Solid-state Quantum Information Using Defects in Semiconductors

Hosung Seo

아주대학교
물리학과

Outline

- **Quantum Technologies using deep-level defects**
  a. Hardware and Software of Quantum Information
  b. The NV center in diamond
  c. Defect spin qubits in wide-gap semiconductors
    1. Quantum decoherence (H. Seo *et al.*, *Nat. Comm.* (2016).)
    2. Design of new spin qubits (H. Seo *et al.*, *arXiv:1709.09818* (2017).)

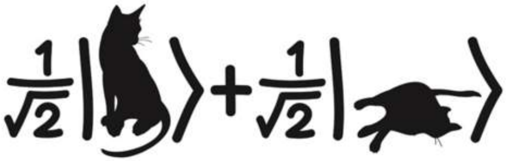

Giulia Galli


David Awschalom

THE UNIVERSITY OF CHICAGO 1890

Argonne NATIONAL LABORATORY

Quantum Age



Understanding of quantum principles

Nano-technology
Microelectronics
Materials science

”The Silicon Age”

Quantum Technologies

”The Quantum Age”

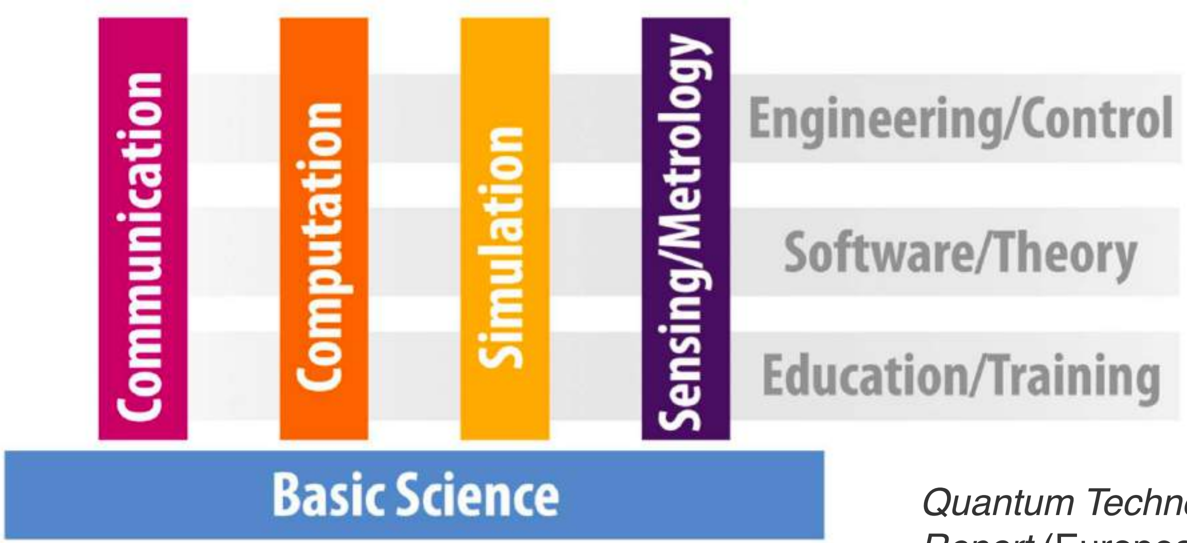
Google IBM
intel

1900~1950 1950 ~ 2020 2020 ~

Quantum Technologies

Technology and science based on quantum principles such as coherence and entanglement.

J. P. Dowling and G. J. Milburn, *Quantum technology: the second quantum revolution*, Phil. Trans. R. Soc. Lond. A 361, 1655 (2003).



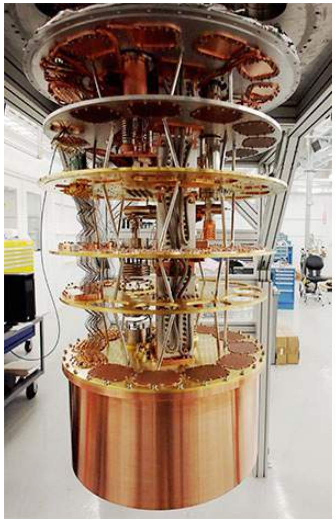
Quantum Technologies Flagship Intermediate Report (European Commission, 2017)
http://go.nature.com/2m48x41

Requirements to Realize Quantum Technologies

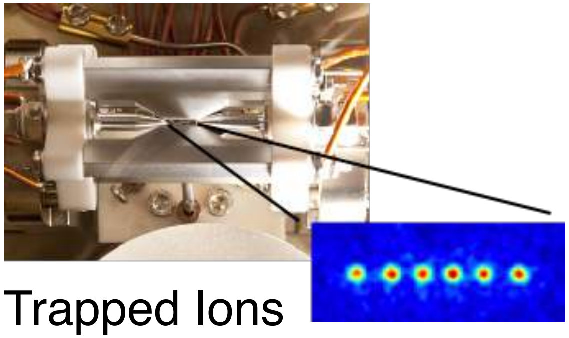
☐ Quantum Software

- **Communication**
  - ✓ BB84
- **Computation**
  - ✓ Shor’s algorithm
  - ✓ Grover’s algorithm
- **Simulation**
  - ✓ Bravyi-Kitaev
- **Sensing/Metrology**
  - ✓ Entanglement-based

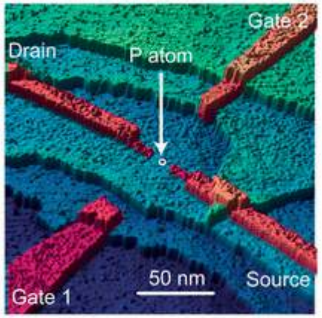
☐ Quantum Hardware



Superconducting Circuits



Trapped Ions

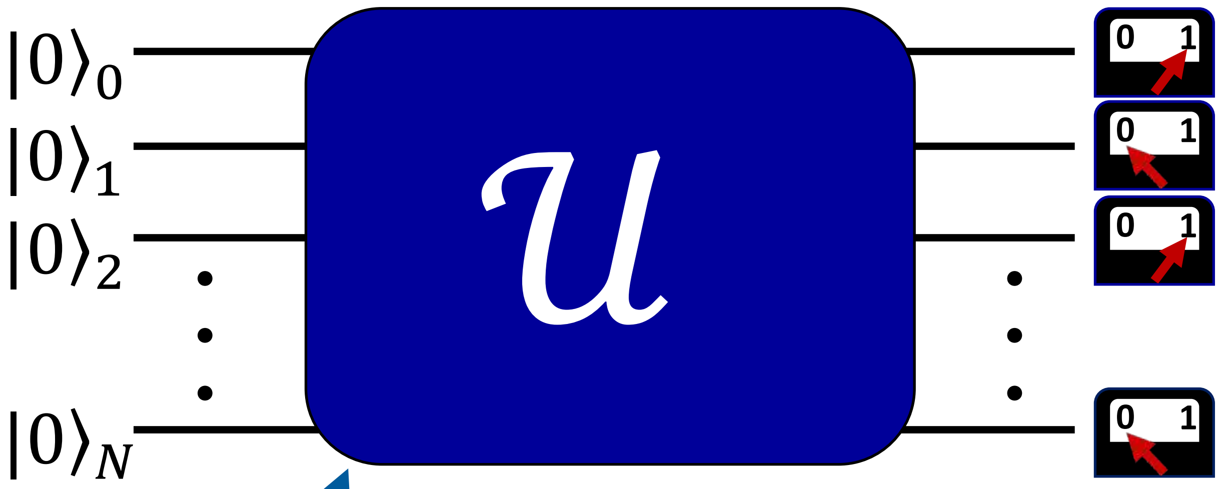


Silicon dopants

Diamond Color Centers

G. Popkin, *Science* **354**, 1090 (2016).

Requirements to be Quantum Bits



Initialization **Manipulation** **Readout**

$|0\rangle_0$

$|0\rangle_1$

$|0\rangle_2$

$|0\rangle_N$

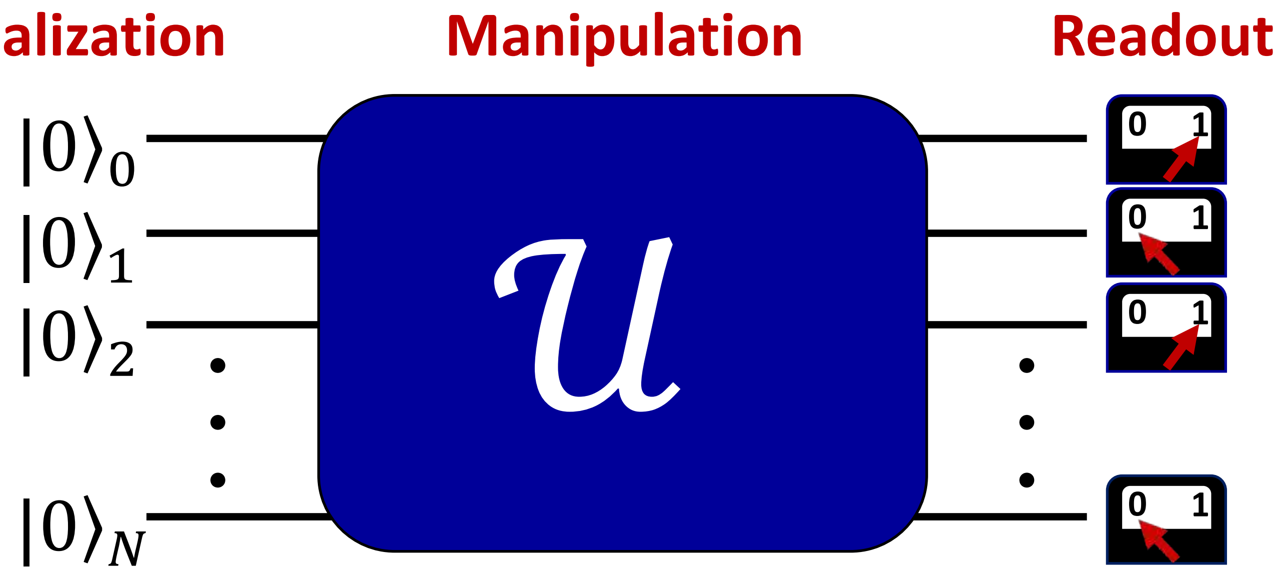
Scalable and well-characterized physical systems

Ideal physical implementation:

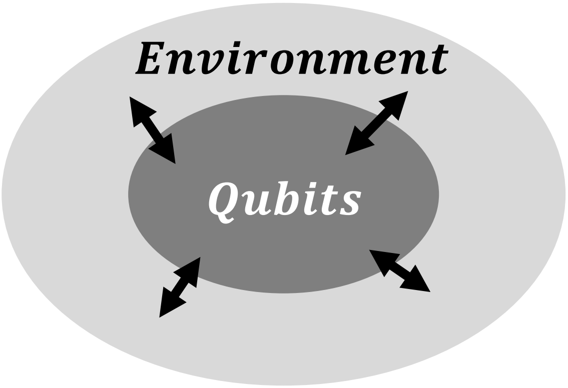
$$\mathcal{U}(t) = \mathcal{T}_{\leftarrow}\exp[-\frac{i}{\hbar}\int_0^t \boldsymbol{\mathcal{H}_s(t')}dt']$$

D.P. DiVincenzo, Fortschritte der Physik **48**, 771 (2000).

Requirements to be Quantum Bits



Initialization **Manipulation** **Readout**

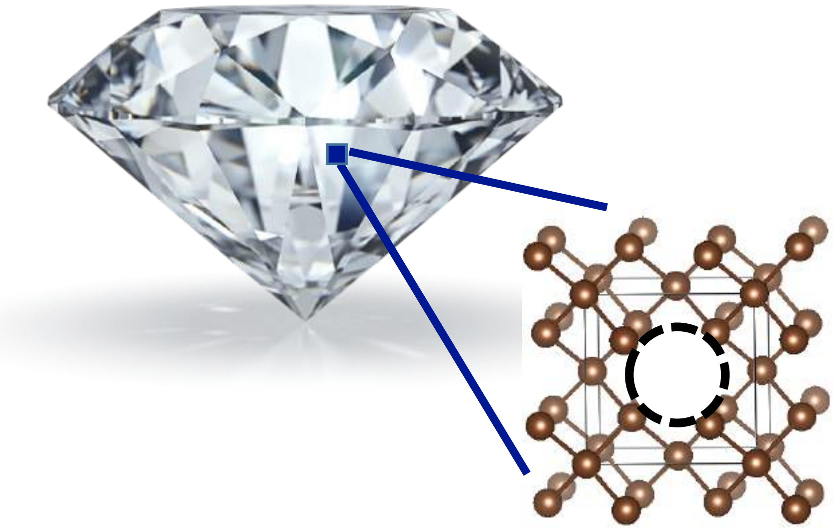


In reality:

$$\mathcal{U}(t) = \mathcal{T}_{\leftarrow}\exp\left[-\frac{i}{\hbar}\int_0^t (\boldsymbol{\mathcal{H}_s(t')} + \boldsymbol{\delta\mathcal{H}_E(t')})dt'\right]$$

➔ (1) Inaccurate control + (2) “Decoherence”

Deep-level defects:
Quantum states trapped in ‘crystal vacuum’

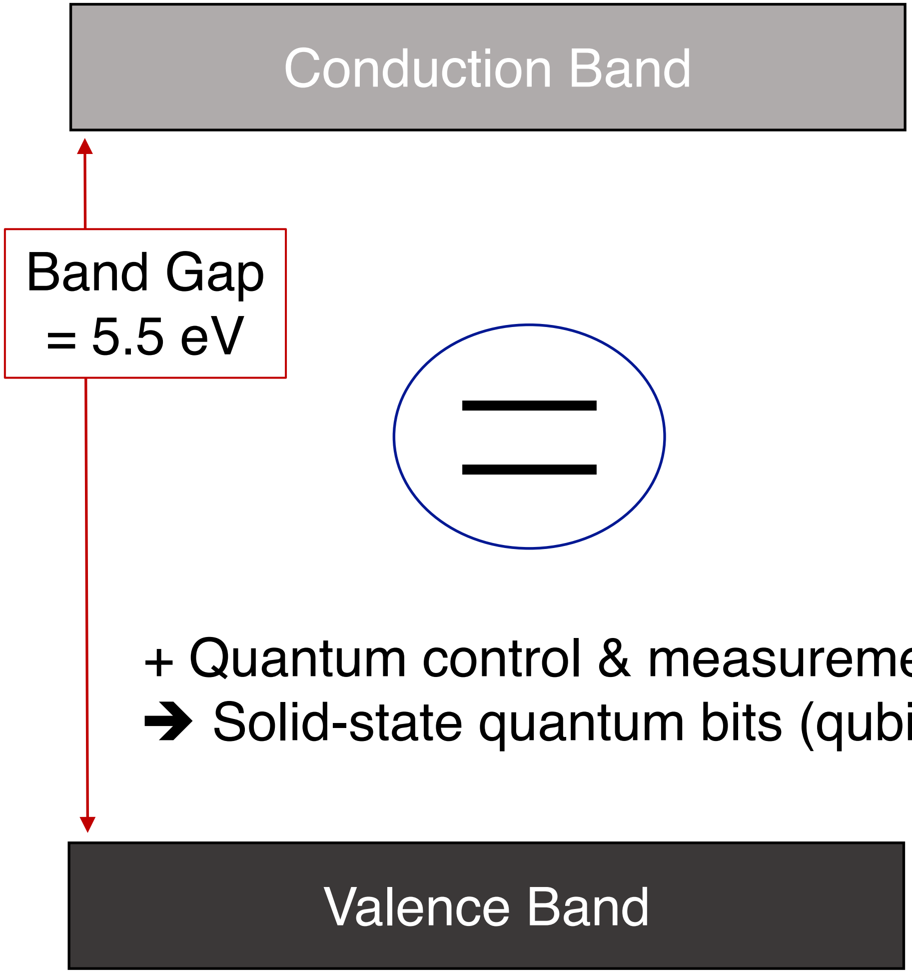


Color Centers


Boron

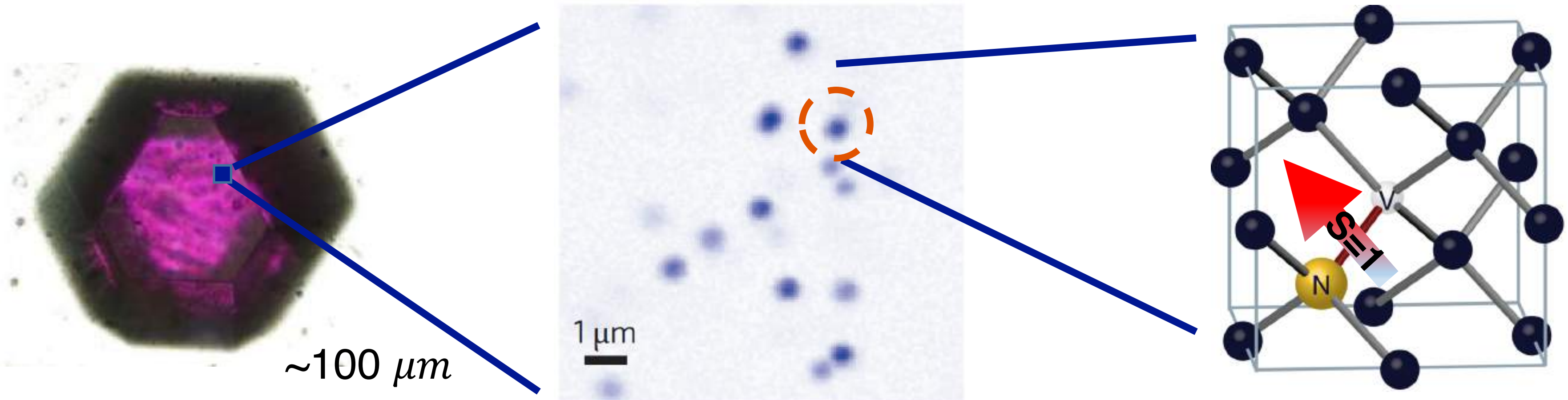

Nitrogen


Nitrogen-vacancy



+ Quantum control & measurement
➔ Solid-state quantum bits (qubits)

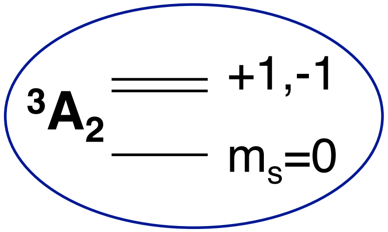
Nitrogen-vacancy (NV) center in diamond



VM Acosta thesis (2011)

WA Gao *et al.*, *Nat. Photon.* (2015).

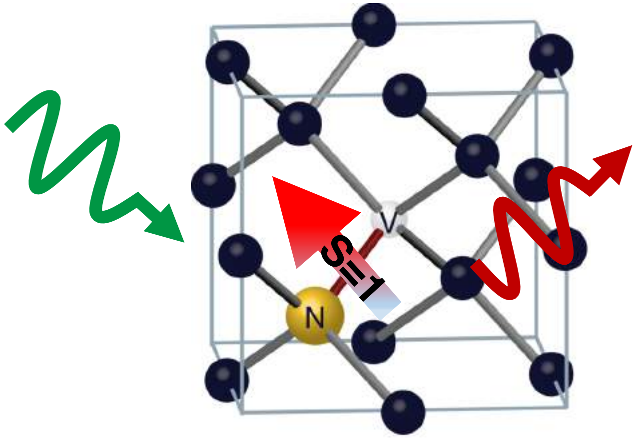
- NV center = artificial ‘atom’ trapped in the solid-state
- Ground state with spin triplet (S=1)
- Long coherence time at ‘room temperature’
- Single-spin optical addressability

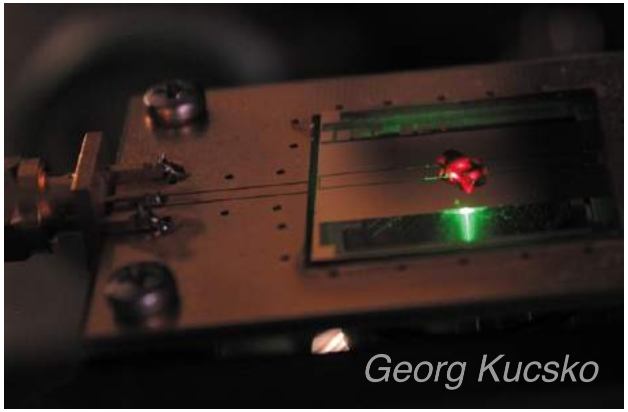


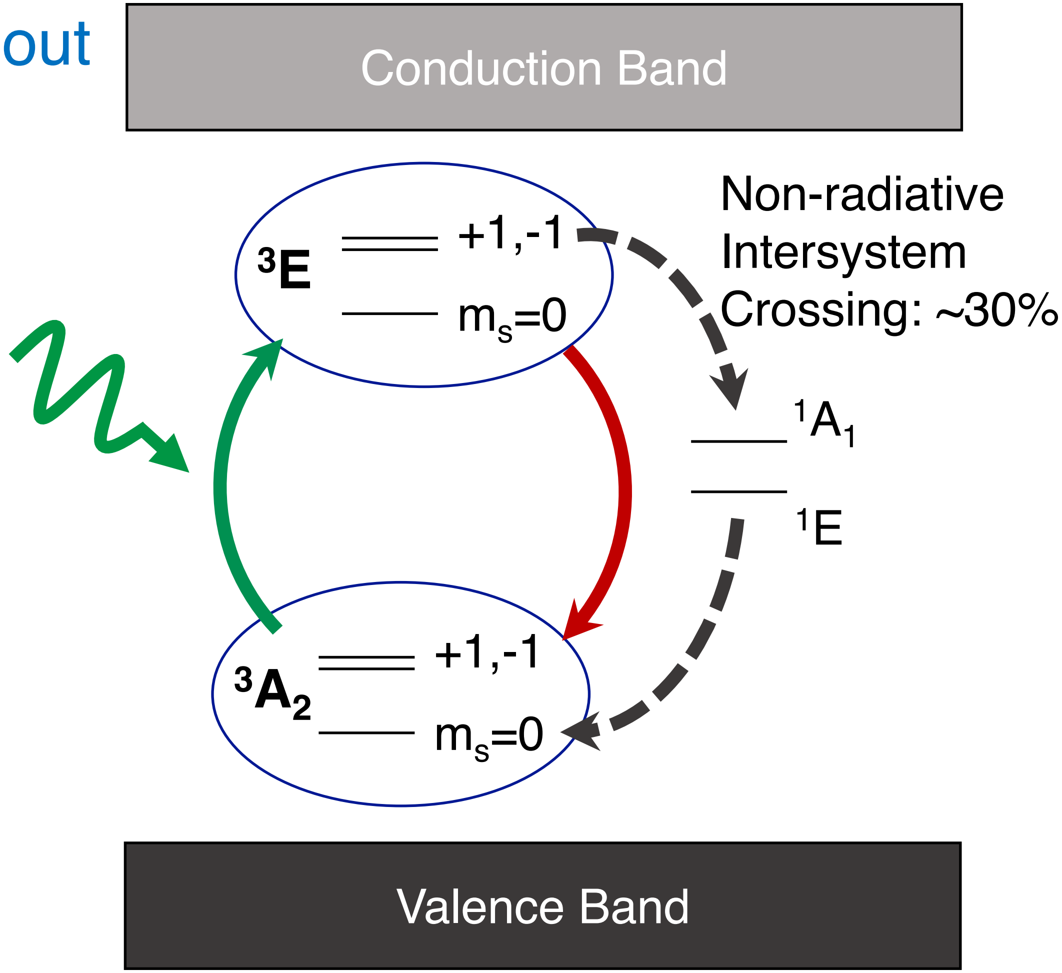
F Jelezko *et al. Phys. Rev. Lett.* (2004).

NV centers as qubits

Optical initialization and readout




Georg Kucsko


Conduction Band

^3E

+1,-1

m_s=0

Non-radiative Intersystem Crossing: ~30%

$^1A_1$

^1E

$^3A_2$

+1,-1

m_s=0

Valence Band

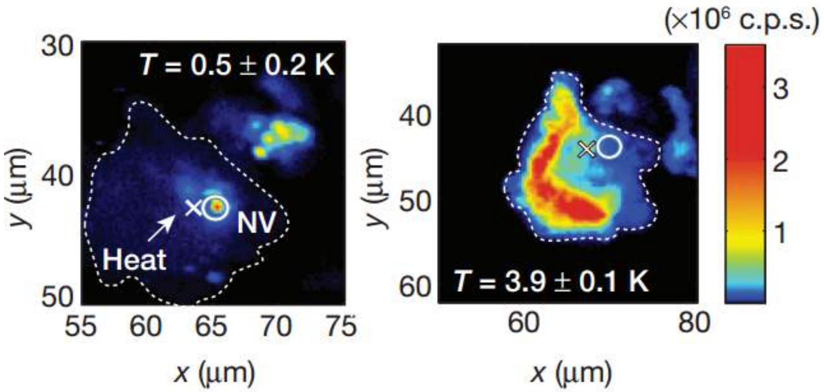
NV centers: applications in science and tech.

- Fundamental Science

Loop-hole free Bell inequality violation
Hensen *et al.*, *Nature* (2015).

Time Crystal
Choi *et al.*, *Nature* (2017).

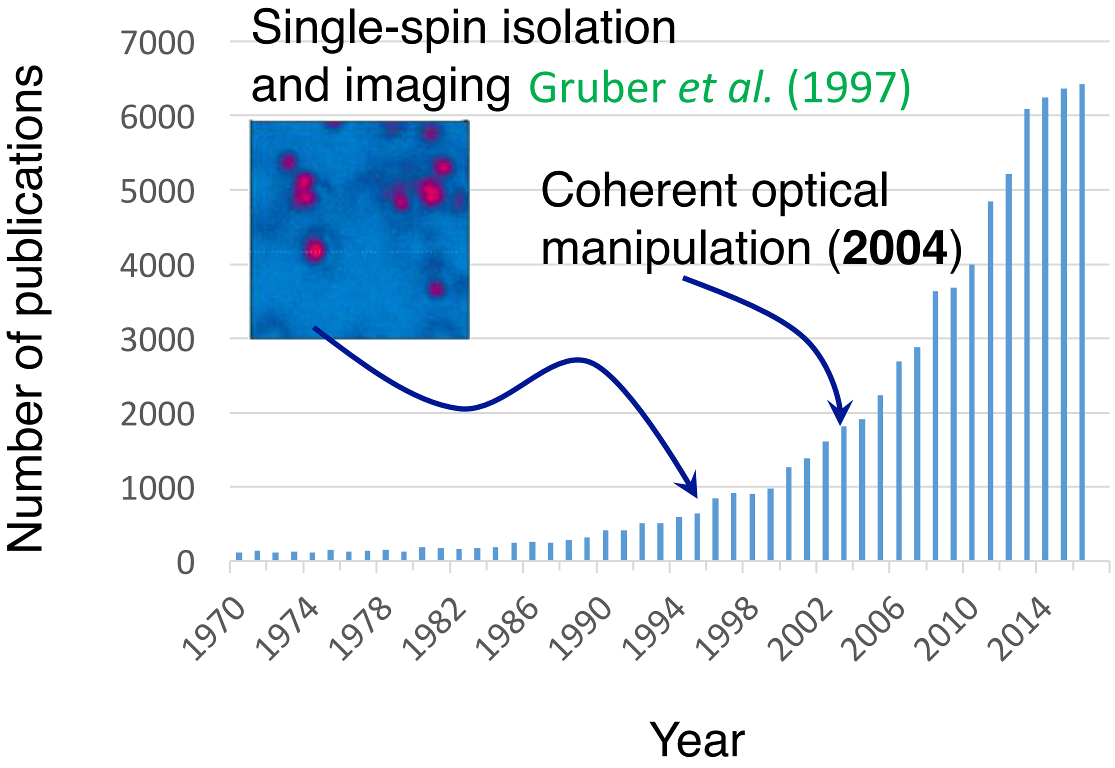
- Applications
  - Quantum Information processing
    van der Sar *et al.*, *Nature* (2012).
  - Nano-scale sensors



Biological thermometer
: Kucsko *et al.*, *Nature* (2013).

NV center in diamond: Continuing Breakthroughs and Innovations

“Nitrogen Vacancy Center in Diamond” searched in Google Scholar

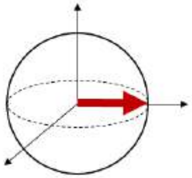


Single-spin isolation and imaging Gruber *et al.* (1997)

Coherent optical manipulation (**2004**)

Going beyond NV:
Spin qubits in wide-gap semiconductors

Qubit properties



- Low Readout fidelity

➔ Hundreds of repetition

- Large phonon side band

➔ Hard to entangle qubits

- Orbital dephasing of the excited states

➔ Limiting high T operation

Host properties

- Chemically inert and durable

➔ Hard to process for device fab.

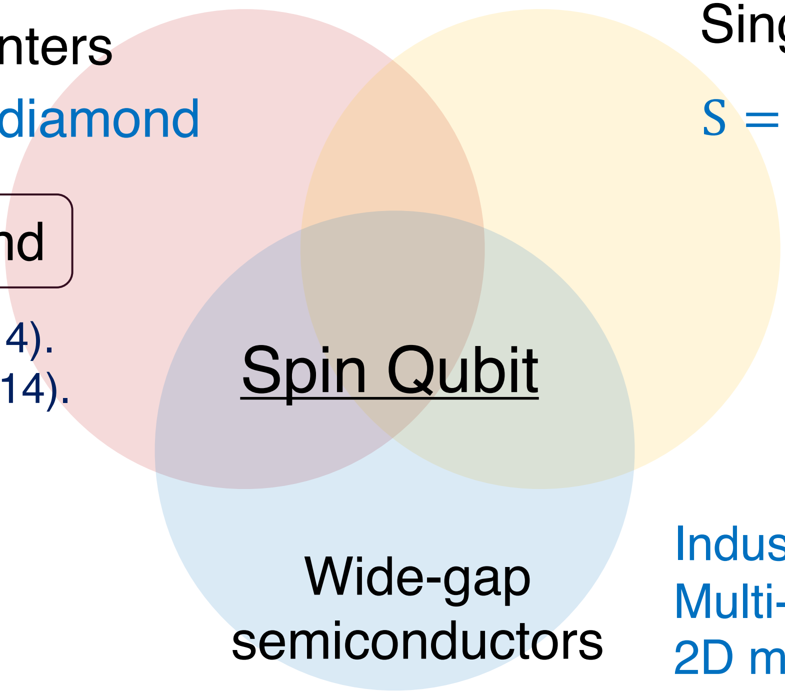
- Growth of eletronic-grade diamond is limited < a few mm

➔ scalable implementations are limited

- No interesting lattice functionality

NV is unique? No other choices?

Going beyond NV:
Spin qubits in wide-gap semiconductors



Color centers

> 500 even in diamond

SiV^- in diamond

Rogers *et al.* (2014).
Pingault *et al.* (2014).

Single spins

$S = \frac{1}{2}, 1, \frac{3}{2}, \dots$

<u>Spin Qubit</u>

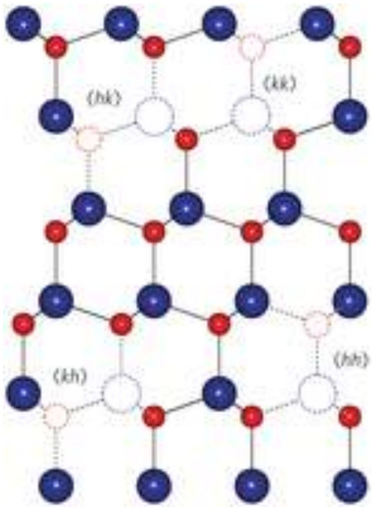
Wide-gap semiconductors

Industrial: SiC, GaN, SiO_2
Multi-functional: AlN, $BaTiO_3$
2D materials: *h*-BN, TMD

W. Koehl, <u>H. Seo</u>, G. Galli, and D.D. Awschalom, MRS Bulletin **40**, 1146 (2015).

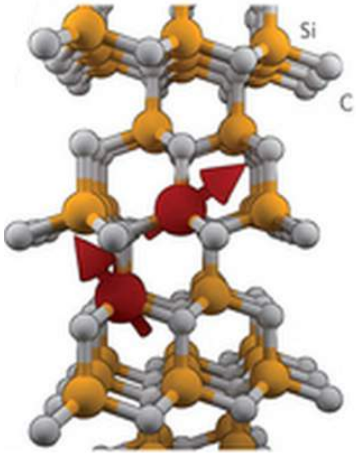
Alternative defect qubits in SiC

Di-vacancy



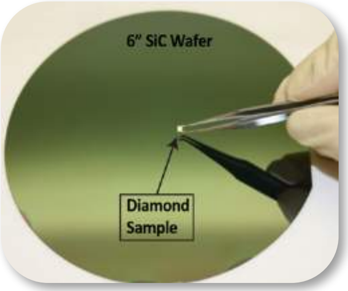
Koehl *et al.* (2011).

Si vacancy



Widmann *et al.* (2015).

✓ SiC-based technologies



SiC LEDs

SiC Power electronics

- The divacancy spins is 'NV-like' qubit in SiC.
- Surprisingly, the coherence time of the divacancy spin has been measured to be much longer than that of the NV center.

Hosung Seo (hseo2017@ajou.ac.kr)

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    1. **Quantum decoherence (H. Seo *et al.*, *Nat. Comm.* (2016).)**
    2. Design of new spin qubits (H. Seo *et al.*, *arXiv:1709.09818* (2017).)


A. Falk (IBM)


P. Klimov (Google)

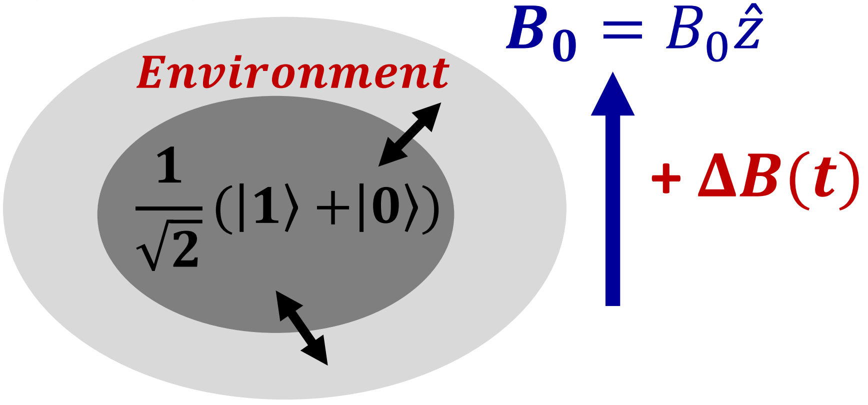

K. Miao


Giulia Galli


David Awschalom

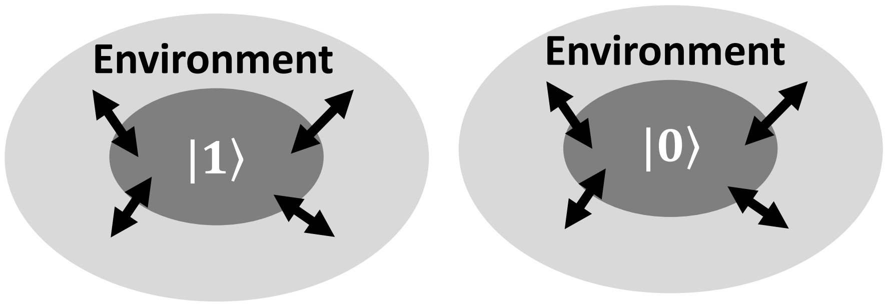
Quantum decoherence: transition from a pure state to a classical mixed state

$t = 0$



$$\mathcal{H} = -\hbar[\omega_0 + \boldsymbol{\Delta\omega_0(t)}]S_z$$

$t \rightarrow \infty$



'classical' mixed state

$$\rho(0) = \frac{1}{2}\begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} \xrightarrow[\mathcal{U}(\tau)]{} \frac{1}{2}\begin{pmatrix} 1 & \frac{1}{2}\exp[-\frac{S}{4}\tau] \\ \frac{1}{2}\exp[-\frac{S}{4}\tau] & 1 \end{pmatrix}$$

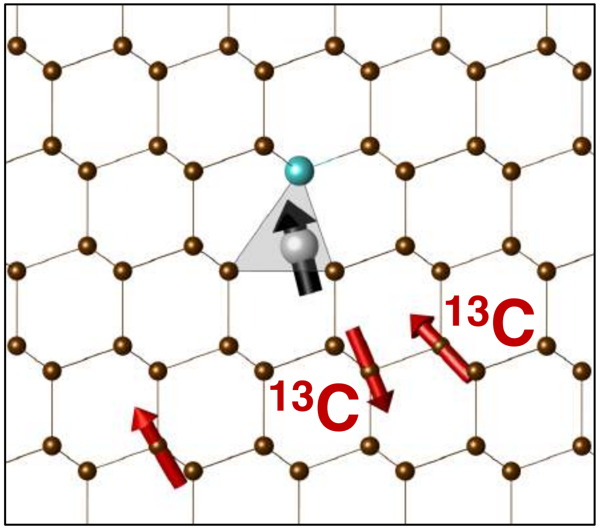
A. Abragam, *Principles of Nuclear Magnetism*, Clarendon Press, Oxford (1961).

Hosung Seo (hseo2017@ajou.ac.kr)

Nuclear spin driven decoherence of spin qubits

Defect spin qubits lose the coherence in the presence of fluctuating nuclear spin bath.

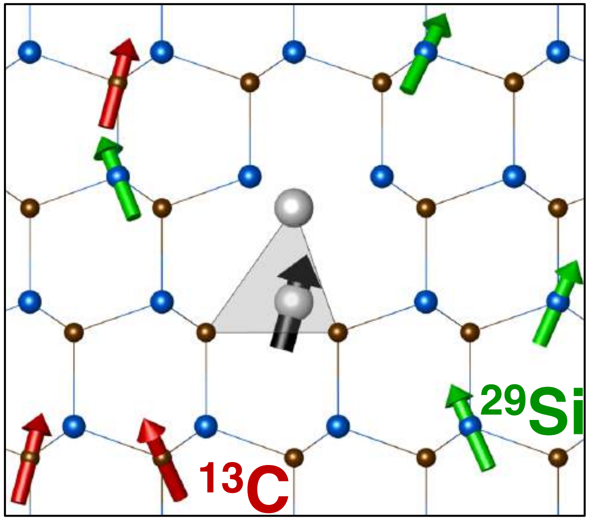
NV center $T_2 \approx 0.6$ ms



1.1% ^{13}C
($I_C = ½$)

Stanwix *et al.*, PRB (2010)
Maze *et al.*, PRB (2008).

Divacancy $T_2 \approx 1.3$ ms



1.1% ^{13}C
($I_C = ½$)

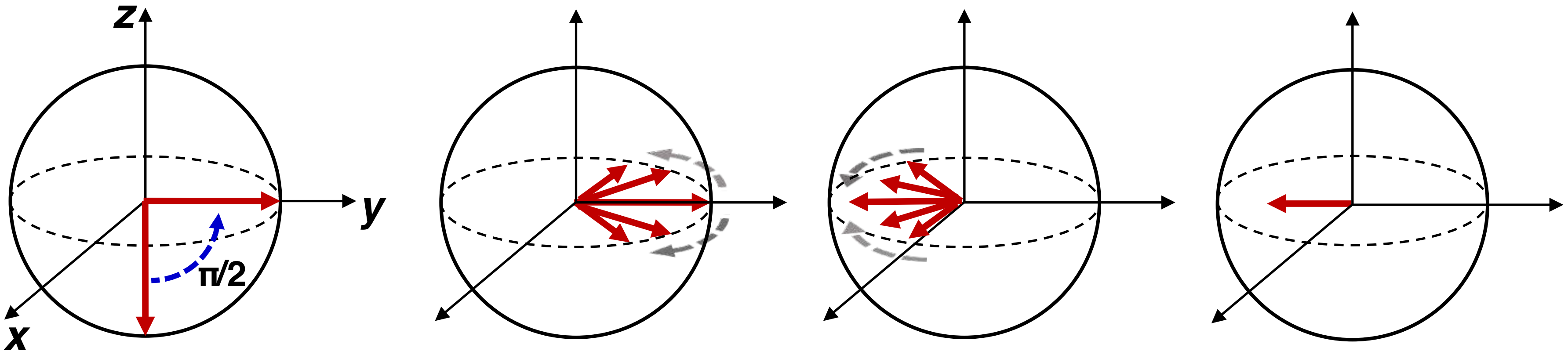
&

4.7% ^{29}Si
($I_{Si} = ½$)

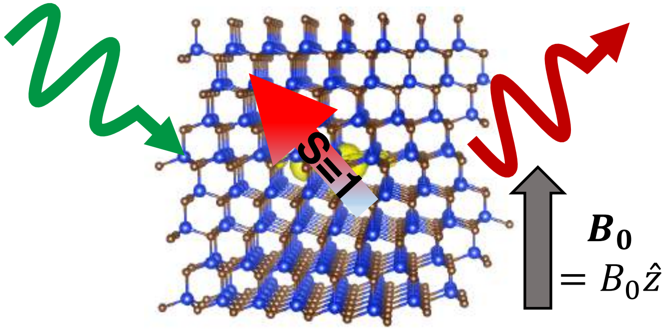
Christle *et al.*, Nat. Mater. (2014).
Seo *et al.*, Nat. Comm. (2016).

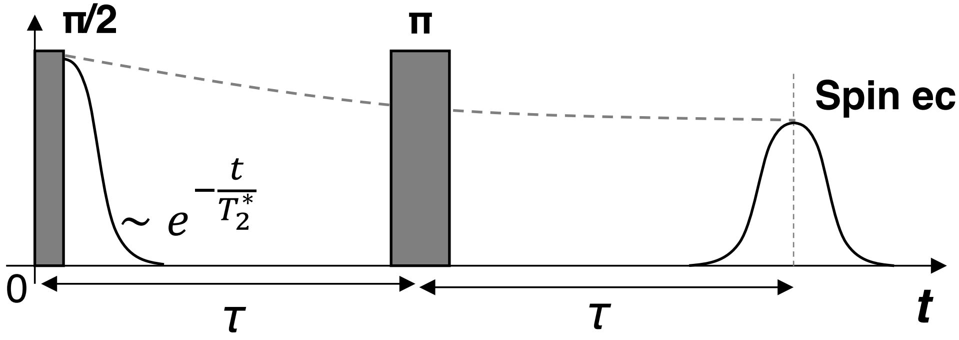
Hahn-echo spin coherence has been measured using optical methods

Hahn echo (*performed by Falk, Klimov, Miao, Awschalom)



ODMR for *(kk)* spin-ensemble

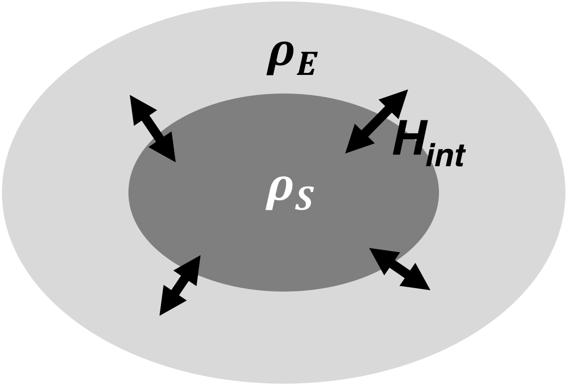




Schweiger and Jeschke (Oxford, 2001)

Two approaches for quantum decoherence

1. Top-down approach (phenomenological): Lindblad (Markovian) Master Eq.



$$\rho_{tot}(t) \approx \rho_S(t) \otimes \rho_E(t)$$

$$\rho_s \rightarrow \Lambda\rho_s$$

$$\frac{d\rho_s}{dt} = \frac{i}{\hbar}[H, \rho_s] + \sum_{\mu>1} \mathbb{D}[L_\mu]\rho_s$$

2. Bottom-up approach (microscopic): Quantum Bath Approach

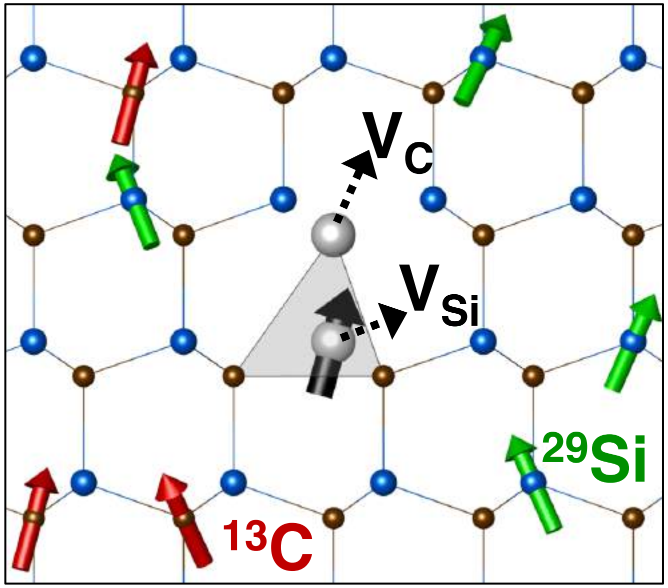
$$\rho_{tot}(t) = U(t)\rho_{tot}(0)U^\dagger(t) \neq \rho_S(t) \otimes \rho_E\ (t) \quad \rightarrow \quad \mathcal{L}(\tau) \equiv \frac{tr[\rho_{tot}(\tau)S_+]}{tr[\rho_{tot}(0)S_+]}$$

Breuer and Petruccione, *The Theory of Open Quantum Systems* (Oxford, 2002)

Quantum bath model for qubit decoherence

Theory: Quantum Bath + Cluster Correlation Expansion

*No adjustable free parameters!



$$\mathcal{H}_{total} = \mathcal{H}_e + \mathcal{H}_n + \mathcal{H}_{e-n} + \mathcal{H}_{n-n}$$

$$|\Psi(0)\rangle = \frac{1}{\sqrt{2}}(|1\rangle + |0\rangle)\otimes|\mathcal{B}(0)\rangle$$

$$|\Psi(\tau)\rangle = \frac{1}{\sqrt{2}}\left(|0\rangle\otimes|\mathcal{B}^{(0)}(\tau)\rangle + |1\rangle\otimes|\mathcal{B}^{(1)}(\tau)\rangle\right)$$

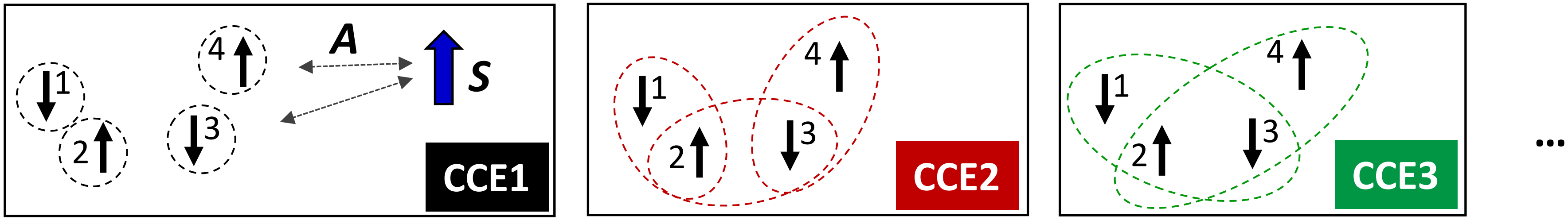
$$\mathcal{L}(\tau) \propto \langle\mathcal{B}^{(1)}(\tau)|\mathcal{B}^{(0)}(\tau)\rangle$$

: off-diagonal element of the reduced density operator.

Breuer and Petruccione (Oxford, 2002)

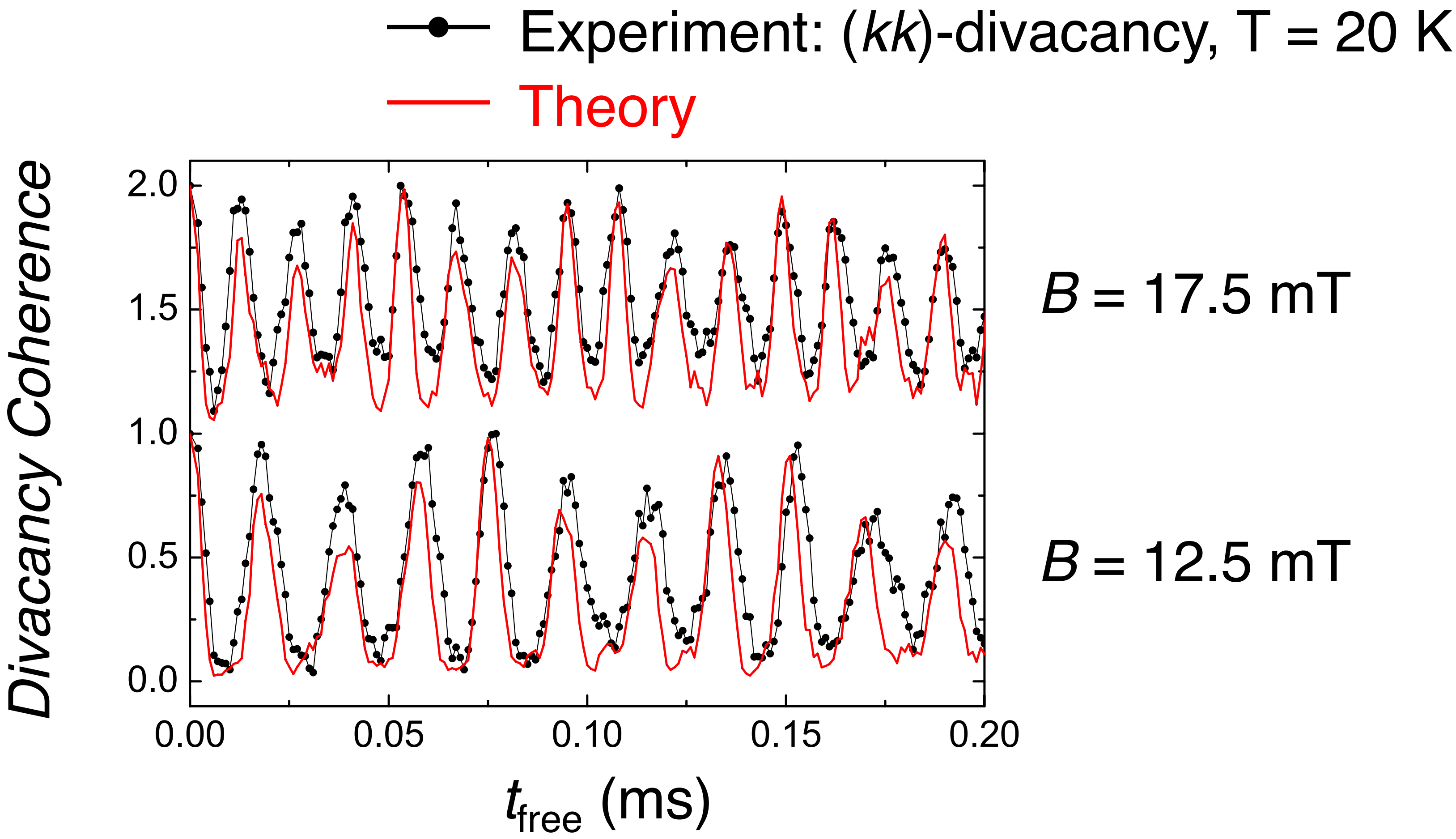
Cluster correlation expansion (CCE)

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



Yang and Liu (2008), Witzel *et al.* (2005)

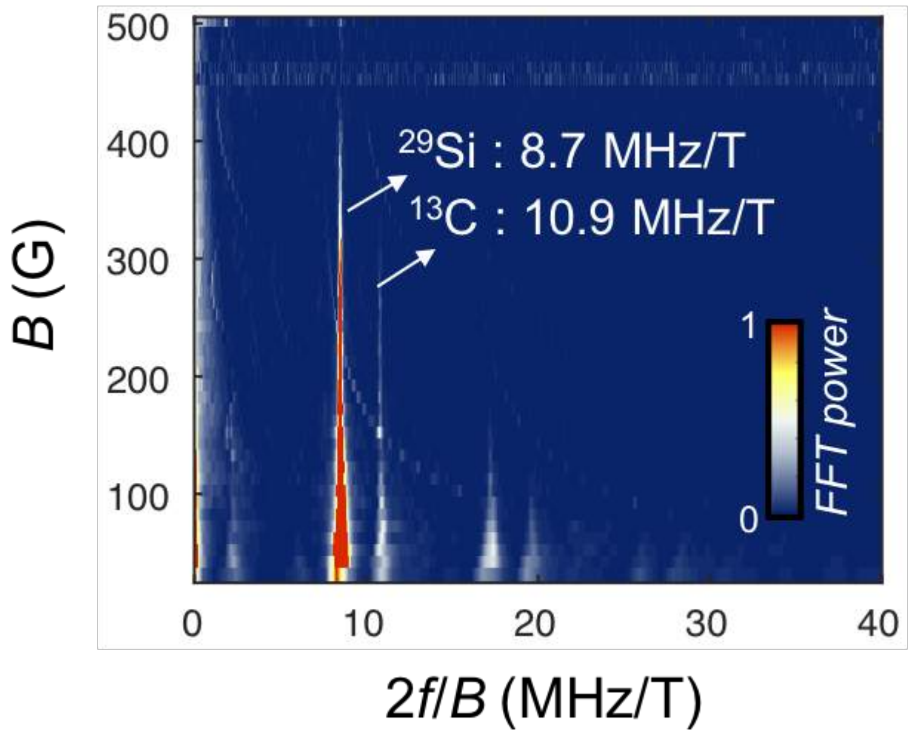
Validation of the theoretical model



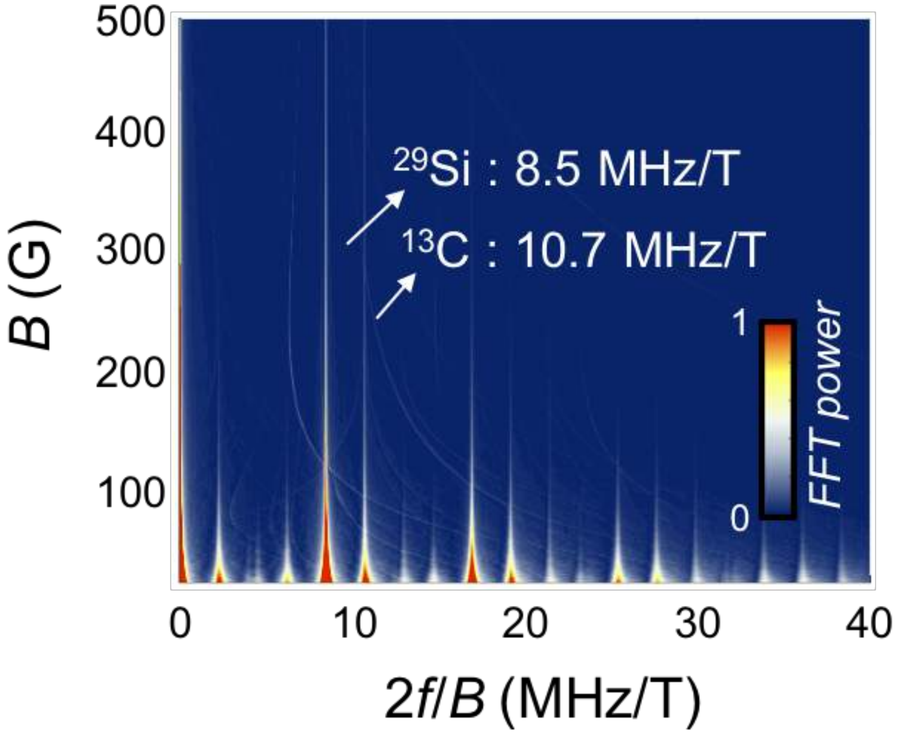
Hosung Seo (hseo2017@ajou.ac.kr)

Validation of the theoretical model

Experiment



Theory

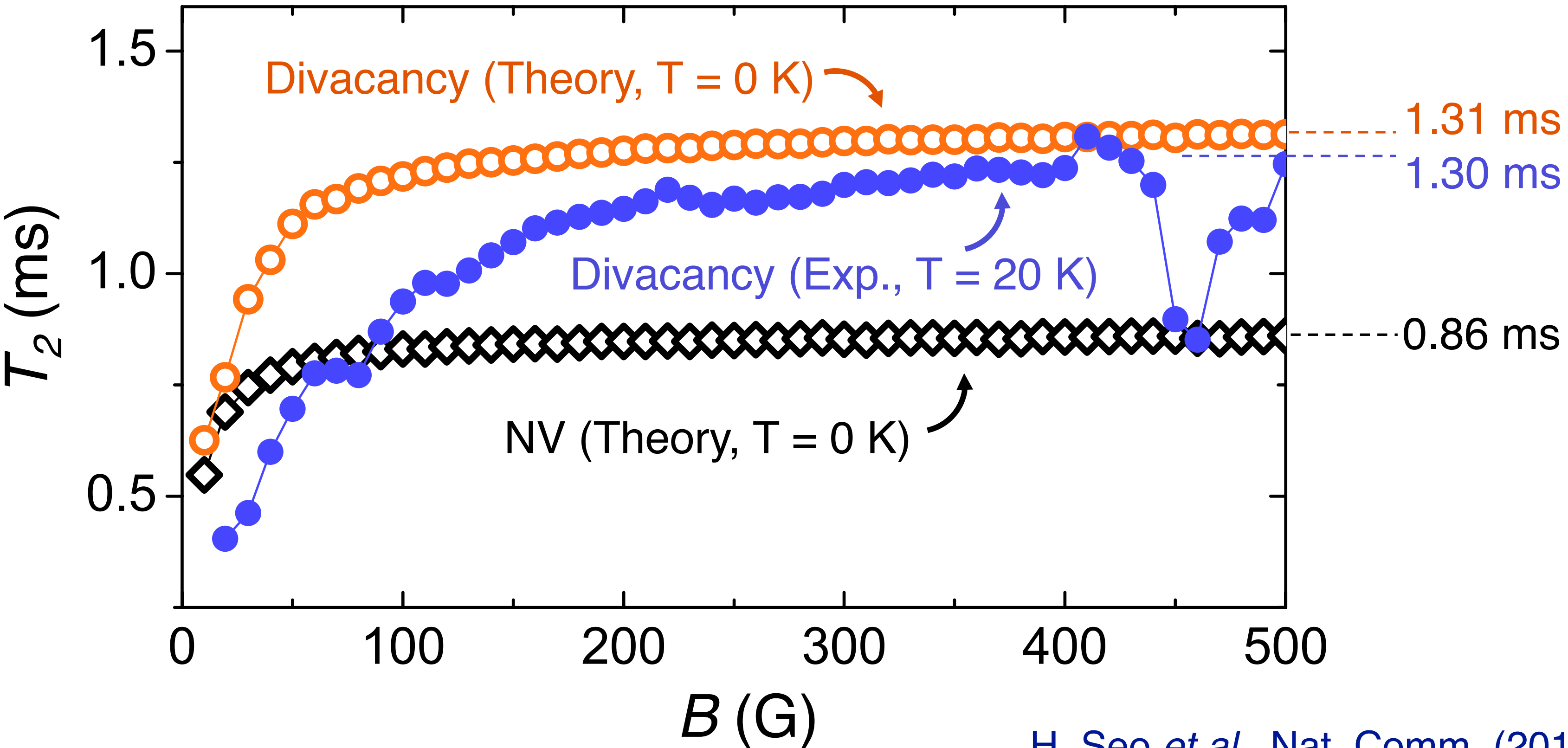


Hosung Seo (hseo2017@ajou.ac.kr)

Hahn-echo coherence time (T_2)

T_2 of the (*kk*)-divacancy and NV ensembles

*in natural 4H-SiC *in natural diamond



H. Seo *et al.*, Nat. Comm. (2016)

Strongly interacting nuclear spin clusters cannot form in SiC

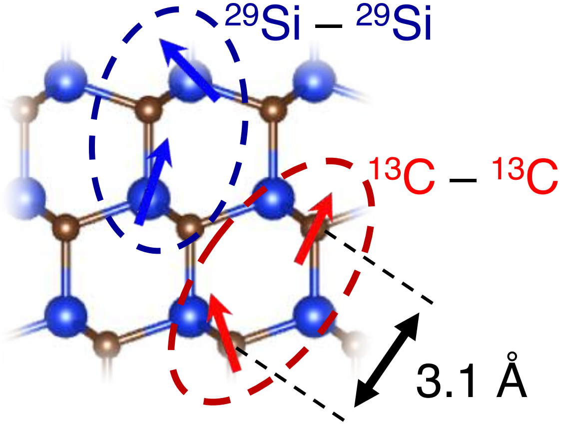
I. The ^{29}Si and ^{13}C nuclear spin baths are decoupled.

Witzel and Das Sarma (2006).
Yang *et al.* (2014).

$$\mathcal{L}_{(kk)}(\tau) = \prod_{\boldsymbol{i}} \widetilde{\boldsymbol{\mathcal{L}}}_{\boldsymbol{i}}(\boldsymbol{\tau}) \prod_{\{i,j\}_{hetero}} \widetilde{\boldsymbol{\mathcal{L}}}_{\boldsymbol{i,j}} \prod_{\{i,j\}_{homo}} \widetilde{\boldsymbol{\mathcal{L}}}_{\boldsymbol{i,j}}$$

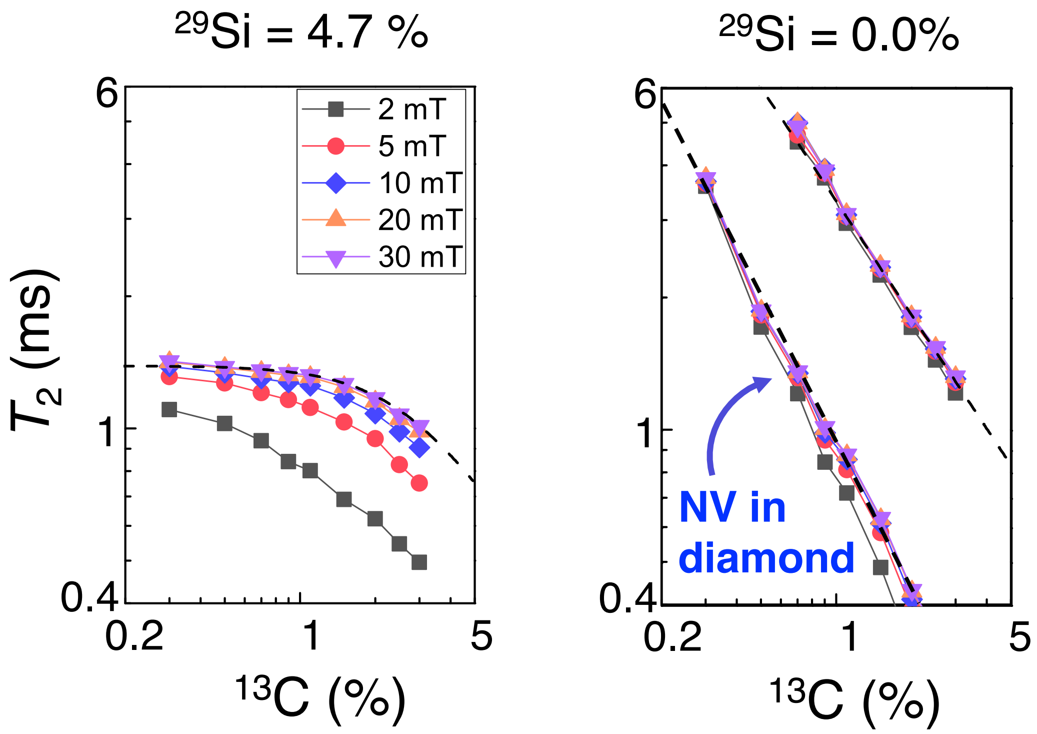
γ_C = 10.71 MHz/T
γ_{Si} = -8.46 MHz/T

II. Homonuclear spin pairs are farther apart. Dipole-dipole ~ $1/r^3$



The heterogeneity of the nuclear spin bath in SiC prohibits formation of strongly interacting nuclear spin pairs.

Outlook: Extension of T_2 by isotopic purification



Experimental collaboration


C. Anderson


A. Bourassa


D. Awschalom

For Divacancy in SiC,

$$T_2 \sim \left[\left(a_{Si} n_{Si}^{N_{Si}}\right)^{-n} + \left(a_C n_C^{N_C}\right)^{-n} \right]^{-1/n}$$

For NV, $T_2 \sim 1/n_C$

Maze *et al.* (2008)
Mizuochi *et al.* (2009)

Hosung Seo (hseo2017@ajou.ac.kr)

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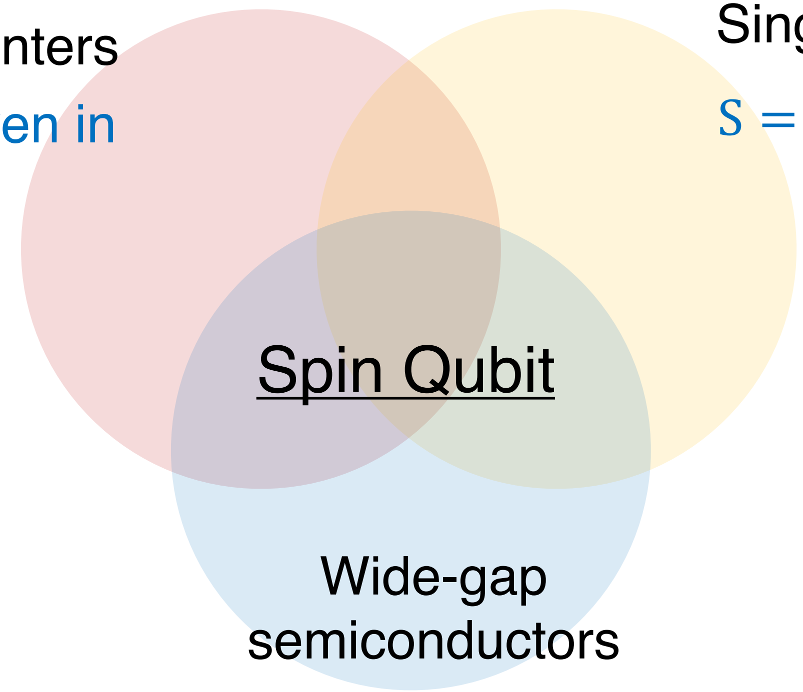
He Ma

Marco Govoni

Giulia Galli

Hosung Seo (hseo2017@ajou.ac.kr)

Alternative qubits in wide-gap semiconductors



Color centers

> 500 even in diamond

Single spins

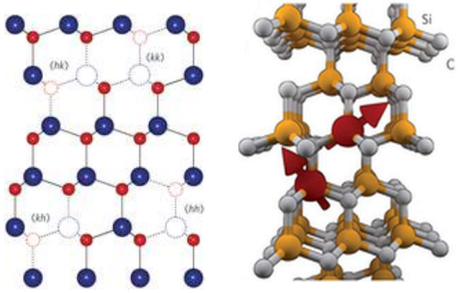
$S = \frac{1}{2}, 1, \frac{3}{2}, \ldots$

Spin Qubit

Wide-gap semiconductors

Industrial: SiC, GaN, SiO_2
Multi-functional: AlN, $BaTiO_3$
2D materials: *h*-BN, TMD

Defects in SiC



Divacancy

Koehl *et al.* (2011).

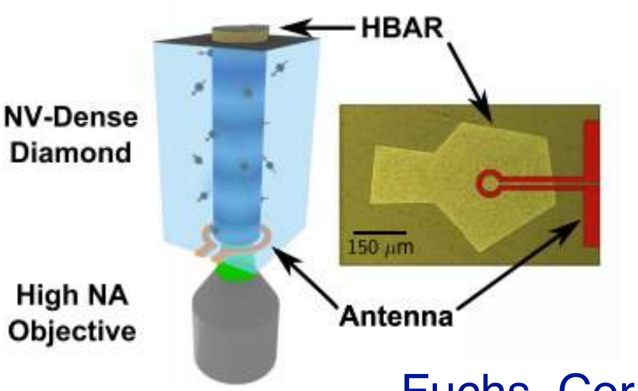
Si vacancy

Widmann *et al.* (2015).

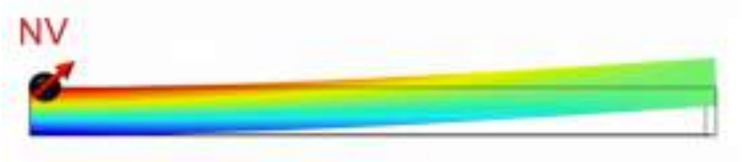
Hybrid quantum systems

Spin qubits in the solid state exhibit interactions with many degrees of freedom in the solid that can be harnessed for quantum applications.

Quntum-state control and sensing

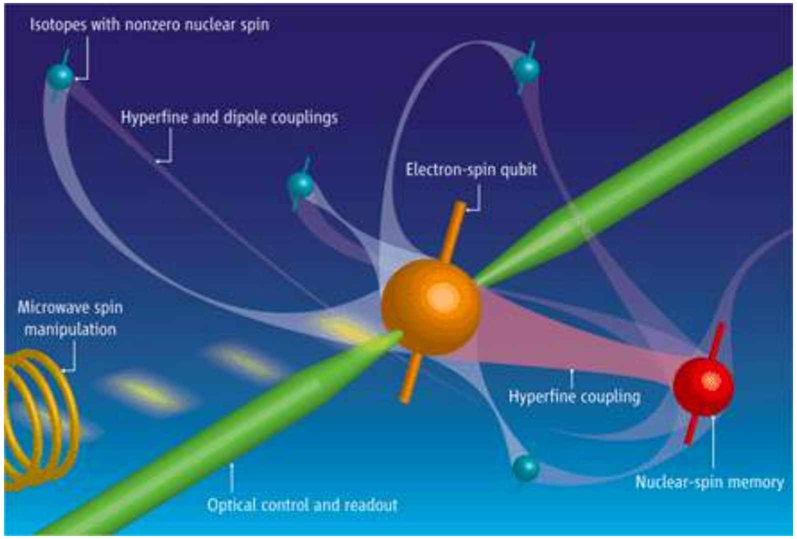


Fuchs, Cornell



Jaychi, UCSB

Quantum memory using nuclear spins

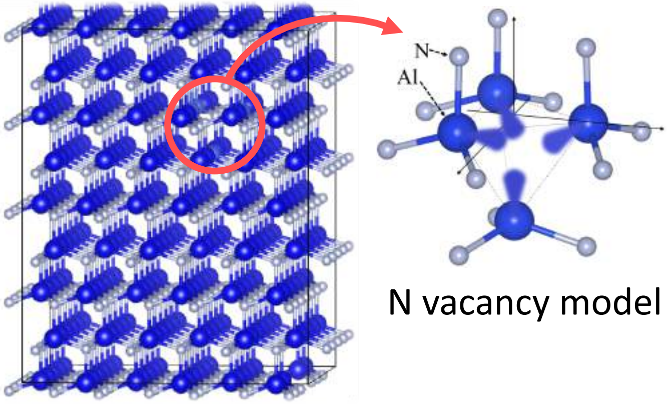


Boehme *et al.*, *Science* (2012)

First-principles design of defect spin qubits

1. Point Defect = Localized quantum system coupled to an infinite lattice

First-principles Computational Methods + Effective Modeling



N vacancy model

AlN crystal

2. Kohn-Sham Density Functional Theory

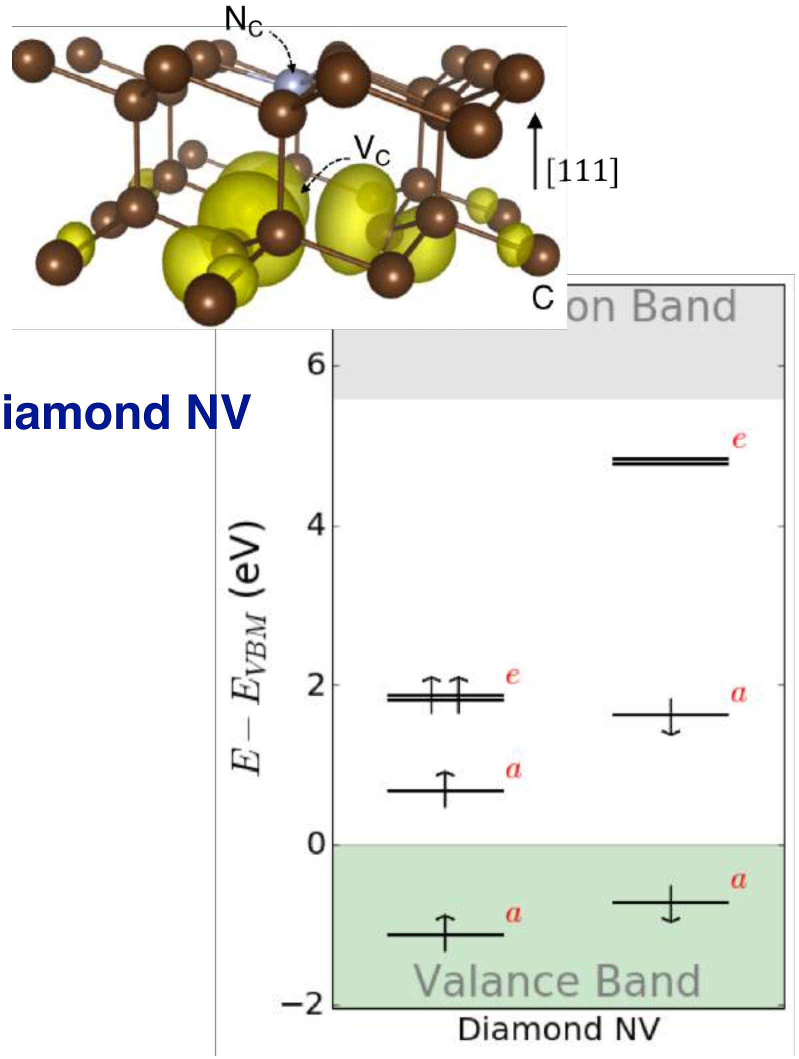
V_{xc}: e.g. LDA or GGA

$$\left[-\frac{\hbar^2}{2m}\nabla^2+e^2\int\frac{\rho(\vec{r}\,')}{|\vec{r}-\vec{r}\,'|}d^3r'+V_{ion}(\vec{r})+\frac{\delta E_{xc}[\rho]}{\delta\rho}\right]\psi_{n,k}(\vec{r})=\varepsilon_{n,k}\psi_{n,k}(\vec{r})$$

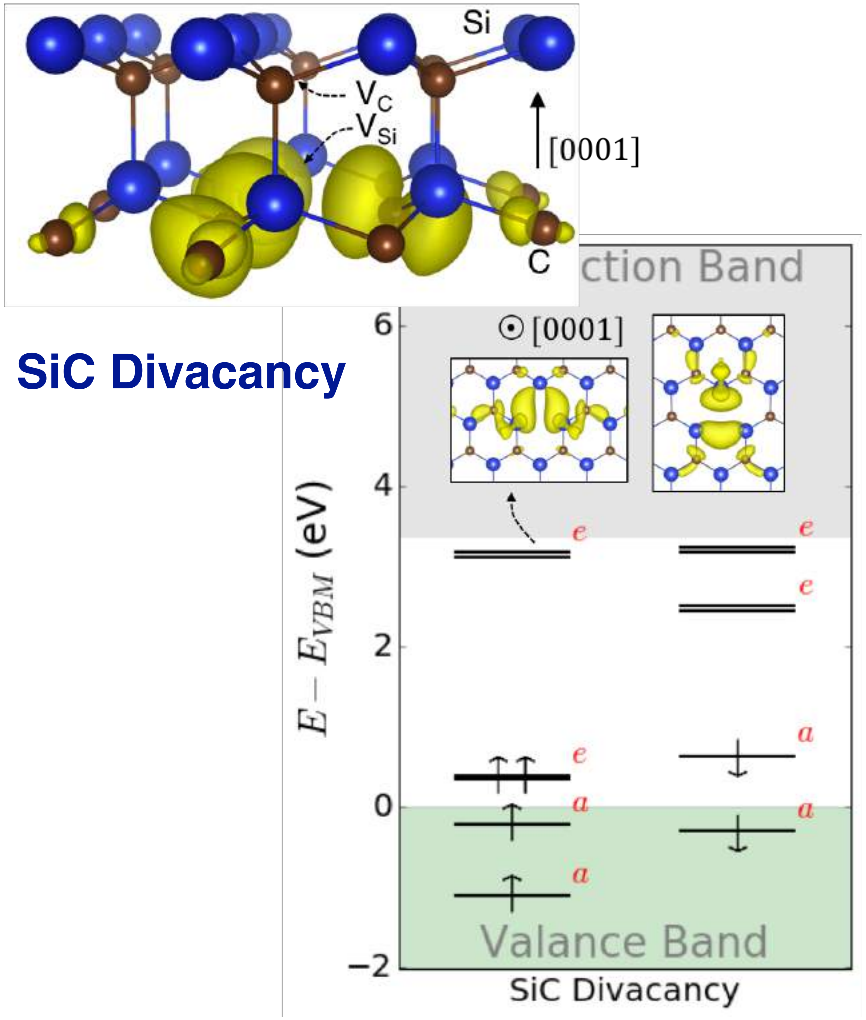
Hosung Seo (hseo2017@ajou.ac.kr)

Traditional design strategy of new defect qubits

Diamond NV



SiC Divacancy



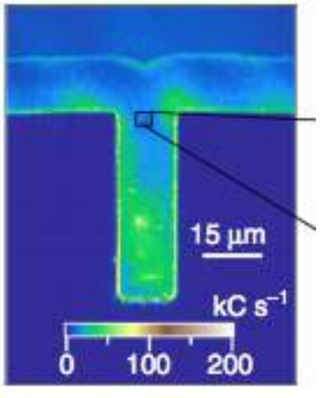
Hosung Seo (hseo2017@ajou.ac.kr)

Traditional design strategy of new defect qubits

NV and divacancy share many common properties. But, is this always good?

1. Low optical read-out fidelity
2. Small spin – transverse coupling constant.

NV center in diamond
($d_\perp$ =10 ~ 20 GHz/strain)



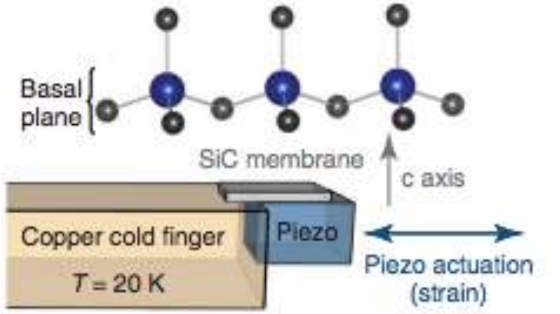
Jayich group, UCSB

Single-phonon cooperativity

$$C = \frac{g^2}{n_{th}\kappa\gamma} \propto \frac{(d_\perp \epsilon_{ZPM})^2 Q T_2}{k_B T}$$

Lee *et al.* J. Opt. (2017).

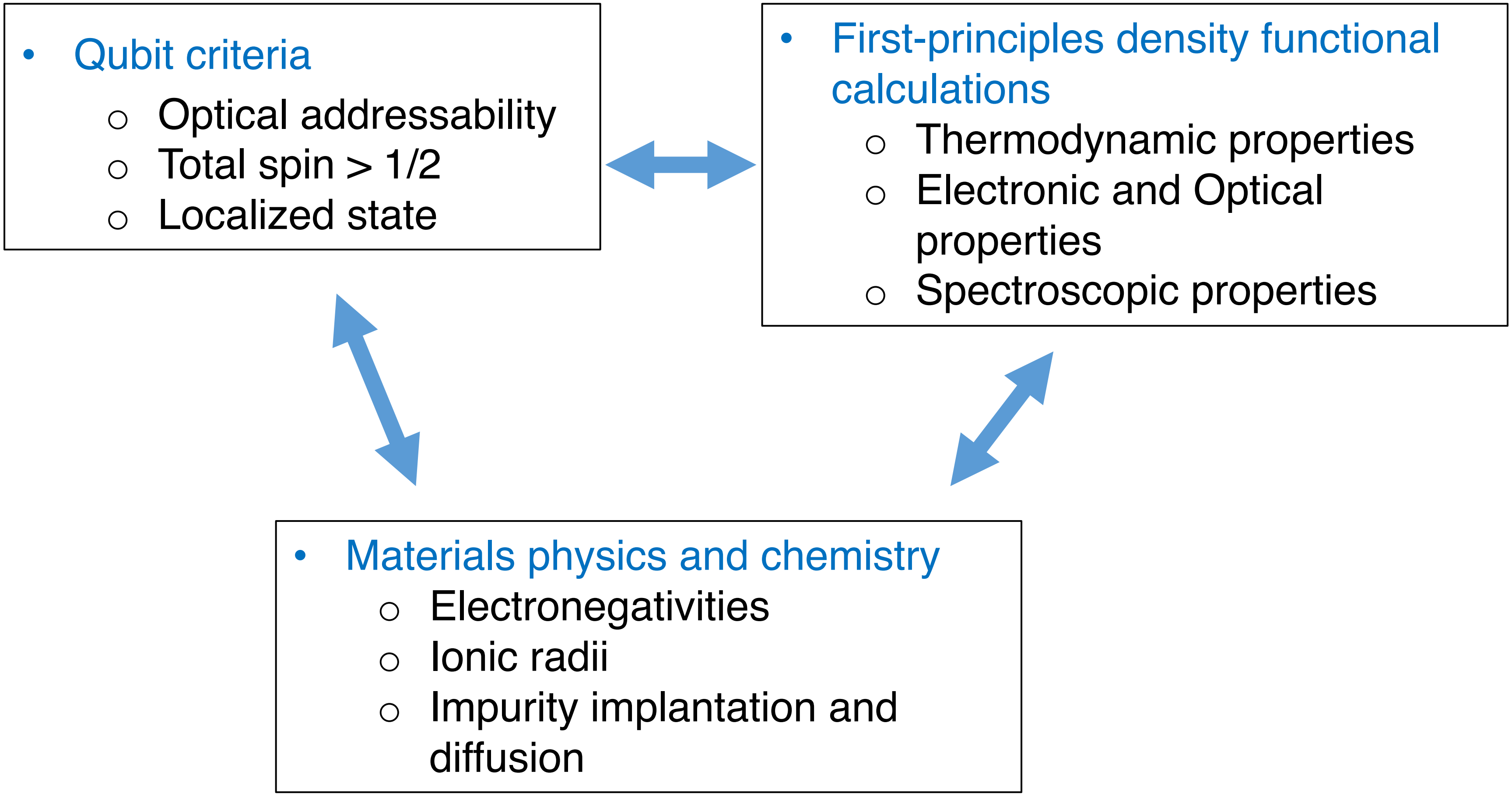
Divacancy in SiC
($d_\perp$ = 5 ~ 10 GHz/strain)



Awschalom group, UChicago

Hosung Seo (hseo2017@ajou.ac.kr)

Computational design of new spin qubits



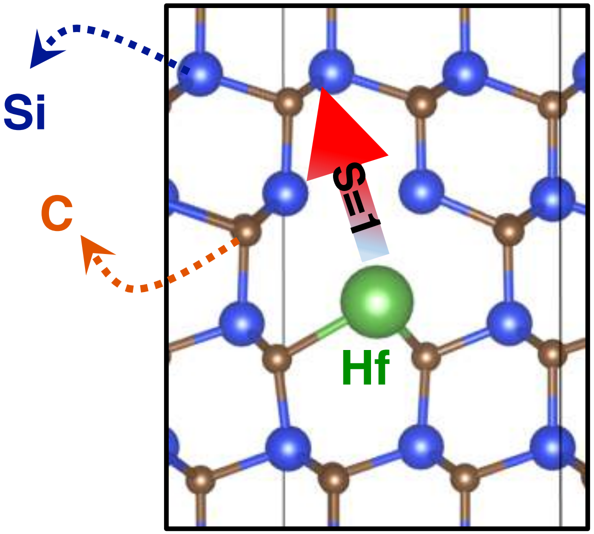
- Qubit criteria
  - Optical addressability
  - Total spin > 1/2
  - Localized state

- First-principles density functional calculations
  - Thermodynamic properties
  - Electronic and Optical properties
  - Spectroscopic properties

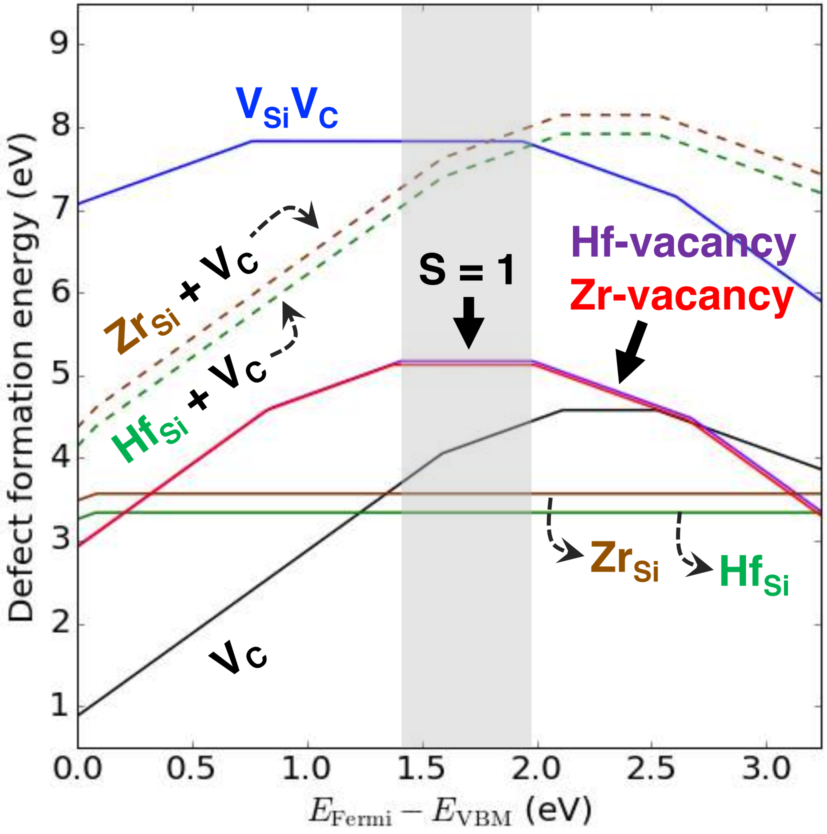
- Materials physics and chemistry
  - Electronegativities
  - Ionic radii
  - Impurity implantation and diffusion

Large-metal ion vacancy complexes

Neutral Hf- or Zr-vacancy complex



Defect Formation Energy



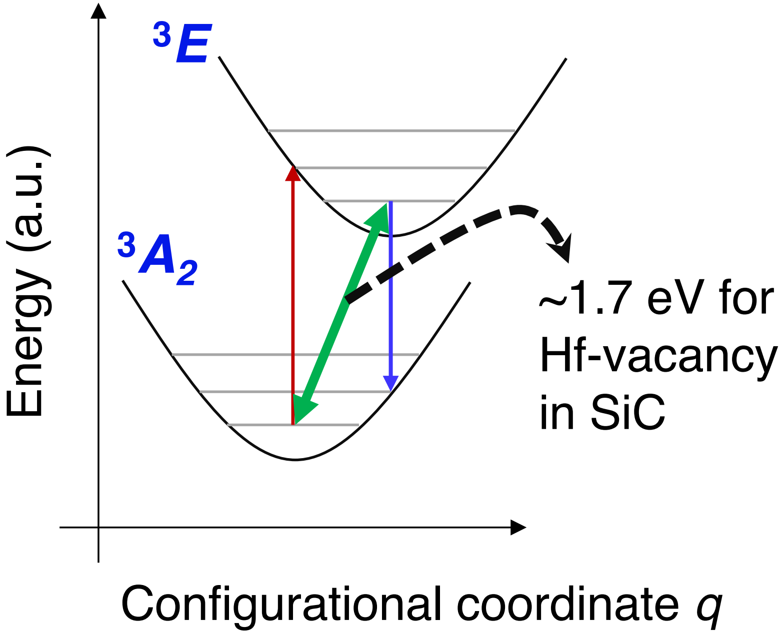
Main points:

1. Formation of the complex
2. Charge-state stability
3. Ionization energy = 2 eV

Hosung Seo (hseo2017@ajou.ac.kr)

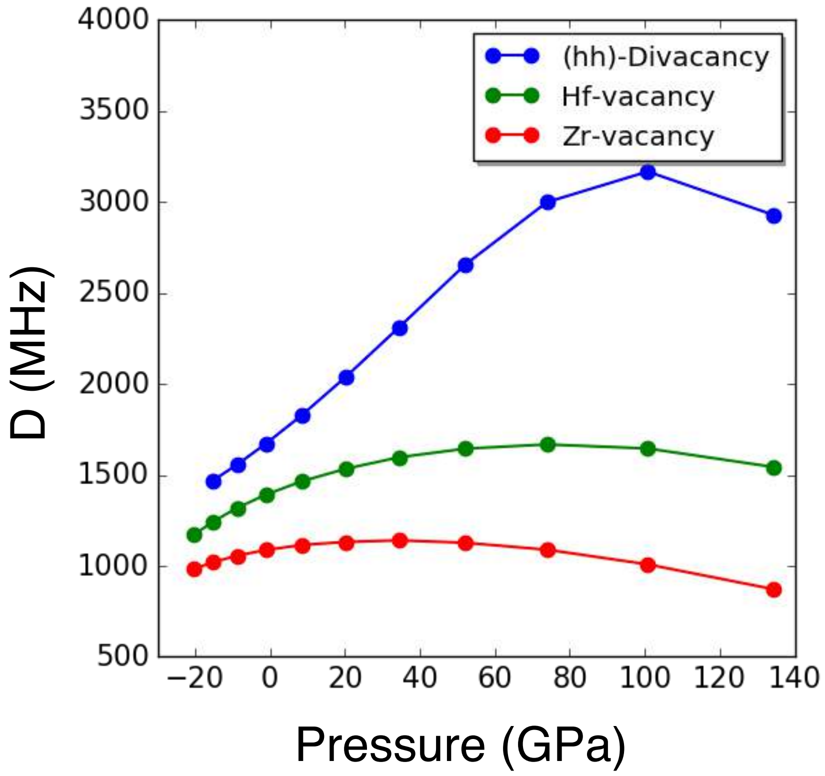
Spectroscopic quantities

1. Optical Zero-phonon Line (ZPL)



2. Hyperfine tensor ($S \cdot A \cdot I$)

3. Zero-field splitting ($S \cdot D \cdot S$)



*Spin-strain coupling

*Stand-alone Python code (developed by H. Ma, H. Seo, and M. Govoni)

Outlook

1. We designed spin qubits in SiC from scratch, which could be suitable for SiC-based hybrid quantum systems

2. The strategy also works for AlN.

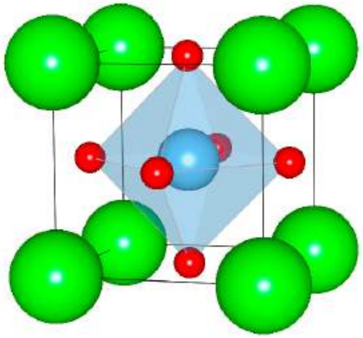
→ Portable qubit platform. Potential interface with AlN-based optomechanics

Experimental collaboration

S. Whiteley

D. Awschalom

Defect qubits in complex oxides

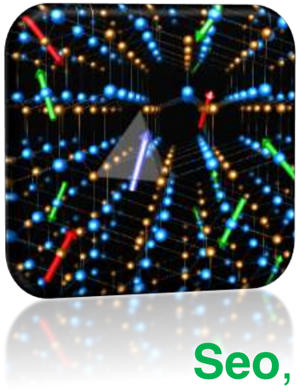


$T_2 \gg 1$ ms (expected)

F.J. Heremans
(Argonne)

Conclusion

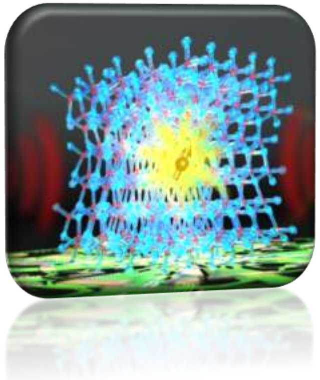
Spin decoherence dynamics



- We showed theoretically and experimentally that T_2 of the divacancy reaches 1.3 ms.
- Our results point to polyatomic crystals as promising hosts for coherent qubits in solids.

Seo, Falk, Klimov, Miao, Galli, and Awschalom, *Nat. Comm.* ***7***, *12935* (2016).

Predictions on new potential defect qubits



- We showed that large-metal-ion (LMI) vacancy complexes could be a 'portable' qubit platform in wide-gap semiconductors potentially with a large spin-strain coupling.

Seo, Ma, Govoni, and Galli, *arXiv:1709.09818* (2017).

Acknowledgement

NATIONAL SCIENCE FOUNDATION MRSEC
MATERIALS RESEARCH SCIENCE & ENGINEERING CENTERS

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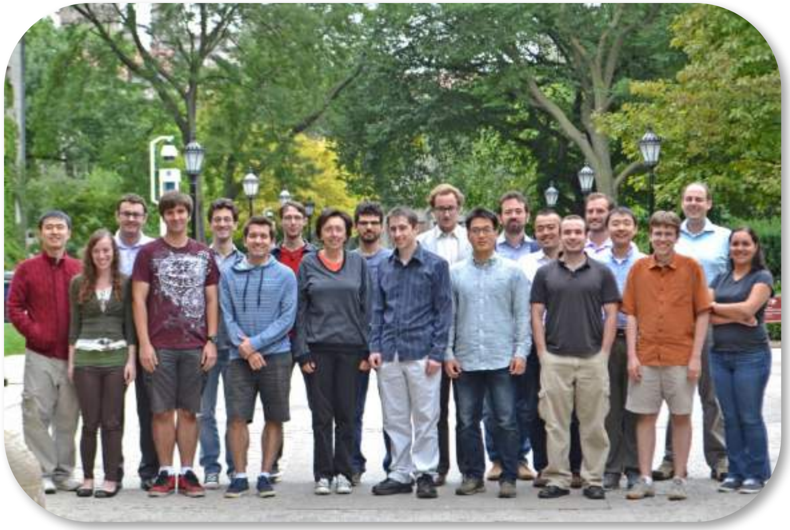
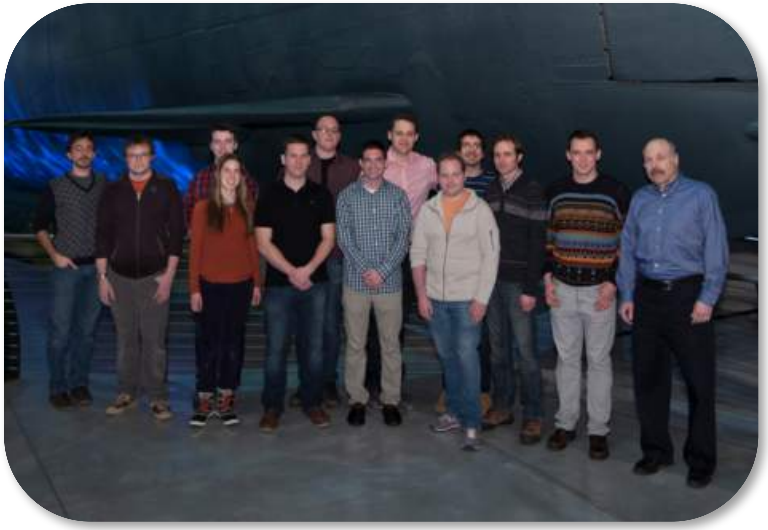
THE UNIVERSITY OF CHICAGO | Research Computing Center

THE INSTITUTE FOR MOLECULAR ENGINEERING

The Giulia Galli group



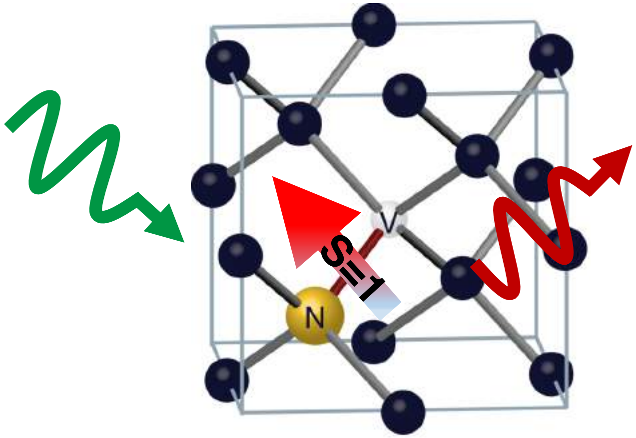
The David Awschalom Group

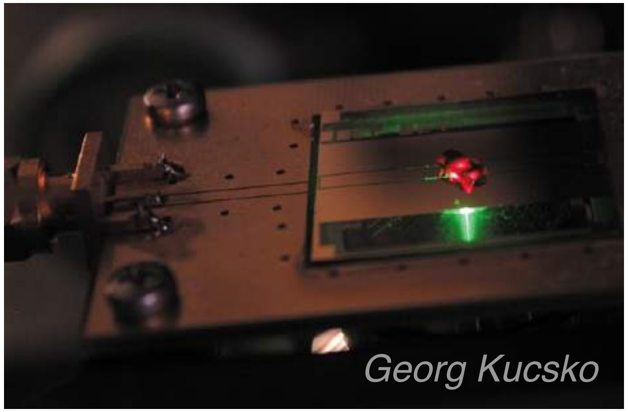


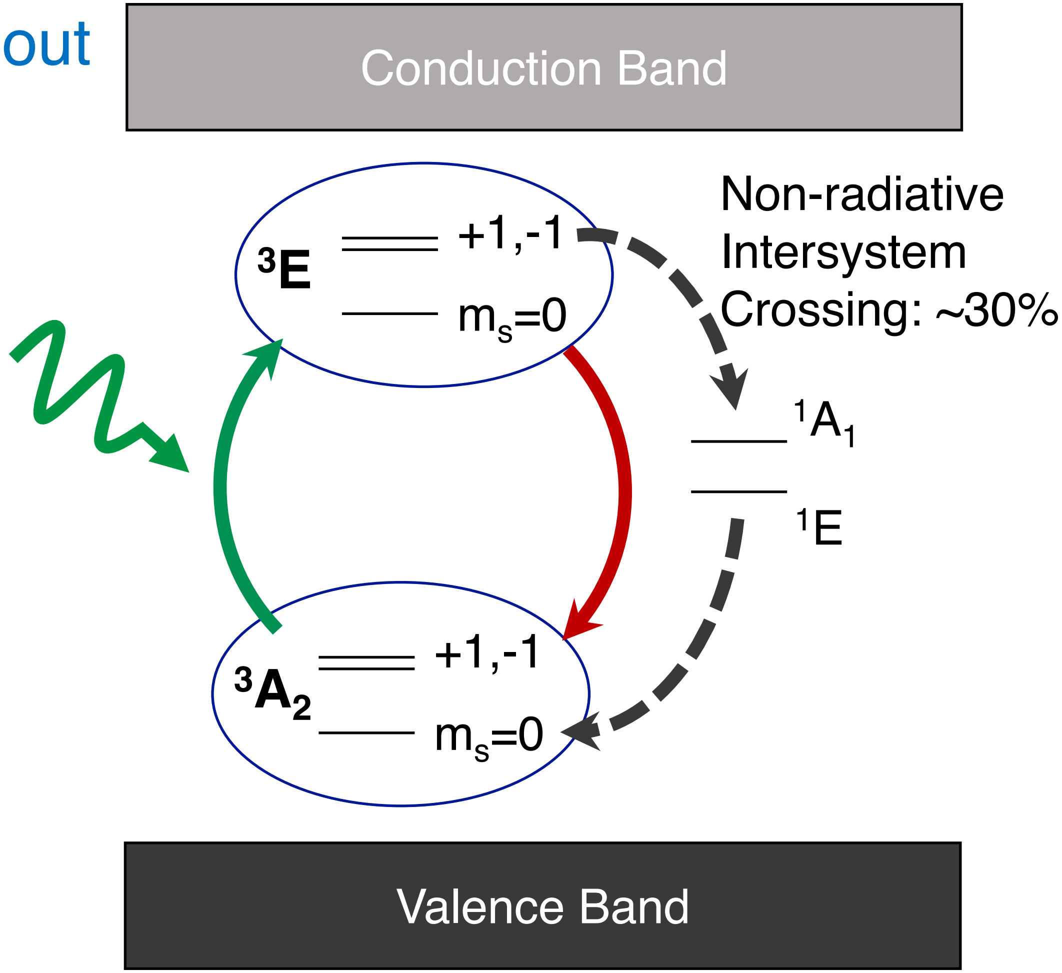
Thank you for your attention!

NV centers as qubits

Optical initialization and readout




Georg Kucsko


Conduction Band

^3E +1,-1 $m_s=0$

Non-radiative Intersystem Crossing: ~30%

$^1A_1$

^1E

$^3A_2$ +1,-1 $m_s=0$

Valence Band