Triplet and Singlet Electronic States of a Local Neutral Defect in wAlN Favorable for Solid State Spin Qubit Applications: An Ab initio Study

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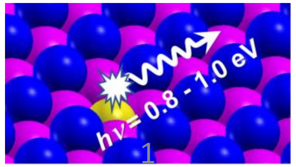

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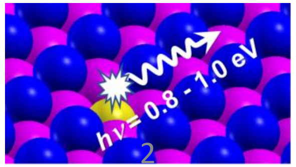
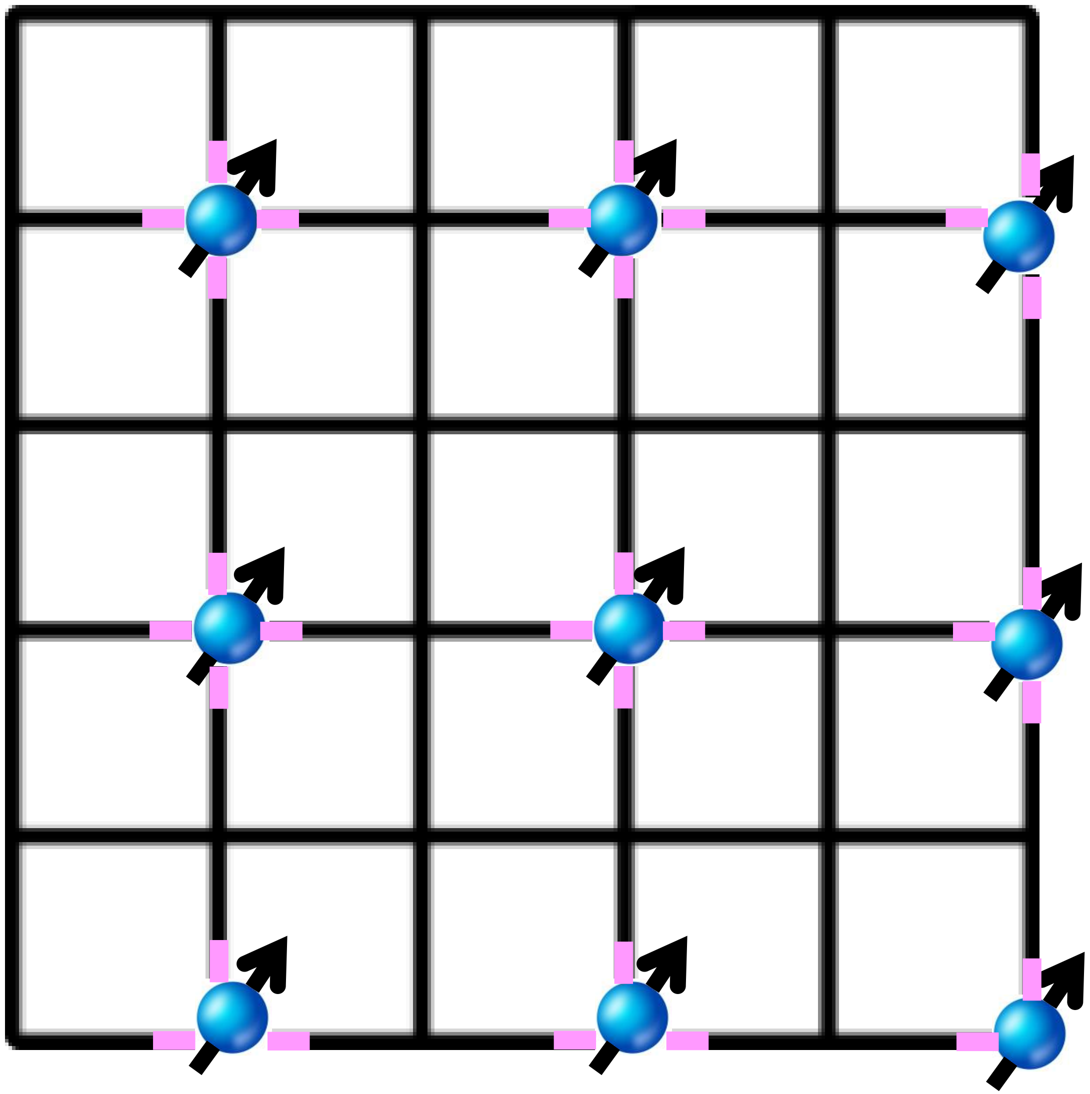


Spin Qubits

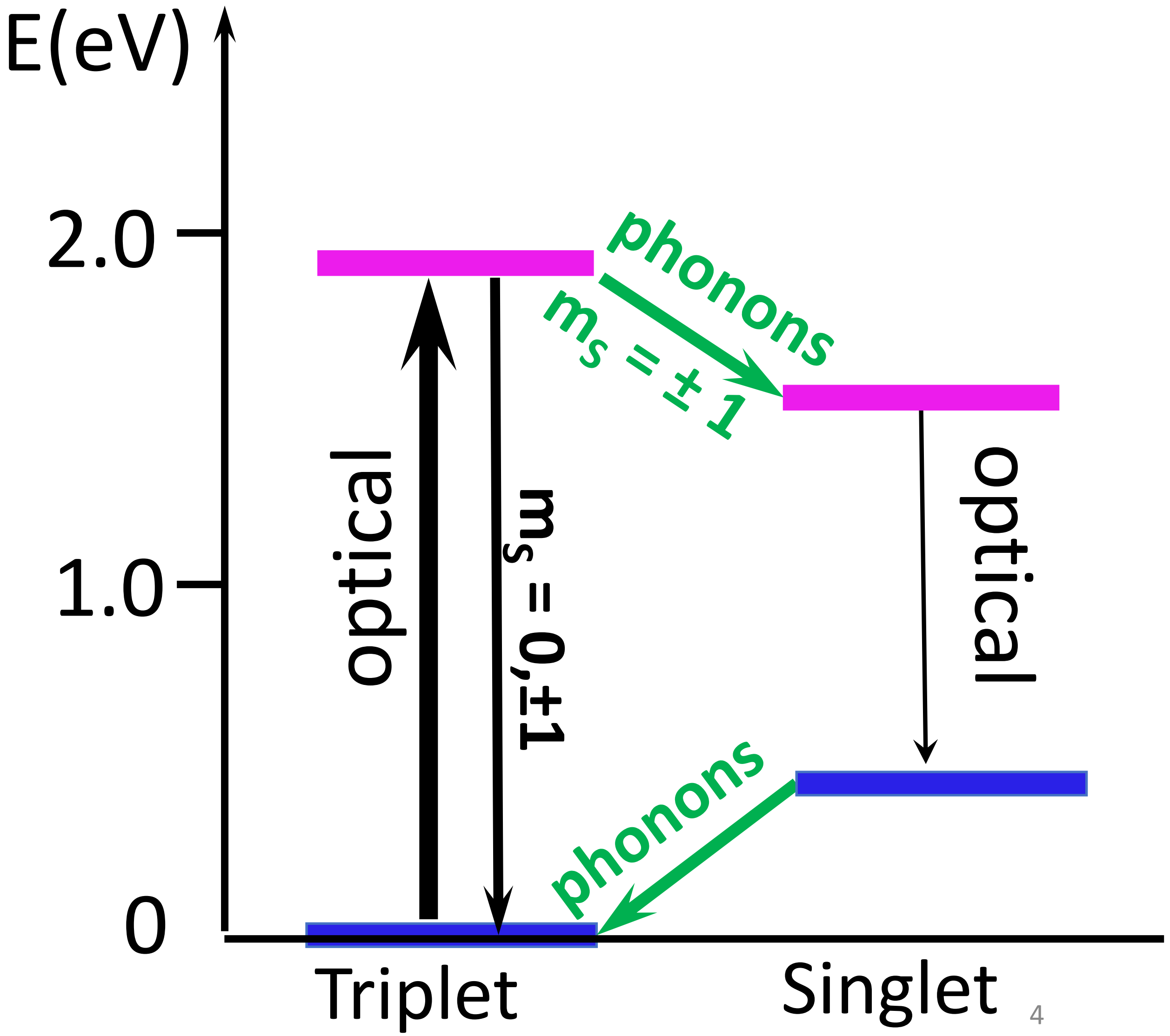
- Potential application in quantum information technology
- Local defects in semiconductors → spin density is localized around the defect
- Prototype: NV^- center in diamond



NV⁻ center in diamond: Prototype of what?

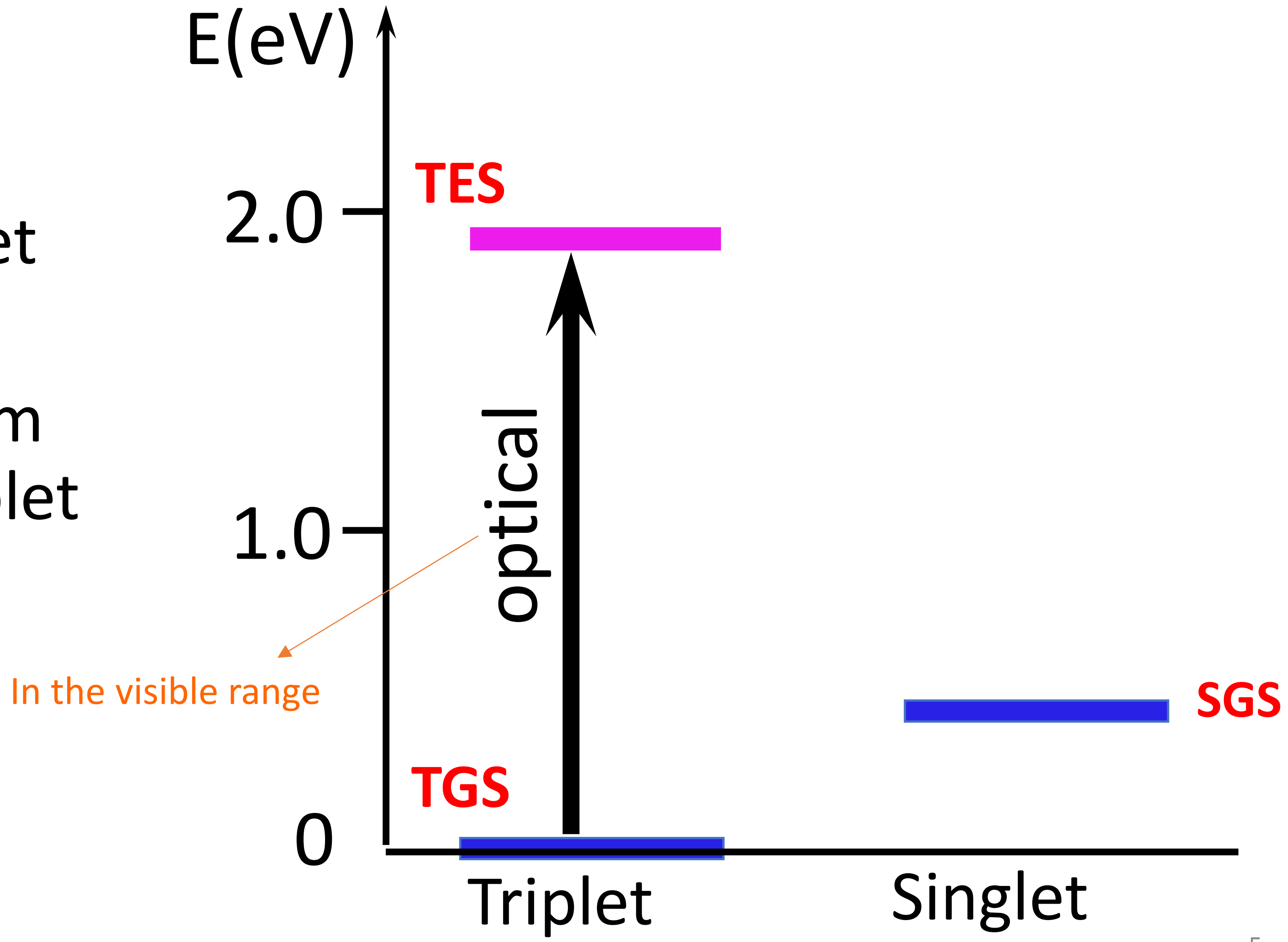
Spin=1

SPIN POLARIZATION CYCLE:
- m_s=0 spin-polarization of TGS
- Needed for Initialization and readout of qubits



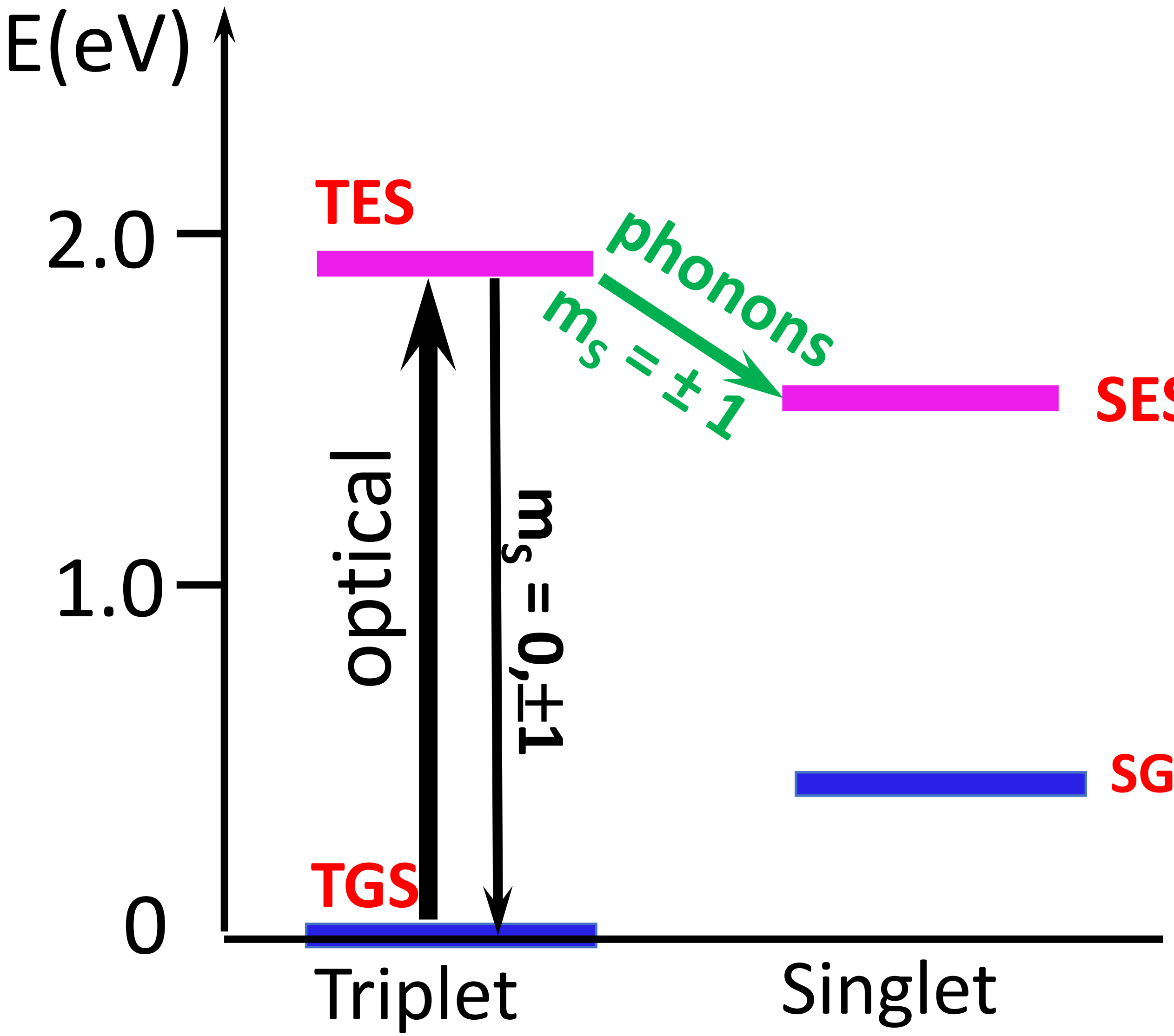
Spin qubits properties based on NV^{-} center in Diamond

- Triplet lowest-energy ground state (TGS)
- Local minimum singlet ground state (SGS)
- Optical transition from TGS to an excited triplet state (TES)



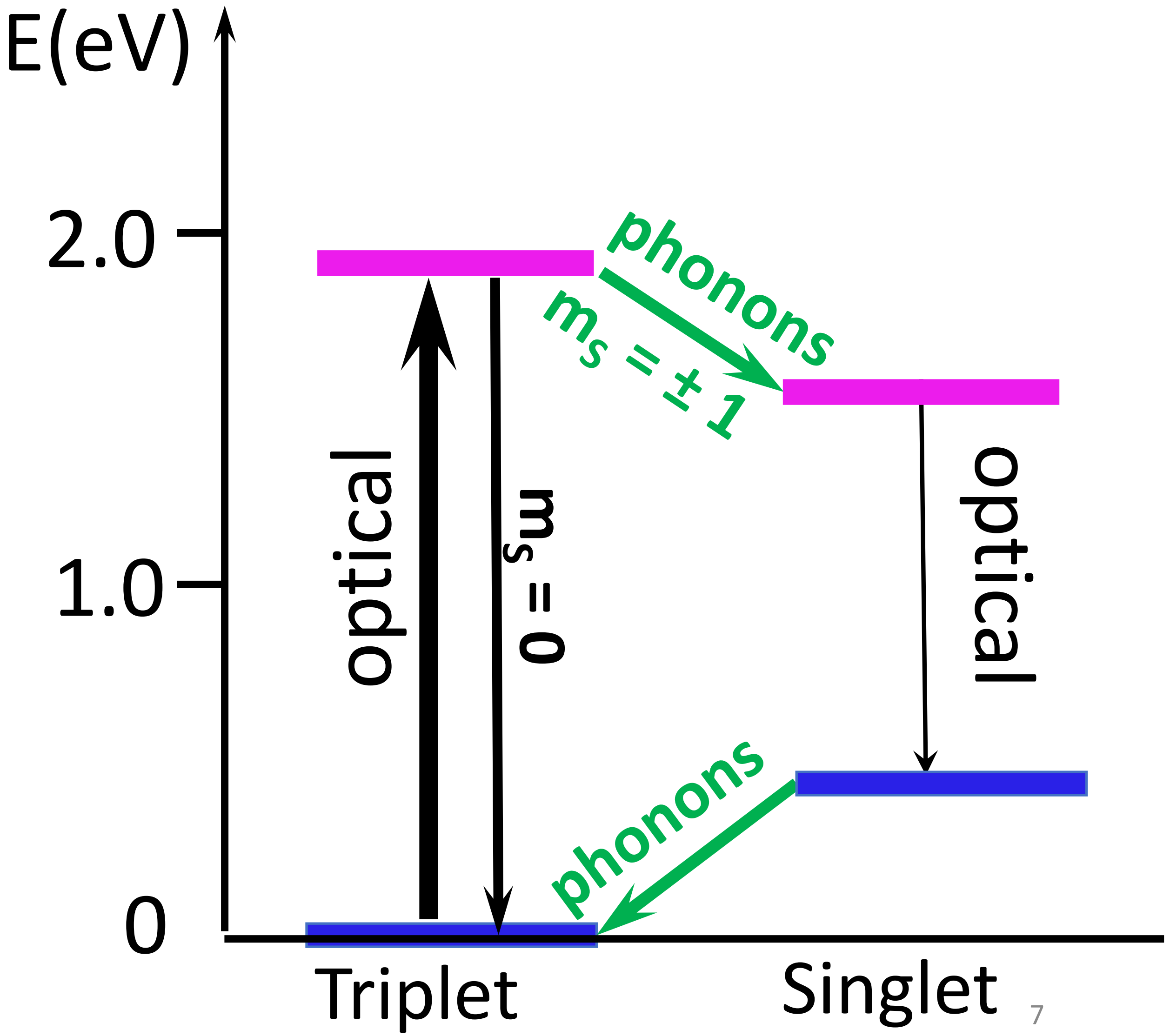
Spin qubits properties based on NV^{-} center in Diamond

- **<u>Phonon-assisted decay</u>** to an excited singlet state (SES).
- **<u>Change of spin</u>**: The TES – SES transition must conserve total angular momentum $\vec{J}$
  $\rightarrow$ spin-orbit coupling
  $\rightarrow$ TES to SES transition only allowed for $m_s=\pm1$
  $\rightarrow$ states spatial localization
  $\rightarrow$ $m_s=0$ and $\lesssim$50% of $m_s=\pm1$ relax back to TGS



Spin qubits properties based on NV⁻ center in Diamond

- Optical SES – SGS emission is available
- Phonon-assisted decay from SGS to TGS is feasible: $\vec{J}$, SOC and energy barrier
- **Electronic states associated with the transitions are localized within the host band gap for long coherence time**



Rational Design of Novel Spin Qubit: $V_{Al}S_N$

Wide band gap semiconductor as host of defects: → **Wurtzite AlN (Band gap ~6 eV)**

Defect must leave dangling bonds (favorable for spin polarization): → **Removal of an Al atom**

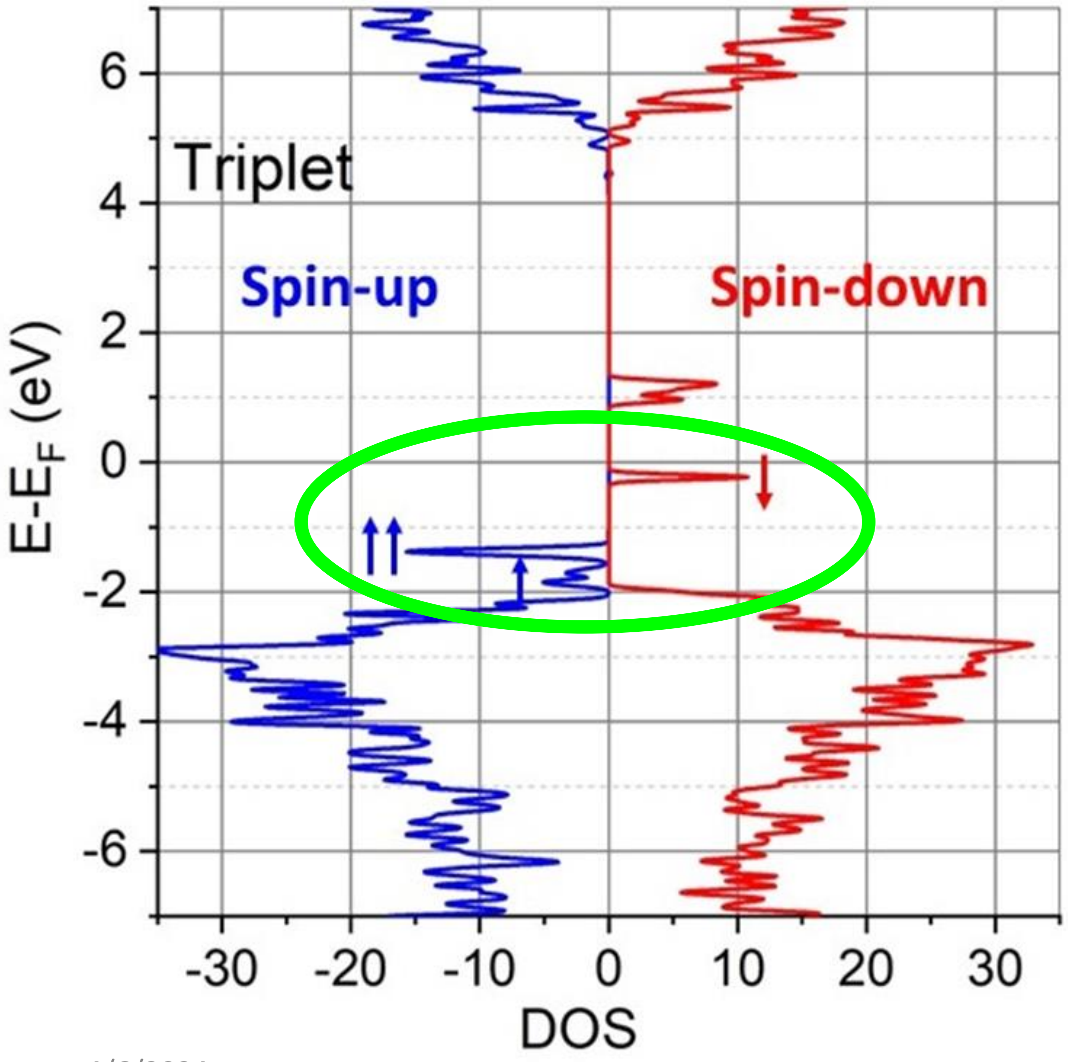
Defect that leaves the number of p-electrons even (favorable for triplet formation) → **Removal of an Al atom decreases the even number of p-electrons by one, but the replacement of N with S brings back one electron**

Substitutional defect does not bind too strongly to host for narrow states in band gap → **The relatively low electronegativity of S hints that it will not bind too strongly**

Ab initio calculations to evaluate Qubit functionality

- **Spin polarization**: Ground state must be a triplet and a local minimum must have be a singlet (DFT, 4x4x3 supercells, 192 atoms)
- **Stability**: Formation energy and phonon spectrum at Γ (DFT, 4x4x3 supercells, 192 atoms)
- **Defect states localized within band gap**: Electronic Structure (DFT followed by self consistent GW method calculations, 3x3x2, 72 atoms)
- **Existence and transition rate of optical excitations**: (Bethe-Salpeter equation method, 3x3x2, 72 atoms)

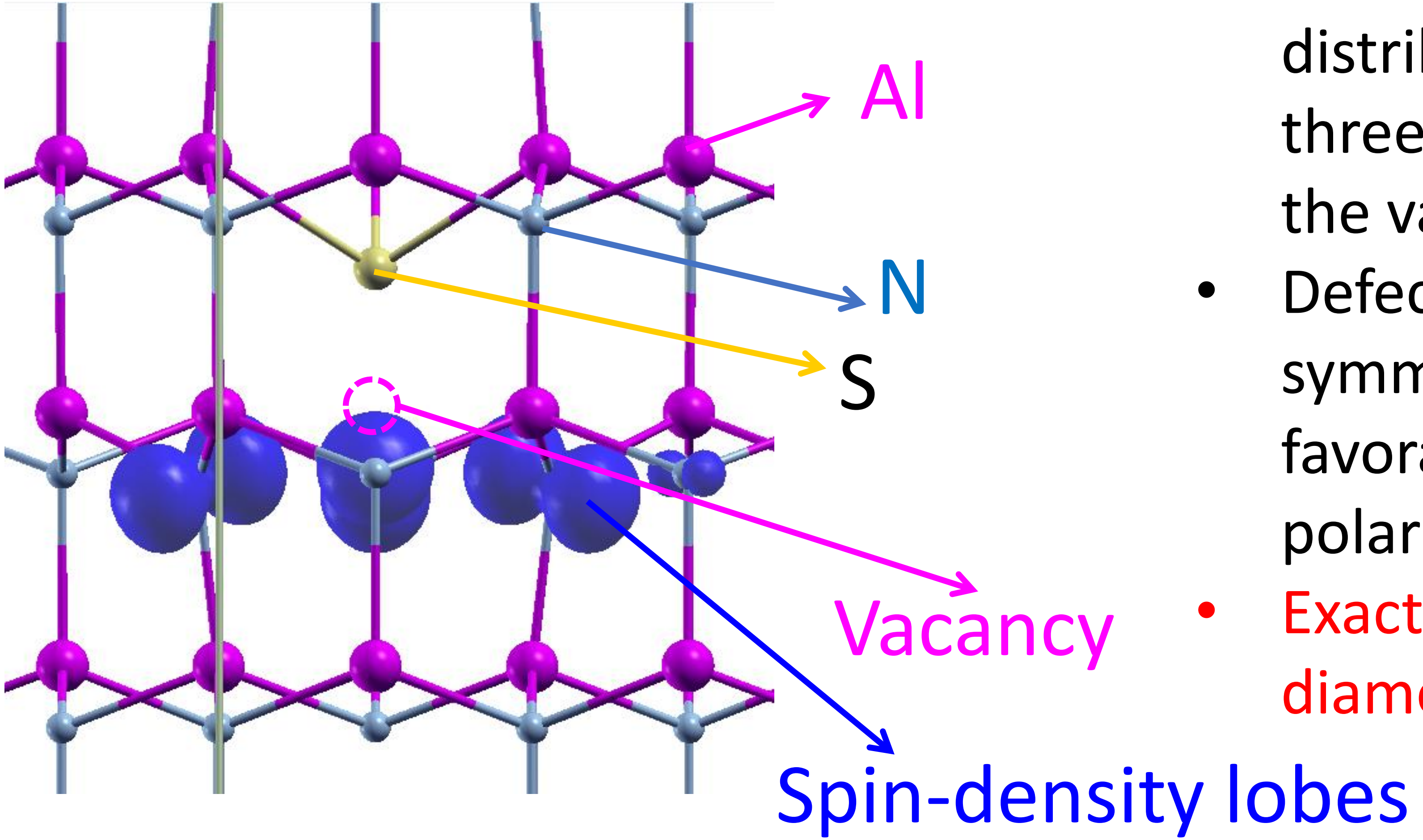
Results: $\mathbf{V_{Al}S_N}$ Triplet Density of States



Densities of the independent quasi-particle states calculated for the triplet state of the $V_{Al}S_N$ defect using the self-consistent GW method.

The four electrons determining the TGS in $V_{Al}S_N$ occupy narrow GW IQP peaks located in the band gap

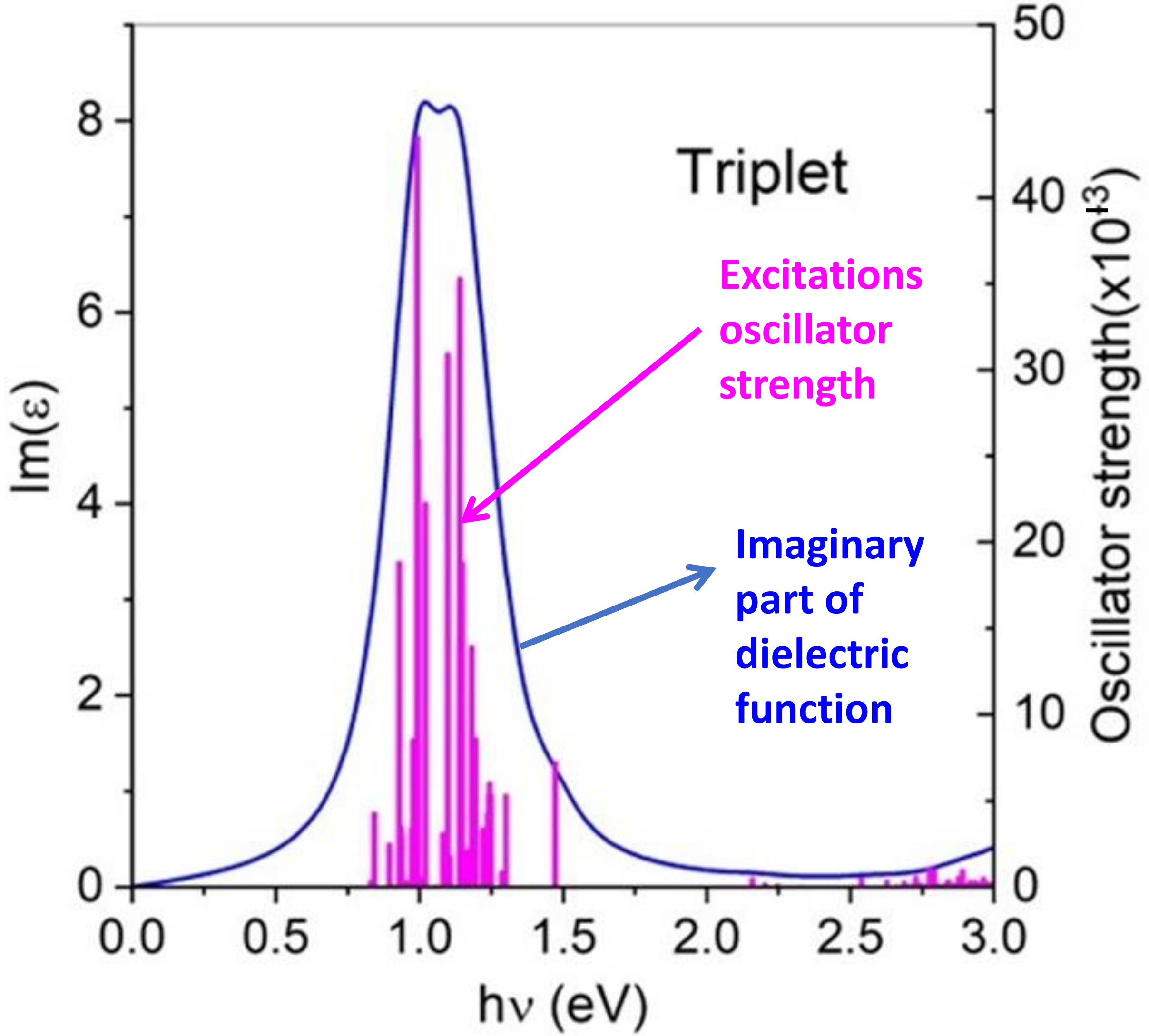
Results: Spin Density



Spin density

- Localized and evenly distributed around the three N atoms next to the vacancy
- Defect has a C_{3v} symmetry, which is favorable for the spin-polarization cycle.
- Exactly like NV^- center diamond, in this regard

Results: Optical excitation TGS → TES



- Optical transitions from TGS to an excited triplet state (TES) do exist (0.85 – 1.3 eV) and they have a pretty large rate (oscillator strength), compared to NV^{-} center
- The two most prominent peaks occur at 1.0 and 1.15 eV
- They optical transition are in the near-infrared range

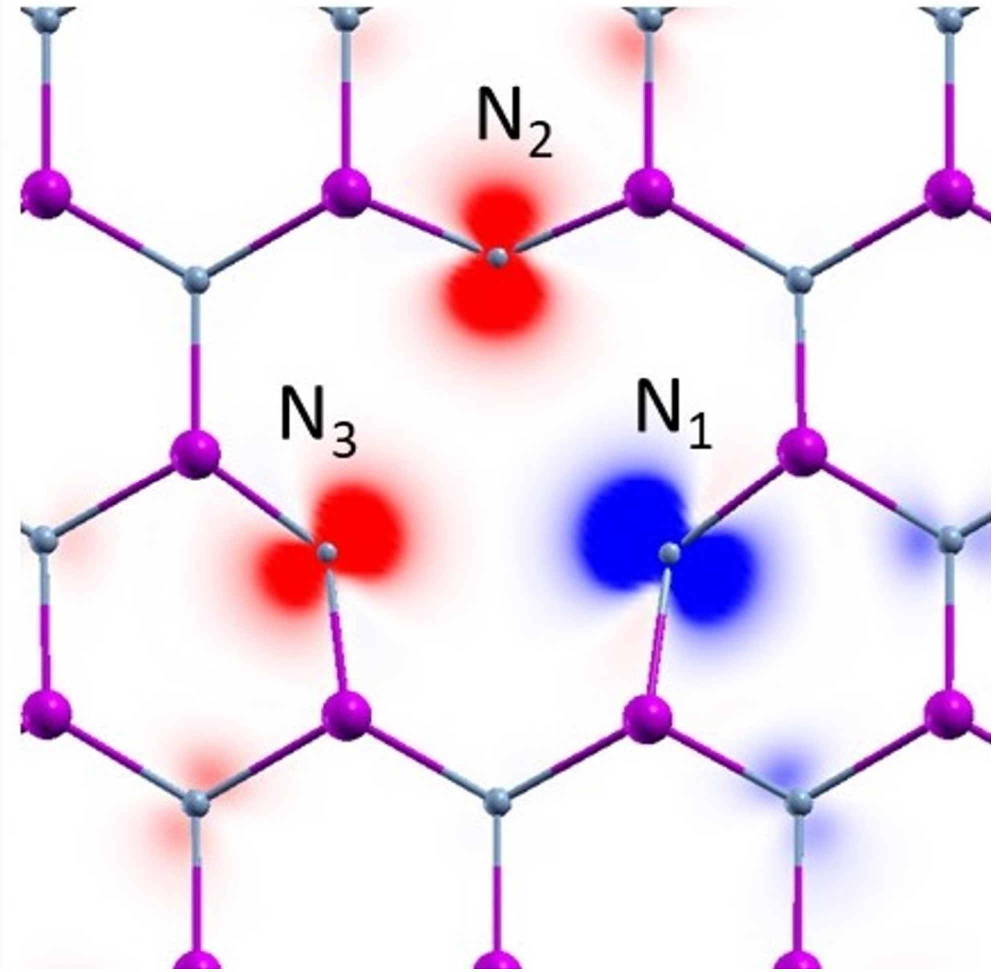
Results: The local minimum singlet state(s)

How to tweak the structure to find a singlet state?

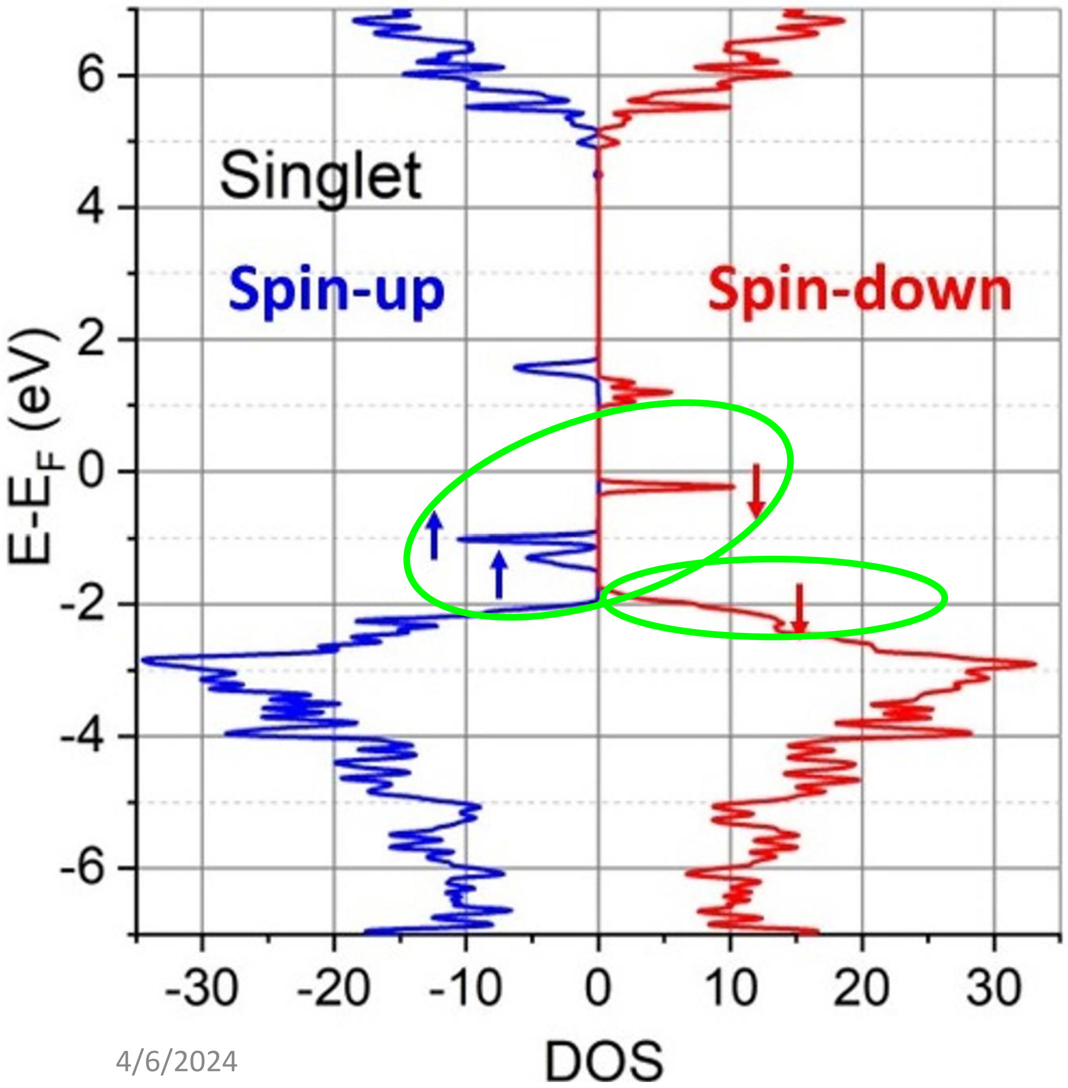
Guided by the changes in electron charge density caused by the optical excitation in the triplet, we searched a singlet ground state (SGS)

We found one (three) only 53 meV above the TGS.

The spin of two N atoms cancel out that of a third one: C_1 symmetry, 3 identical singlets rotated by 120°



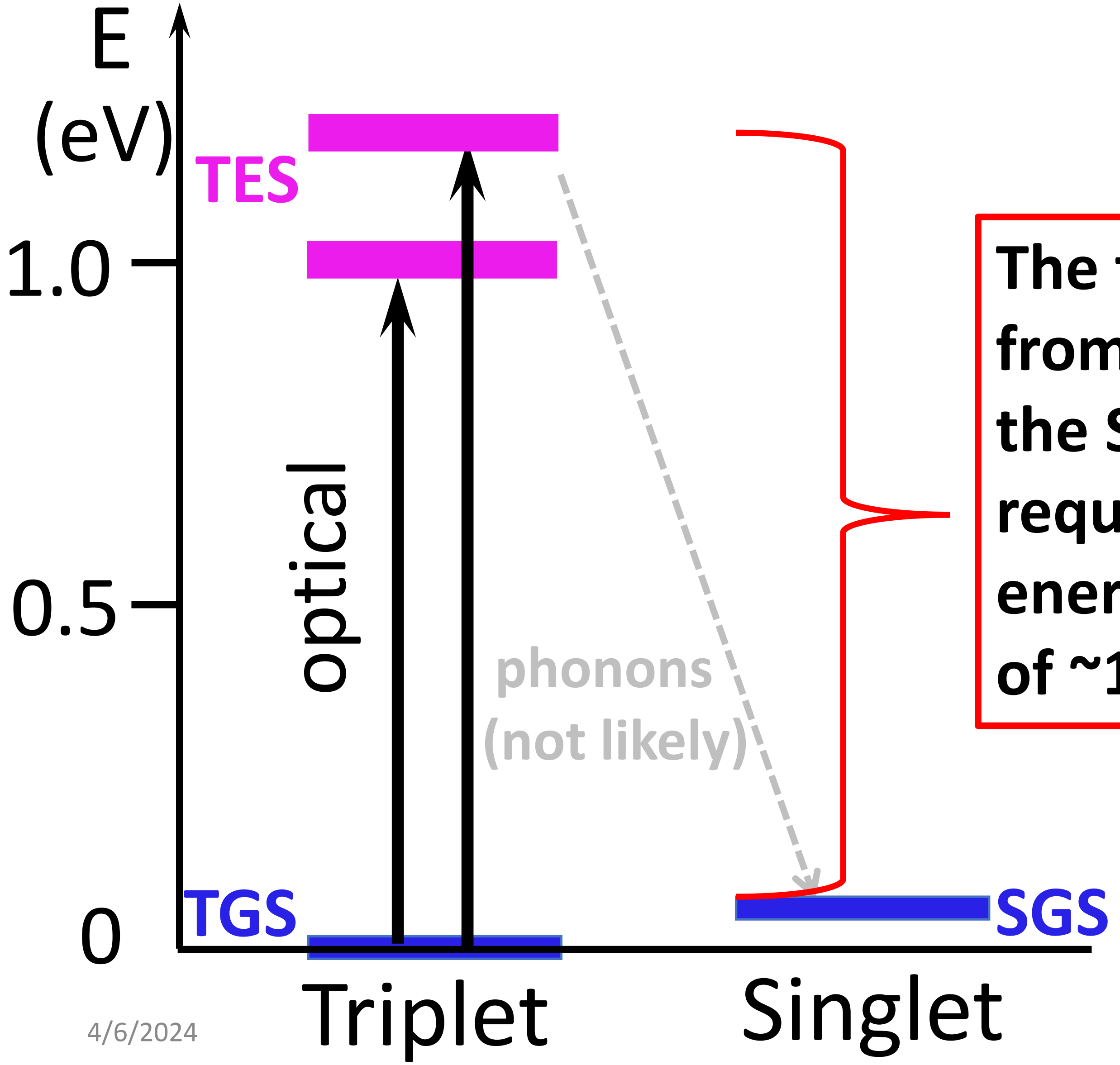
Results: $V_{Al}S_N$ Singlet Density of States



Three of the states determining the spin state of the defect occupy local narrow peaks located in the band gap.

One of the electrons occupies an IQP peak that merges the top of the VB, but it does not play a role because it is too deep.

Is it possible a direct TES → SGS transition?



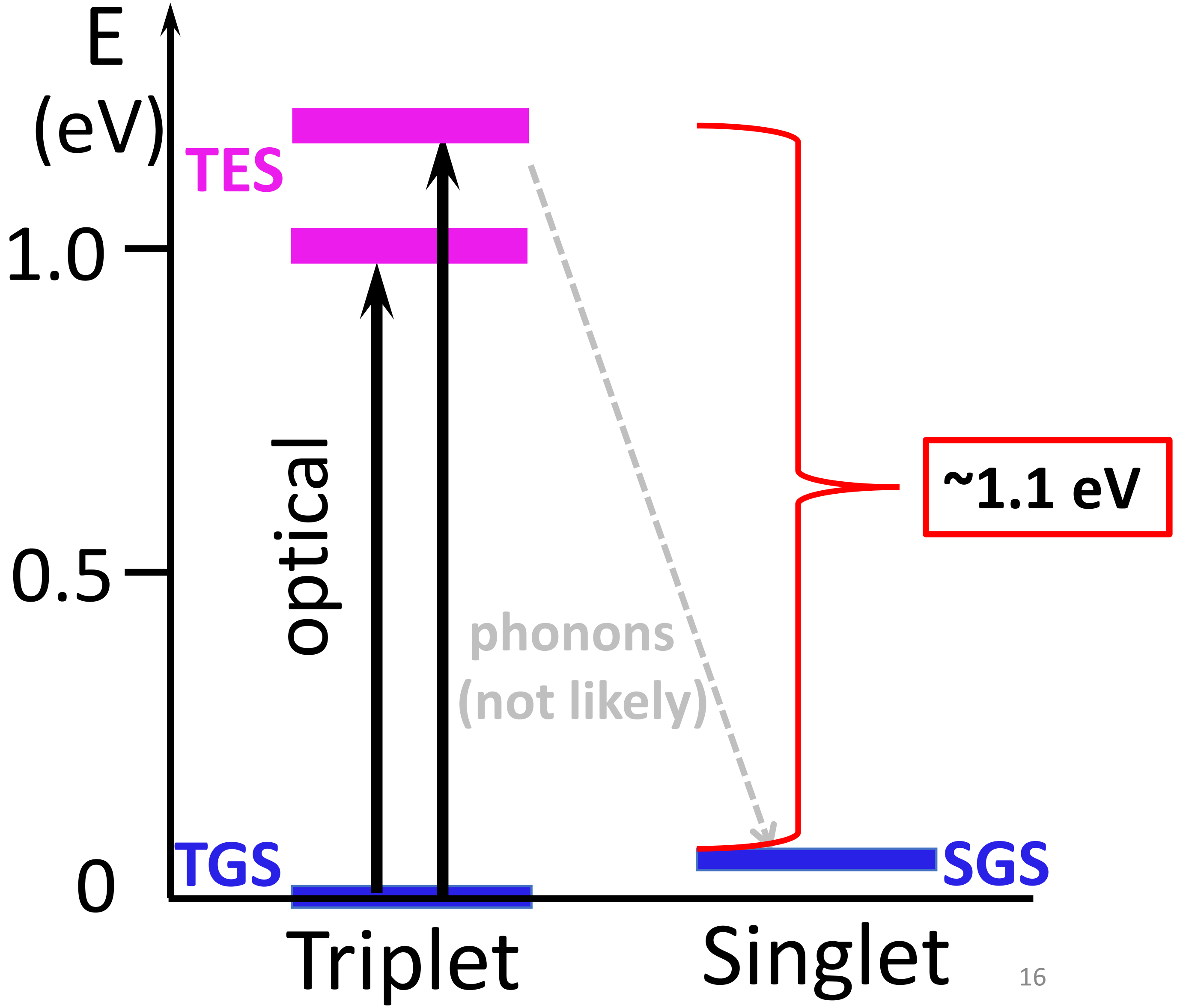
The transition from the TES to the SGS requires an energy decay of ~1.1 eV

A phonon-assisted transition is unlikely or at least unlikely to be efficient because the maximum phonon frequencies do not exceed ~100 meV

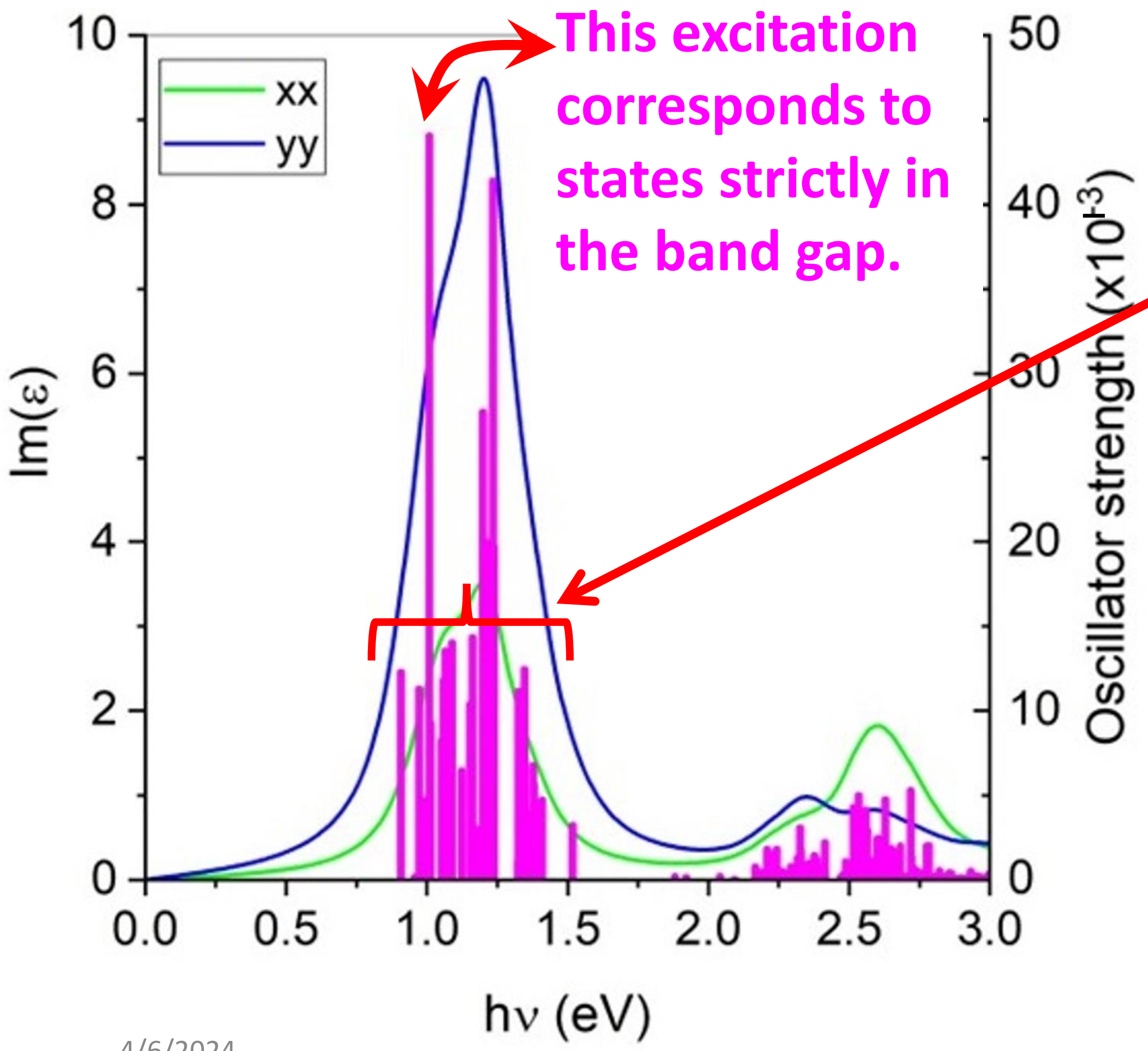
Is it possible a direct TES → SGS transition?

Is there any singlet excited state that is:

- **1 eV or less above SGS that may serve as an intermediate state?**
- **With high optical transition rate?**



Results: Optical excitation SES→SGS



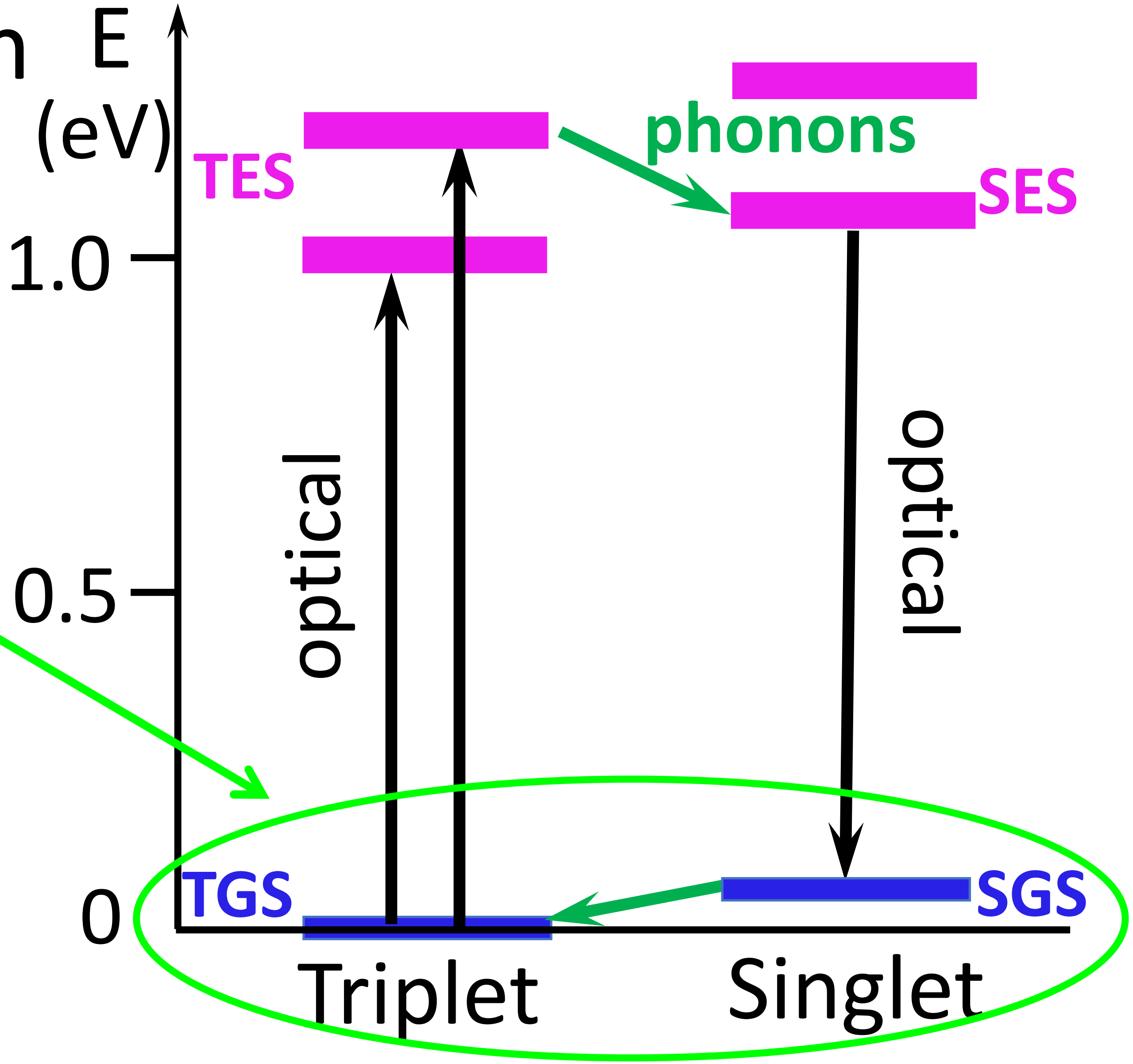
The low energy excitation group (from 0.9 to 1.55 eV) could serve as a the intermediate state to relax most of the energy optically

Unlike the case of the NV^{-} center in diamond, <u>the transition rate of the singlet excitation is large, as large as that of the triplet which is favorable for the spin-polarization cycle</u>

The SGS → TGS transition

Spin-changing transition:
SOC must allow $\vec{J}$ conservation

Yet it is a transition between two minima: **we can estimate the energy barrier**



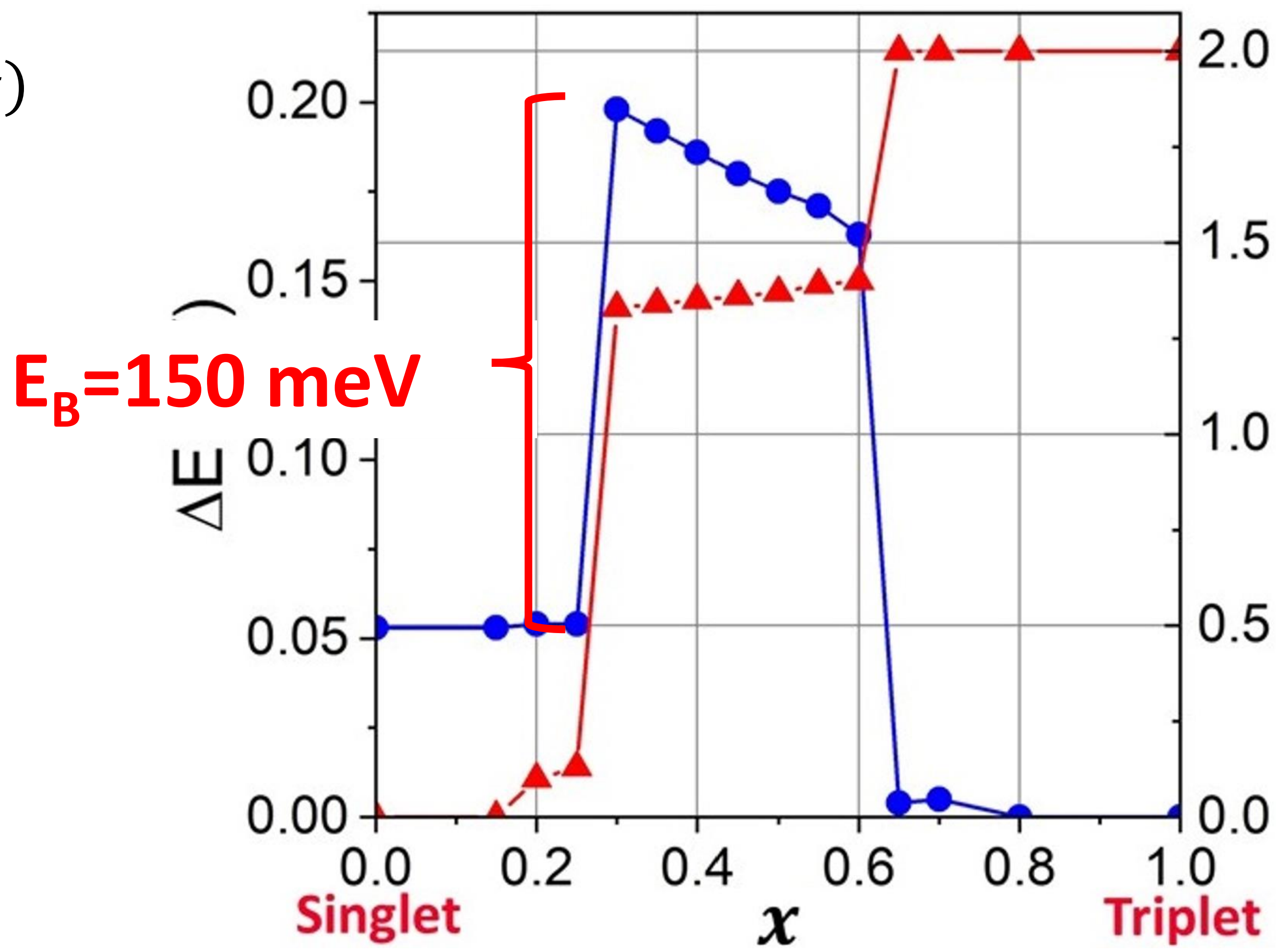
Results: The SGS → TGS transition

Simple transformation:

$$\vec{\mathbf{r}}_{\mathbf{i}}(x) = (1-x)\vec{\mathbf{r}}_{\mathbf{i}}(singlet) - x\vec{\mathbf{r}}_{\mathbf{i}}(triplet)$$

yields that the barrier should not exceed 150 meV.

Thus, **the transition can be phonon mediated**



Conclusion

The $V_{Al}S_N$ defect in wurtzite AlN has a spin-polarization cycle

- qualitatively identical to that of diamond
- that could perform as good as or better than the NV^- center in diamond