Quantum spin defects in industrially important semiconductors for scalable solid-state quantum technology

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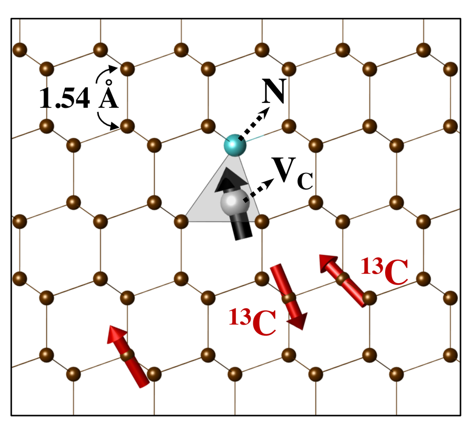
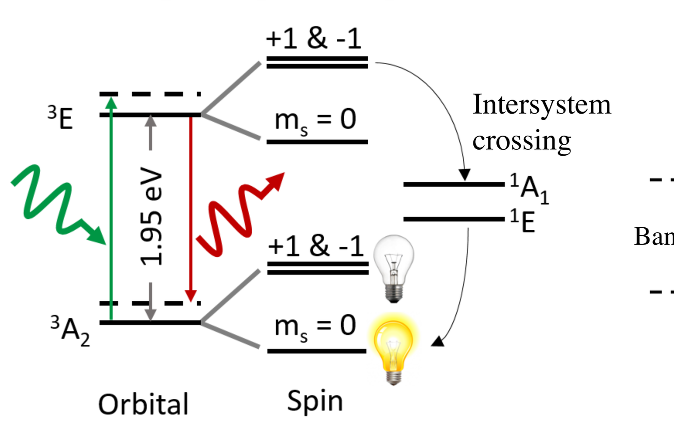
Defect qubits in solids: NV in diamond vs. Divacancy in SiC

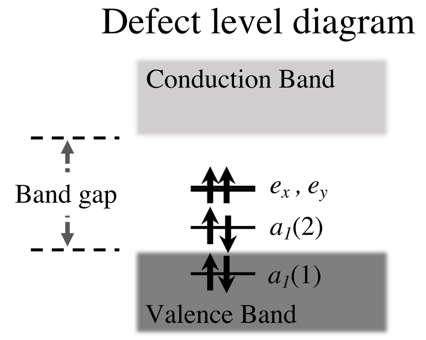
Nitrogen-vacancy (NV) center in diamond: optically addressable single spin[1]

Electronic structure
- Robust S=1 spin-triplet ground state
- Zero-field splitting (2.9 GHz)

Optically detected magnetic resonance (ODMR)
- Intra-defect excitation and intersystem crossing
- Microwave control of spin transitions
- Spin-dependent photoluminescence

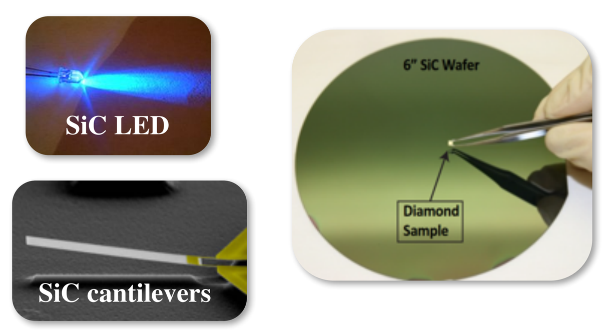




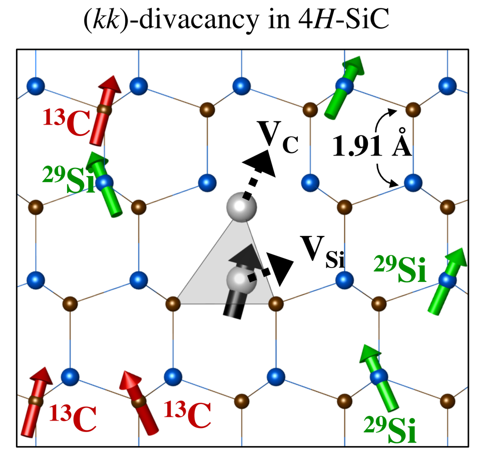


- The NV center is being developed for quantum information processing, quantum sensors in physical and biological systems, and hybrid quantum systems[2].

Divacancy in SiC: NV-analogs in industrial materials[3-9]



(*kk*)-divacancy in 4*H*-SiC



Common features
- C_{3v}, 6 e^-, S=1[4]
- Optically addressable[6]
- Single-spin isolation[9]
- Room-T operation[6]

✓ Excellent materials systems for optoelectronic devices, micromechanical systems and power electronics industry.

✓ Advanced material technologies are available.
- Wafer-scale growth
- Doping and device fabrication

- Recent developments of SiC divacancy includes: dynamic nuclear polarization and electrical control, etc[7].
- Related systems in SiC include silicon vacancy[8].

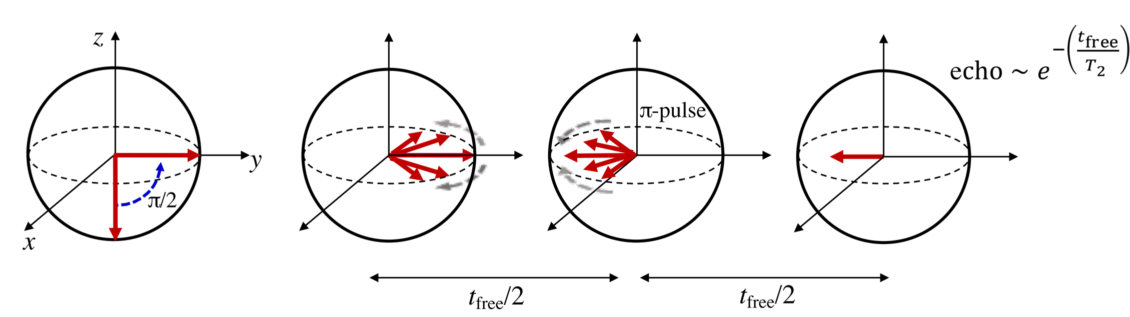
The divacancy coherence times have been measured much longer than that of the NV center in diamond: 1.2 ms for divacancy in SiC[9] (T = 20 K) and 0.63 ms for NV in diamond[10] (Room T.).

Quantum decoherence dynamics of divacancy spins in SiC: Theory and experiment

We combine theory and experiment to investigate the origin of the robust coherence of divacancy qubits in naturally isotopic 4*H*-SiC[11].

*Natural SiC contains ^{29}Si (4.7%) and ^{13}C (1.1%) spin-1/2 nuclear spins. In diamond, only ^{13}C exists.

Experiment: ODMR + Hahn-echo pulse sequence for the (*kk*)-divacancy spin ensemble in 4*H*-SiC

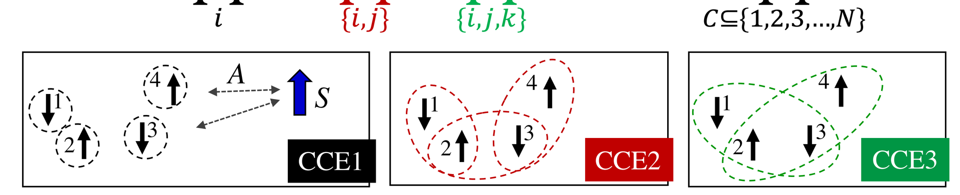


Coherence function:

$$\mathcal{L}(t) \equiv \frac{tr[\rho_{tot}(t)S_+]}{tr[\rho_{tot}(0)S_+]}$$

Theory: Quantum-bath model + Cluster correlation expansion[12] (CCE)

$$\mathcal{L}(\tau) = \prod_i \tilde{\mathcal{L}}_i(\tau) \prod_{\{i,j\}} \tilde{\mathcal{L}}_{i,j} \prod_{\{i,j,k\}} \tilde{\mathcal{L}}_{i,j,k} \cdots = \prod_{C \subseteq \{1,2,3,\ldots,N\}} \tilde{\mathcal{L}}_C(\tau)$$



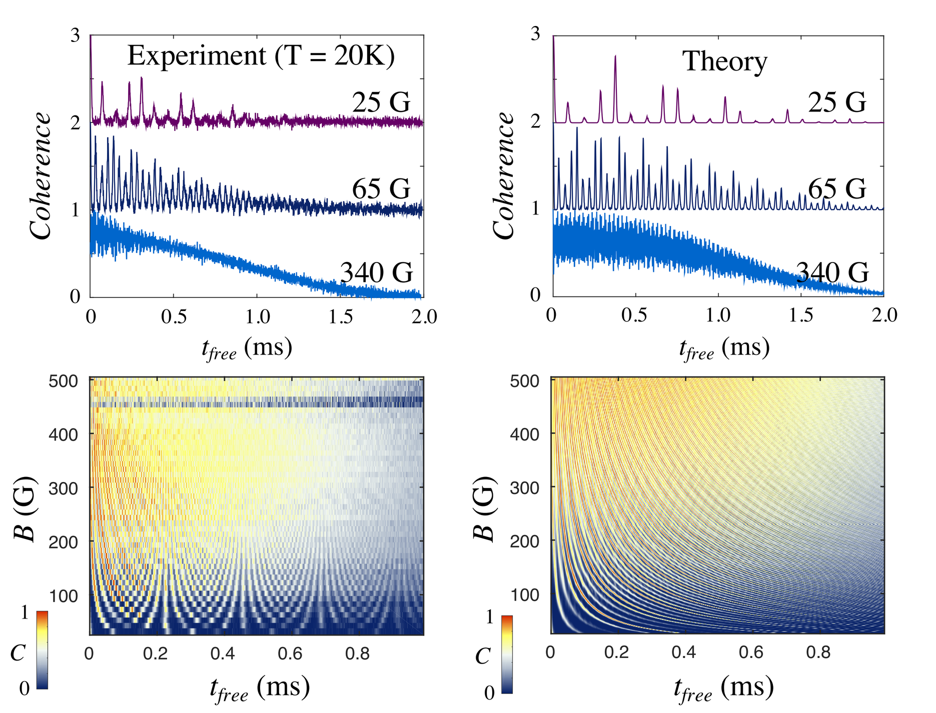
$H = H_S + H_B + H_{int}$, where

$$\mathcal{H}_S = -\gamma_e \vec{B} \cdot \vec{S} + \Delta S_z^2$$

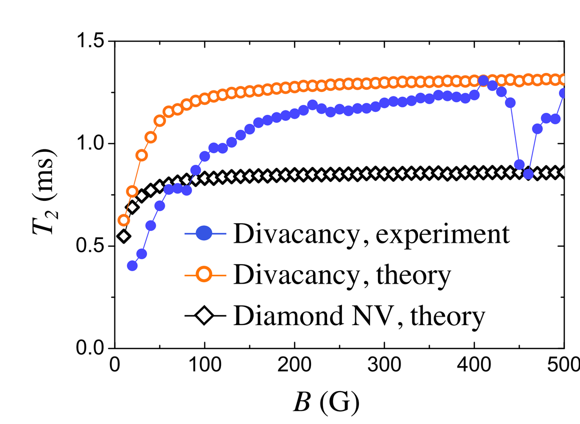
$$\mathcal{H}_B = -\vec{B} \cdot \sum_i \gamma_i \vec{I}_i + H_{n-n}$$

$$\mathcal{H}_{int} = \vec{S} \cdot \sum_i \vec{A}_i \cdot \vec{I}_i$$

Hahn-echo coherence of the divacancy ensemble in 4*H*-SiC



T_2 as a function of magnetic field

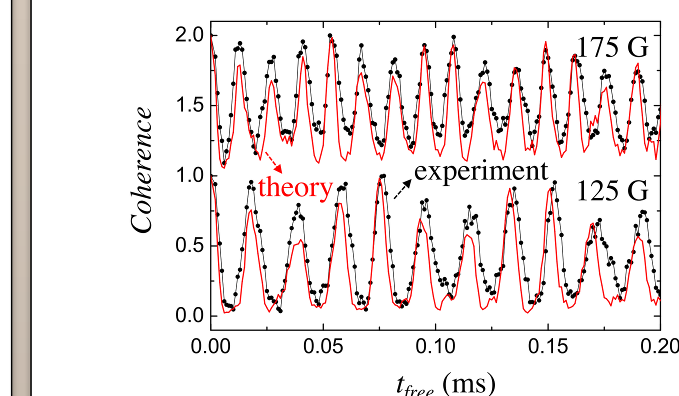


- The coherence time (T_2) increases as a function of *B*-field.
- T_2 of 1.3 ms is significantly longer than that of NV (≈ 0.8 ms in theory)

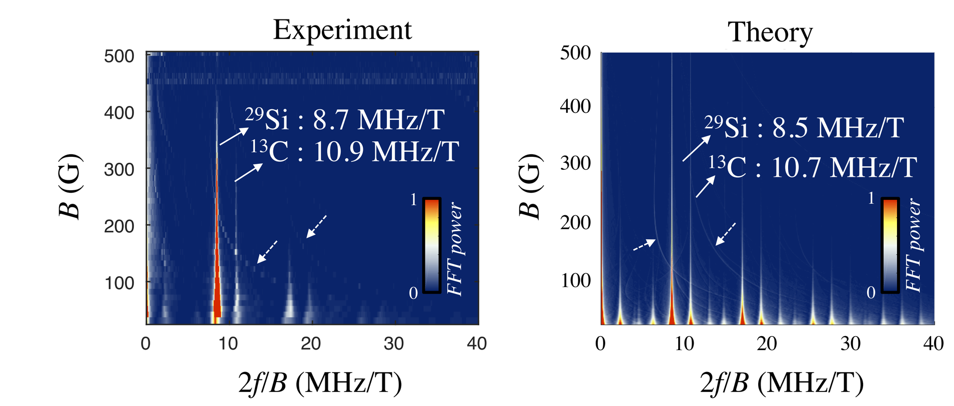
Quantum decoherence dynamics of divacancy spins in SiC: Analysis

The origin of the robust divacancy coherence is identified.

Direct comparison between theory and exp.



Fast Fourier Transform (FFT) power spectra



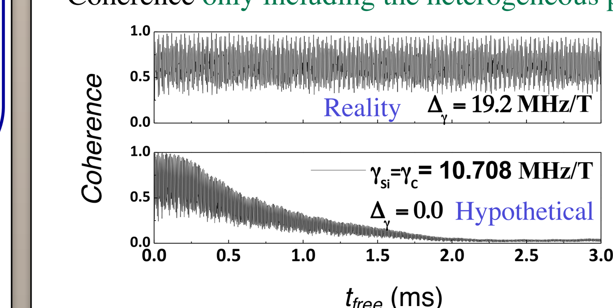
Coherence function factorization

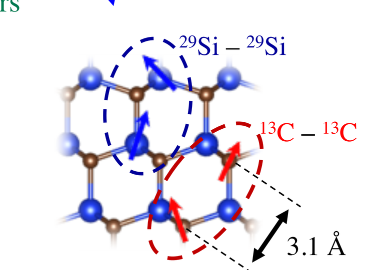
$$\mathcal{L}_{(kk)}(\tau) = \prod_i \tilde{\mathcal{L}}_i(\tau) \prod_{\{i,j\}_{hetero}} \tilde{\mathcal{L}}_{i,j} \prod_{\{i,j\}_{homo}} \tilde{\mathcal{L}}_{i,j}$$

γ_C = 10.71 MHz/T
γ_{Si} = -8.46 MHz/T

Homonuclear spin pairs

Coherence only including the heterogeneous pairs



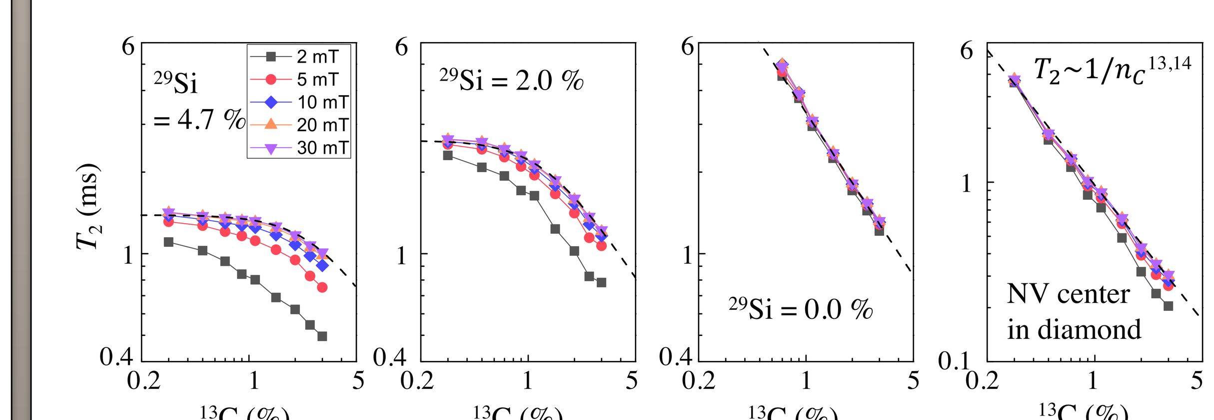


✓ Strongly coupled nuclear spin clusters cannot be formed in SiC in the presence of $\Delta\gamma$.
- Smaller γ_{Si} than γ_C.
- Larger a_{SiC} than a_C.
- Binary crystal structure.

✓ On the other hands, NV center coherence[13] is mainly driven by a few strongly coupled homonuclear spin paris.

Less fluctuating intra-crystalline magnetic environment in SiC → a longer T_2 time in SiC than in diamond.

T_2 can be enhanced by isotopic purification (Theory)



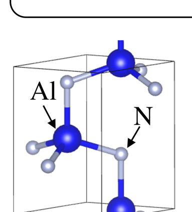
✓ A longer qubit coherence time could be achieved if the ^{29}Si or ^{13}C nuclei are removed by isotopic purification.

First-principles design of new spin qubits in piezoelectric AlN: Motivation and methods

Industrial materials + coherent spin qubits → scalable solid-state quantum devices[3,7]

Material	Band gap (eV)
Diamond	5.5
4*H*-SiC	3.3
w-AlN	6.2
GaN	3.4
ZnO	3.4
Si	1.1

We consider *w*-AlN as a new potential host material for defect qubits[15,16].



✓ Strong piezoelectricity
✓ Strong candidate for hybrid quantum systems.
✓ Material for Optomechanics

Question: Can we identify a NV-analog in piezoelectric *w*-AlN?
Method: Materials design from first-principles!

We use density functional theory to predict and design new defect spin qubits.

Qubit Requirements
Triplet spin,
Localization,
Intra-defect excitation, …

Materials Issues
doping,
defect stability,
strain, …

First-principles Computational Modeling

Spectroscopy
EPR,
Photoluminescence,
…

Experimental Issues
Optical transition energies,
Reasonable parameter range,
…

- **Hybrid density functional theory[17]**

$$v_{xc}(\boldsymbol{r},\boldsymbol{r}') = \alpha v_x^{ex}(\boldsymbol{r},\boldsymbol{r}') + (1-\alpha)v_x(\boldsymbol{r}) + v_c(\boldsymbol{r})$$

Which α for solids? Relation ship $\alpha = \epsilon_\infty^{-1}$ is inferred from:
1. Empirical observations[18]
2. Many-body perturbation theory[17] (static COHSEX approximation).

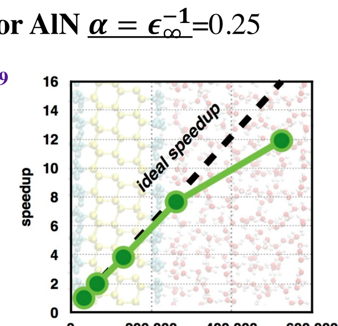
→ *α is self-consistently determined*[17] **:For AlN** $\alpha = \epsilon_\infty^{-1}$=0.25

- **GW many-body perturbation theory[19]**

$$E_n^{QP} = \epsilon_n^{KS} + \langle\Sigma(E_n^{QP})\rangle_n - \langle V_{xc}\rangle_n$$
$$\Sigma^{GW} = iGW$$

⟨WEST!⟩

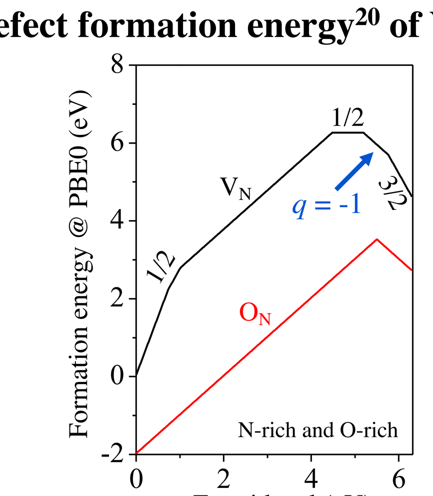
http://www.west-code.org/



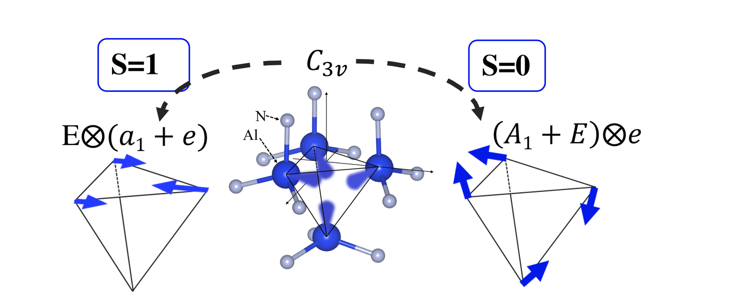
First-principles design of new spin qubits in piezoelectric AlN: Results

Negatively charged N vacancy under strain is a strong candidate for qubits in AlN.

Defect formation energy[20] of V_N and O_N

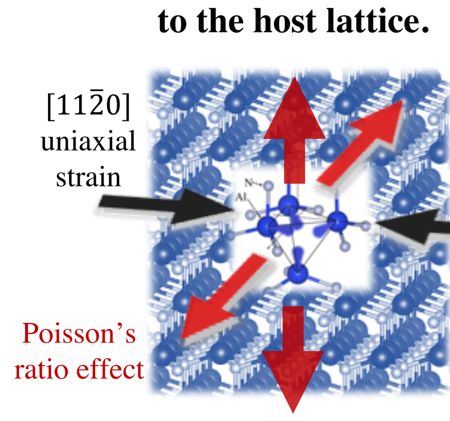


Spin-dependent Jahn-Teller distortion in V_N^-

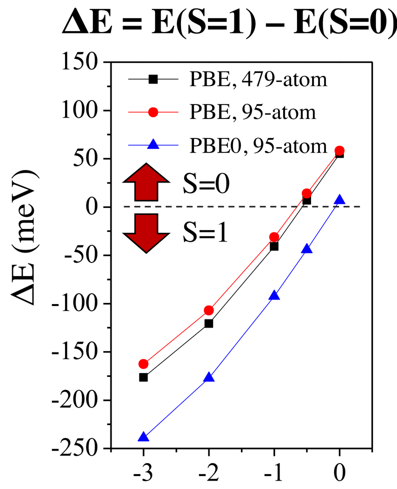


- V_N^0 has been detected using electron paramagnetic resonance.
- V_N^- has two nearly degenerate S=0 and S=1 states.

Uniaxial strain is applied to the host lattice.

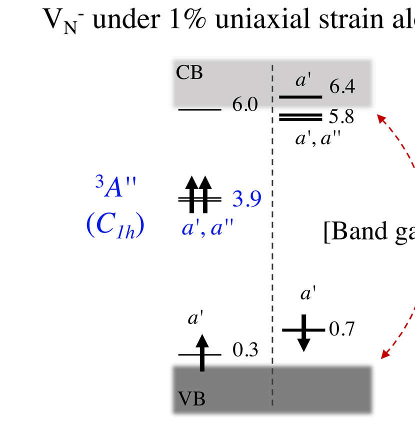


ΔE = E(S=1) – E(S=0)



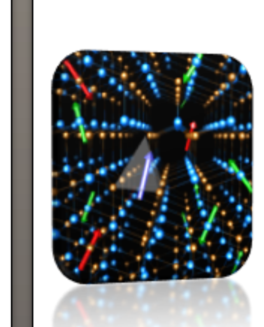
Defect level diagram (G_0W_0@PBE)

V_N^- under 1% uniaxial strain along [11$\bar{2}$0]



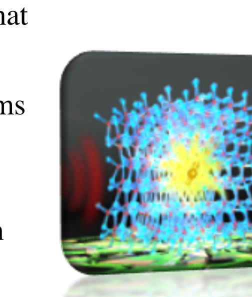
Conclusions, Reference, and Acknowledgement

Quantum decoherence dynamics of divacancy spins in 4*H*-SiC[11]



- We showed theoretically and experimentally that the T_2 time of the divacancy reaches 1.3 ms.
- We found that the robust coherence in SiC stems from the combination of the two effects:
  1. The ^{29}Si and ^{13}C spin baths are decoupled.
  2. The active homonuclear spins are are much further apart than those in diamond.
- Our results point to polyatomic crystals as promising hosts for coherent qubits in solids.

Design of new defect qubits in aluminum nitride[15]



- We found that negatively charged N-vacancy (V_N^-) have $^3A''$ and $^3A_2$ spin-triplet ground states under uniaxial and biaxial strain, respectively.
- The ground states of Al-related defects may not be suitable as spin qubits as they undergo strong resonance with the valence states of AlN.

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Acknowledgements

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U.S. DEPARTMENT OF ENERGY | Office of Science
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The computing resources were provided by: THE UNIVERSITY OF CHICAGO Research Computing Center, NERSC, Argonne NATIONAL LABORATORY