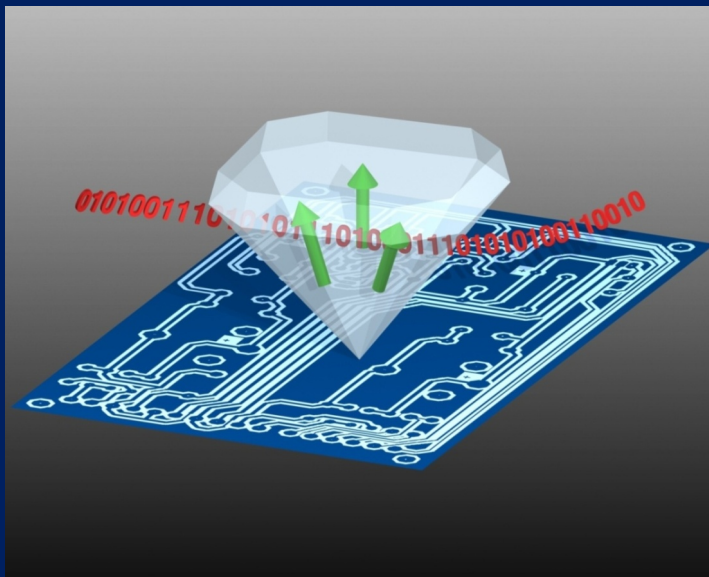


Single spin, photon, and charge manipulation of NV center in diamond

Norikazu MIZUOCHI

Engineering Science, Osaka University, Japan



Collaborators and Acknowledgements

- Prof. Suzuki and group members (Osaka Univ.)
- Dr. Yamasaki and group members (AIST)
- Prof. Hatano (Univ. of Tokyo Inst. Tech.), RENESAS
- Dr. Saito, Dr. Matsuzaki, Dr. Munro, Dr. Zhu (NTT)
- Prof. Kosaka (Yokohama Univ.)
- Dr. Semba, (NICT)
- Prof. Nemoto (NII)
- Prof. J. Wrachtrup and group members (Stuttgart Univ.)
- Prof. F. Jelezko (Ulm Univ.)
- Dr. A. Gali (Wigner Research center.)

Osaka Univ



Content

1. Introduction

NV center in diamond

2. Electric field effect on spin coherence of NV

S. Kobayashi, NM, et al., unpublished.

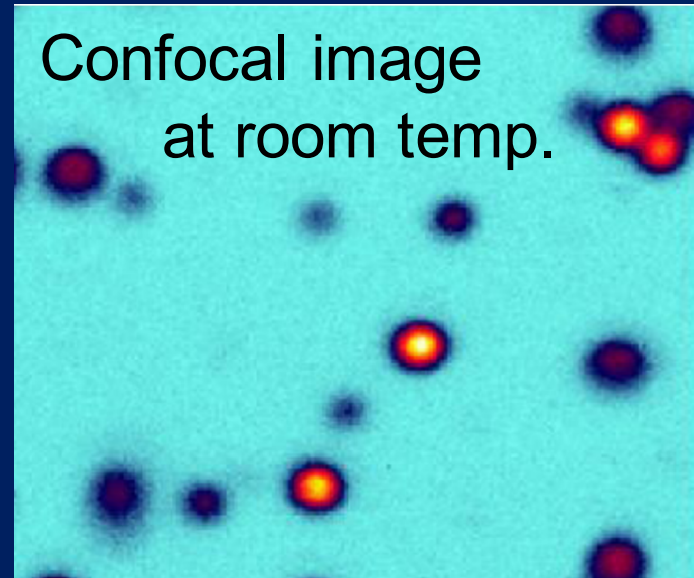
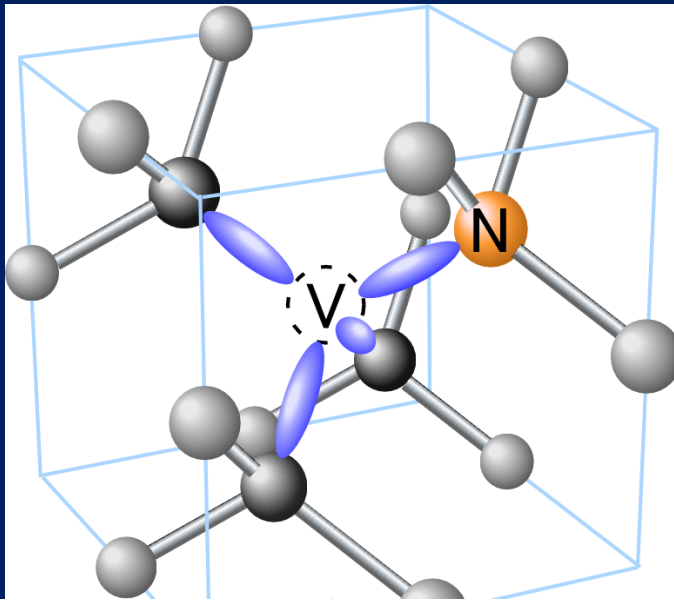
3. Selective alignment of N-V axis

Four-fold improvement of the magnetic field sensitivity

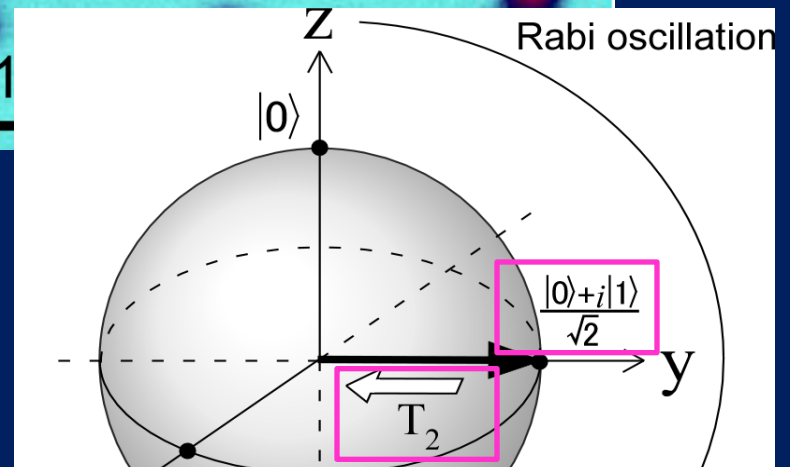
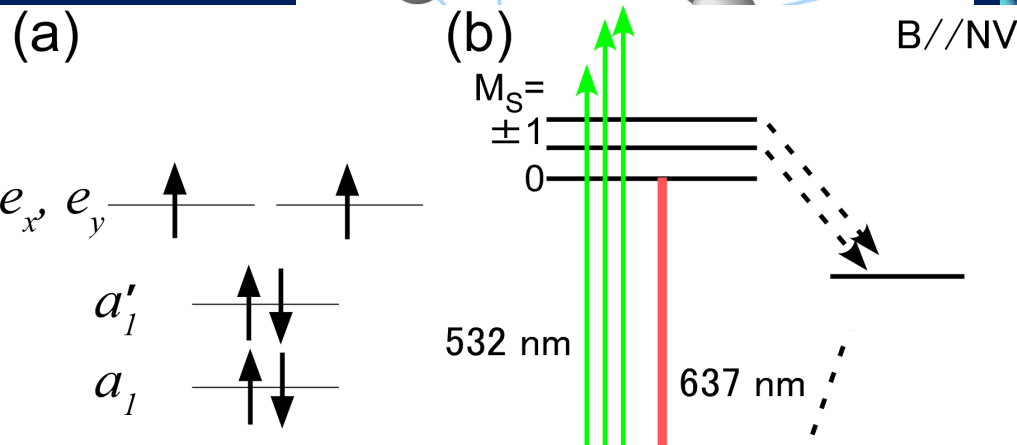
T. Fukui, NM, et al., *Appl. Phys. Exp.* 7, 055201 (2014), selected in “Spotlights”

T. Miyazaki, NM, et al., *Appl. Phys. Lett.* 105, 261601 (2014).

NV center in diamond



Confocal image
at room temp.



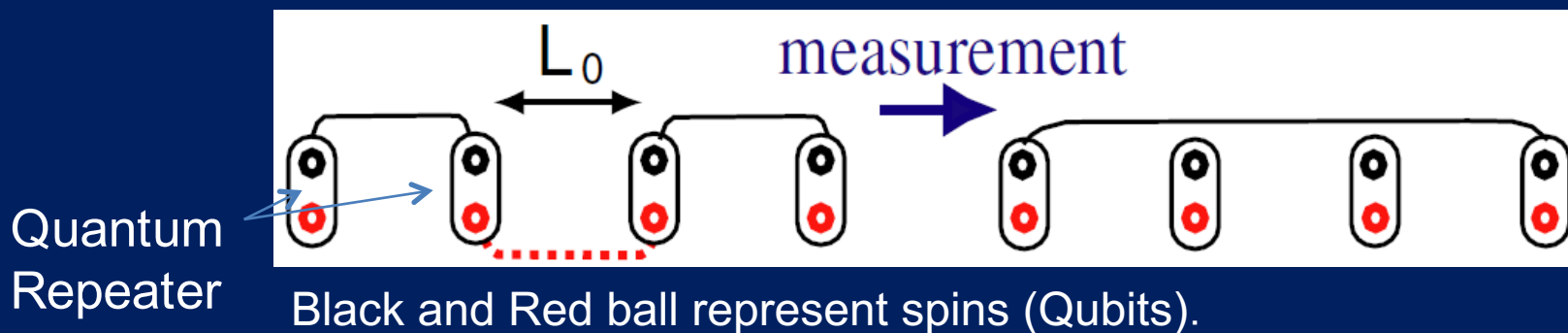
Coherent Control and detection of Single spin at RT
Unique character among solid state material

Quantum Cryptography

BB84: completely secure communication
by using single photon

Single photon source, Quantum repeater

(“spin” for processing and memory, “Photon” for communication)

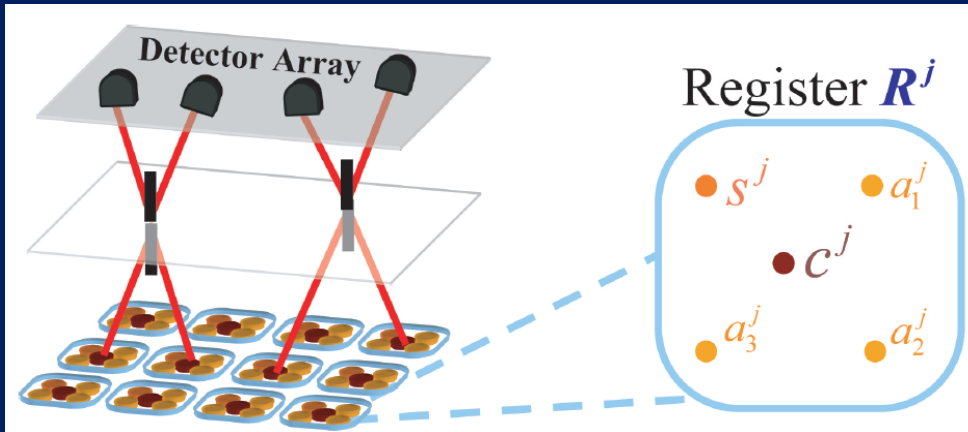


Black and Red ball represent spins (Qubits).

L. Childress, et al., PRL 2006.

Quantum Computing

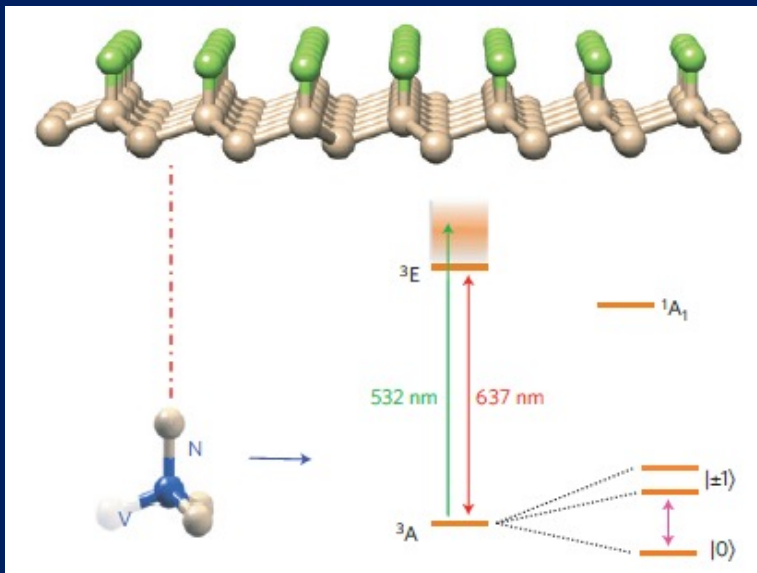
Distributed scalable quantum computer



It consists of 5 qubits quantum registers.

Jiang et al., PRA 2007

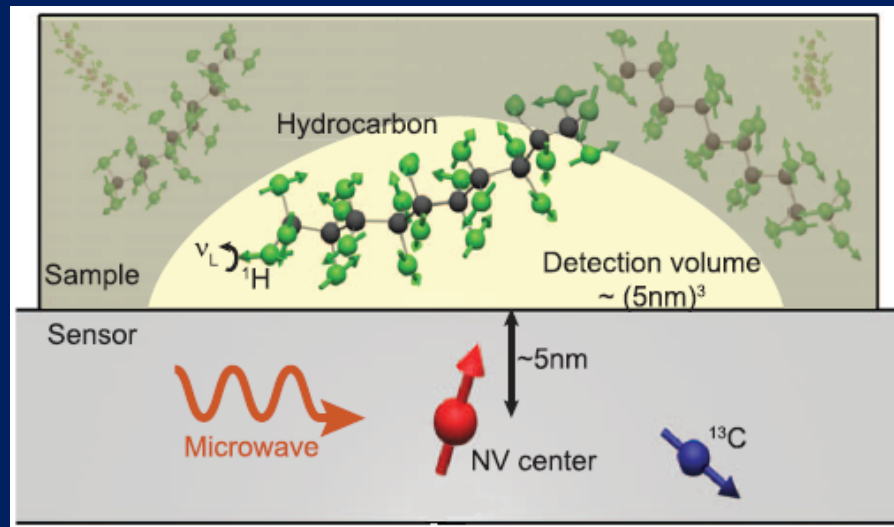
Quantum Simulation



Quantum simulation by NMR on the diamond surface: Initialization and readout is carried out by NV center
Cai et al., Nat. Commun. 2014

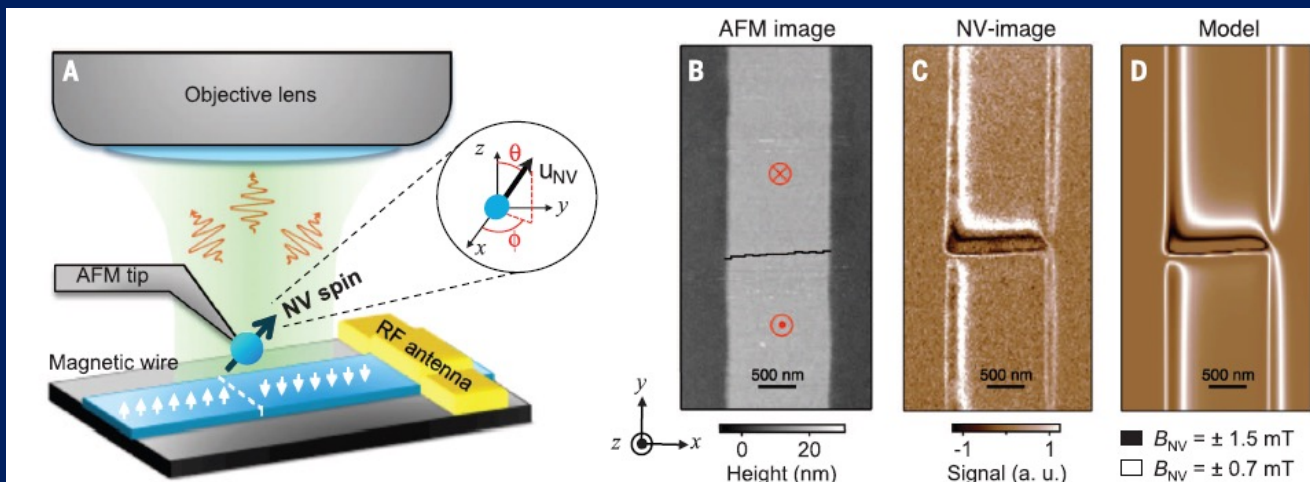
Sensor (Magnetic, Electric field, Temperature...)

High resolution and high sensitive sensor



NMR of molecules
on diamond surface

Science 2013



Nanoscopy of
domain walls

Science 2014

Scanning probe magnetometry (magnetic sensor)

Minimum detectable magnetic field

$$\delta B \approx \frac{1}{g_s \mu_B R \sqrt{\eta}} \frac{1}{\sqrt{NtT_2^*}},$$

R : Measurement contrast
 h : detection efficiency
 N : number of spin centers
 t : integration time

Single (RT)

$$B_{AC} = 4.3 \text{ nT Hz}^{-1/2}$$

Nature Mat. 2009

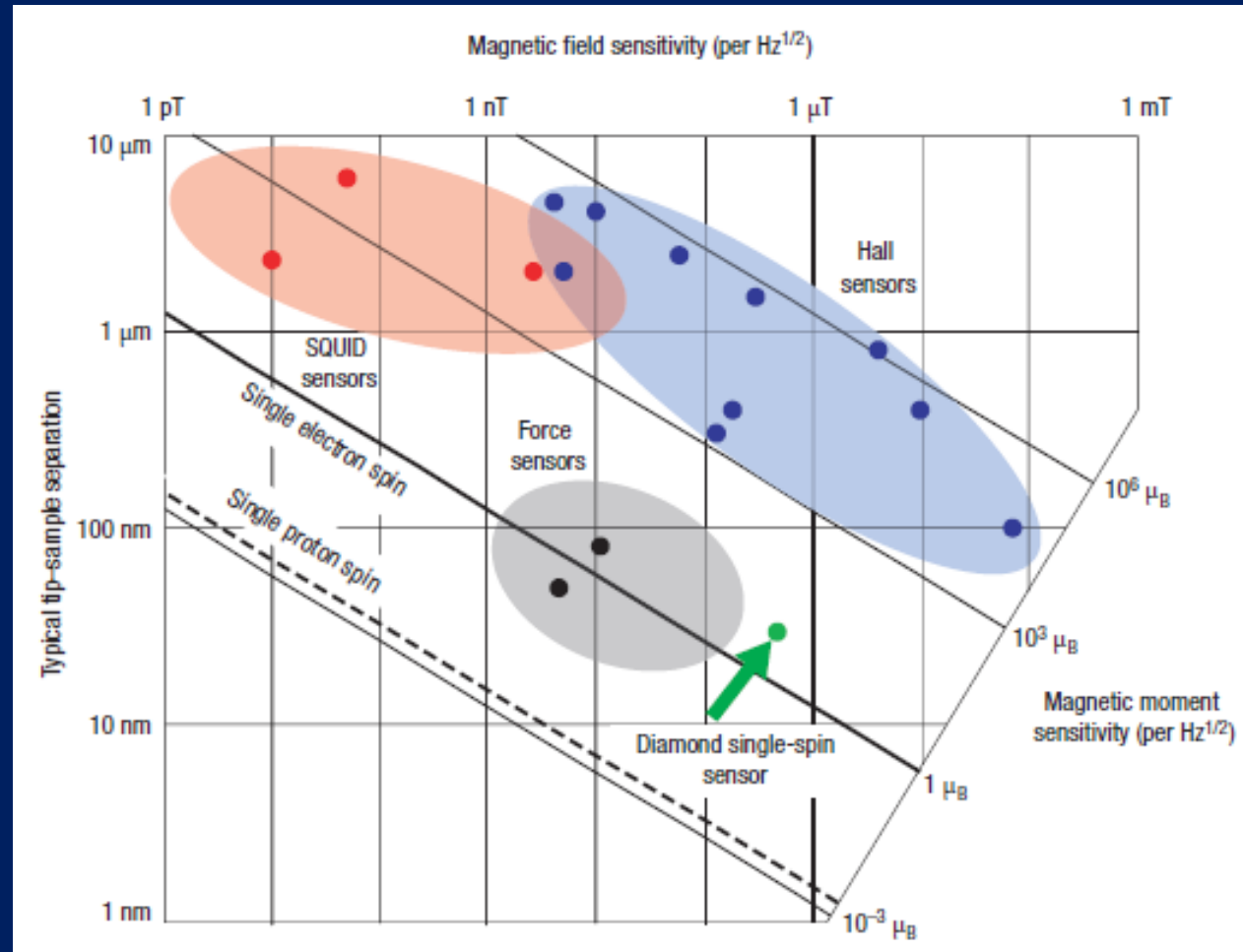
$$B_{DC} = 0.3 \text{ mT Hz}^{-1/2}$$

PRB 2009

Ensemble (RT)

$$B_{AC} \approx \sim 100 \text{ pT Hz}^{-1/2}$$

PRB 2012

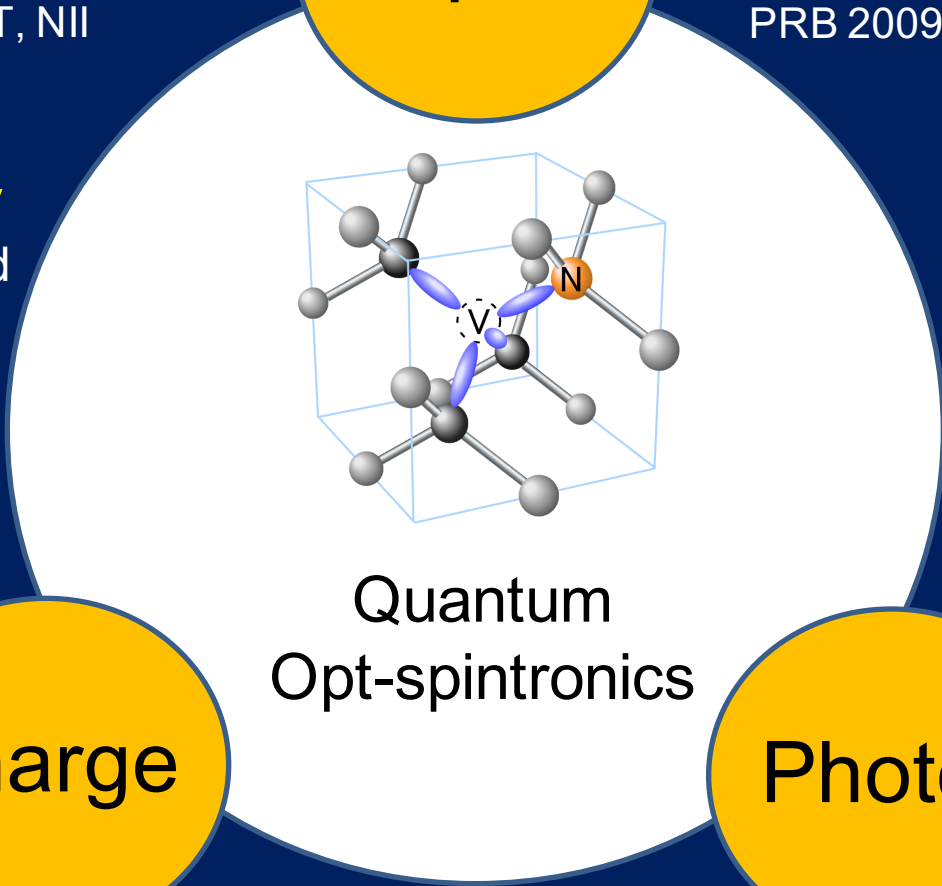
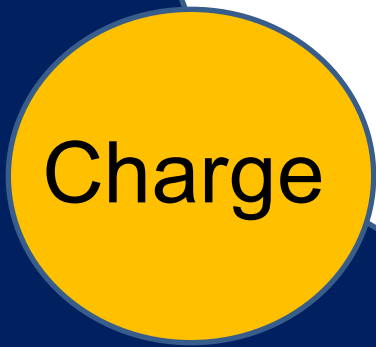


C. Degan, Nature nanotechnology, 3, 643 (2008).

The previous and recent our researches

Quantum hybrid system with
superconducting flux qubit
Nature 2011, Nature commun. 2014
Collaboration with NTT, NII

QIP by single spins
Science 2008,
Nature Materials 2009,
PRB 2009



Quantum
Opt-spintronics

Extension of T_2 by
E field, unpublished

Electrical control
of charge state
PRX 2014

Electrically driven single photon source
Nature Photonics 2012

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S. Kobayashi, NM, et al., unpublished.

3. Selective alignment of N-V axis

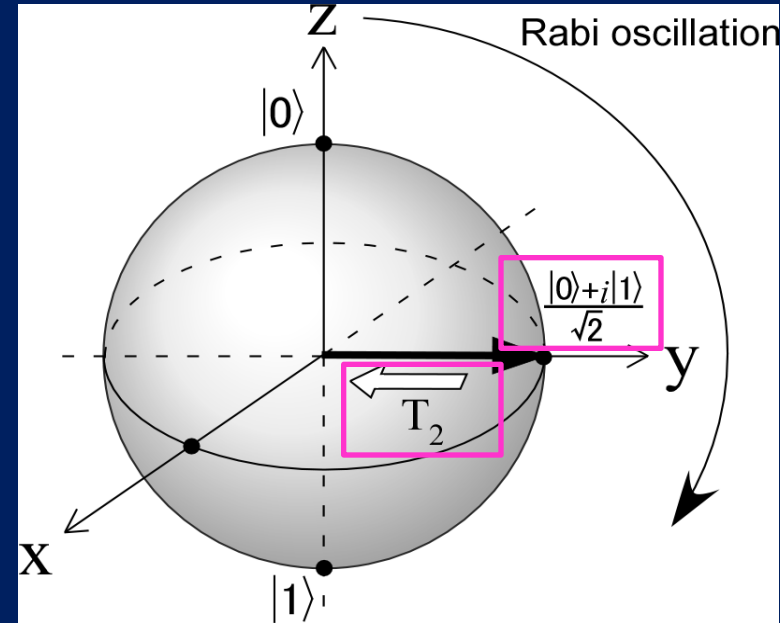
Four-fold improvement of the magnetic field sensitivity

T. Fukui, NM, et al., *Appl. Phys. Exp.* 7, 055201 (2014), selected in “Spotlights”

T. Miyazaki, NM, et al., *Appl. Phys. Lett.* 105, 261601 (2014).

Coherence time (T_2) and Readout Contrast

- Quantum Memory time: Time to maintain superposition state
- Sensitivity (sensor)



Minimum detectable B (sensitivity) : η

$$\eta \propto \frac{1}{C \sqrt{n_{NV} \tau T_2}}$$

C : readout contrast

n_{NV} : The number of NV

τ : Measurement time

L. M. Pham, et. al., Phys. Rev. B **86**, 121202 (2012)

Alignment of N-V axis, APEX 7, 055201 (2014) "Spotlights",

T₂ of electron spin of NV at RT

T₂ (electron spin in natural abundance of ¹³C (I=1) diamond)

2003 50 ms T. A. Kennedy, et al., *APL*, 2003

2006 200 ms L. Childress, et al., *Science*, 2006

350 ms T. Gaebel, et al., *Nature physics*, 2006

2009 650 ms N. Mizuochi, et al., *PRB*, 2009

(77 K 2012 0.6 s B. Grotz, *Nat. Commun.* 2012)

T₂ (electron spin in ¹²C (I=0) enriched diamond)

2009 1.8 ms G. Balasubramanian, et al., *Nat. Mater.* 2009.

Contributions from Material Science

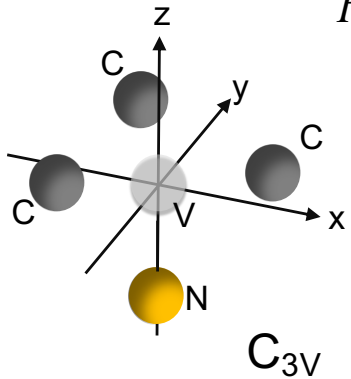
Extension by Dynamical decoupling

(Magnetic Resonance Technique)

Extension by other technique? Electrical?

Electric field effect on spin coherence of NV

■ Hamiltonian



$$H_{gs} = hD_{gs} S_z^2 - \frac{1}{3} S_x S_x + S_y S_y$$

Zero field split D_{gs} : 2870 MHz

$$B g_e S \cdot B$$

Zeeman effect : 2.7 MHz/G

$$d_{gs}^{\parallel} E_z S_z^2 - \frac{1}{3} S_x S_x + S_y S_y$$

Electrical effect : $d_{gs}^{\parallel} / h = 0.35$ Hz cm/V, $d_{gs}^{\perp} / h = 17$ Hz cm/V

$$d_{gs} E_x S_x S_y - S_y S_x - E_y S_x^2 - S_y^2$$

Electron spin interaction between unpaired spin are affected by E.

F. Dolde *et al.*, Nature Phys. 7, 459–463 (2011)

■ Change of Resonant frequency

$$D_{gs} + B g_e B_z + d_{gs} E$$

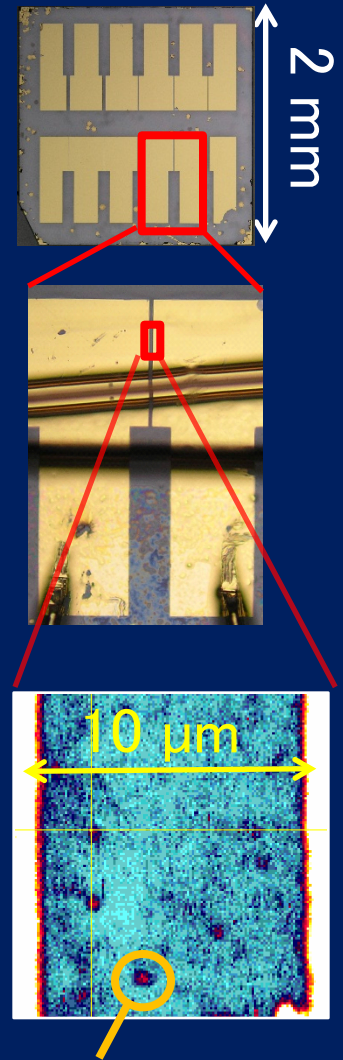
$$\sqrt{\frac{2}{B g_e} B_z^2 + d_{gs}^2 E^2} / \hbar$$

Magnetic noise may be suppressed in case $\frac{2}{B g_e} B_z^2 \gg d_{gs}^2 E^2$

Extension of T_2 by applying Electric field.

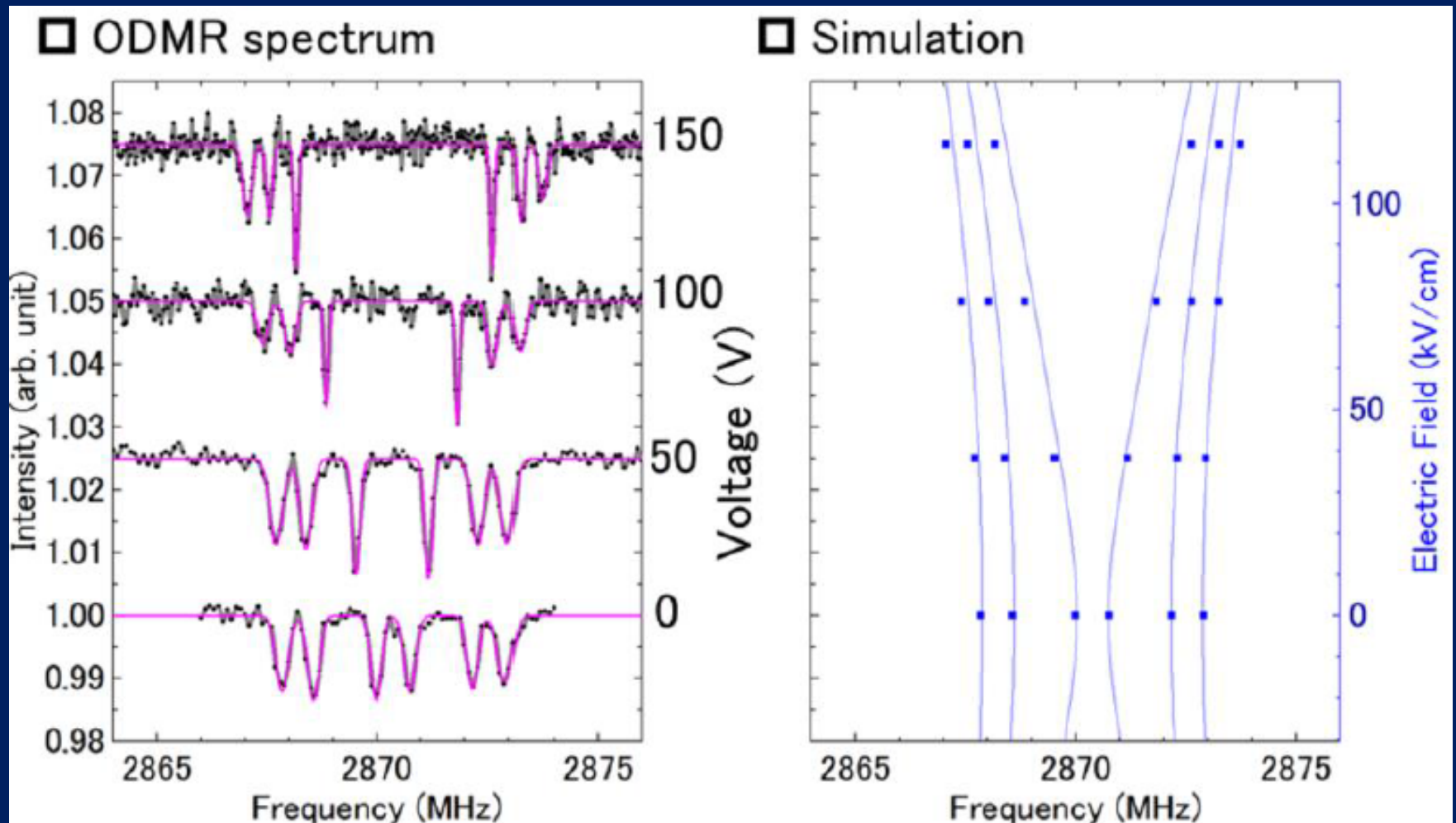
Sample and measurement

- NV center in diamond
 - Ion implantation (30 keV, 600 °C, 6×10^{10} 1/cm² , Au mask was used for small hole)
 - Anneal (1000 °C, 120 min)
- Electrode
 - EB lithography
 - Diamond(001) / Ti (10 nm) / Au (100 nm)
- Home built confocal microscope



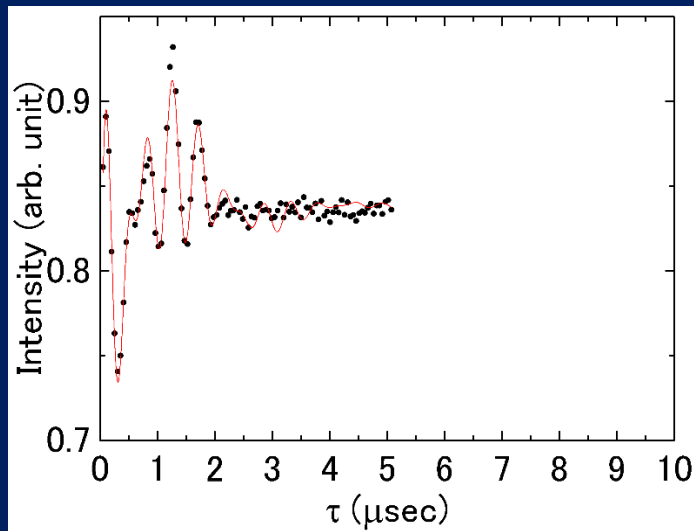
NV center ¹⁴

ODMR spectra and simulation



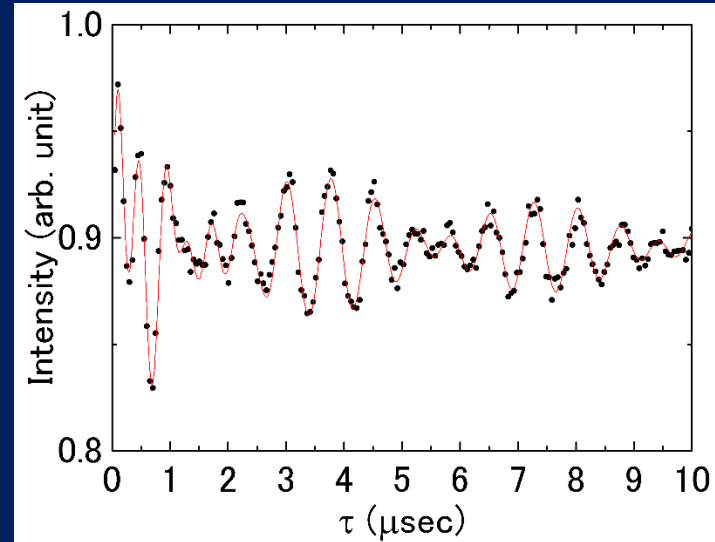
Free induction decay (T_2^*)

0 kV/cm



$$T_2^* = 1.2 \mu\text{sec}$$

80 kV/cm

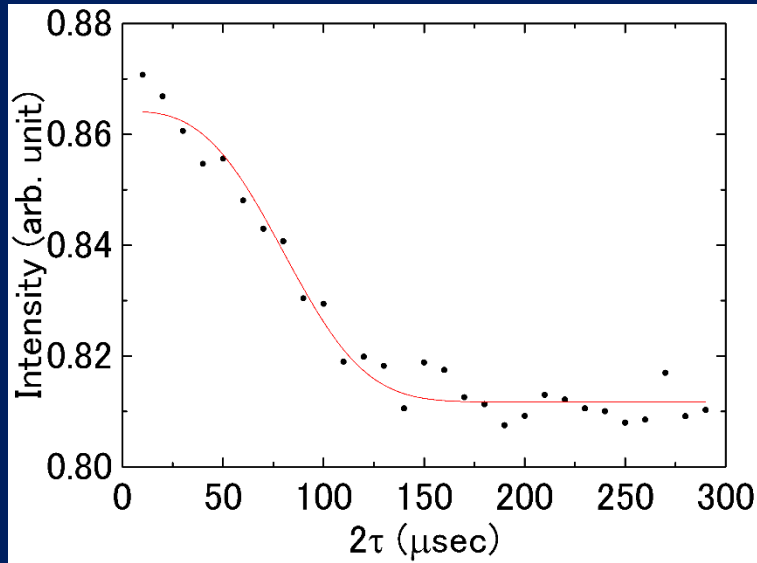


$$T_2^* = 11 \mu\text{sec}$$

Extension of T_2^* increased by E field !

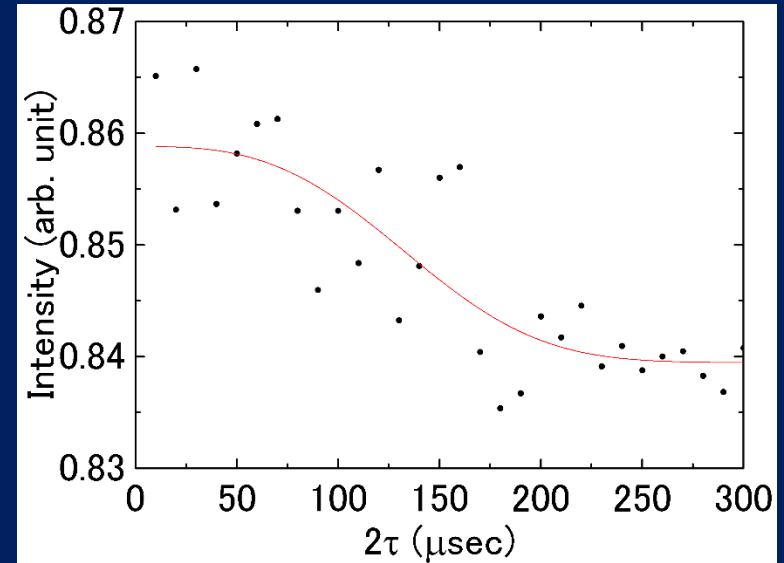
Hahn Echo meas. (T_2)

0 kV/cm



$$T_2 = 92 \pm 3 \mu\text{sec}$$

80 kV/cm



$$T_2 = 152 \pm 19 \mu\text{sec}$$

Extension of T_2 by E field !

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S. Kobayashi, NM, et al., unpublished.

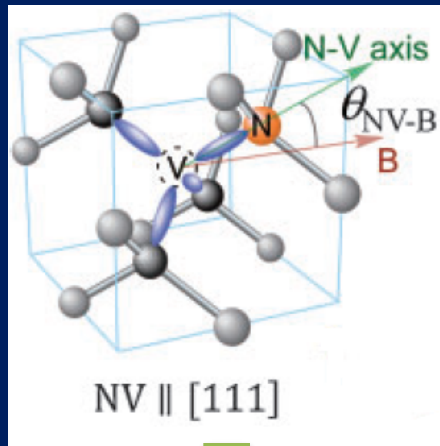
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Four-fold improvement of the magnetic field sensitivity

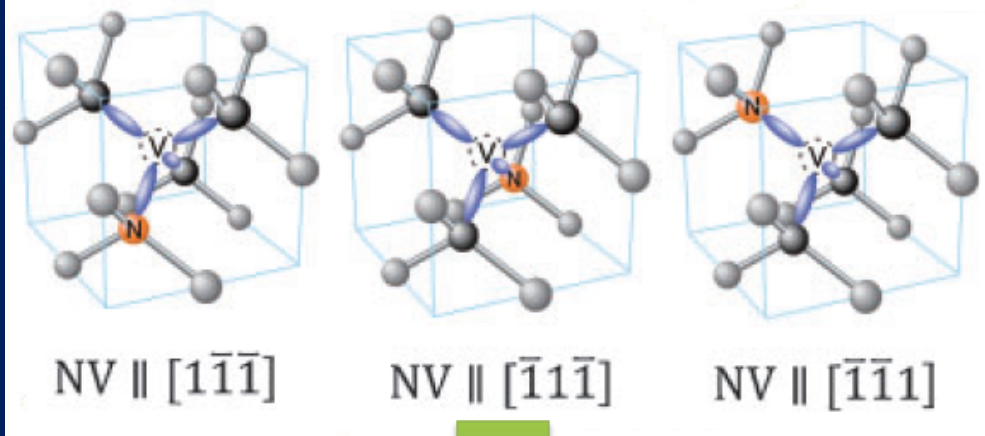
T. Fukui, NM, et al., *Appl. Phys. Exp.* 7, 055201 (2014), selected in “Spotlights”

T. Miyazaki, NM, et al., *Appl. Phys. Lett.* 105, 261601 (2014).

Magnetic field sensor sensitivity



the readout signal



the background signal

Magnetic field sensitivity : η

$$\eta \propto \frac{1}{C \sqrt{n_{NV} \tau T_2}}$$

C : readout contrast

n_{NV} : The number of NV

τ : Measurement time

L. M. Pham, et. al., Phys. Rev. B **86**, 121202 (2012)

η

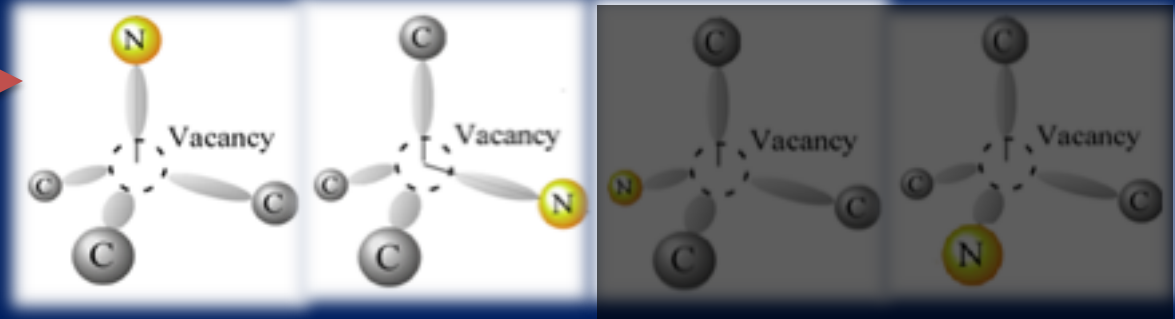
Four-fold enhancement due to four possible orientation

It is important to control NV orientation for magnetic sensor

Previous research

CVD layer

(110) and (100)
diamond substrate



NV centers aligned along two of the four orientation.

L. M. Pham, et. al., Phys. Rev. B **86**, 121202(2012)

A. M. Edmonds, et. al., Phys. Rev. B **86**, 035201(2012)

using (100)-CVD diamond, the readout contrast
and magnetic field sensitivity can be enhanced
by a factor of two.

L. M. Pham, et. al., Phys. Rev. B **86**, 121202 (2012)

$$\frac{1}{C\sqrt{n_{NV}}}$$



However, perfect alignment has not yet been realized....

**We will show the perfect alignment of NV
centers in (111)-CVD diamond**

Angular dependence of resonance

Spin Hamiltonian of NV center under magnetic field (B)

$$\mathcal{H}_S = DS_z^2 + E(S_x^2 - S_y^2) + g_s\mu_B\vec{B} \cdot \vec{S}$$

D, E : zero field splitting parameter
 g_s : electron g-value
 μ_B : Bohr magneton

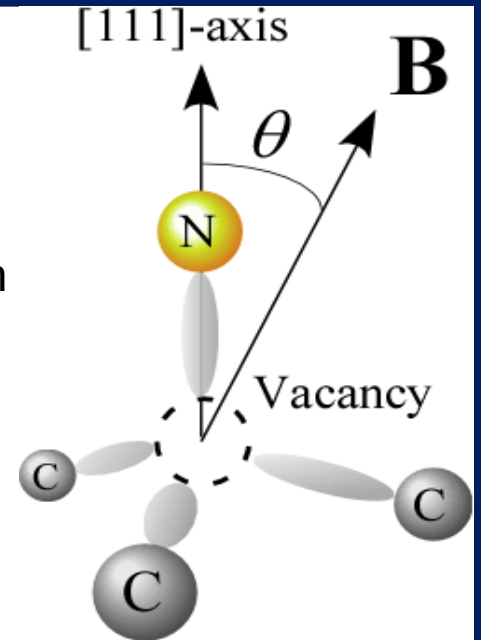
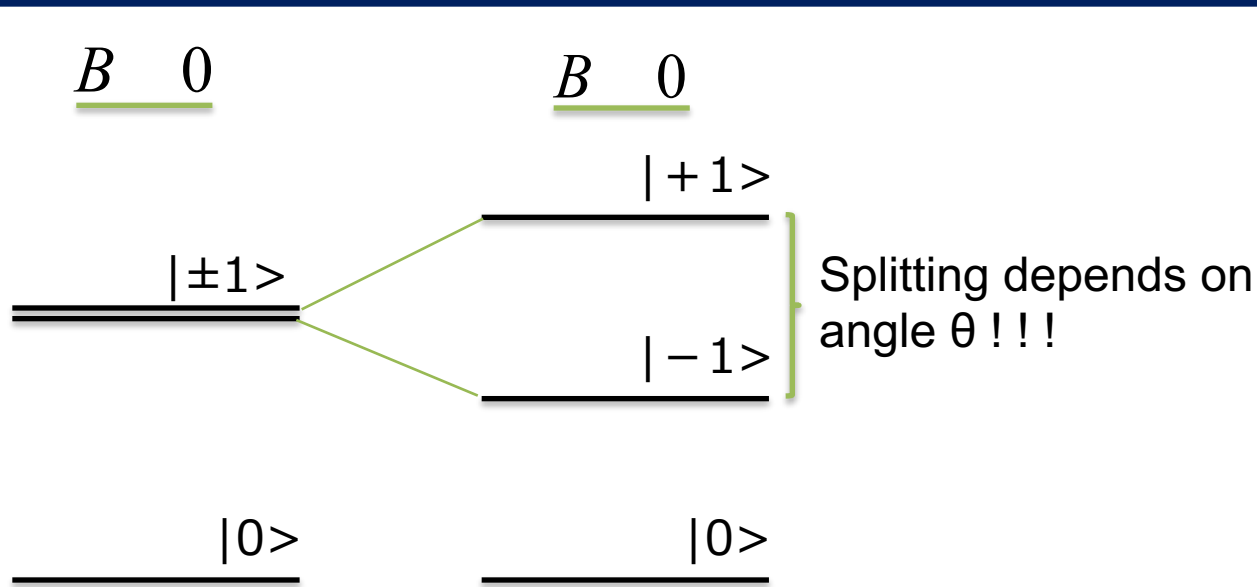
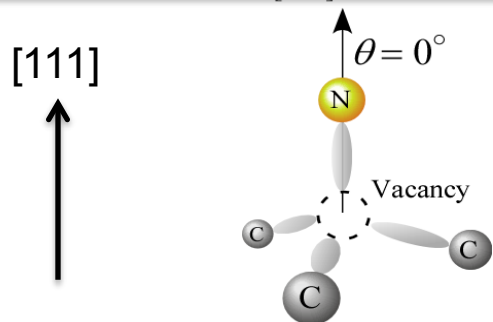


Fig. electronic spin structure of ground state of NV center

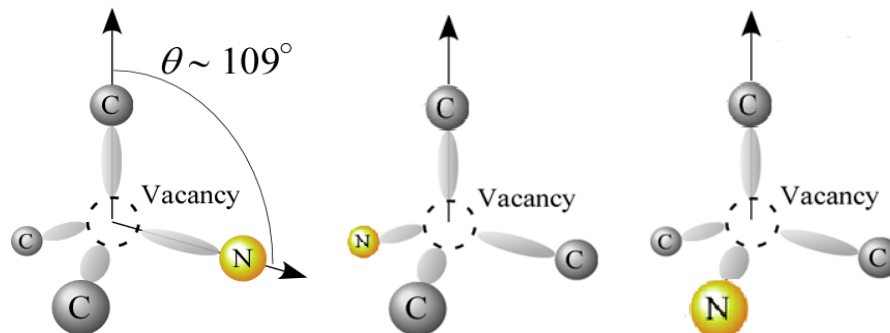
Splitting width depends on angular θ between the direction of NV axis and magnetic field

Investigation of the NV orientation by ESR

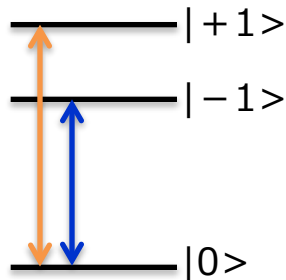
In the case of $B // [111]$



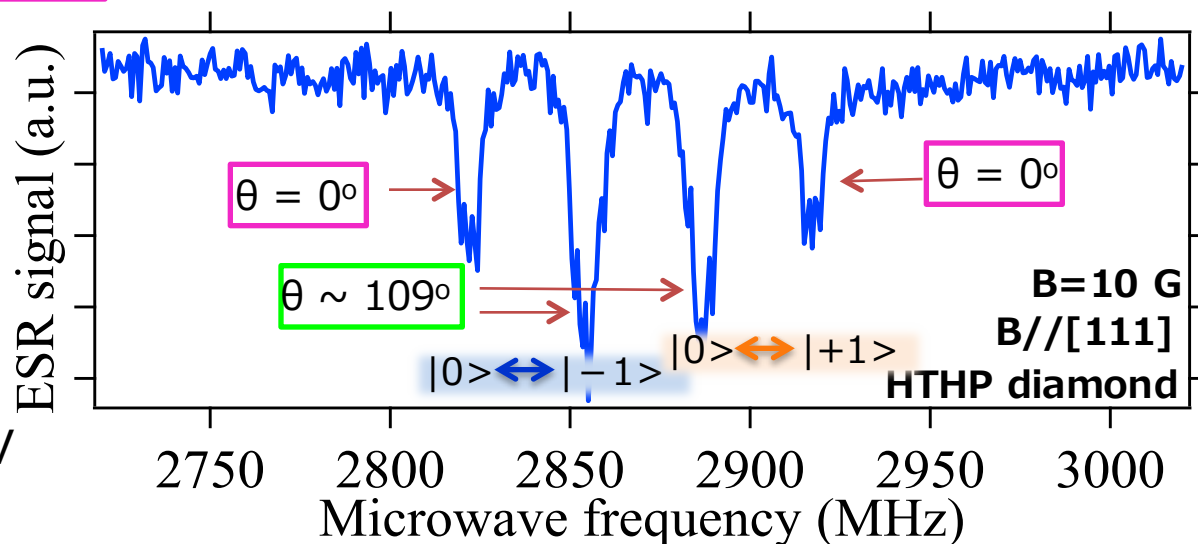
When NV orientation is along $[111]$ direction, $\theta = 0^\circ$



When NV orientation is along except for $[111]$ direction, $\theta \sim 109^\circ$

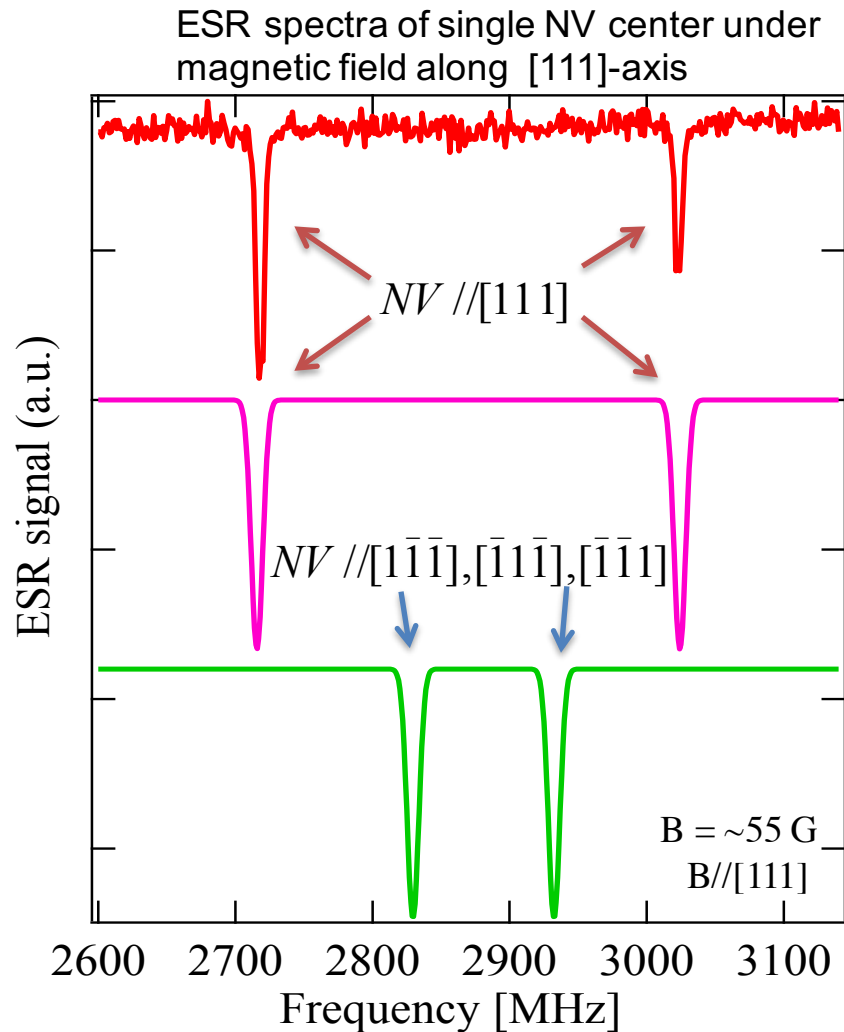


Electronic spin structure of NV center under B



Assignment of the NV orientation by magnetic resonance

Measurement result ~single~



— Single NV in (111)-CVD high quality diamond

— Simulation $\theta = 0^\circ$

— Simulation $\theta = 109.47^\circ$

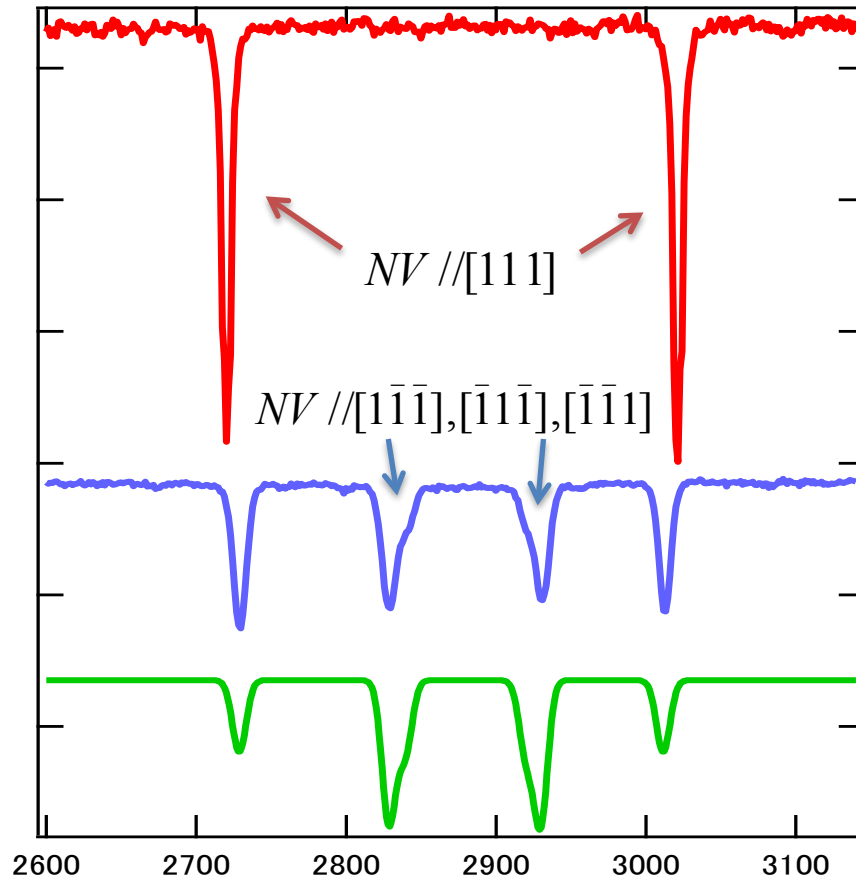
We randomly investigated 100 single NV centers

All NV centers have the same resonance frequency as red spectrum

The orientation of more than 99% of the single NV centers in (111)-CVD diamond were aligned along the [111] axis

Measurement result ~ensemble ~

Fig. ODMR spectra of ensemble NV centers under magnetic field along $[111]$ -axis



- in (111)-CVD high quality diamond
- in (111)-HTHP diamond (NV is incorporated by e-irradiation)
- Simulated blue spectrum

Only two peaks $NV//[111]$ appear !

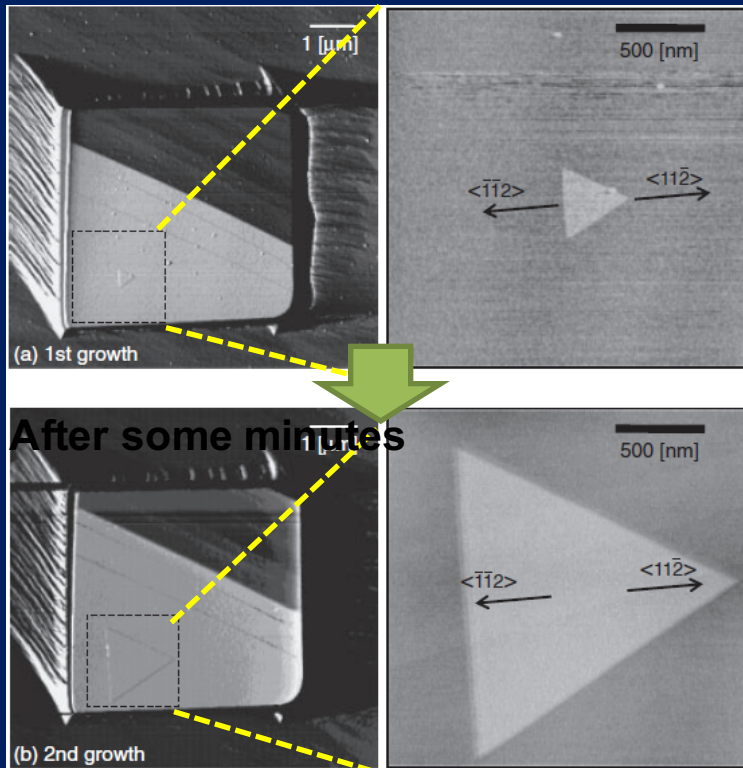
We randomly investigated more than 10 locations

All NVs exhibit an $NV//[111]$ orientation

We show the orientation of ~99% of the NV centers in (111)-CVD diamond can be aligned along the $[111]$ axis

(111)-CVD diamond growth ~Kink flow~

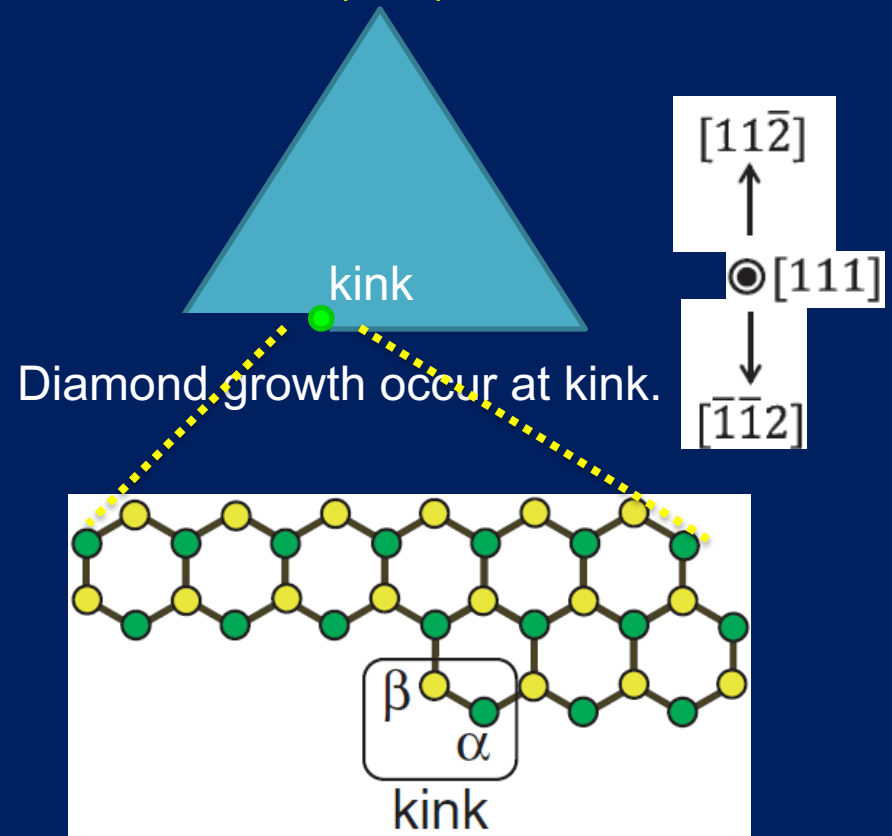
How to grow (111)-diamond by CVD



AFM image of diamond surface

N. Tokuda, et al., Jpn. J. Appl. Phys. (2014)

kink flow of (111)-diamond island



(111)- diamond bi-layer structure

α site is the top C atom in bi-layer

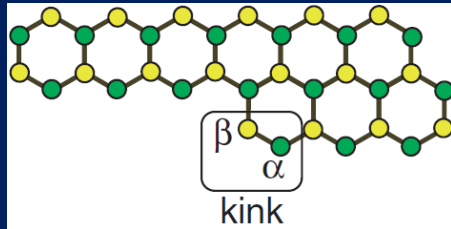
β site is the second C atom in bi-layer

(111)-diamond grows by C atom incorporation to α and β site in kink

Theoretical study

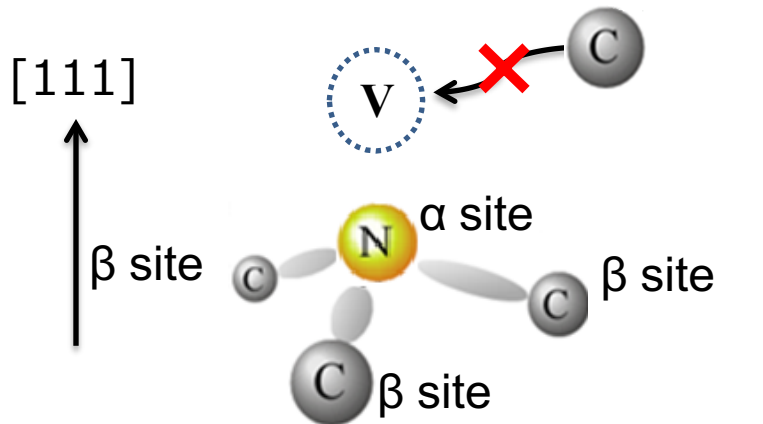
We calculated which N is easy to be incorporated at α or β site

(With first-principles calculation)

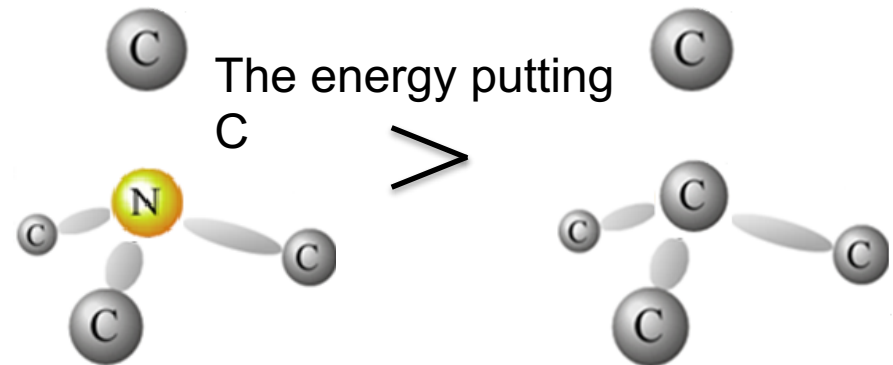


The N at the α kink site is energetically favorable.
(0.51 eV)

When an N atom is incorporated at α site



N atom doesn't have lone pairs.
So, C is difficult to put on N atom



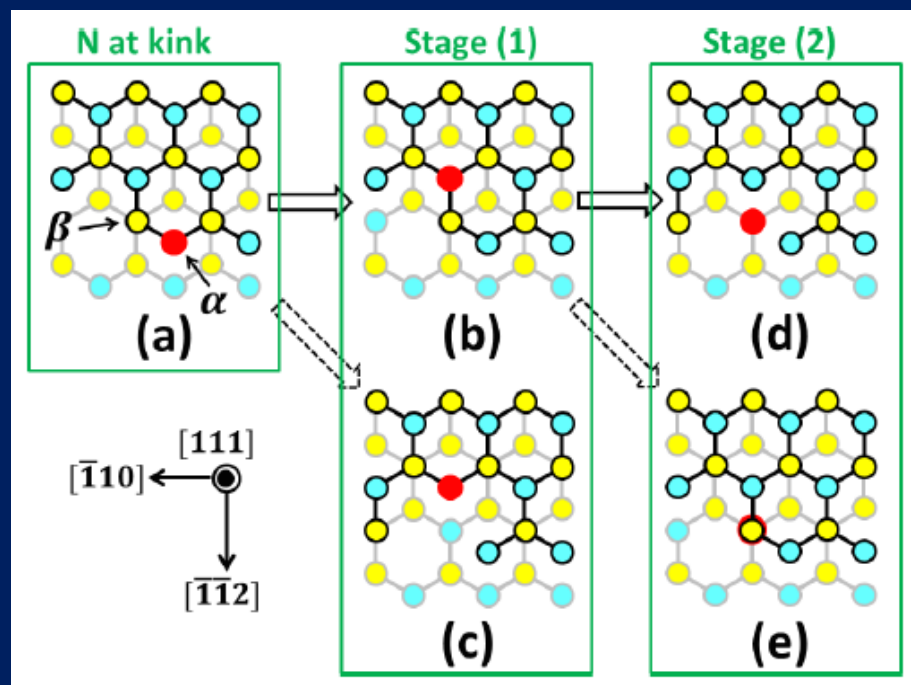
The energy putting C on N atom is larger (3 eV) than ones on C itself

Atomistic mechanism of perfect alignment of nitrogen-vacancy centers in diamond

Takehide Miyazaki^{1,6}, Yoshiyuki Miyamoto^{1,6}, Toshiharu Makino^{2,6}, Hiromitsu Kato^{2,6}, Satoshi Yamasaki^{2,6}, Takahiro Fukui³, Yuki Doi³, Norio Tokuda⁴, Mutsuko Hatano^{5,6}, and Norikazu Mizuochi^{3,6}

first-principles calculation

T. Miyazaki, NM, et al., *Appl. Phys. Lett.* 105, 261601 (2014).



If N atom is incorporated in this mechanism, perfect alignment by CVD can be realized!

Summary

- Extension of coherence time by electric field.

S. Kobayashi, NM, et al., unpublished

- The orientation of more than 99% of the NV centers could be aligned along the [111] axis. The mechanism was discussed.

T. Fukui, NM, et al., *Appl. Phys. Exp.* 7, 055201 (2014), selected in “Spotlights”

T. Miyazaki, NM, et al., *Appl. Phys. Lett.* 105, 261601 (2014).

Important in application for
quantum information, sensing, ...

