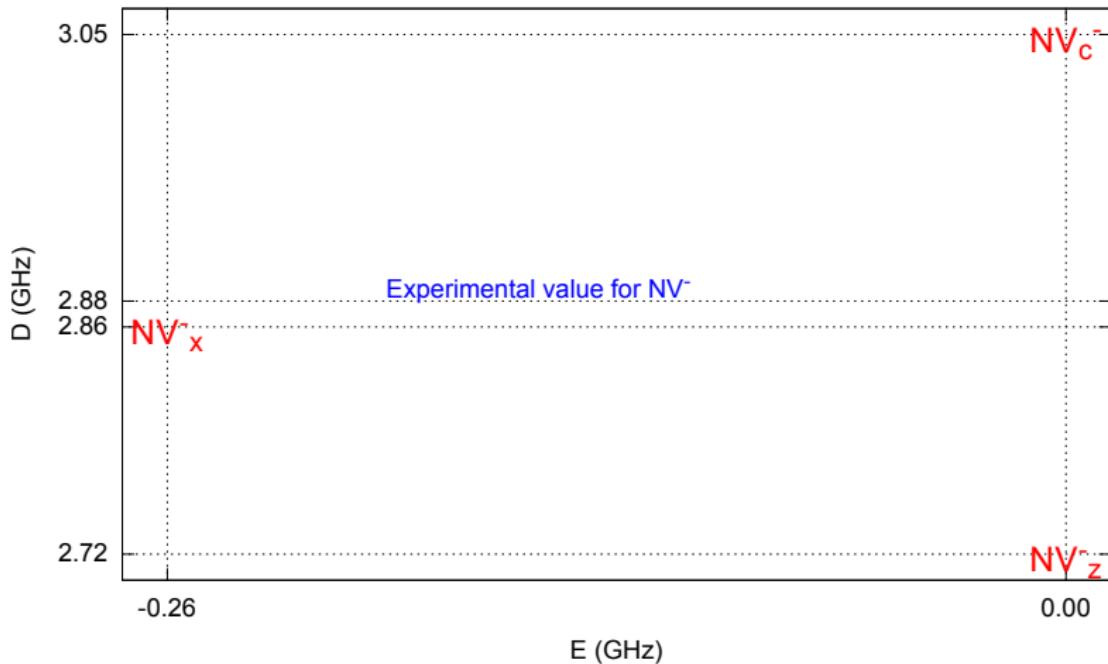


Results

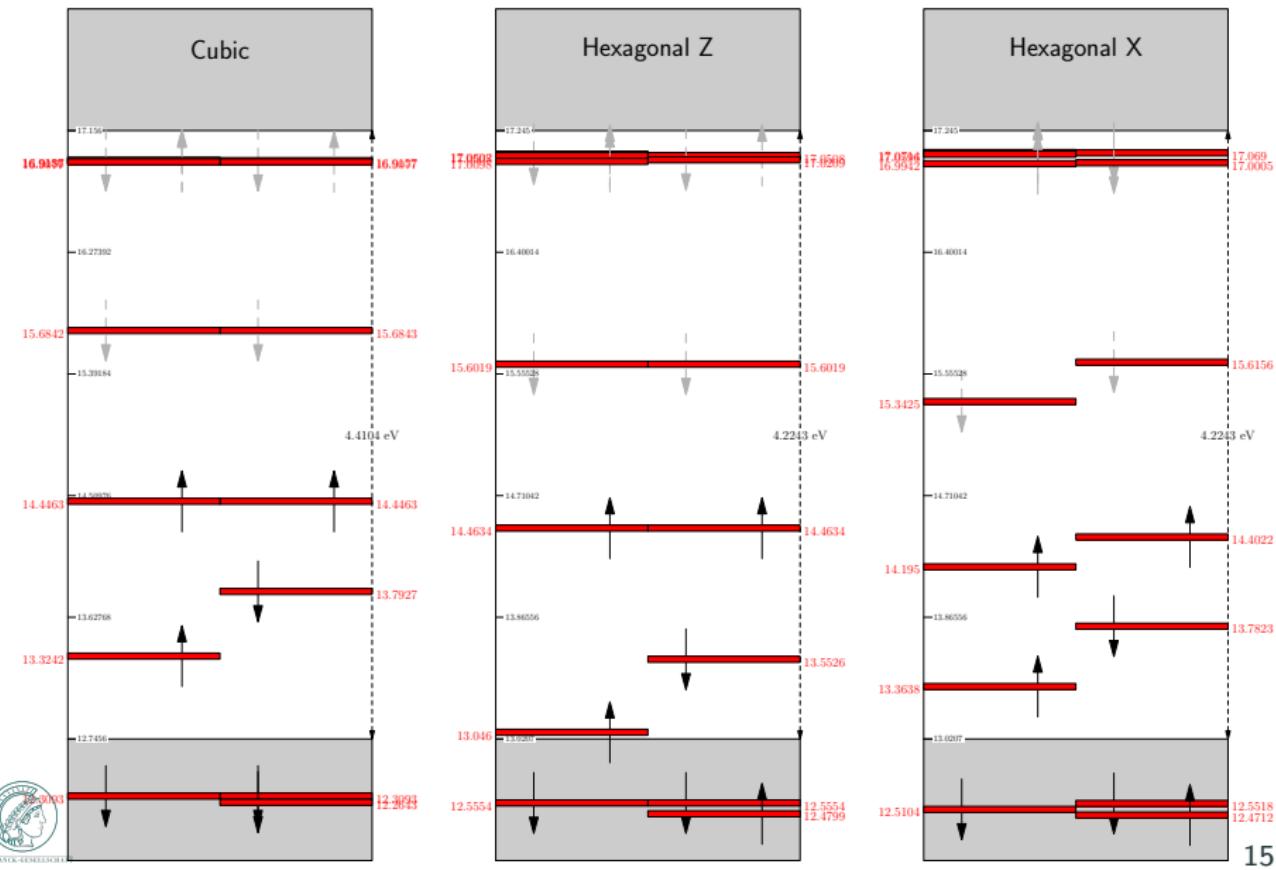
- Cubic diamond, convergence and comparison with the experimental result.



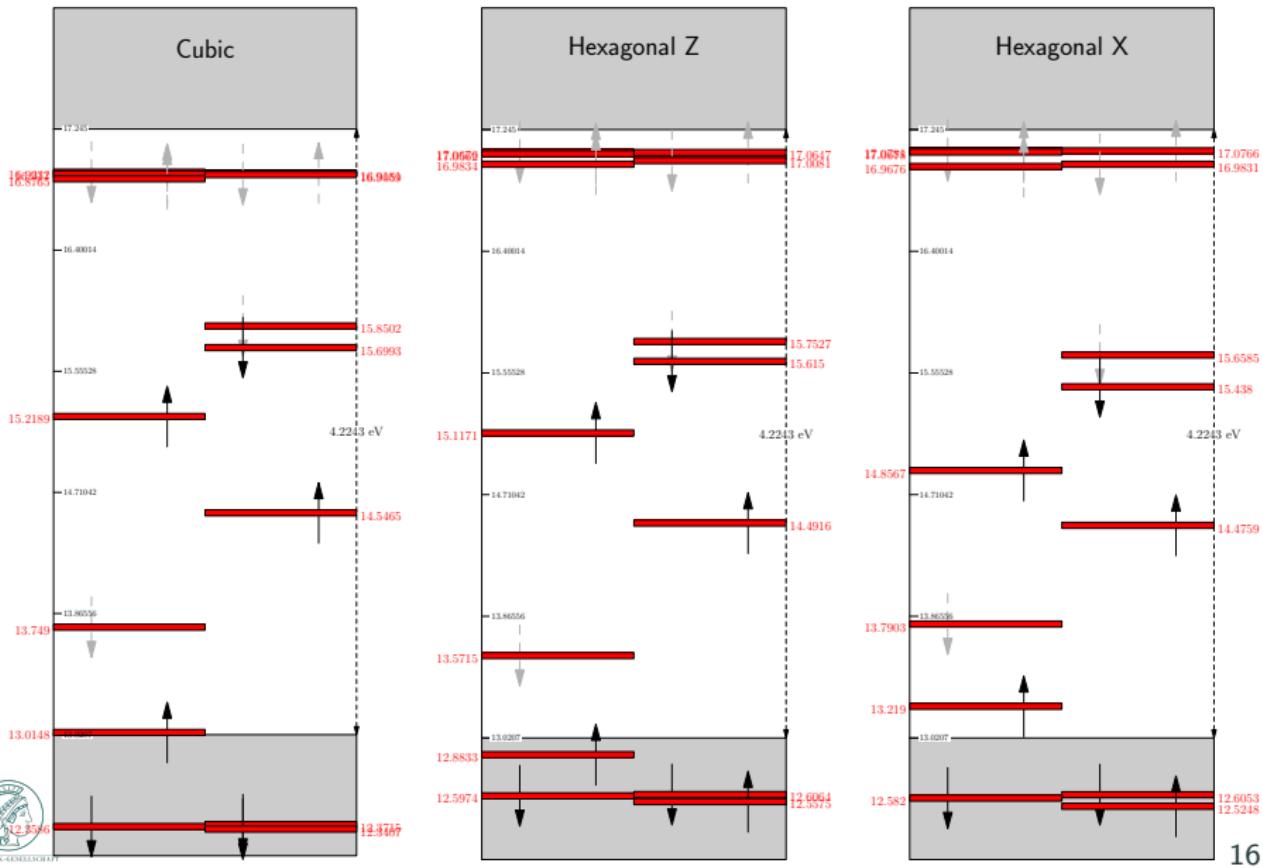
NV*: ZFS cubic (NV_c^*), Hexagonal x, z ($\text{NV}_{x,z}^*$)



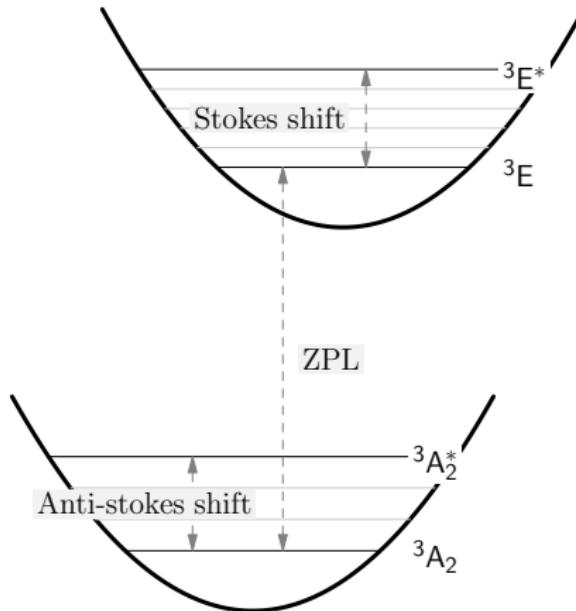
NV⁻: Ground state 3A_2



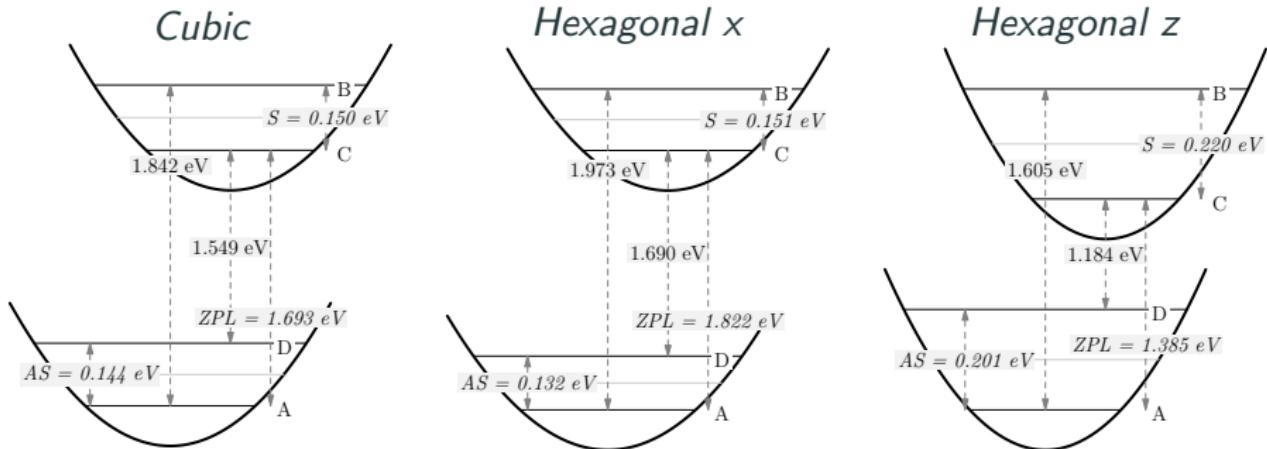
NV⁻: Excited state 3E



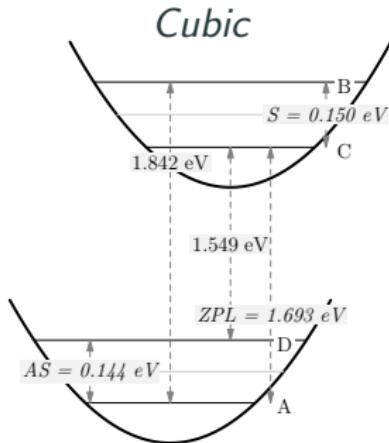
NV⁻: Vibronic scheme (PBE + 128 atomic cell)



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 | | |



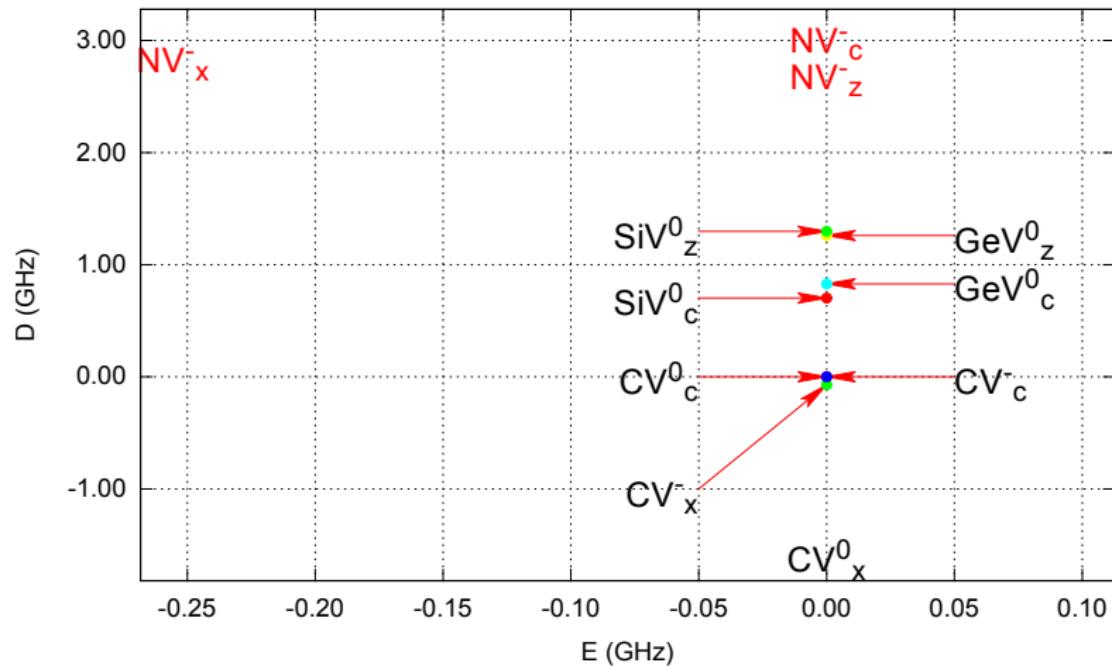
Expanding the defect

14 IVA 15 VA

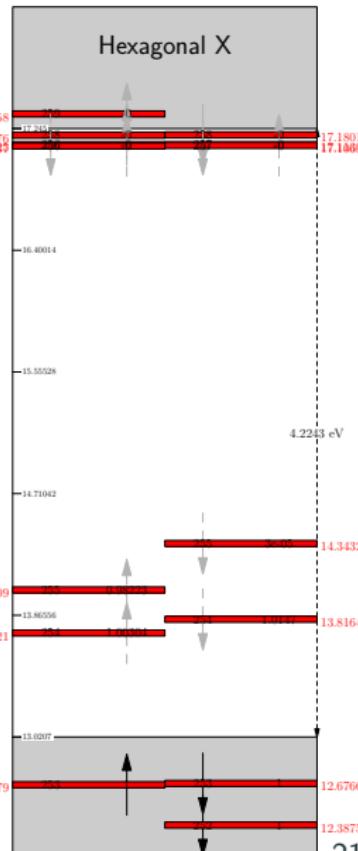
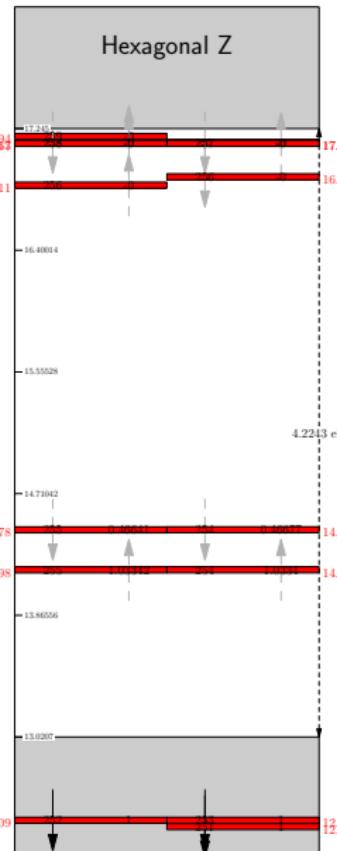
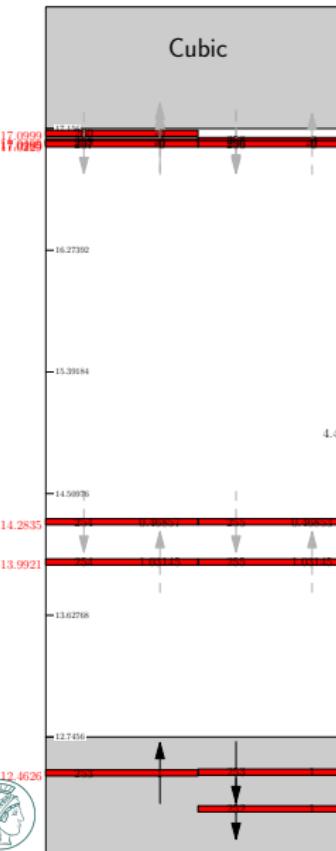
| | | | |
|-------------------------------------|--------|--|--------|
| 6 | 12.011 | 7 | 14.007 |
| $\text{CV}_{\textcolor{red}{a}}^q$ | | $\text{NV}_{\textcolor{red}{a}}^q$ | |
| Carbon | | Nitrogen | |
| 14 | 28.086 | 15 | 30.974 |
| $\text{SiV}_{\textcolor{red}{a}}^q$ | | $\text{PV}_{\textcolor{red}{a}}^q$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



ZFS map



Ground state example: GeV^-



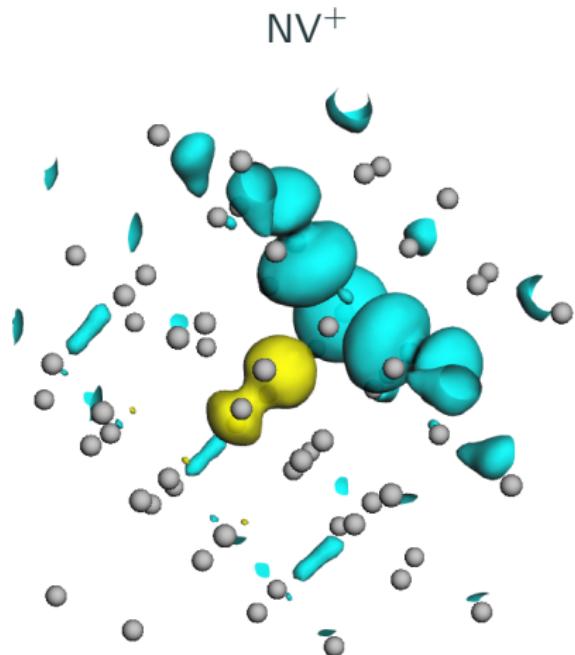
Summary and outlook

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!

