“Towards sensitivity and resolution enabling single-molecular detection”
LIMITATIONS:

Sensitivity
- Measurement of single spin

Resolution
- Variation of the temperature inside the cell
QUANTUM SENSING

QUANTUM PRINCIPLES:
- Measurements at \textit{nanoscale} or/and with high \textit{sensitivity}!!!
DEFINITION:
- Describes the use of a quantum system, quantum properties, or quantum phenomena to perform a measurement of a physical quantity.

1. Using quantum object
   - Electronic, magnetic or vibrational states of superconducting or spin qubits, neutral atoms, or trapped ions

2. Using quantum coherence
   - Wavelike spatial or temporal superposition states

3. Using quantum entanglement
   - Two or more sensing qubits entangled together for improved sensitivity and precision

Types 1 and 2 - close to application - nanoscale sensors
Type 3 - exploits the full power of quantum mechanics - researched

Degen C. L., Reinhard F., Cappellaro P., Reviews of Modern Physics, 2017
Attributes of quantum system:

I. It has discrete, resolvable energy levels
   - E.g. two-level system with lower \(|0\rangle\) and upper \(|1\rangle\) energy state separated by transition energy \(E = \hbar \omega_0\)

II. It is possible to initialize it to known quantum state and to readout its state

III. It can be coherently manipulated
   - Not required for all protocols - exceptions: CW spectroscopy or relaxation rate measurements

IV. It interacts with relevant physical quantity \(V(t)\)
   - This interaction is quantified by transduction parameter - \(\gamma = \partial^q E / \partial V^q\), where q is typically q = 1 (linear interaction) or q = 2 (quadratic interaction)
   - This leads to shift of the energy levels or to transitions between energy levels

Quantum sensing employs the change in the transition frequency \(\omega_0\) or the transition rate \(\Gamma\)

Sensitivity is limited by the decoherence (or relaxation) time \(T_x\)
- i.e. immunity of the quantum sensor to the external noise

\[ \text{sensitivity} \propto \frac{1}{\gamma \sqrt{T_x}} \]

\(\Rightarrow T_x\) must be made as long as possible

Degen C. L., Reinhard F., Cappellaro P., Reviews of Modern Physics, 2017
**QUANTUM SENSORS**

- Quantum sensors are one the most promising application of quantum mechanics (together with quantum computers and quantum cryptography)
- Thanks to high-sensitivity (even single spins) and nanometric resolution they are believed to bring ground-breaking discoveries (MRI of single proteins etc.)

**Sensors of:**
- Electric and magnetic fields
- Time and frequency
- Rotation
- Pressure, force
- Temperature
- Gravity
- Displacement
- Etc.

**Initialization:**
- Optical
- Electrical
- Thermal
- Etc.

**Readout:**
- Optical
- Electrical
- Pickup coil (e.g. for NMR)
- Etc.

**Possible drawbacks:**
- Specific working conditions are often required (e.g. Liquid He temperature, vacuum)
- Long acquisition times (short coherence times)
QUANTUM SENSORS

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- Thanks to high-sensitivity (even single), they are believed to bring ground-breaking developments.

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Degen et al., Rev. of Mod. Ph., 2017
- Quantum sensors are one of the most promising technologies (together with quantum computers).
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**Sensors of:**
- Electric and magnetic fields
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- Pressure, force
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- Gravity
- Displacement
- Etc.

**Possible drawbacks:**
- Specific working conditions are often required
- Long acquisition times (short coherence times)

**NV centers in diamond**
- Room temperature qubit
- Work at ambient conditions
- Easy optical initialization and readout
- Long coherence times
- Scalable (1D, 2D)
- Demonstrated sensitivity of 1 pT/√Hz

(Wolf et al., 2015)
- **MATERIAL:**
  - Synthetic diamond in form of nanodiamonds (1D) or a diamond layer (2D)

- **QUBIT:**
  - Nitrogen-vacancy (NV) center - defect in diamond lattice
- MATERIAL:
  - Synthetic diamond in form of nanodiamonds (1D) or a diamond layer (2D)

- QUBIT:
  - Nitrogen-vacancy (NV) center - defect in diamond lattice
Nitrogen containing nanodiamonds or diamond crystals (HPHT Iib diamond)

Irradiation by high energy ions - protons, electrons, ...

Creation of vacancies

By thermal annealing, the luminescent nitrogen-vacancy centres are formed

OTHER OPTIONS:
- Nitrogen implantations of pure Ila diamond
- Chemical Vapor Deposition (CVD) growth with nitrogen added to the feed gas
Defect in diamond lattice
- Two adjacent carbons replaced by Nitrogen and Vacancy

Charge states:
- $\text{NV}^+$ (4 electrons)
- $\text{NV}^0$ (5 electrons)
- $\text{NV}^-$ (6 electrons)

- $\text{NV}^0$:
  Two electrons from N, three electrons from C around the vacancy

- $\text{NV}^-$:
  1 additional electron (e.g. from electron donor defects such as $N_s^0$)

Quantum sensing
NV CENTERS IN DIAMOND

Conduction Band

Defect in diamond lattice
- Two adjacent carbons replaced by Nitrogen and Vacancy

Charge states:
- $\text{NV}^+ (4 \text{ electrons})$
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Valence Band

Schafer-Nolte, Universität Stuttgart, 2014
NV PROPERTIES

- **Photostable Single-Photon Luminescence**
  (READOUT)

- **Sensitivity** to Surrounding (e.g. Small Magnetic Fields)
  (SENSING)

- **Bio-compatibility** and **Non-toxicity**
  (BIO-APPLICATIONS)

- **Possible Surface Modification**
  (SPECIFICITY)

- **Operating at Room Temperature**
  (SIMPPLICITY)
NV LUMINESCENT PROPERTIES

Photostable Single-Photon Luminescence

Excitation:

$E_0 \rightarrow 532 \text{ nm}$

Luminescence:

$575 \text{ nm}$

$637 \text{ nm}$

Graph:

Intensity (a. u.)

Wavelength (nm)
**QUANTUM PROPERTIES OF NV⁻**

- **Sensitivity to Surrounding**
- **Electronic structure of NV⁻**

![Diagram showing electronic transitions in NV⁻](image)

- 532 nm to 637 nm transition
- Energy levels: 532 nm (GS) → 637 nm (ES)
- Magnetic sublevels: $m_s = \pm 1$
- Zero-field splitting: $D = 2.87 \text{ GHz}$
- Other properties:
  - 2 unpaired electrons
  - GS triplet (3 states)
  - Spin preserving excitation
  - Dark transition
  - Optical readout
  - Spin initialization
  - Zero-field splitting
  - MW excitation
QUANTUM PROPERTIES OF NV⁻

**Sensitivity to Surrounding**

- Electronic structure of NV⁻

**Attributes of quantum system:**

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---

**Diagram with Energy Levels**

- 532 nm
- 637 nm
- $m_s = \pm 1$
- $D = 2.87 \text{ GHz}$
- $m_s = 0$

- 2 unpaired electrons
- GS triplet (3 states)
- Spin preserving excitation
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- Spin initialization
- Zero-field splitting
- MW excitation

Bradic, Macquarie University, 2012
QUANTUM PROPERTIES OF NV⁻

**Sensitivity** to Surrounding

- **Electronic structure of NV⁻**

- 2 unpaired electrons
- GS triplet (3 states)
- Spin preserving excitation
- Dark transition
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- Zero-field splitting
- MW excitation
QUANTUM PROPERTIES OF NV

Sensitivity to Surrounding

- Optically detected magnetic resonance (ODMR)

QUANTUM PROPERTIES OF NV⁻

Sensitivity to Surrounding

- Electronic structure of NV⁻

**Magnetometry:**
The energy difference of Zeeman split is often expressed in terms of frequency shift between $m_s = -1$ and $m_s = +1$ sublevels as $\Delta = 2\gamma B$, where $\gamma$ is the electron gyromagnetic ratio and it is equal to:

$$\gamma = \frac{g_e \mu_B}{\hbar} = 2.80 \text{ MHz/G} = 28 \text{ MHz/mT}$$

($g_e$ is electron spin factor, $\mu_B$ is Bohr magneton, $\hbar$ is reduced Planck constant)
**Quantum Properties of NV**

**Sensitivity to Surrounding**

- Optically detected magnetic resonance (ODMR)

![Graph showing microwave frequency and magnetic field effects on fluorescence.](image)

QUANTUM PROPERTIES OF NV⁻
QUANTUM PROPERTIES OF NV⁻

Pham, Linh My, Harvard University, 2013
NV based sensor can act as a vector magnetometer!
QUANTUM PROPERTIES OF NV⁻

Sensitivity to Surrounding

- Electronic structure of NV⁻
QUANTUM PROPERTIES OF NV

THERMOMETRY:
The crystal field parameter $D$ depends on temperature $T$, axial electric field, and strain.

Under ambient conditions the temperature dependence is:

$$c_T = \frac{dD}{dT} = -74.2 \text{ kHz/K}$$

Neumann et al., Nano Letters, 2013
Attributes of quantum system:

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Neumann et al., Nano Letters, 2013
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Quantum sensing employs the change in the transition frequency $\omega_0$ or the transition rate $\Gamma$

$E = \hbar \omega_0$

$\Rightarrow$ PULSED MEASUREMENTS

Neumann et al., Nano Letters, 2013
PULSED MEASUREMENTS

- For single proton we need 3 nT at 10 nm
- Sensitivity can be improved either by increasing the detection efficiency or by decreasing the ODMR linewidth $\Delta \nu$
- Fundamental limit $\Delta \nu \sim 1/T_2^*$
- Separation of spin manipulation, spin readout and phase accumulation in time to prevent laser and MW power broadening

Pulsed sequence are necessary for NMR sensing of external spins!

- RF field
- NMR $\pi$ pulses
- MW pulses
- NV spin echo
- (532 nm laser)


DC magnetometry

PULSED MEASUREMENTS

\[ m_s = \pm 1 \quad \text{MW} \quad m_s = 0 \]

\[ m_s = 0 \quad m_s = \pm 1 \]

The diagram illustrates the transition of spin states with microwave (MW) pulses, showing the change from \( m_s = 0 \) to \( m_s = \pm 1 \) and vice versa.
PULSED MEASUREMENTS

BLOCH SPHERE

\[ m_s = \pm 1 \]

MW

\[ m_s = 0 \]

\[ m_s = \pm 1 \]

MW
PULSED MEASUREMENTS

Initialization \( \rightarrow \Delta \tau \rightarrow \) Detection

- LASER
- MICROWAVE
- LASER

BLOCH SPHERE

\[ |0\rangle \]

\[ |1\rangle \]

\[ x \]

\[ y \]

\( m_s = \pm 1 \)

MW

MW

\( m_s = 0 \)

\( m_s = 0 \)
PULSED MEASUREMENTS

 Initialization $\quad \Delta \tau \quad$ Detection

 Laser $\quad$ Microwave $\quad$ Laser

time

BLOCH SPHERE

\[ |0\rangle \quad |1\rangle \]

RABI OSCILLATIONS
- Exponential decay
- Spin relaxation times - $T_\chi$

MW pulse duration $\sim \tau$ ($\mu$s)

MW

$|m_s = \pm 1\rangle$

$m_s = 0$
PULSED MEASUREMENTS

Initialization $\rightarrow \tau \rightarrow$ Detection

- **LASER**
- **MICROWAVE**
- **LASER**

Attributes of quantum system:

I. It has discrete, resolvable energy levels
II. It is possible to **initialize** it to known quantum state and to **readout** its state
III. It can be **coherently manipulated**
IV. It **interacts** with relevant physical quantity $V(t)$

---

**RABI OSCILLATIONS**
- Exponential decay
- Spin relaxation times $- T_x$

---

**MW** pulse duration $\sim \tau$ (µs)

$\pi$, $\pi/2$, $2\pi$

---

$m_s = \pm 1$

---

$m_s = 0$
PULSED MEASUREMENTS

BLOCH SPHERE

Initialization

LASER

MICROWAVE

\[ \pi/2 \]

\[ \pi \]

\[ 2\pi \]

Luminescence intensity (a.u.)

- 90°
- 180°
- echo

MW pulse duration \( t \) (\( \mu s \))

\[ |0\rangle \]

\[ |1\rangle \]
PULSED MEASUREMENTS

Initialization

\[ \pi/2 \quad \tau \quad \pi/2 \]

Detection

\[ \text{LASER} \quad \text{MW} \quad \text{MW} \quad \text{LASER} \]

\[ \text{time} \]

\[ T_2^* \text{ spin DEPHASING - Ramsey sequence} \]

- External inhomogeneities
- Coupling with nearby $^{13}\text{C}$ nuclear spin
- Typical for NMR, EPR, MRI
PULSED MEASUREMENTS

\[ T_2^* = 180 \pm 30 \text{ ns} \]
PULSED MEASUREMENTS

\[ T_2^* \text{ spin DEPHASING} - \text{Ramsey sequence} \]
- External inhomogeneities
- Coupling with \(^{13}\text{C}\) nuclear spin
- Typical for NMR, EPR, MRI

\[ T_2 \text{ spin DECOHERENCE} - \text{Hahn-echo sequence} \]
- Intrinsic properties
- Spin-spin coupling
- Also called spin echo sequence
PULSED MEASUREMENTS

Initialization

LASER

Initialization

LASER

\[ T_2 = 397 \pm 5 \mu s \]
**PULSED MEASUREMENTS**

<table>
<thead>
<tr>
<th>Time</th>
<th>Initialization</th>
<th>Detection</th>
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<tbody>
<tr>
<td></td>
<td>LASER</td>
<td>π/2</td>
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<tr>
<td>π/2</td>
<td>MW</td>
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**$T_2^*$ spin DEPHASING - Ramsey sequence**
- External inhomogeneities
- Coupling with nearby $^{13}$C nuclei
- Typical for NMR, EPR, MRI

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**$T_2$ spin DECOHERENCE - Hahn-echo sequence**
- Intrinsic properties
- Spin-spin coupling
- Also called spin echo sequence

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<td>τ</td>
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</table>

**$T_1$ spin RELAXATION - Relaxation sequence**
- Interaction with lattice phonons
- Shows population decay
- Not used for magnetometry
Initialization

LASER

Initialization

LASER

Initialization

LASER

PULSED MEASUREMENTS

$T_1 = 6.0 \pm 0.3 \text{ ms}$
PULSED MEASUREMENTS

**Initialization**

- Laser
- Pulse $\pi/2$
- Delay $\tau$
- Pulse $\pi/2$

**Detection**

- Laser
- Microwave

$T_2^*$ spin DEPHASING - Ramsey sequence

- External inhomogeneities
- Coupling with nearby $^{13}$C nuclei
- Typical for NMR, EPR, MRI

$T_2$ spin DECOHERENCE - Hahn-echo sequence

- Intrinsic properties
- Spin-spin coupling
- Also called spin echo sequence

$T_1$ spin RELAXATION - Relaxation sequence

- Interaction with lattice phonons
- Shows population decay
- Not used for magnetometry

$T_1 >> T_2 >> T_2^*$
PULSED MEASUREMENTS

**DC MAGNETOMETRY**

- Initialization
- \( \pi \)
- Time \( \tau \)
- Detection
- Laser
- Microwave

**AC MAGNETOMETRY**

- Initialization
- \( \pi/2 \)
- Time \( \tau \)
- Detection
- Laser
- Microwave

**T\(_2^*\) spin DEPHASING** - Ramsey sequence
- External inhomogeneities
- Coupling with nearby \( ^{13}\)C nuclear
- Typical for NMR, EPR, MRI

**T\(_2\) spin DECOHERENCE** - Hahn-echo sequence
- Intrinsic properties
- Spin-spin coupling
- Also called spin echo sequence

**T\(_1\) spin RELAXATION** - Relaxation sequence
- Interaction with lattice phonons
- Shows population decay
- Not used for magnetometry

\[ T_1 \gg T_2 \gg T_2^* \]
DC MAGNETOMETRY

CONTINUOUS WAVE (CW) ODMR

- Easier implementation
- Lower sensitivity defined as:

ODMR resonance linewidth (limited by $1/T_2^*$ and power broadening)

$$\eta \sim \frac{h}{g_e \mu_B} \frac{\Delta f}{C_{CW} \sqrt{R}}$$

CW contrast

Photon collection rate

RAMSEY PULSE SEQUENCE

- More complicated implementation
- Higher sensitivity defined as:

$$\eta \approx \frac{\hbar}{g_e \mu_B} \frac{1}{C e^{-\left(\frac{\tau}{T_2^*}\right)} \sqrt{R t_R}} \frac{\tau}{\sqrt{t_I + \tau + t_R}}$$

Initialisation time

Precession time

Readout time

Ramsey contrast

Dephasing contrast loss

Number of collected photons

AC signal

Insensitive to AC

DC signal

Sensitive to DC

$\tau$
AC MAGNETOMETRY

HAHN-ECHO PULSE SEQUENCE

- High sensitivity to AC fields with frequency of $1/\tau$
- Single spin NMR - with additional RF field to manipulate the targeted spin

OTHER PULSING TECHNIQUES
- Multi-pulse sequences like CPMG, XY
SENSITIVITY AND RESOLUTION

EXPERIMENTAL VALUES FOR NV MAGNETOMETRY

- **SENSITIVITY:**
  - Ensembles: $\sim 1 \text{ pT/Hz}^{1/2}$, DC: $\sim 10 \text{ pT/Hz}^{1/2}$
  - Single NV: $\sim 1 \text{ nT/Hz}^{1/2}$, DC: $\sim 100 \text{ nT/Hz}^{1/2}$

- **RESOLUTION:**
  - Ensembles: $30 \text{ nm}$ (with superresolution techniques)
  - Single NV: $1 \text{ Å}$

---

**PROJECTED VALUES**

- Magnetic sensitivity vs. Spatial resolution
- Data from various technologies:
  - MRFM, Hall probe, Single electron, Single protein, Early solar system, Meteorite, GMR/TMR, SQUID, Diamond, Atomic vapor cell

Acosta et al., Optical magnetometry with nitrogen-vacancy centers in diamond, 2013
• DETECTION OF LOCALIZED CHEMICAL EVENTS BY T1 SENSING

Rendler, Neburkova et al., Nature Comm., 2017
• DETECTION OF LOCALIZED CHEMICAL EVENTS BY T1 SENSING
• QUANTUM SENSING INSIDE CELLS
  • Magnetometry - Based on ODMR detection together with quantum sensing protocols
APPLIcATIONS

NANODIAMONDS

- DETECTION OF LOCALIZED CHEMICAL EVENTS BY T1 SENSING
- QUANTUM SENSING INSIDE CELLS
  - Magnetometry
  - Thermometry

Neumann et al., Nano Letters, 2013

Kucsko et al., Nature, 2013
• DETECTION OF LOCALIZED CHEMICAL EVENTS BY T1 SENSING
• QUANTUM SENSING INSIDE CELLS
  • Magnetometry
  • Thermometry
  • Detection of short-living superoxide radicals inside cells with confocal resolution using spin-traps
  • Measuring membrane potentials (e.g. for neuronal action potential)
• And more
• NMR and MRI
SINGLE NV

Sensor for 3D MRI mapping with atomistic resolution

Direct monitoring and imaging of single molecular objects
Even resolving their atomic structure
Important new tool for drug discovery!

CONVENTIONAL COIL-BASED MRI

Insensitive to sample at nanometer scale
- $10^{12}$ - $10^{18}$ nuclei to generate observable signal[1]
- With resolution of $3 \ \mu m^2$ at its best[2]

Bigger magnets - better performance
- However the price and complexity does not scale up

Sensor for 3D MRI mapping with atomistic resolution

Direct monitoring and imaging of single molecular objects
Even resolving their atomic structure
Important new tool for drug discovery!

DIAMONDS-BASED MRI

Sensitivity \( \sim nT/Hz^{1/2} \)
- For single proton we need \( 3 \) nT at 10 nm

Atomistic resolution achievable
- Voxel volume of \((5-nm)^3\) has been demonstrated\(^{[1]}\)

NMR obtained from external single electron\(^{[2]}\) and nuclear\(^{[3]}\) spin

**APPLICATIONS**

**SINGLE NV**

Sensor for 3D MRI mapping with atomistic resolution

---

**DIAMONDS-BASED MRI**

Sensitivity $\sim nT/Hz^{1/2}$
- For single proton we need $3 \text{nT at } 10 \text{ nm}$

Atomistic resolution achievable
- Voxel volume of $(5-\text{nm})^3$ has been demonstrated\(^1\)

NMR obtained from external single electron\(^2\) and nuclear\(^3\) spin

The 3D-MRI principle
- Demonstrated recently\(^4\)
- The theoretical acquisition time: $\sim \text{weeks! (bad detection rates)}$

Novel approach:
Photoelectric detection of Magnetic Resonance (PMR)
$\sim \text{tens of minutes!}$

---

SINGLE NV

- NMR and MRI
- MAGNETIC IMAGING WITH DIAMOND AFM TIPS

Maletinsky et al., Nature Nanotechnology, 2012

NV magnetic field image of bit tracks on a magnetic memory
• NMR and MRI
• MAGNETIC IMAGING WITH DIAMOND AFM TIPS
• CAVITY QUANTUM ELECTRODYNAMICS
• And more
ENSEMBLE OF NVs

- MAGNETIC IMAGING OF LIVING CELLS

(LEFT) Bright-field optical image of magnetotactic bacteria (MTB) adhered to the diamond surface. (RIGHT) Image of magnetic field projection determined from NV ODMR.

Le Sage et al., Nature, 2013
APPLICATIONS

ENSEMBLE OF NVs

- MAGNETIC IMAGING OF LIVING CELLS
- LAB ON CHIP

Kehyias et al., Nature Comm., 2017
ENSEMBLE OF NVs

- MAGNETIC IMAGING OF LIVING CELLS
- LAB ON CHIP
- MAGNETIC IMAGING OF ANCIENT ROCK AND METEORITES
- QUANTUM ENTANGLMENT OF FEW NVs
- And more
PHOTOELECTRIC DETECTION OF MAGNETIC RESONANCES

E. Bourgeois, M. Gulka et al.,
Nature Communications, 6 (2015)
PHOTOELECTRIC DETECTION OF MAGNETIC RESONANCES


Rabi PDMR on 5 NVs:

PROS:
- EFFICIENT READOUT
- Better detection rates (300x higher compare to ODMR)
- SPATIAL RESOLUTION
- SMALL DESIGN
- ON-CHIP

CONS:
- LOWER CONTRAST
- NITROGEN BACKGROUND

THANK YOU FOR YOUR ATTENTION!