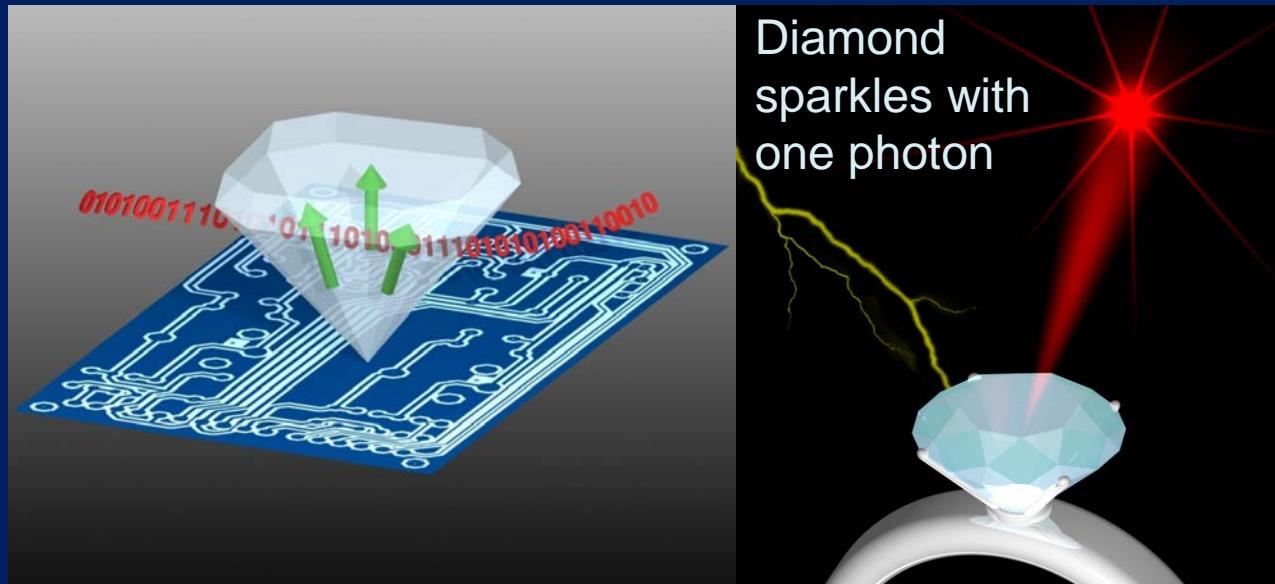


First夏季研修会

ダイヤモンド中のNV中心を用いた量子情報処理

Norikazu Mizuuchi

Engineering Science, Osaka University



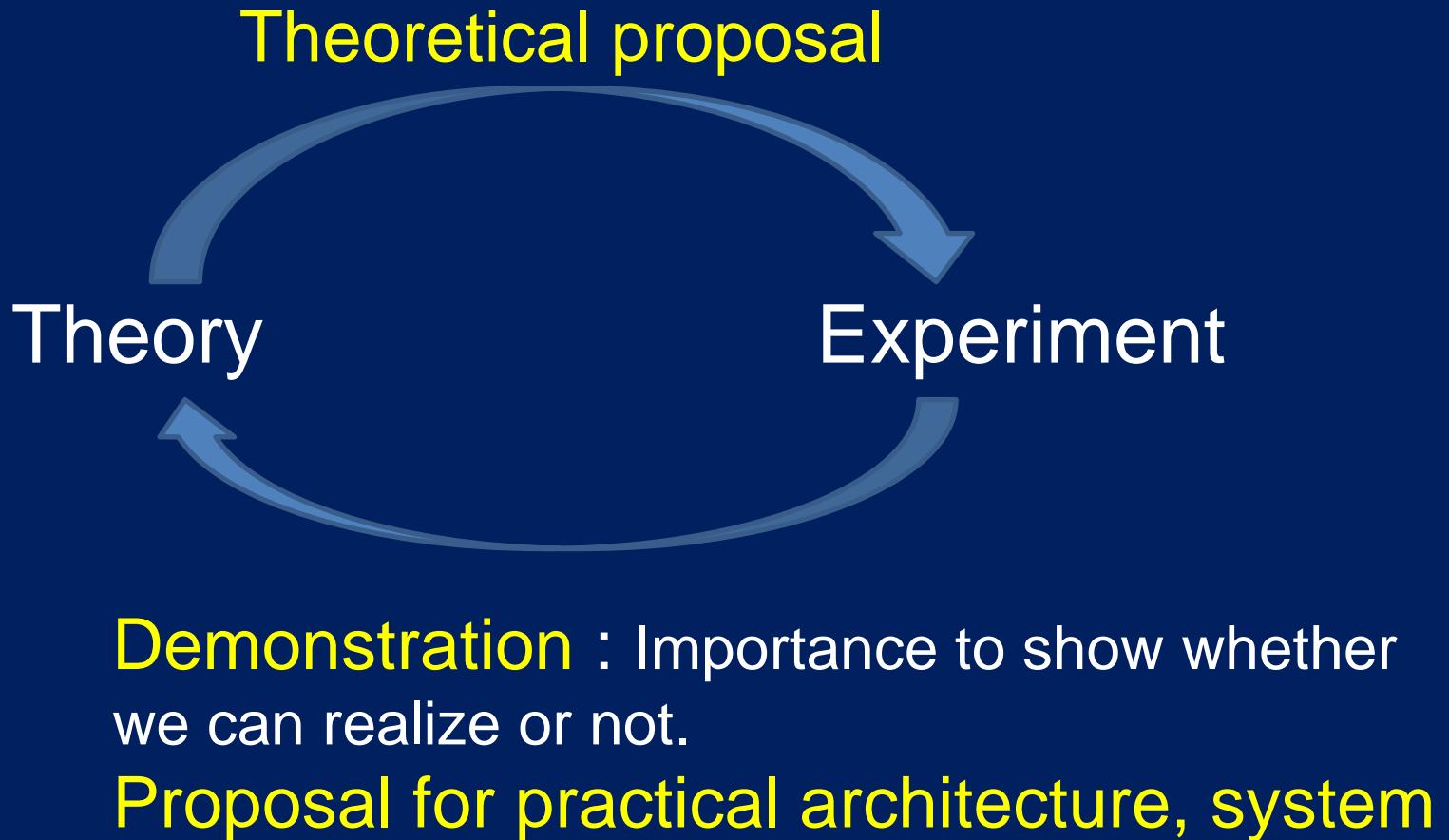
Collaborators and Acknowledgements

- Prof. Suzuki and group members (Osaka Univ.)
- Dr. Yamasaki and group members (AIST)
- Prof. Semba and group members (NTT)
- Prof. Kosaka (Tohoku Univ.)
- Prof. J. Wrachtrup and group members (Stuttgart Univ.)
- Prof. F. Jelezko (Ulm Univ.)
- Dr. A. Gali (Budapest Univ.)



Osaka Univ

Quantum information science and technology



Contents

Introduction of NV center in diamond

Historical view

Electronic states of NV center

Previous and Recent topics in NV center

何がなされてきたか？ NV中心で何ができるか？

Recent our results

Multi-qubit by Nuclear spins

Poster session (By Shimo-oka)

Electrical control

Electrically driven single photon source at room temperature based on NV center. *Nature Photonics*, 6, 299, 2012

Recent results

Diamond



近年、きれいで大きなダイヤモンドが人工的に合成されるように！

既にいくつかの企業から販売！

デビアスの子会社からもCVDで合成されたダイヤモンドを販売

既に1cm角が売られ、近い将来、1インチ角の試料も販売されるという。

Synthetic CVD Diamond

elementsix.

[CVD Diamond eShop](#) [Technology & Materials](#) [About eShop](#) [About E6](#)

Electronic Grade Plates

Element Six offers two types of Electronic grade diamond material - Single Crystal and Polycrystalline.

Single crystal Electronic grade is Element Six's highest purity commercially available material in plate sizes up to 4.5 x 4.5 mm. Larger plate sizes are available in Polycrystalline material.

These standard plates give researchers easy access to an exciting and enabling material technology.

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[Electronic Grade Polycrystalline Plate](#)

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[How To Buy](#)

CVD Single Crystal Electronic Grade Plates

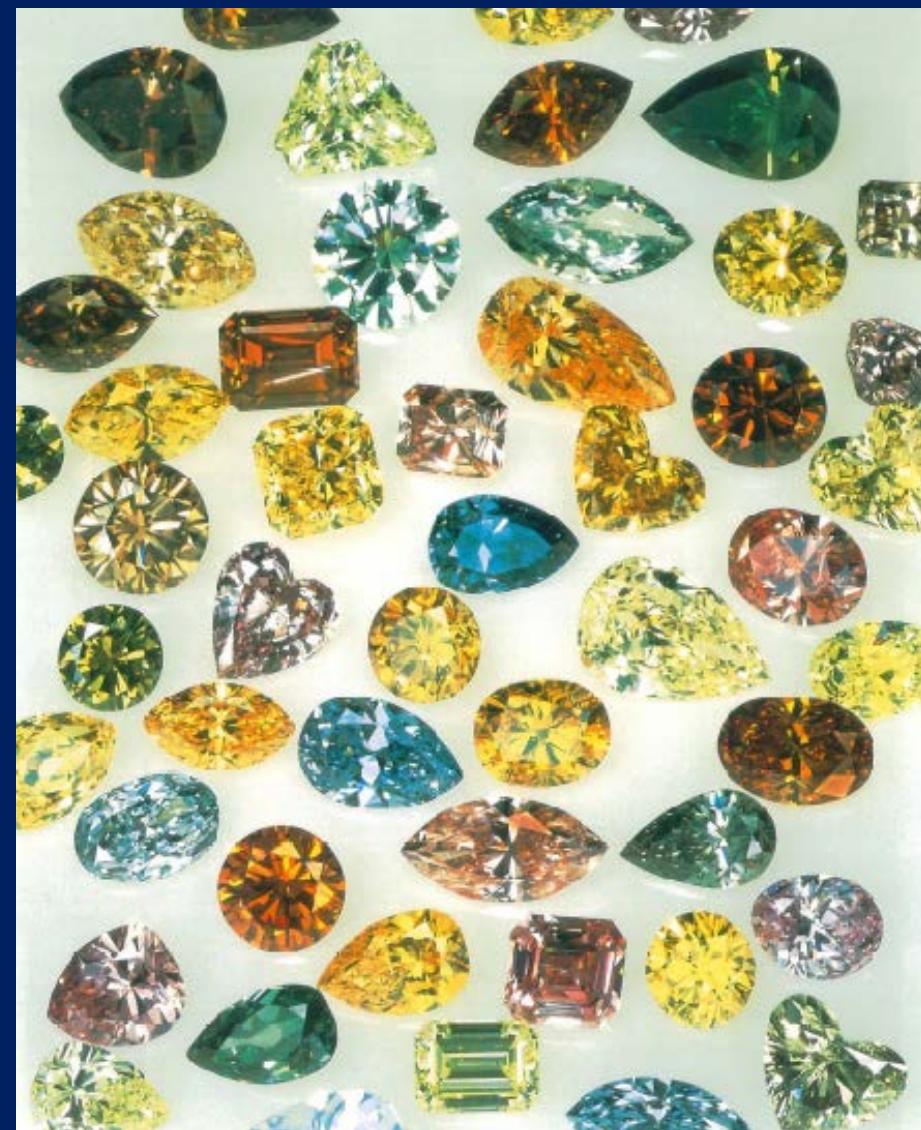
High purity CVD diamond plate suitable for electronic applications and quantum optics.

Plates are supplied with high quality, low damage polished surfaces.

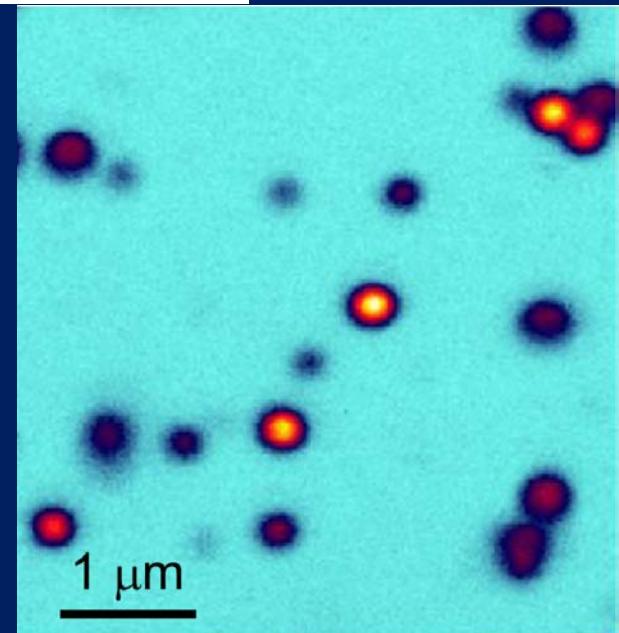
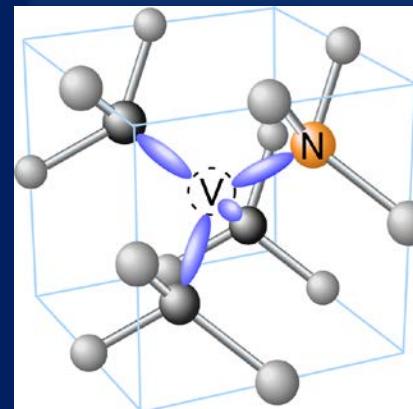
These plates have the lowest NV centre density of all commercially available Element Six materials.



Diamond



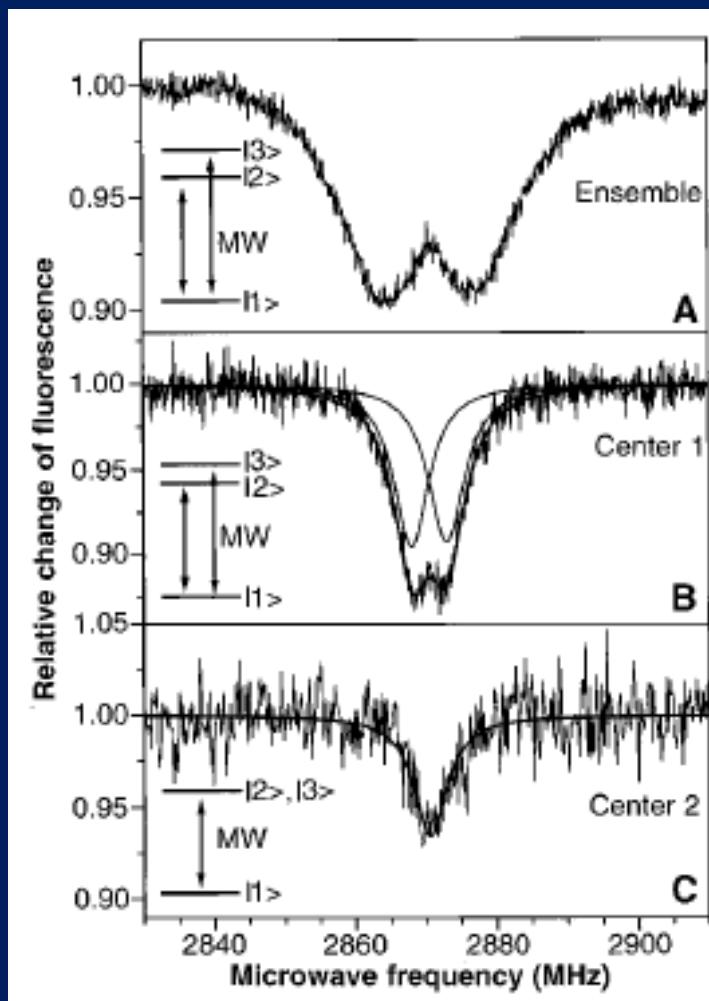
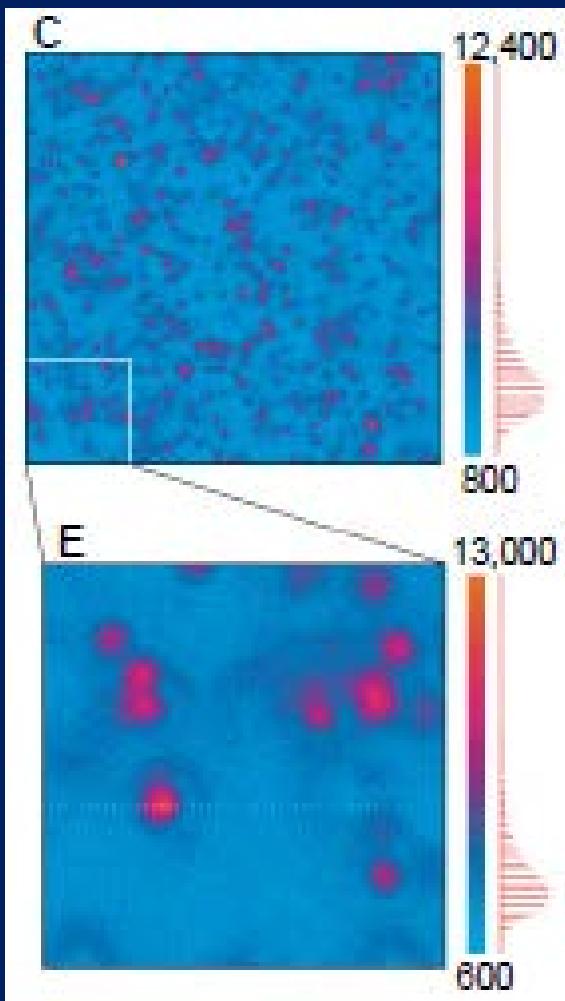
Impurities can incorporate :
Various colors



Historical view (1/3)

“Scanning confocal optical microscopy and magnetic resonance on single defect centers: Single NV center”

A. Gruber, A. Dräbenstedt, C. Tietz, L. Fleury, J. Wrachtrup,* C. Borczyskowski, *Science*, 276, 2012, 1997



Prof. Dr. J. Wrachtrup

Historical view (2/3)

VOLUME 65, NUMBER 21

PHYSICAL REVIEW LETTERS

19 NOVEMBER 1990

Single Pentacene Molecules Detected by Fluorescence Excitation in a *p*-Terphenyl Crystal

M. Orrit and J. Bernard

Centre de Physique Moléculaire Optique et Herzienne, Centre National de la Recherche Scientifique et Université de Bordeaux I,
351, Cours de la Libération, F-33405 Talence CEDEX, France

(Received 9 July 1990)



Prof. M. Orrit

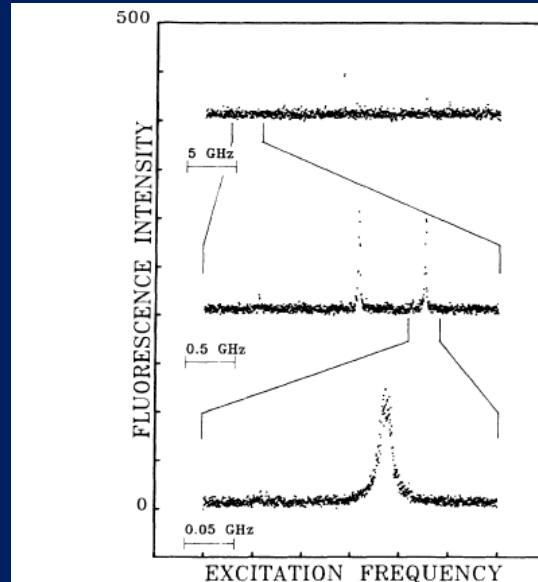
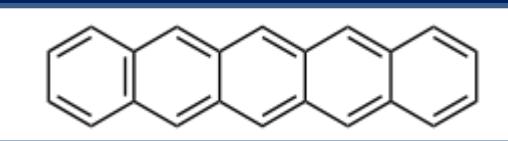


FIG. 2. Shape of a single molecule's excitation peak at different frequency scales. The bottom spectrum is approximately Lorentzian with FWHM about 12 MHz. The vertical scale is in counts/channel.

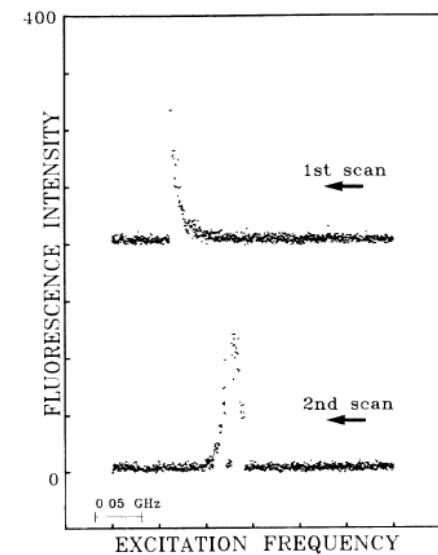


FIG. 4. Two successive scans of the excitation spectrum of a single molecule suggesting a photophysical hole-burning process. The sudden intensity falls and surges might arise from the flip-flops of a two-level system in the neighborhood of the molecule. The time per channel was 0.08 s and a scan lasted about 1 min. The vertical scale is in counts/channel.

Historical view (3/3)

Optical detection of magnetic resonance in a single molecule

J. Wrachtrup*, C. von Borczyskowski*, J. Bernard†,
M. Orrit† & R. Brown†

* Fachbereich Physik, Freie Universität Berlin, Arnimallee 14,
1000 Berlin 33, Germany

† Centre de Physique Moléculaire Optique et Hertzienne, u.a. 283 du
C.N.R.S., Université Bordeaux I, 33405 Talence cedex, France

NATURE · VOL 363 · 20 MAY 1993

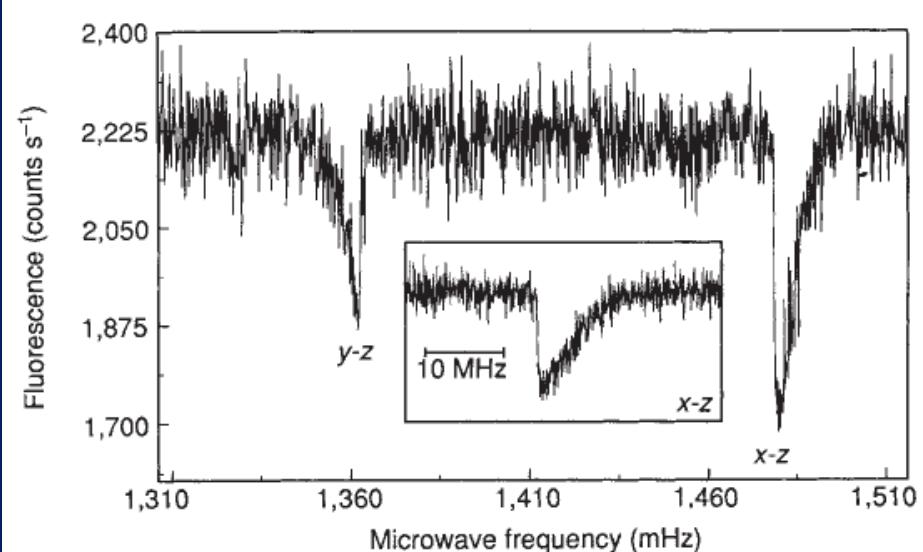


FIG. 1 Decrease of the fluorescence intensity of a single pentacene molecule when the microwave field is resonant with the $y-z$ or the $x-z$ transition. The inset shows the $x-z$ transition on an enlarged frequency scale, on reducing the microwave power by a factor of ~ 30 .

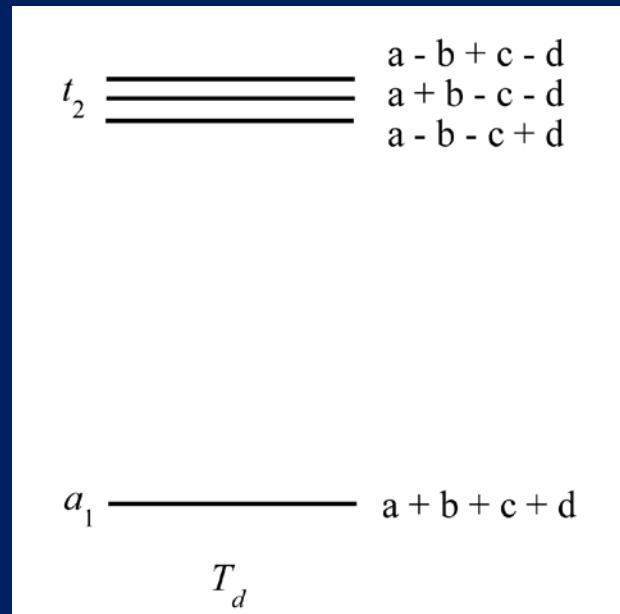
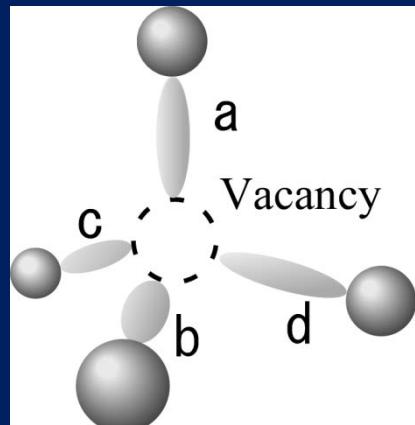
Electronic states of NV center (1/5)

Why it has S=1 ? About optical property.

ダングリングボンド軌道(原子軌道)の線形結合
:LCAO分子軌道による理解
各エネルギーレベルの性質を定性的に理解

C. A. Coulson and M. J. Kearsley, Proc. R. Soc. Lond. A 1957 241, 433.
G. D. Watkins, "Deep defects in semiconductors"

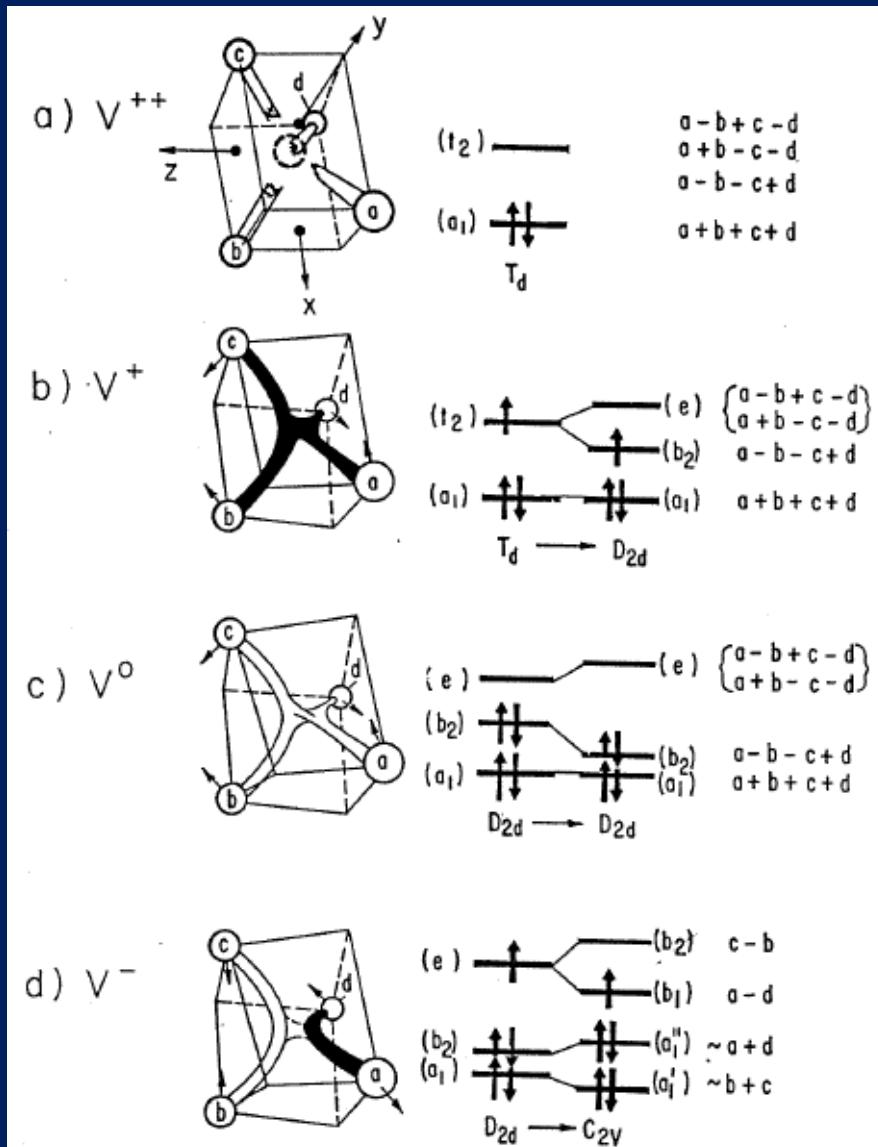
四面体対称(T_d)の場合



Electronic states of NV center (2/5)

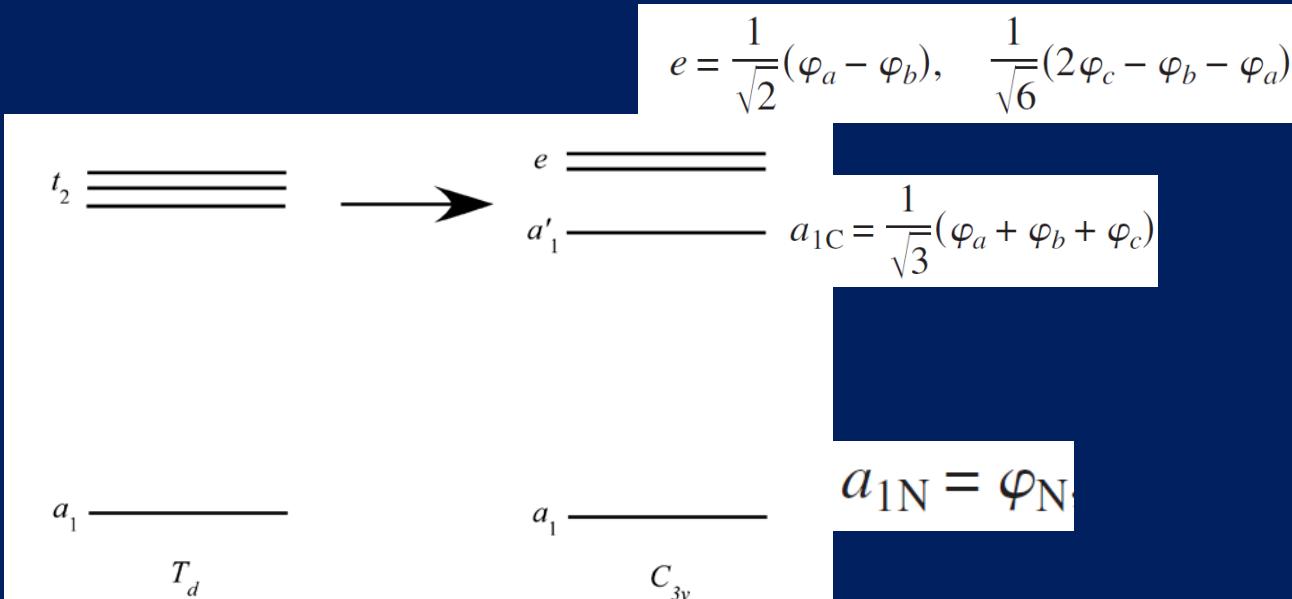
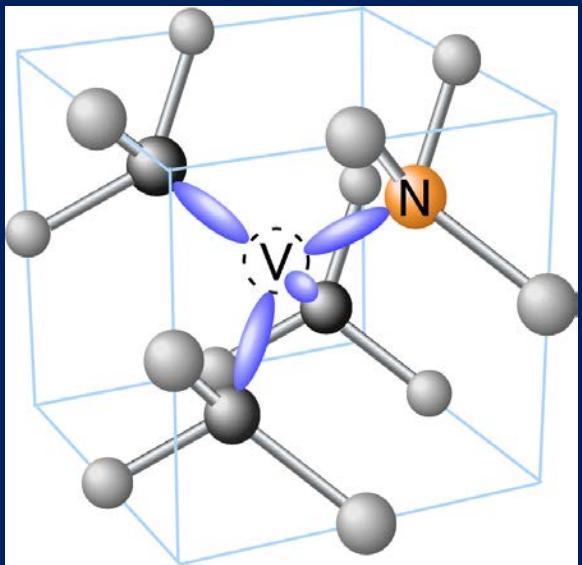
Example in Si

In Si, the structure distorts, but does not in diamond.

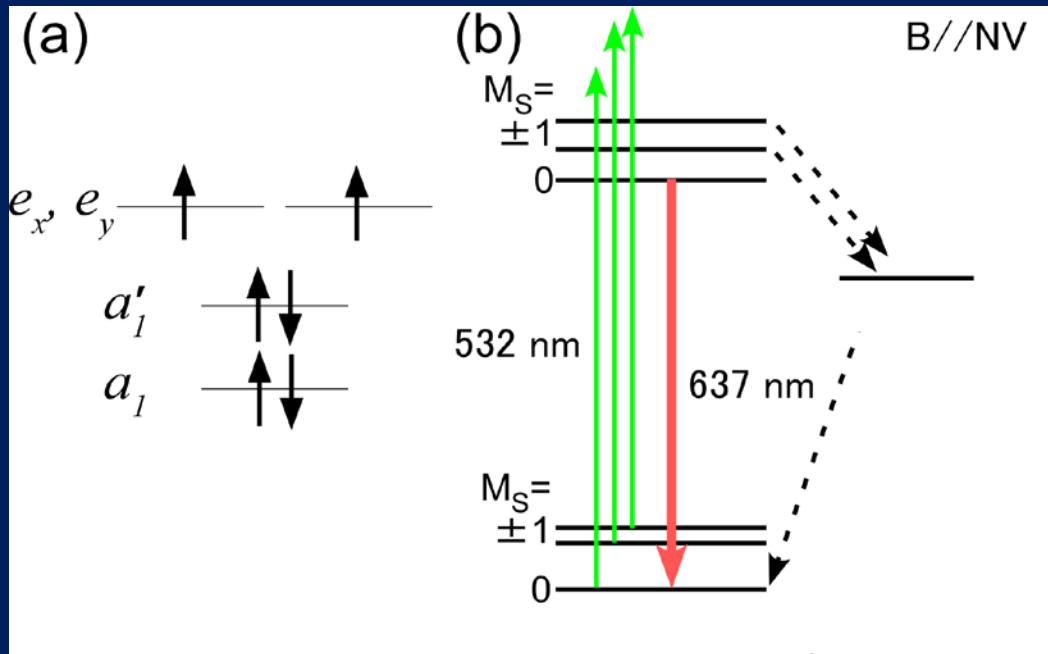
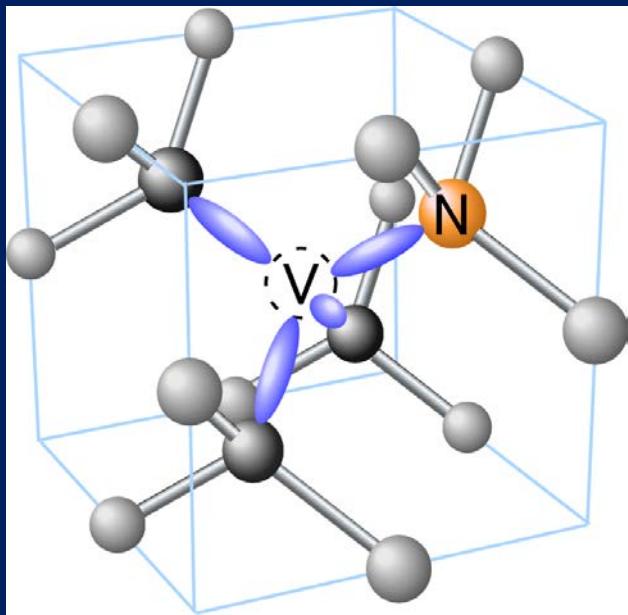


Electronic states of NV center (3/5)

S. Felton, et al., PRB 77, 081201, 2008.



Electronic states of NV center (4/5)

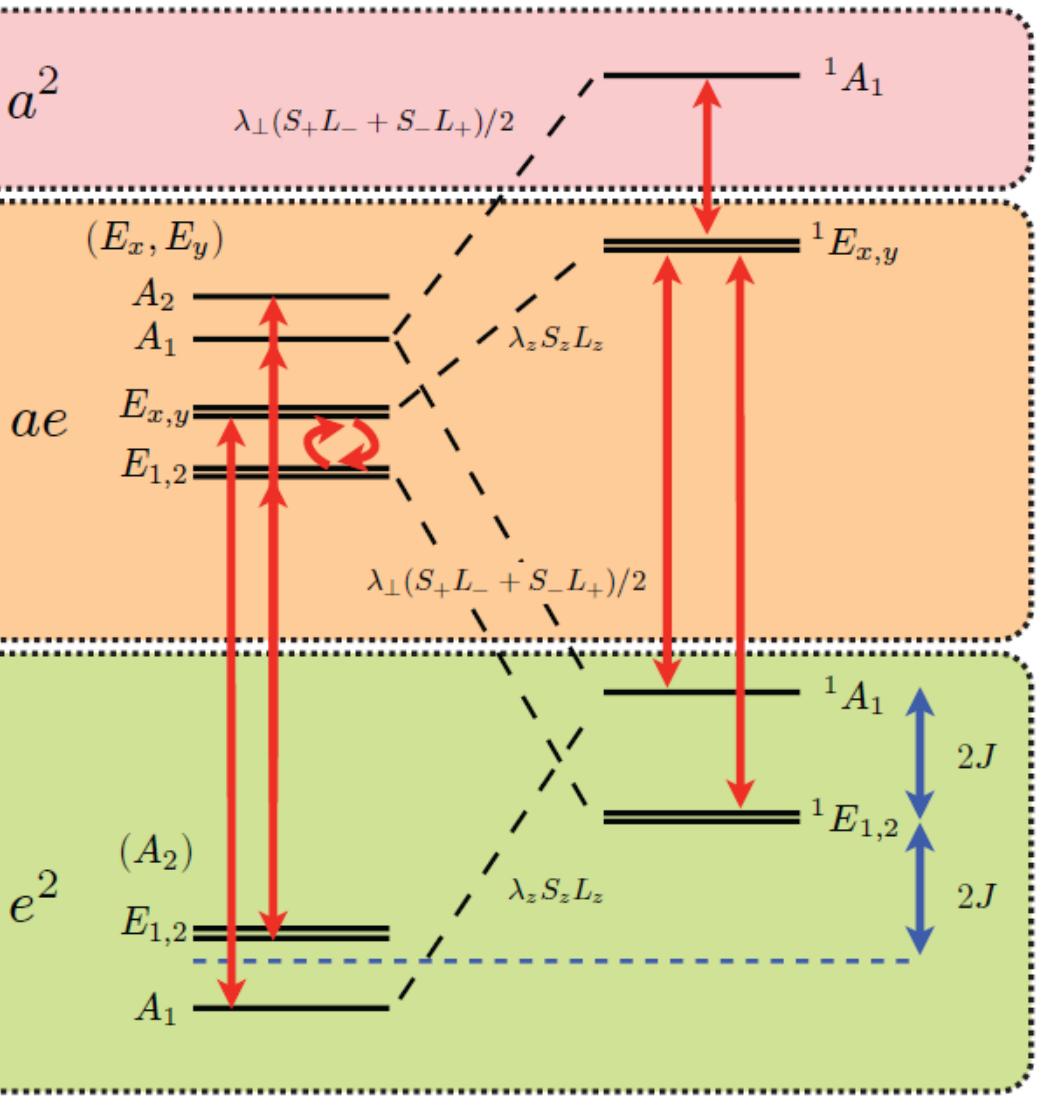


Electronic states of defect in deep level within band-gap

- Spin ($S=1$) with long spin coherence time
- Strong and stable emission (Fluorescence)

Control and detection of single spin at RT

Electronic states of NV center (5/5)

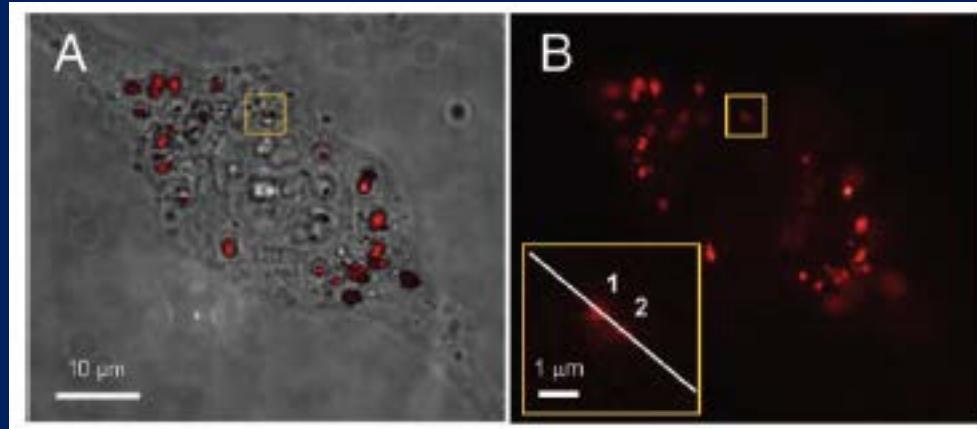


- ・スピン-スピン相互作用、スピン軌道相互作用を考慮した詳細な電子状態
- ・群論(対称性)による定性的な理解だけでなく、ab initio法による定量的な議論

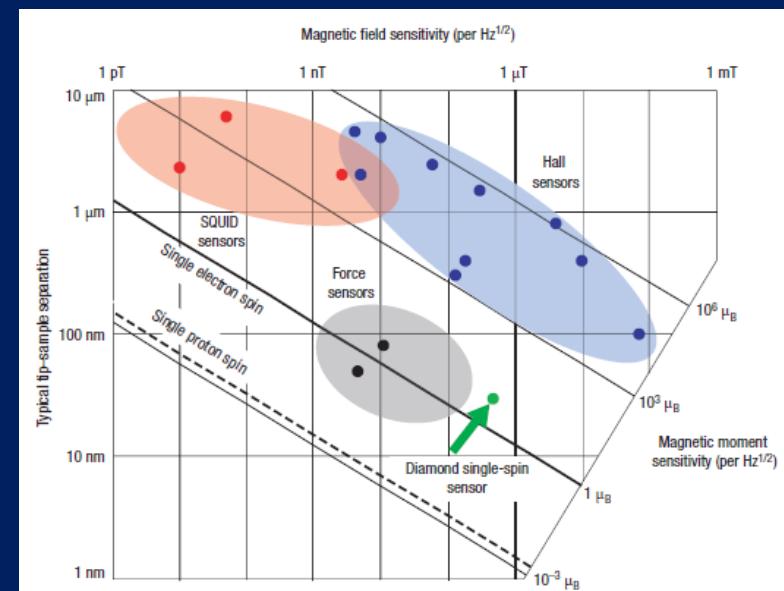
J. Maze et al., NJP, 13
(2011) 025025.

Topic (other than quantum information science)

Single fluorescent nanodiamonds as cellular biomarkers
(PNAS 2007)



Scanning probe magnetometry (magnetic sensor)
(Nature, 455, 648, 2008)

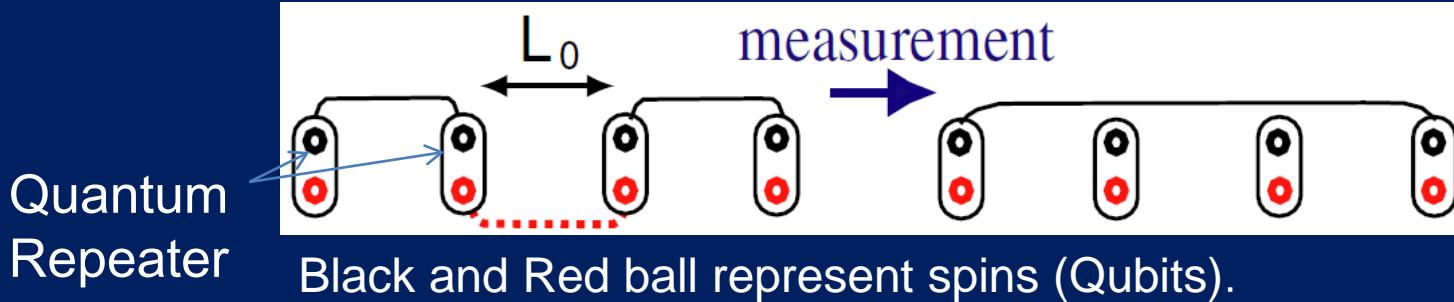


Characteristics of NV center (quantum information science)

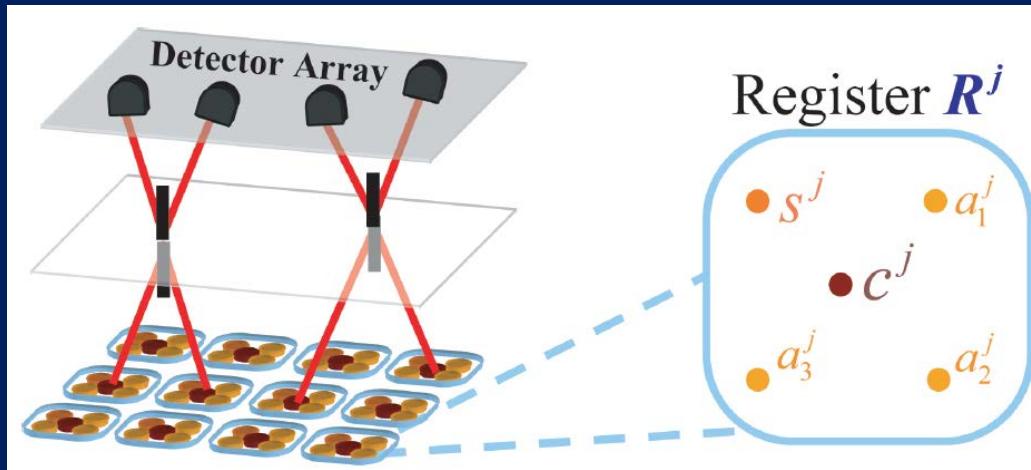
- Ground triplet state (Electron spin quantum number $S=1$)
- In solid material, coherent control of single spin at RT is only possible in NV center. (Science 1997)
- Long coherence time ($T_{2e} > 1$ ms at RT, Nature Material 2009, $T_{2n} > 1$ s at RT, Science 2012).
- Optical initialization of electron spin and nuclear spin
- Entanglement between photon and spin (Nature 2010)
- QND meas. of nuclear spin at RT. (Science 2010)
- Single shot meas. of electron spin. (Nature 2011)
- Multi-qubits by nuclear spins (N. Mizuuchi et al., PRB 2009 ...)
- Bell state among single nuclear spins $T_2 = 5\text{ms}$ at RT.
(P. Neumann, N. Mizuuchi et al., Science 2008)

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

Quantum Cryptography (single photon source, quantum repeater)



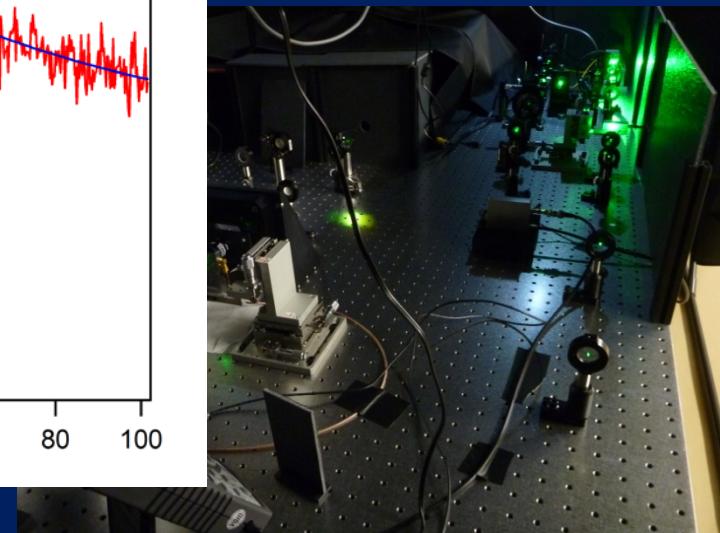
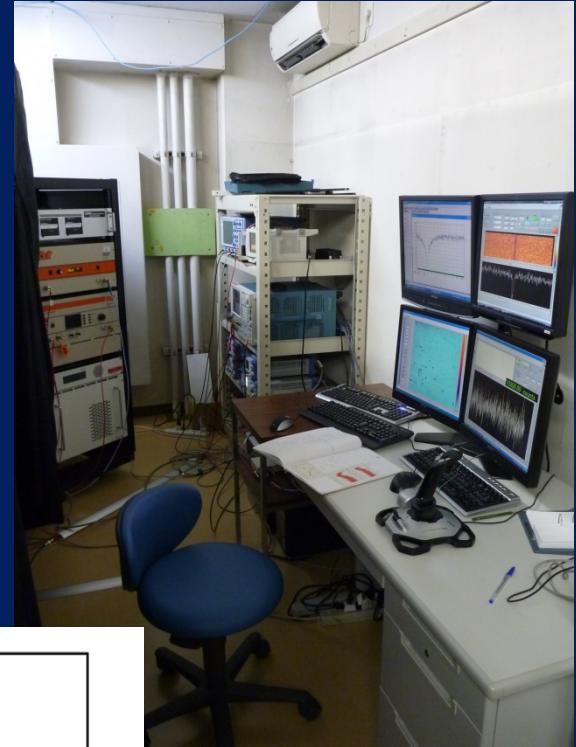
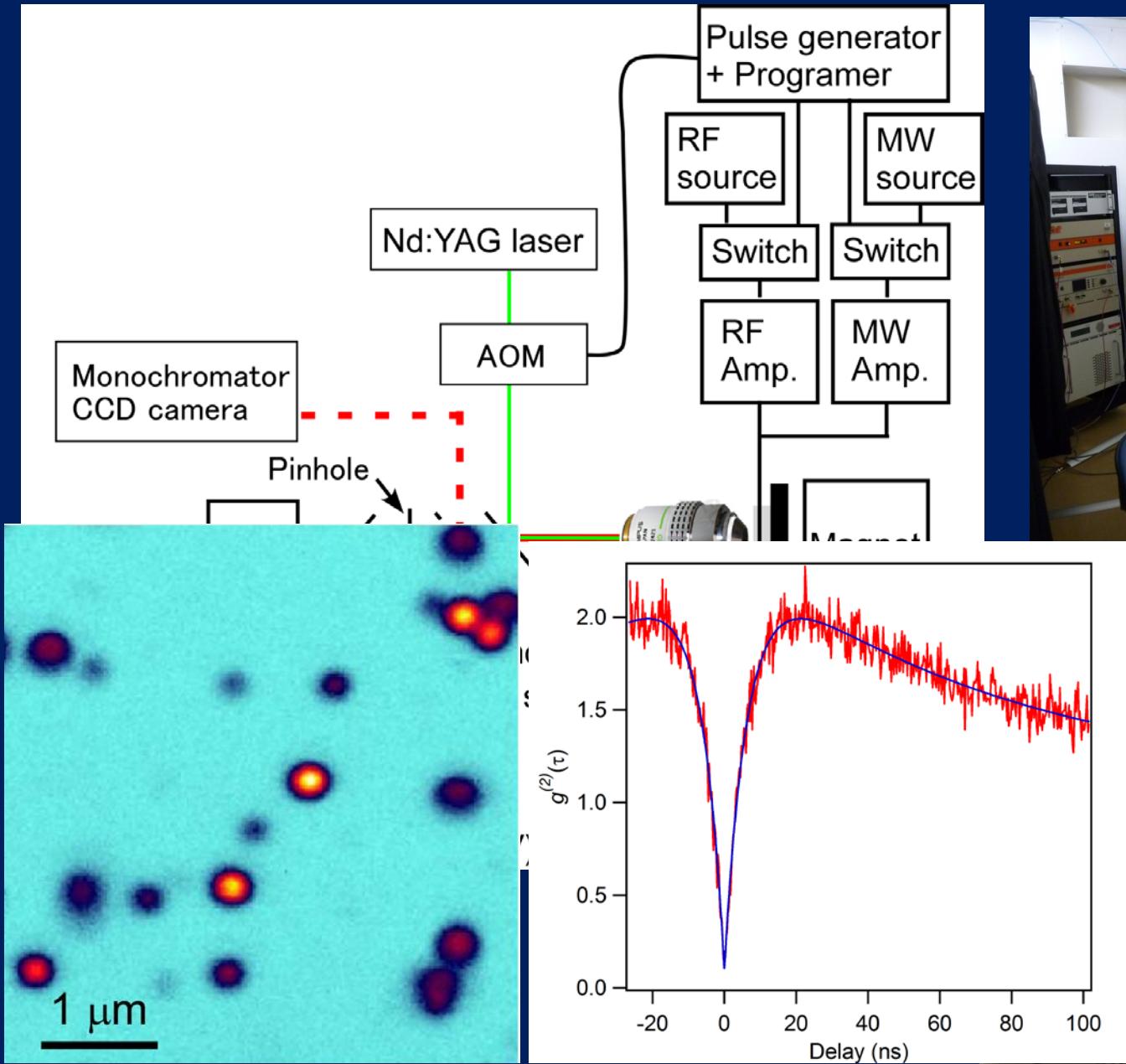
Distributed scalable quantum computer



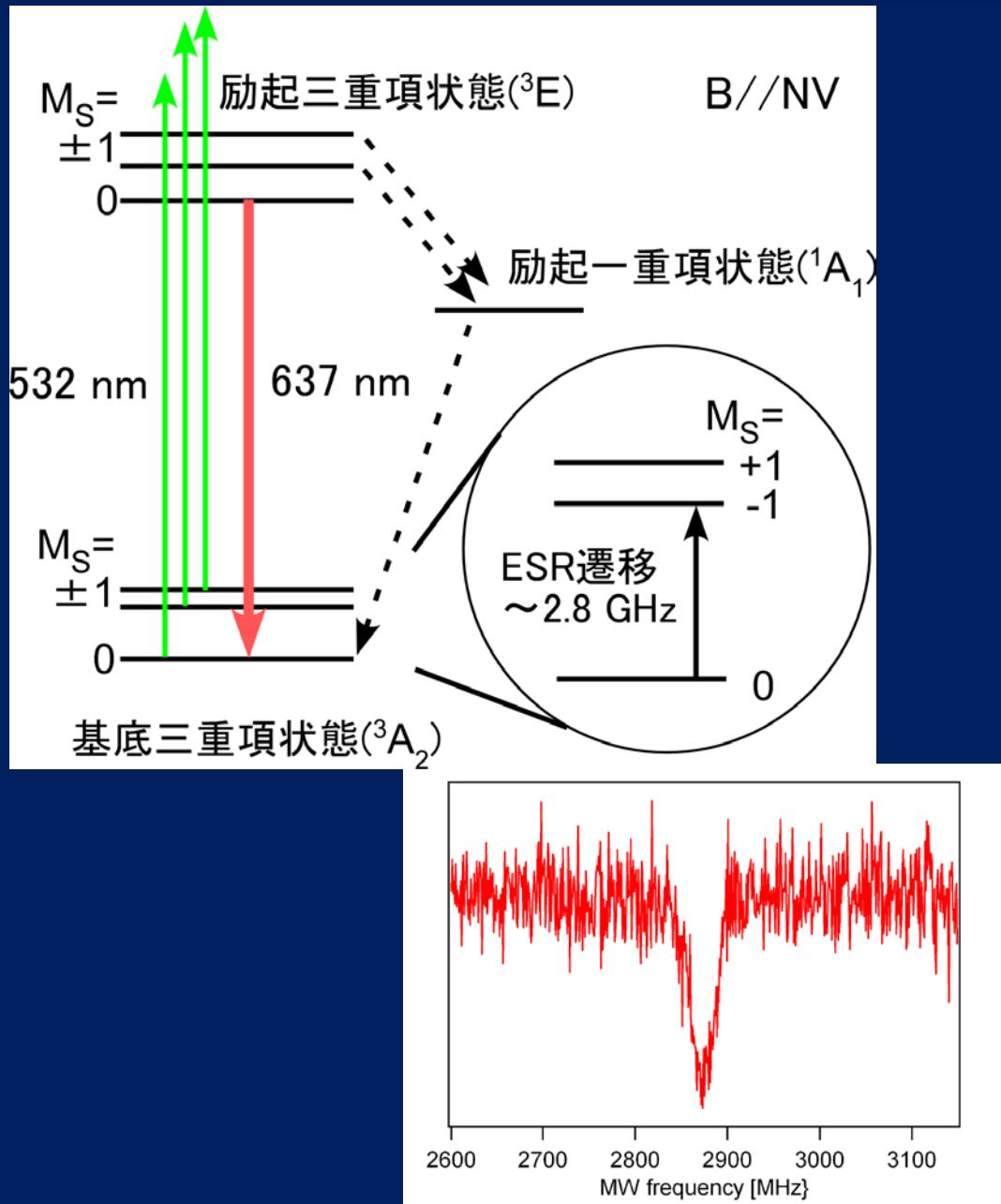
It consists of 5 qubits quantum registers. Jiang et al., PRA 2007

Several-qubit solid system with optical accessibility

Measurement system (in Osaka Univ.)



Optically detected magnetic resonance (ODMR)



Laser excitation (532 nm) plays very important roles as initialization and read out the spin states.

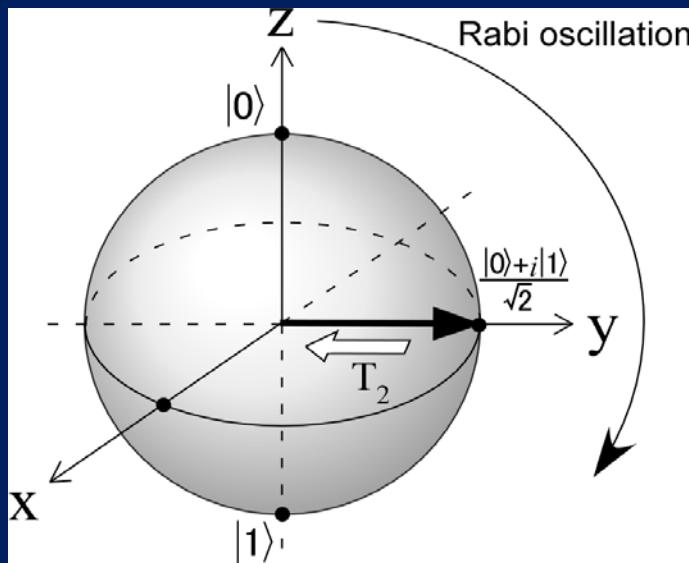
Pure state can be prepared.

We detect Single NV and repeat detection cycles.

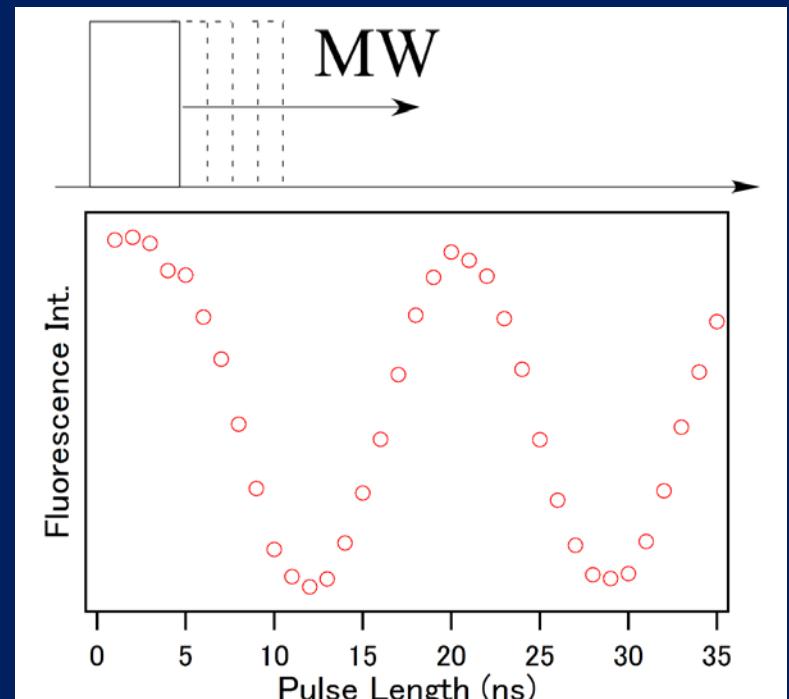
Control of spin

By spin resonance

$$\mathbf{H} = g\beta \mathbf{S} \cdot \mathbf{B}_1$$



$\pi/2$ pulse : coherence generation
 π pulse : inversion



Previous and Recent topics in NV center (1)

Demonstration of single photon source at RT

Stable Solid-State Source of Single Photons

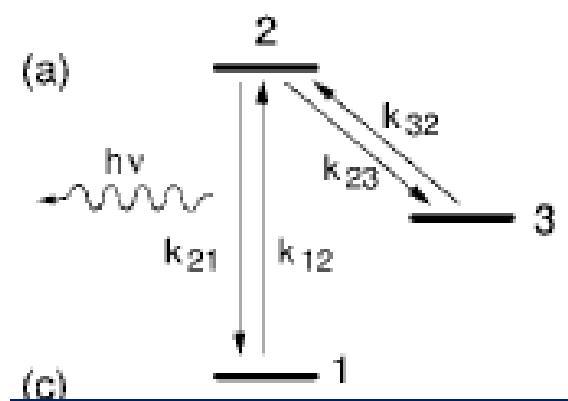
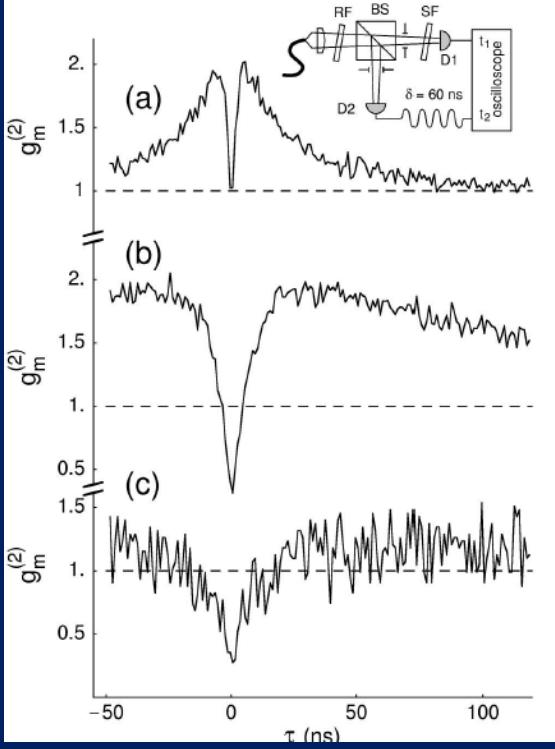
Christian Kurtsiefer,¹ Sonja Mayer,¹ Patrick Zarda,² and Harald Weinfurter^{1,2}

¹*Sektion Physik, Ludwig-Maximilians-Universität, D-80799 München, Germany*

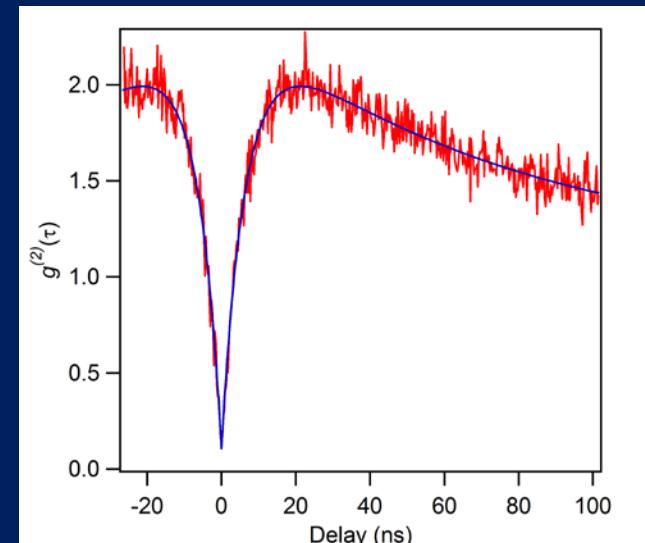
²*Max-Planck-Institut für Quantenoptik, D-85748 Garching, Germany*

PHYSICAL REVIEW LETTERS

10 JULY 2000



Our measurement

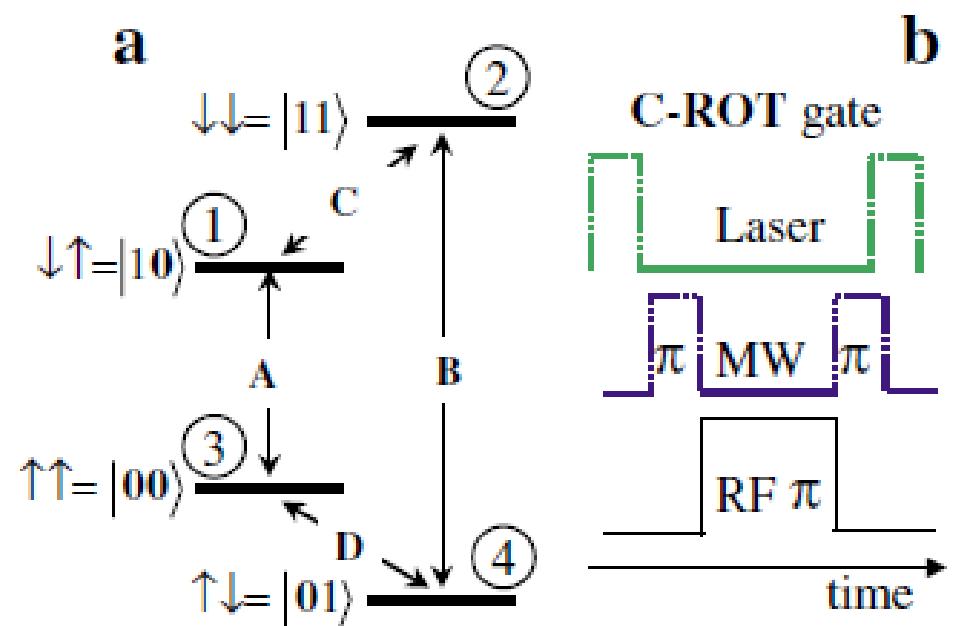
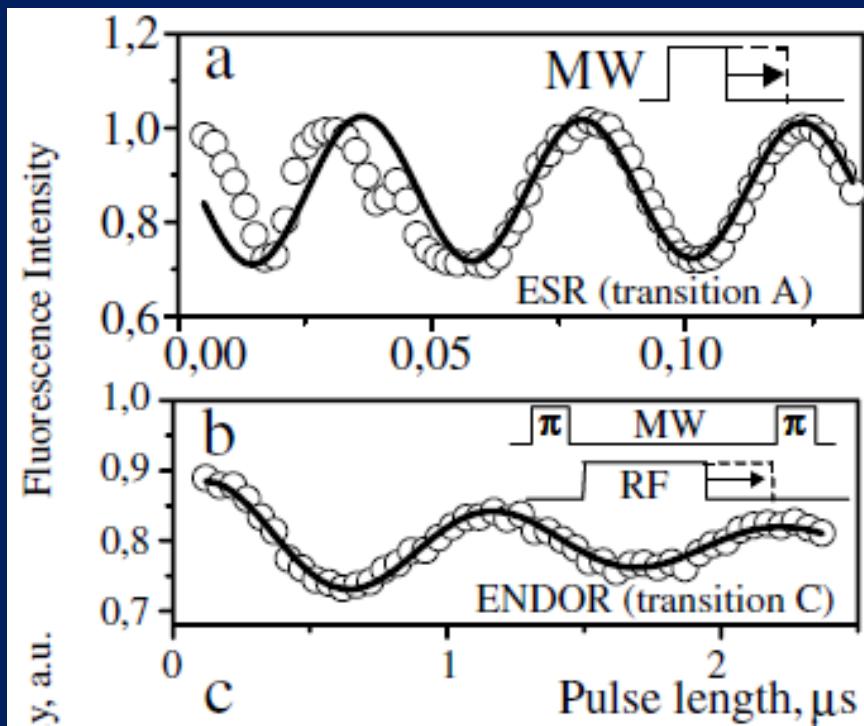


645 nm ~ 800 nm

Previous and Recent topics in NV center (2)

CNOT gate of two qubit (NV- ^{13}C , electron spin and ^{13}C nuclear spin)

F. Jelezko et al., *Phys. Rev. Lett.*, 93, 130501, 2004



(CROT is equivalent to CNOT gate except for a $\pi/2$ rotation of the nuclear spin around z-axis.)

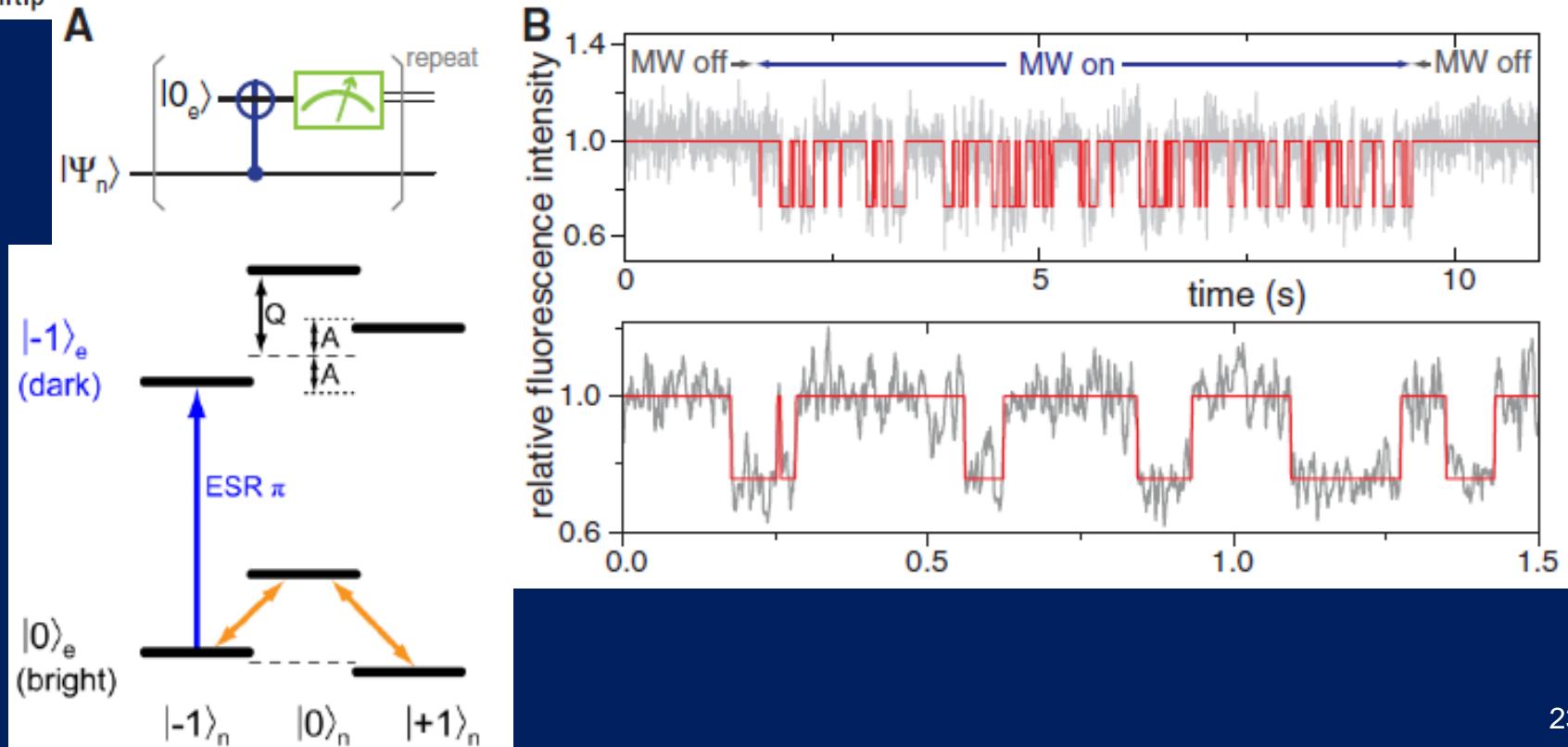
Previous and Recent topics in NV center (3)

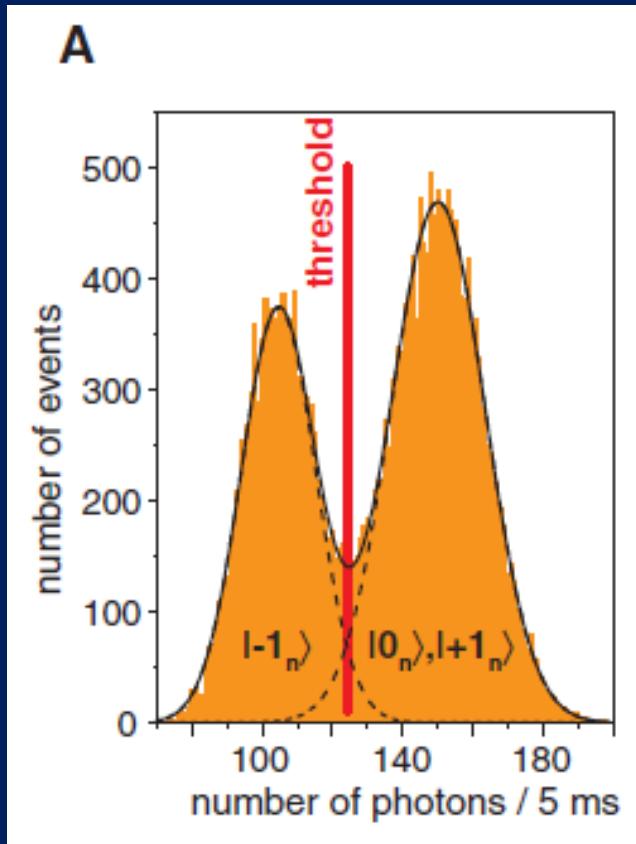
Quantum non-demolition measurement, Projective measurement of a single nuclear spin at RT

Single-Shot Readout of a Single Nuclear Spin

Science 2010

Philipp Neumann,¹ Johannes Beck,¹ Matthias Steiner,¹ Florian Rempp,¹ Helmut Fedder,¹
Philip Schneidewind², Christian T. Spiegel², and Ulrich Gähler^{1,3}





The jumps occurs nuclear-electron flip-flop process.

$$\text{hyperfine Hamiltonian } H_A = (\hat{S}_+ \hat{I}_- + \hat{S}_- \hat{I}_+) A_{\perp} / 2 + \hat{S}_z \hat{I}_z A_{\parallel}$$

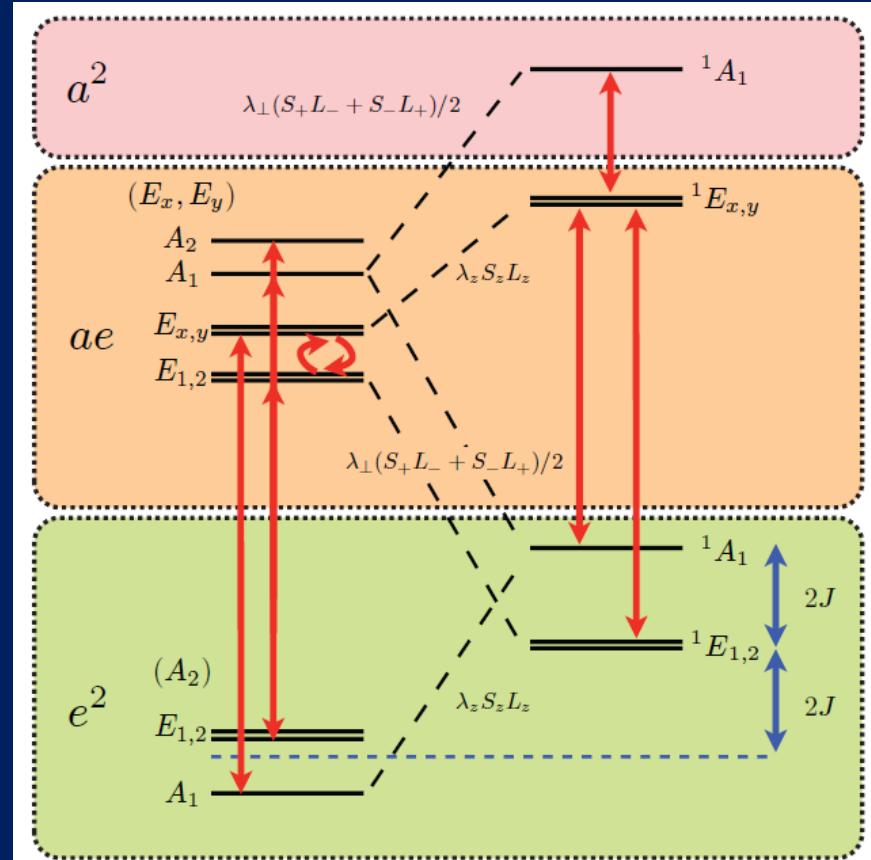
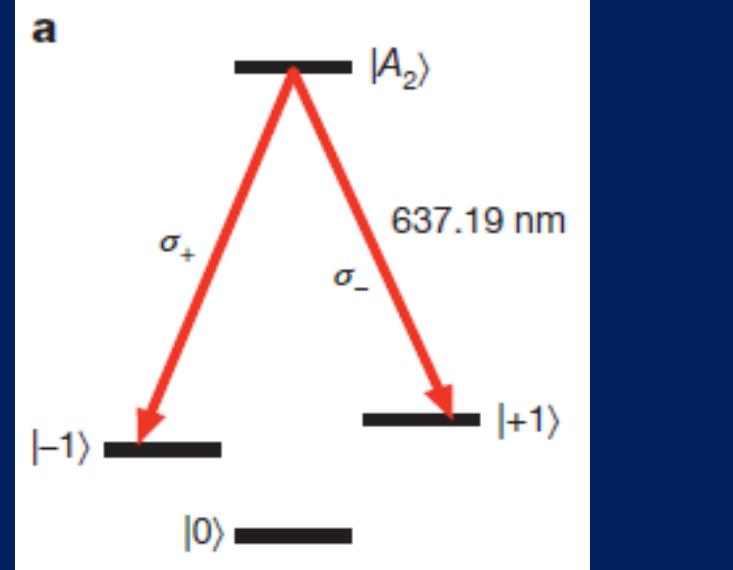
Previous and Recent topics in NV center (4)

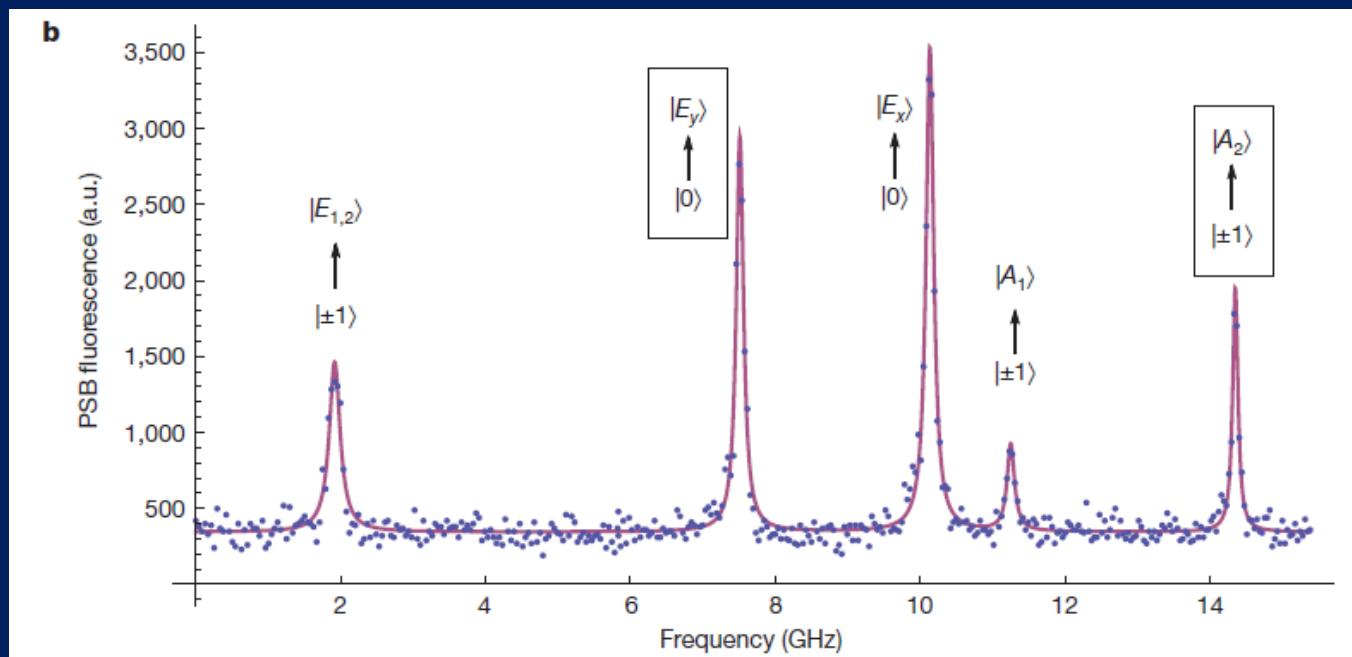
Entanglement between spin and photon (Low temp.)

Quantum entanglement between an optical photon and a solid-state spin qubit

E. Togan^{1*}, Y. Chu^{1*}, A. S. Trifonov¹, L. Jiang^{1,2,3}, J. Maze¹, L. Childress^{1,4}, M. V. G. Dutt^{1,5}, A. S. Sørensen⁶, P. R. Hemmer⁷, A. S. Zibrov¹ & M. D. Lukin¹

Nature 2010



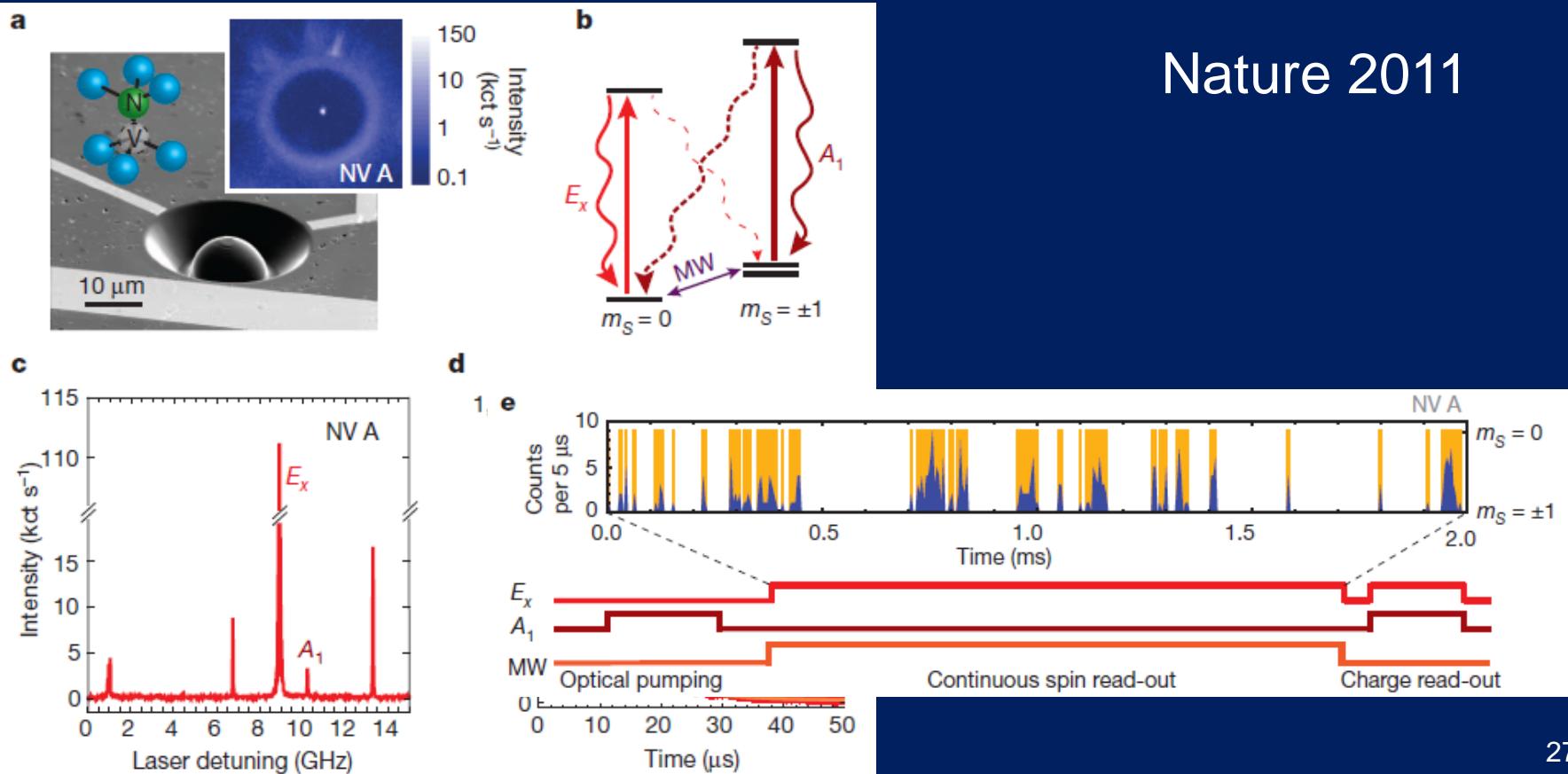


Previous and Recent topics in NV center (5)

QND measurement, Projective measurement of a single electron spin (Low temp.)

High-fidelity projective read-out of a solid-state spin quantum register

Lucio Robledo^{1*}, Lilian Childress^{2*}, Hannes Bernien^{1*}, Bas Hensen¹, Paul F. A. Alkemade¹ & Ronald Hanson¹



Previous and Recent topics in NV center (6)

Two-photon quantum interference (Low temp.)

Two-Photon Quantum Interference from Separate Nitrogen Vacancy Centers in Diamond

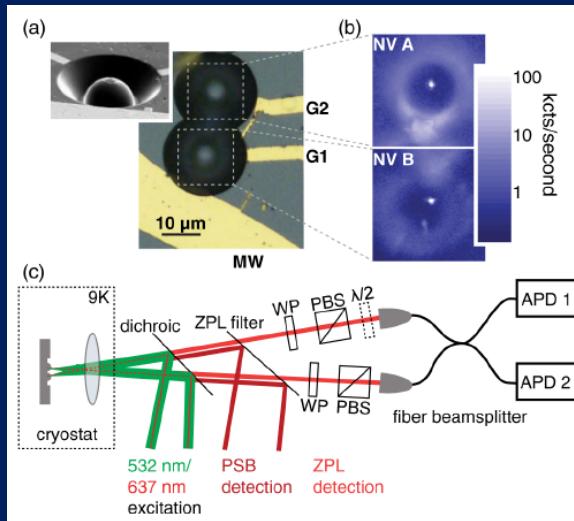
Hannes Bernien,^{1,*} Lilian Childress,² Lucio Robledo,¹ Matthew Markham,³ Daniel Twitchen,³ and Ronald Hanson¹

¹*Kavli Institute of Nanoscience Delft, Delft University of Technology, PO Box 5046, 2600 GA Delft, The Netherlands*

²*Department of Physics and Astronomy, Bates College, 44 Campus Avenue, Lewiston, Maine 04240, USA*

³*Element Six, Ltd., Kings Ride Park, Ascot, Berkshire SL5 8BP, United Kingdom*

(Received 14 October 2011; published 26 January 2012)



Quantum Interference of Single Photons from Remote Nitrogen-Vacancy Centers in Diamond

A. Sipahigil,¹ M. L. Goldman,¹ E. Togan,¹ Y. Chu,¹ M. Markham,² D. J. Twitchen,² A. S. Zibrov,¹ A. Kubanek,^{1,*} and M. D. Lukin¹

¹*Department of Physics, Harvard University, Cambridge, Massachusetts 02138, USA*

²*Element Six Ltd, Kings Ride Park, Ascot SL5 8BP, United Kingdom*

(Received 17 December 2011; published 3 April 2012)

Previous our researches

Nuclear spin

Prototype systems for engineering quantum states
Small interaction with environment

- Long coherence time at RT.
- ✗ It is not easy to access.

In our research



Optical detection and manipulation of single nuclear spins through Single NV center in diamond

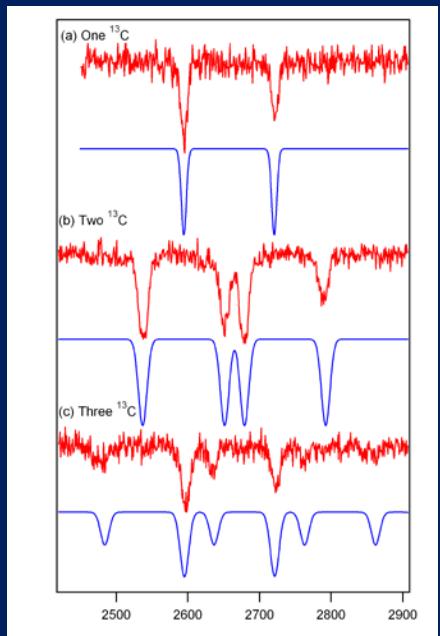
^{13}C ($I=1/2$) conc. Natural abundance = 1.1 %

Multi-qubits system by ^{13}C enriched high quality diamond

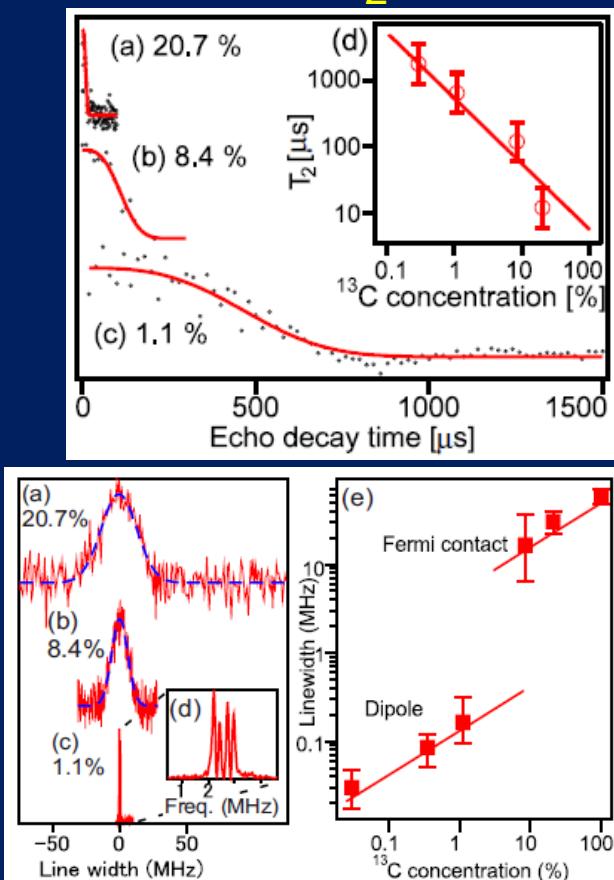
Effects on coherence time by nuclear spin

Previous our researches

Multi-qubit systems by ^{13}C nuclear spin

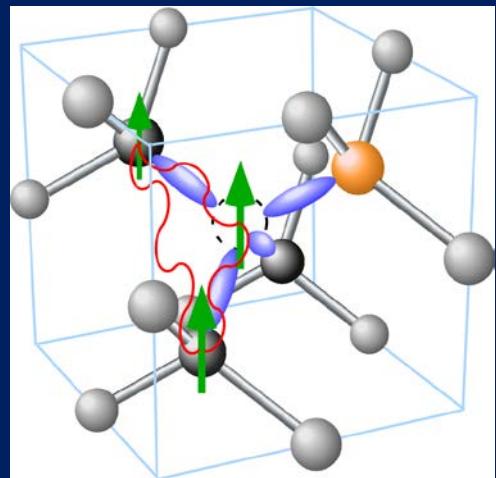


Elucidation and control of T_2 and T_{2^*}



N. Mizuuchi, et al., *PRB* 2009 (Editors' suggestion)
G. Balasubramanian, et al., *Nature materials*, 2009.

Multipartite entanglement among single spins in Diamond



Entanglement of 3 qubit :
Largest number in solid state!

P. Neumann*, N. Mizuuchi*, et
al., *Science*, (2008)

T_2 of electron spin of NV in natural abundance diamond at RT

T_2 (electron spin)

2003	50 μ s	T. A. Kennedy, et al., <i>APL</i> , 2003
2006	200 μ s	L. Childress, et al., <i>Science</i> , 2006
	350 μ s	T. Gaebel, et al., <i>Nature physics</i> , 2006
2009	700 μ s	N. Mizuuchi, et al., <i>PRB</i> , 2009

T_2 of electron spin is increasing development of diamond growth technique. It was dominated by electron spin of impurities and/or defects.

In ^{12}C enriched diamond, $T_2 = 1.8$ ms

G. Balasubramanian, et al., *Nature materials*, 2009.
N. Mizuuchi, et al., *PRB* 2009 (Editors' suggestion)

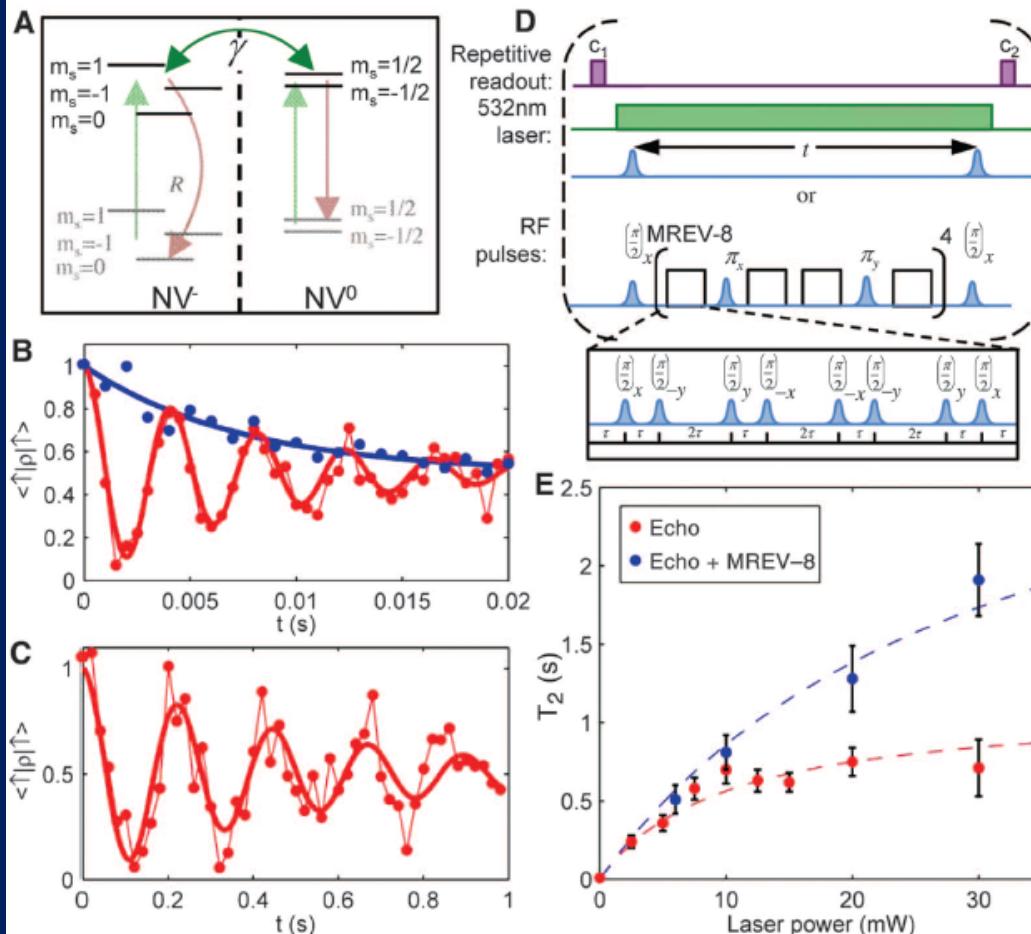
Previous and Recent topics in NV center (7)

Long coherence time (>1 s) of single nuclear spin at RT

Room-Temperature Quantum Bit Memory Exceeding One Second

Nature 2012

P. C. Maurer,^{1*} G. Kucsko,^{1*} C. Latta,¹ L. Jiang,² N. Y. Yao,¹ S. D. Bennett,¹ F. Pastawski,³ D. Hunger,³ N. Chisholm,⁴ M. Markham,⁵ D. J. Twitchen,⁵ J. I. Cirac,³ M. D. Lukin^{1†}



Ionization and deionization



When $\gamma \gg$ Hyperfine int.

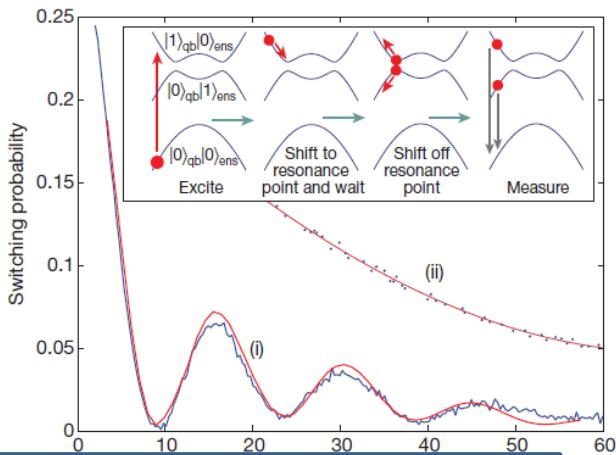
Interaction between electron and nuclear spins is strongly suppressed.

**“Electrically driven single photon source
at room temperature in diamond”**

Nature Photonics, 2012
Collaboration with AIST and Stuttgart Univ.

Coherent coupling of a superconducting flux qubit to an electron spin ensemble in diamond

Xiaobo Zhu¹, Shiro Saito¹, Alexander Kemp¹, Kosuke Kakuyanagi¹, Shin-ichi Karimoto¹, Hayato Nakano¹, William J. Munro¹, Yasuhiro Tokura¹, Mark S. Everitt², Kae Nemoto², Makoto Kasu¹, Norikazu Mizuochi^{3,4} & Kouichi Sembai¹

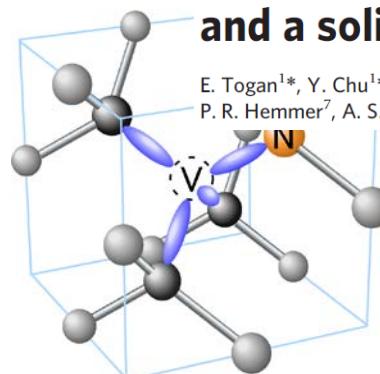


Nature 2011
(Collaboration with NTT
and NII)



Nature Materials 2009,
PRB 2009

Quantum entanglement between an optical photon and a solid-state spin qubit



E. Togan^{1*}, Y. Chu^{1*}, A. S. Trifonov¹, L. Jiang^{1,2,3}, J. Maze¹, L. Childress^{1,4}, M. V. G. Dutt^{1,5}, A. S. Sørensen⁶, P. R. Hemmer⁷, A. S. Zibrov¹ & M. D. Lukin¹

$$|\Psi\rangle = \frac{1}{\sqrt{2}} (|\sigma_-\rangle|+1\rangle + |\sigma_+\rangle|-1\rangle)$$

Nature 2011, Harvard

Quantum

Electrically driven single-photon source at room temperature in diamond

Nature Photonics 6, 299-303, 2012

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Quantum interface among single spin, photon, and charges
In NV center, field of electrical control is frontier.

Toward electrical control of qubit

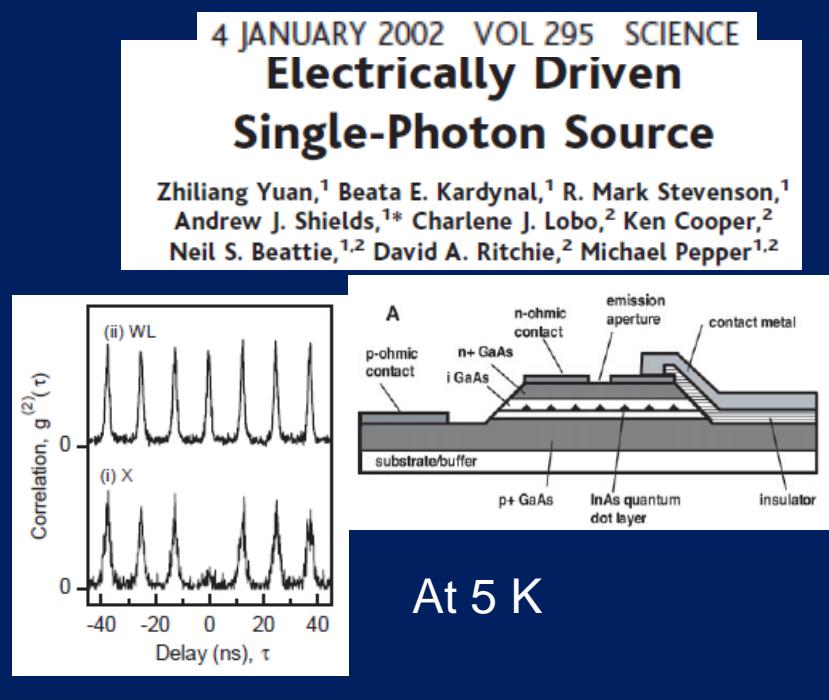
Electrical control is interested in the points...

- Electrically driven single photon source @RT
- Electrical control of spin and photon: Can optical excitation, detection, and manipulation be replaced by electrical ones in NV?
- Parallel, independent Local operation within optical diffraction limit. (impossible only in optical operation)
- Integration into on-chip quantum devices.

Single photon source by QD

See, C. Santori, D. Fattal, Y. Yamamoto,
"Single-photon Devices and Applications (Wiley, 2010)

Electrically Driven Single photon Source by quantum dot at 5K

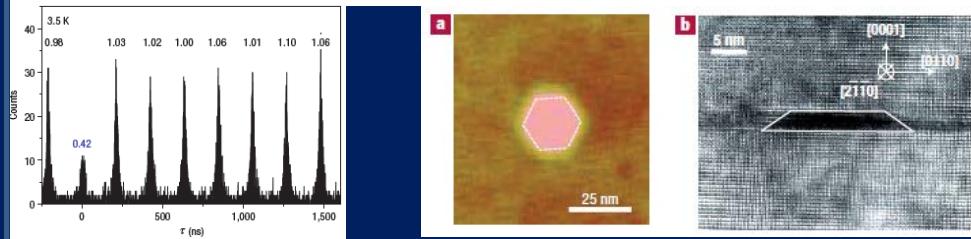


In quantum dot at 200K, PL (not EL)

Nature Materials, 5, 887 2006

A gallium nitride single-photon source operating at 200 K

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In quantum dot (CdSe) at RT, PL (not EL)

Quantum correlation among photons from a single quantum dot at room temperature

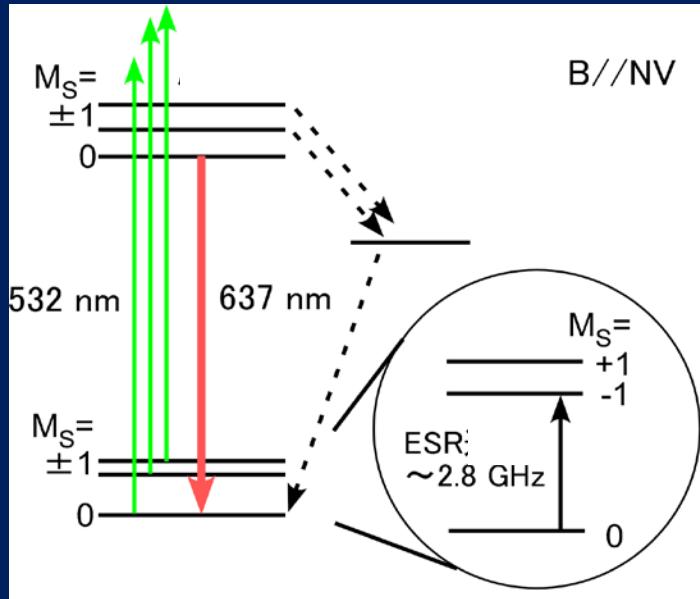
Nature
2000

P. Michler*, A. Imamoğlu*, M. D. Mason†, P. J. Carson†, G. F. Strouse† & S. K. Buratto†

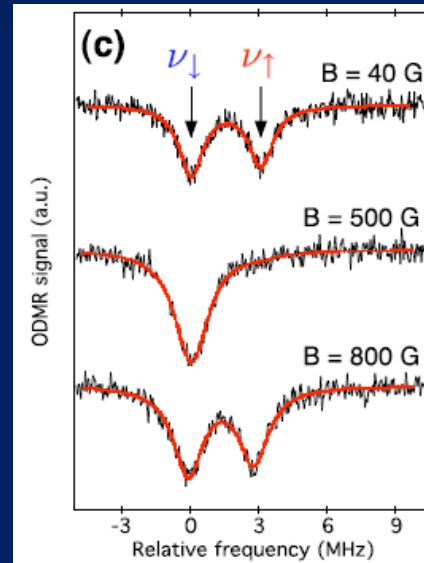
Requirement of cryogenic temperatures due to the necessity to confine carriers within dot
The electrically driven single photon source at RT: Challenging

Polarization of spin (= control of spin state) by optical pumping

Electron spin



Nuclear spin



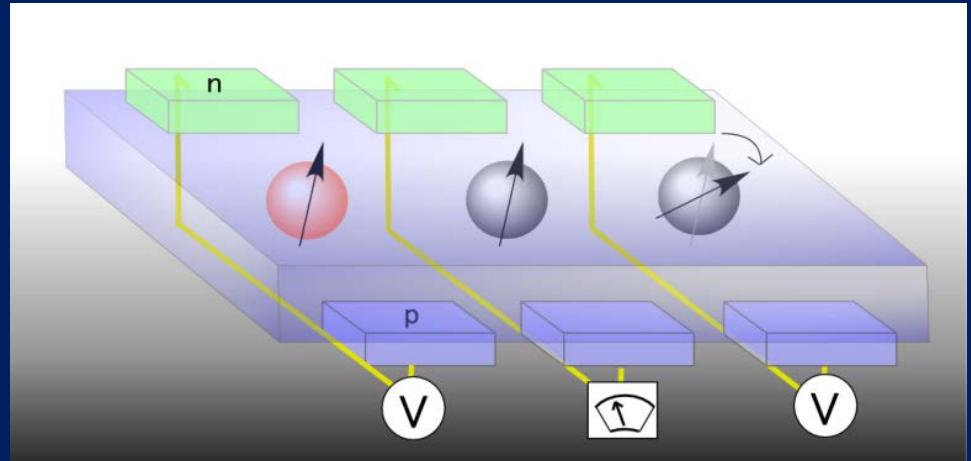
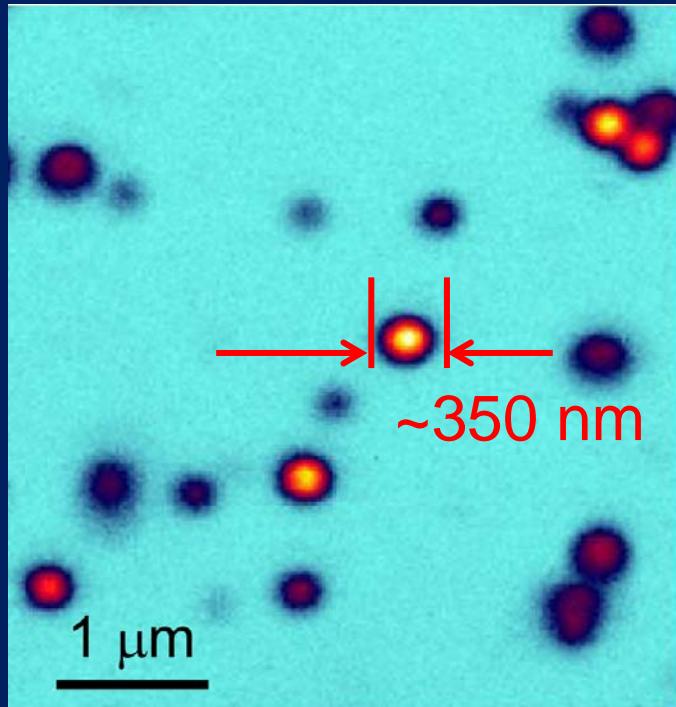
By spin selective deactivation due to SOC.

By mixing of the excited states
V. Jacques et al., *PRL*, 102, 057403 (2009)

Nuclear spin polarization > 98%

These control can be realized by electrical excitation in principle.

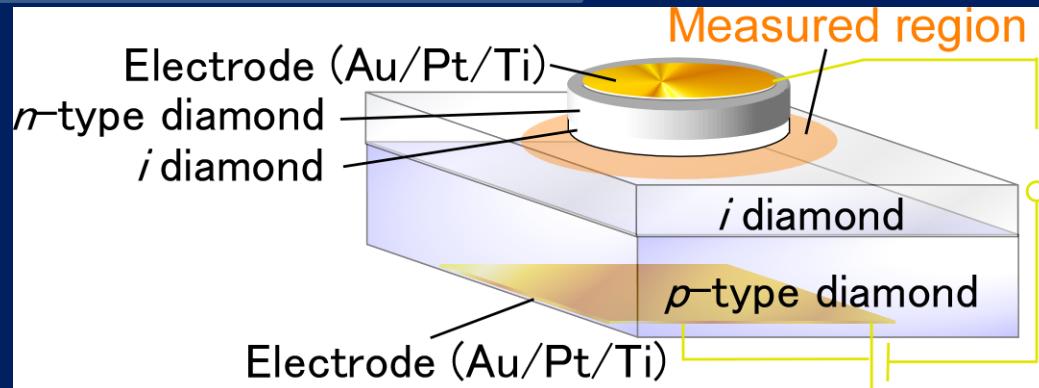
Toward parallel and independent local operation within optical resolution



Importance for quantum information processing in principle
and for integration into elaborated device

Device structure

Overview of procedure



Dopings of P and B cause color centers, so we introduced ultra-pure i-layer ($<10^4/\text{cm}^2$, $<10^7/\text{cm}^3$)

Mesa structure was fabricated to detect NV center in i-layer

N was doped by ion-implantation because of extremely small number of native NV center in i-layer

Electrode (Au/Pt/Ti) was deposited.

Synthesis condition

PIN

p-type Sub, $[B] \sim 10^{19} \text{ cm}^{-3}$

(001) substrate

l : 10mm

n : $[P] \sim 10^{18} \text{ cm}^{-3}$ ($\sim 500 \text{ nm}$)

Electrode:

Top: f200nm dots, Ti(30nm)/Pt(30nm)/Au(100nm)

Bottom:

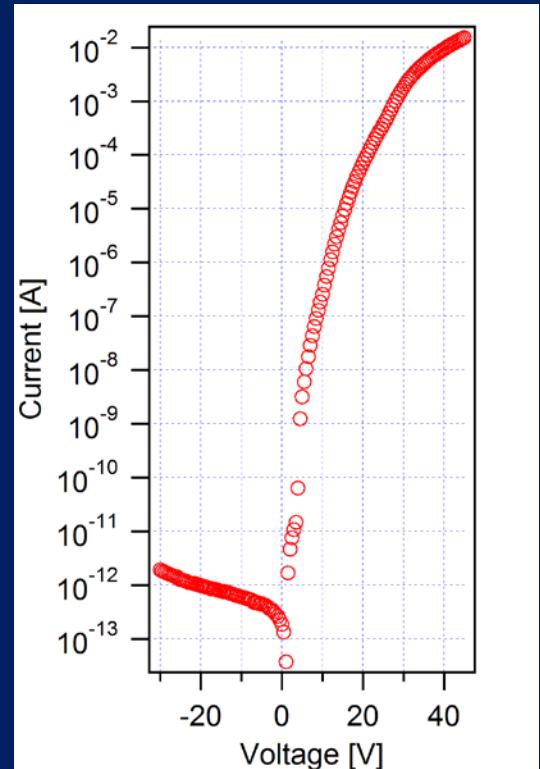
1.5mmx1.5mm Square, Ti(30nm)/Pt(30nm)/Au(100nm)

Hydrogenation, hot filament.

Plasma treatment (Oxygen) to clean the surface

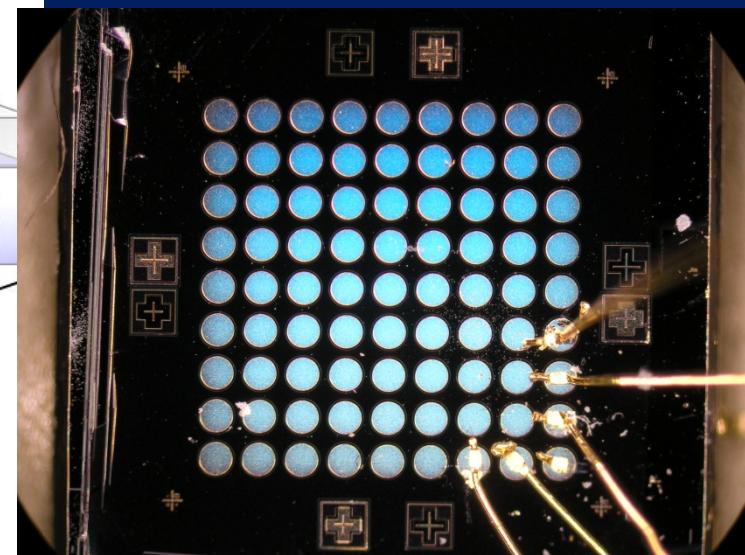
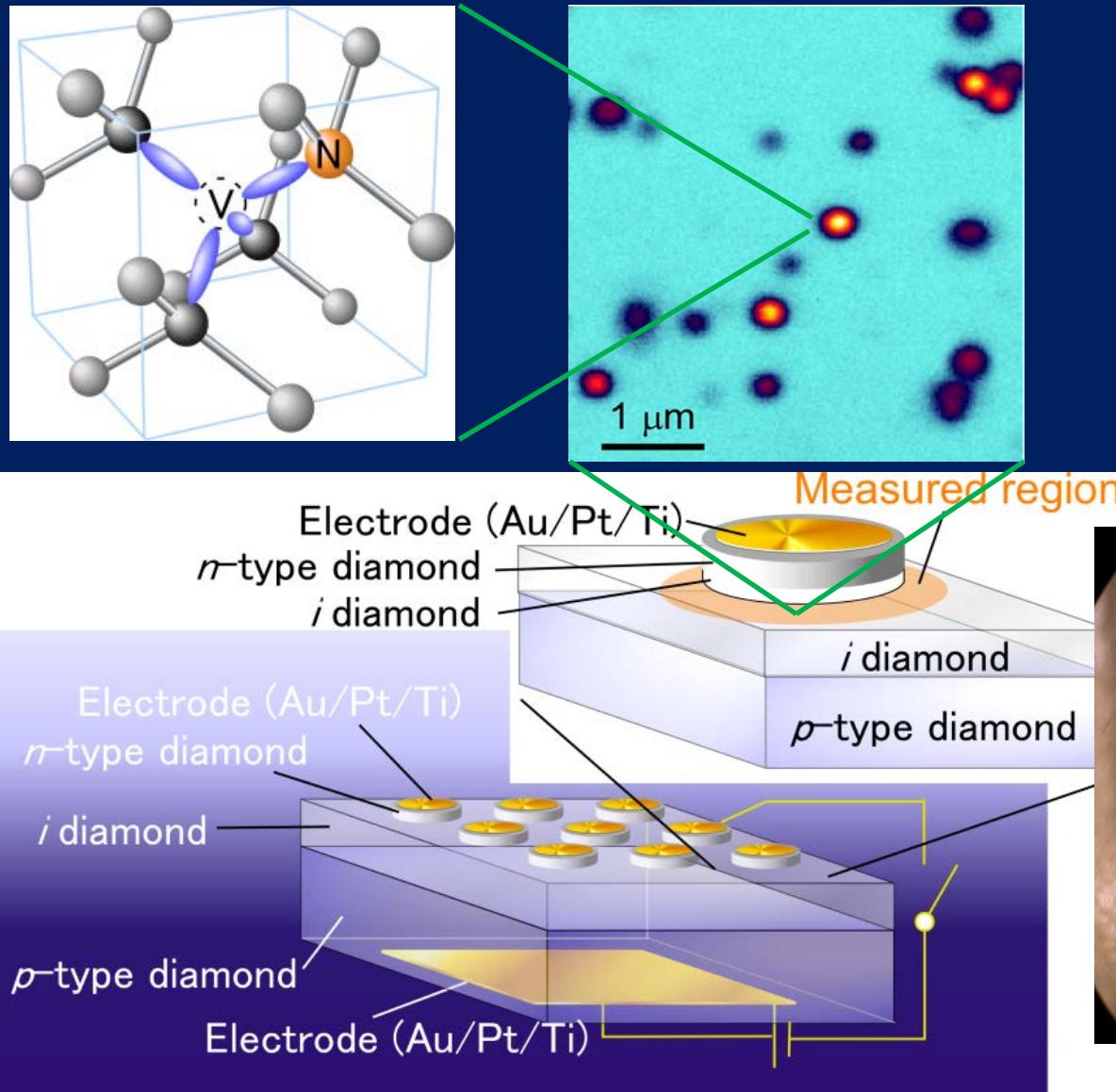
Post Annealing: 420°C, Ar, 30min

Nitrogen was ion-implanted after depositing N film.
 $(180 \text{ keV}, 1 \times 10^9 \text{ atom/cm}^2)$



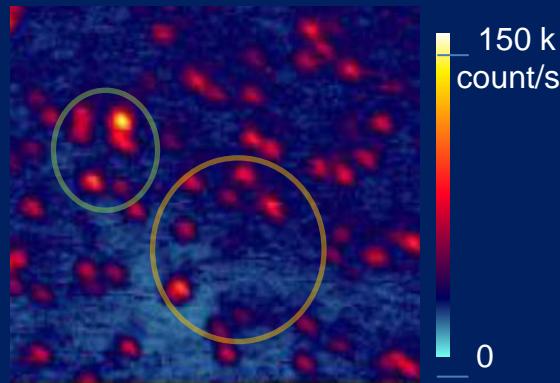
IV characteristic
(rectification ratio of
about 10^9 at 30V)

Device structure

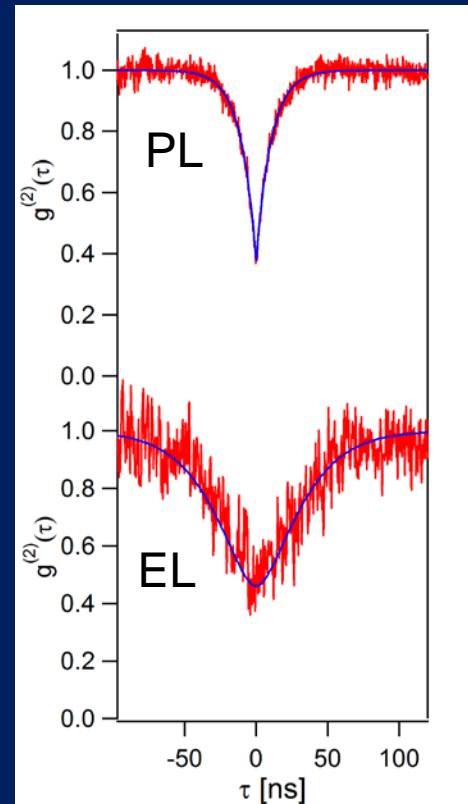
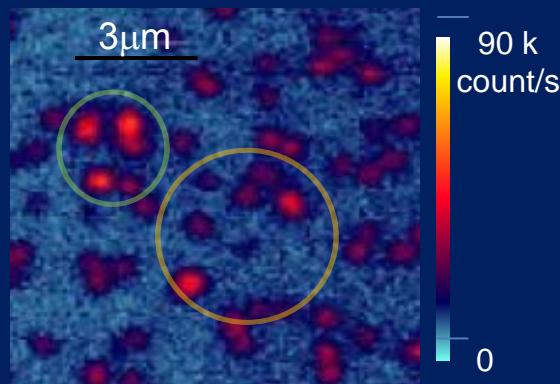


Results

PL image



EL image

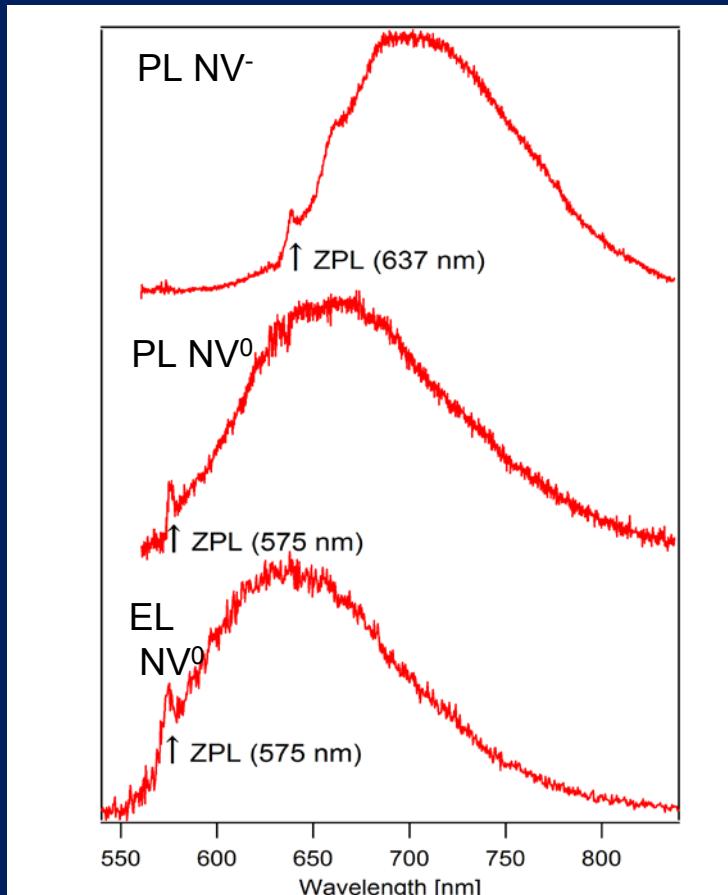


EL from the same region as PL!

The first observation of EL of single quantum system
and photon statistics at room temperature !

See also researches submitted after our submission. (1) diamond LED (APL , 99, 251106, 2011), (2) Organic LED (Nature Comm. 2012, doi 10.1038)

Spectra and charge state



Oxidized surface
PL: NV⁻ (ZPL: 637 nm)

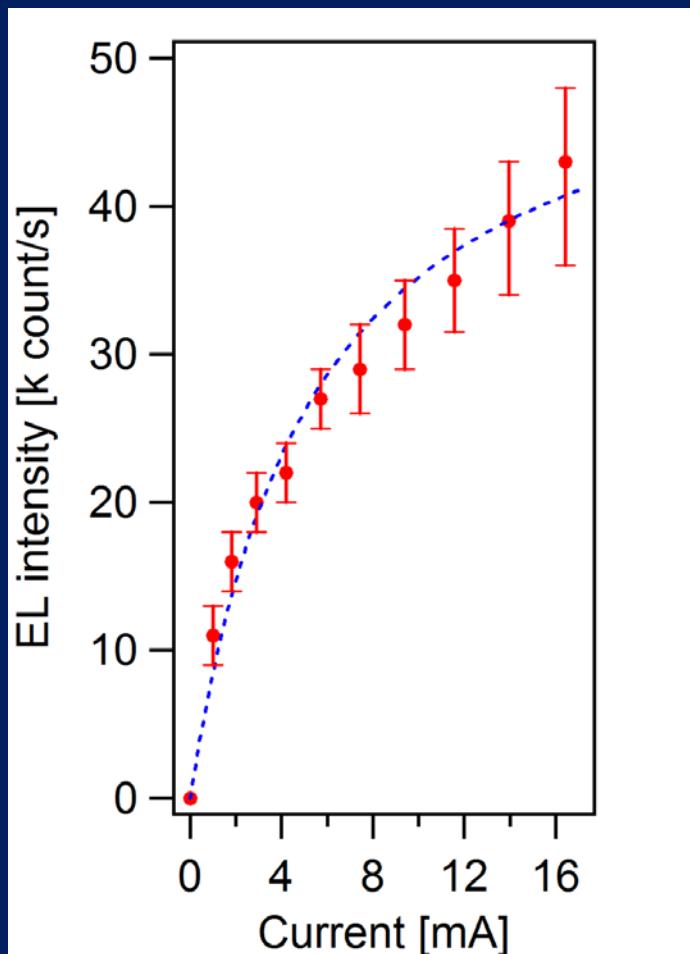
Hydrogenated surface
PL: NV⁰ (ZPL: 575 nm)

EL: NV⁰ (ZPL : 575 nm)

EL was observed from NV⁰.

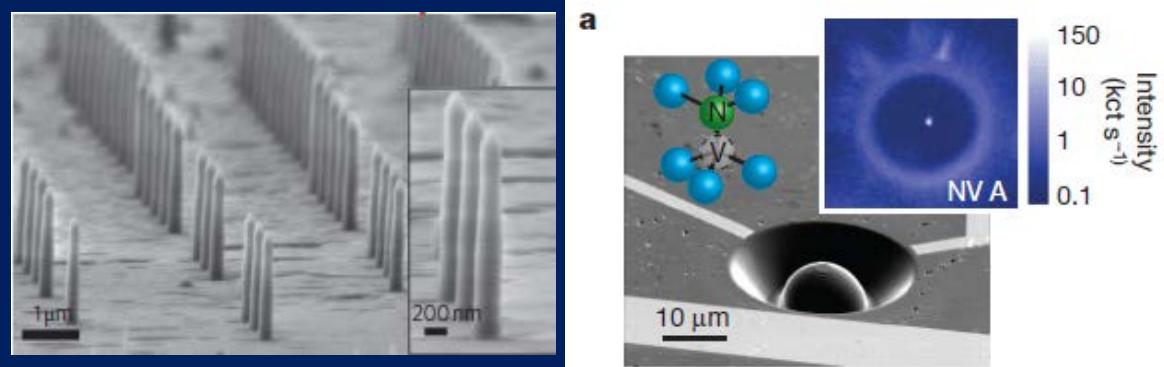
A significant number of holes due to the higher concentration of B ($\sim 10^{19}/\text{cm}^3$) compared to that of P ($\sim 10^{18}/\text{cm}^3$).

EL intensity



5×10^4 count/s at RT

Limit by high temperature due to high current

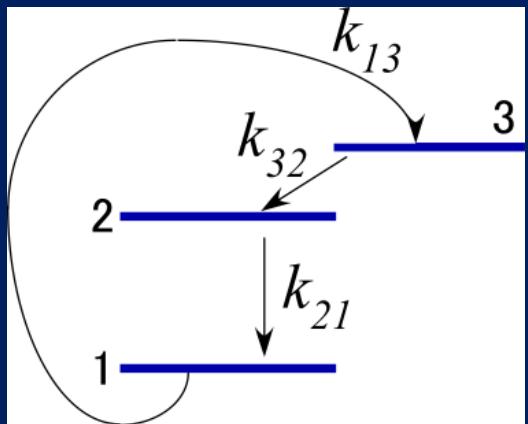


Nanowire, SIL: 10 times

By revising structures, the emission intensity will approach strongest levels (around 10^7 count/s, $T \ll RT$, Nature Photo. 2007).

Mechanism of excitation

Simulation of autocorrelation function



$$\begin{pmatrix} \dot{\rho}_1 \\ \dot{\rho}_2 \\ \dot{\rho}_3 \end{pmatrix} = \begin{pmatrix} -k_{13} & k_{21} & 0 \\ 0 & -k_{21} & k_{32} \\ k_{13} & 0 & -k_{32} \end{pmatrix} \begin{pmatrix} \rho_1 \\ \rho_2 \\ \rho_3 \end{pmatrix}$$

$$g^{(2)}(\tau) = \rho_2(\tau)/\rho_2(\infty) = 1 + c_2 e^{\lambda \tau} + c_3 e^{\gamma \tau}$$

The 3rd state :
additional rate
determining step in
consecutive reactions

P. W. Watkins, *Physical Chemistry*,
Oxford University press (1989).

$$\lambda = (-b - \sqrt{b^2 - 4c})/2, \quad \gamma = (-b + \sqrt{b^2 - 4c})/2$$

$$b = k_{13} + k_{21} + k_{32},$$

$$c = k_{13}k_{21} + k_{21}k_{32} + k_{13}k_{32}$$

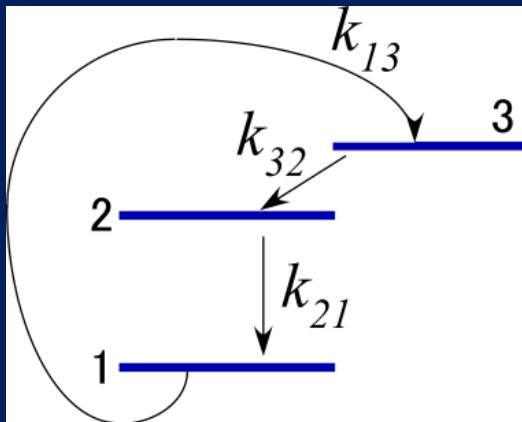
$$c_2 = (k_{13} + \lambda)(k_{32} + \lambda)(k_{32} + \gamma + k_{13})/c_1(\gamma - \lambda)\lambda k_{13}$$

$$c_3 = (k_{13} + \gamma)(k_{32} + \gamma)(k_{32} + \lambda + k_{13})/c_1(\gamma - \lambda)\gamma k_{13}$$

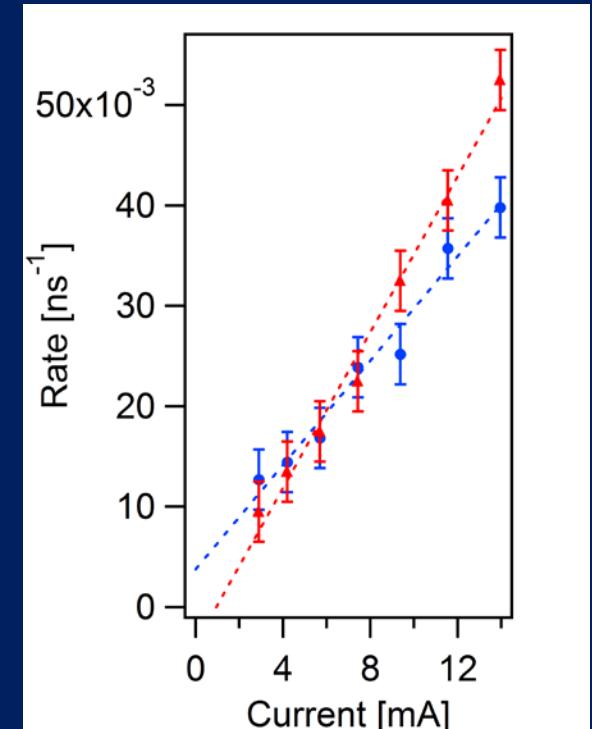
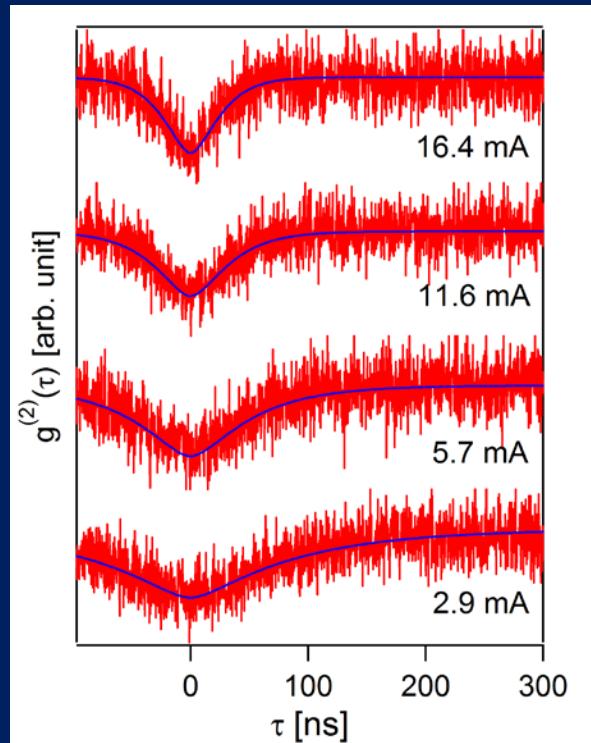
$$c_1 = -c_2 - c_3 + 1$$

Mechanism of excitation

Simulation of autocorrelation function



$$1/k_{32} = 266 \text{ ns}$$



$(k_{13})^{-1}$ at 4 mA is measured to be about 80 ns

Mechanism of excitation

Recombination rate by the Shockley-Read-Hall statistics

$$U = \sigma_n \sigma_p v_{th} N_t (pn - n_i^2) / \{ \sigma_n (n + n_i) + (p + n_i) \},$$

$$U/N_t \approx \sigma_n v_{th} \Delta n .$$

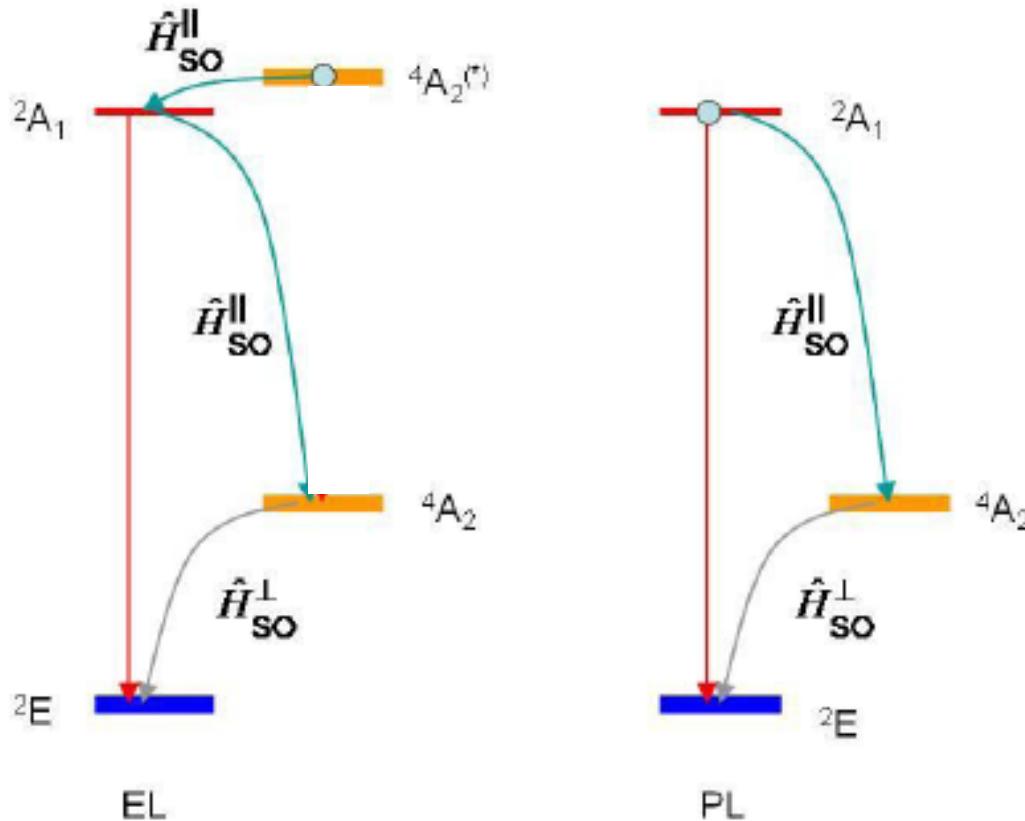
$$k_{13}^{-1} = 20 \text{ ns (4 mA)}$$

EL is generated by electron-hole recombination at the defect

NV act as interface between carrier and photon

A tentative explanation for the different kinetics of EL and PL

Group theory considerations and *ab initio* calculations for excitation energies



an $S=3/2$ excited state of a loosely bound hole and a strongly bound electron on NV⁰

The excited quartet state $^4A_2^{(*)}$ is 0.05 eV higher.

Summary

- Recent topics were presented.
- The first observation of EL of single quantum system and photon statistics at room temperature.
- It provides new function and degree of freedom with promising applications not only as single photon source but also as spintronics for quantum communication and processing.

Open Position (Post-Doctorial fellow) in our group

Thank you very much for your attention!