

# Quantum Techniques for Precision Measurements with NV-Diamond

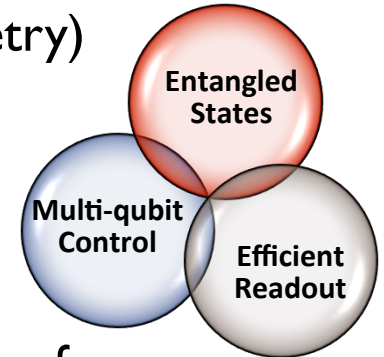
Ronald Walsworth

Harvard-Smithsonian Center for Astrophysics  
Harvard University, Department of Physics



# Outline

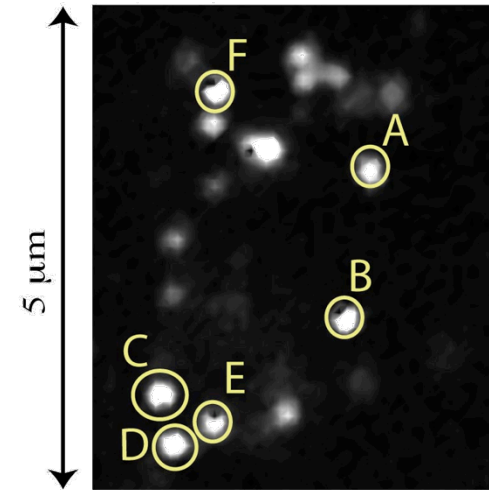
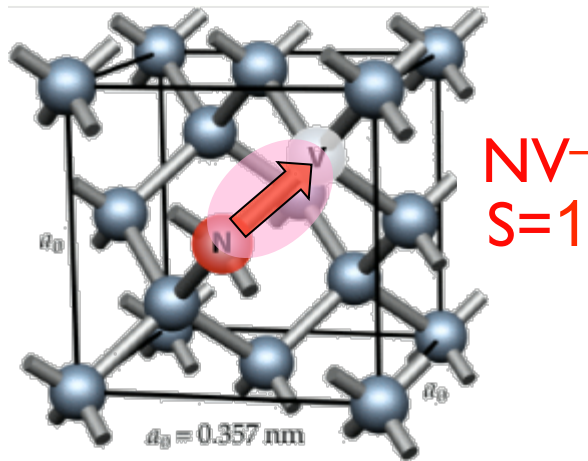
- A little background on NV-diamond physics & precision spin measurements (magnetometry)
- My group's role as part of QuISM team
- Some recent results:
  - Successful application of dynamic decoupling to wide range of NV-diamond samples =>  $T_2 > 2$  ms, improved magnetometry FOM
  - Identified suppression of electronic (N) spin-bath dynamics by nuclear ( $^{13}\text{C}$ ) spin impurities => unexpectedly long NV  $T_2$
  - Side-collection technique => NV optical collection efficiency  $> 50\%$
  - AR-coating of diamond => improved NV excitation/detection efficiency



# Nitrogen Vacancy (NV) color centers in diamond



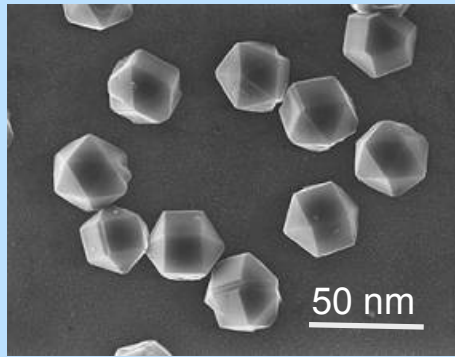
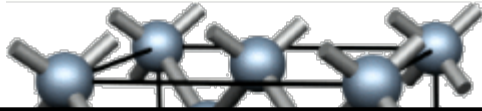
Atom-like defect with special properties:



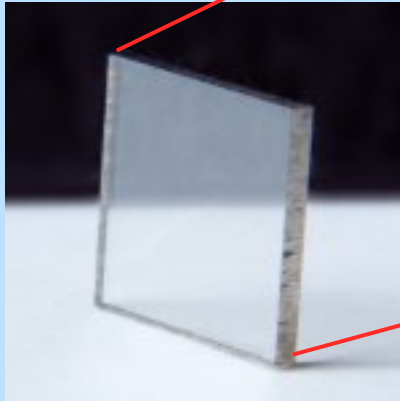
Confocal: individual NVs in diamond

- $NV^-$ : localized electronic spin state ( $S=1$ ) with  $T_2 \sim ms$ , large Zeeman shift
- Optical pumping  $\Rightarrow$  spin-state preparation
- Coherent spin manipulation with microwave pulses
- Optical read-out of spin-state
- Diamond: transparent, robust solid; stiff lattice; small spin-orbit
- NV density  $\sim 10^{12} - 10^{18} \text{ cm}^{-3}$

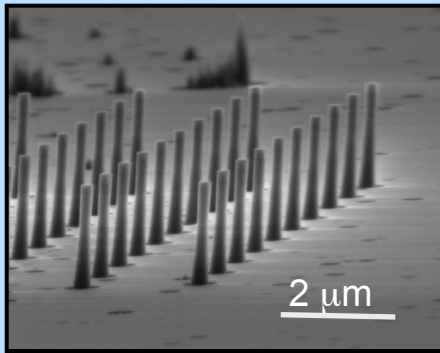
# Nitrogen Vacancy (NV) color centers in diamond



Diamond nanocrystals

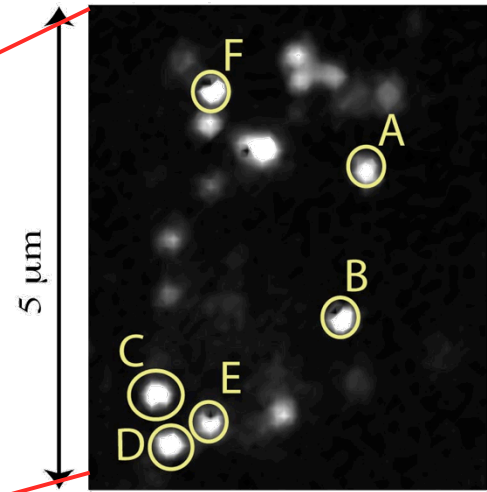


Single crystal diamond



Diamond nanopillars

NV centers created by ion implantation & annealing; also CVD techniques

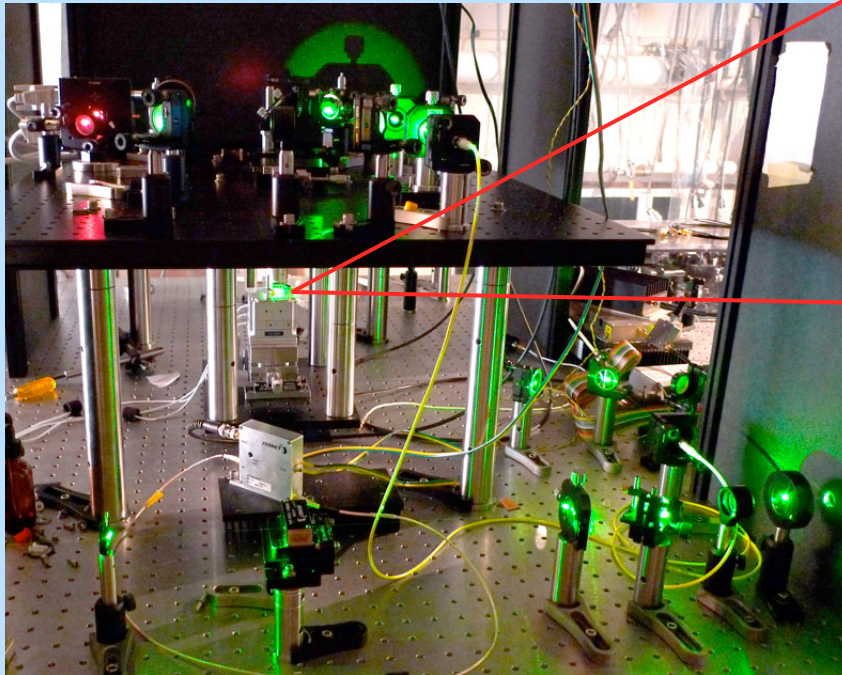


Confocal: individual NVs in diamond

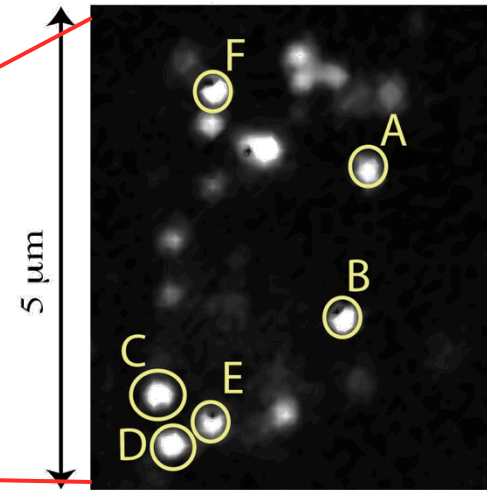
ms, large Zeeman shift

l spin-orbit

# Nitrogen Vacancy (NV) color centers in diamond



Relatively simple apparatus (confocal)

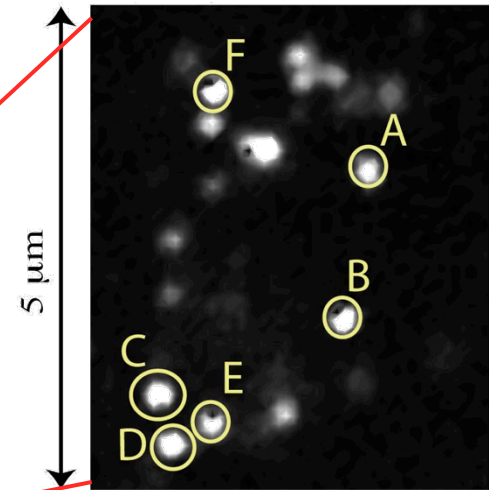
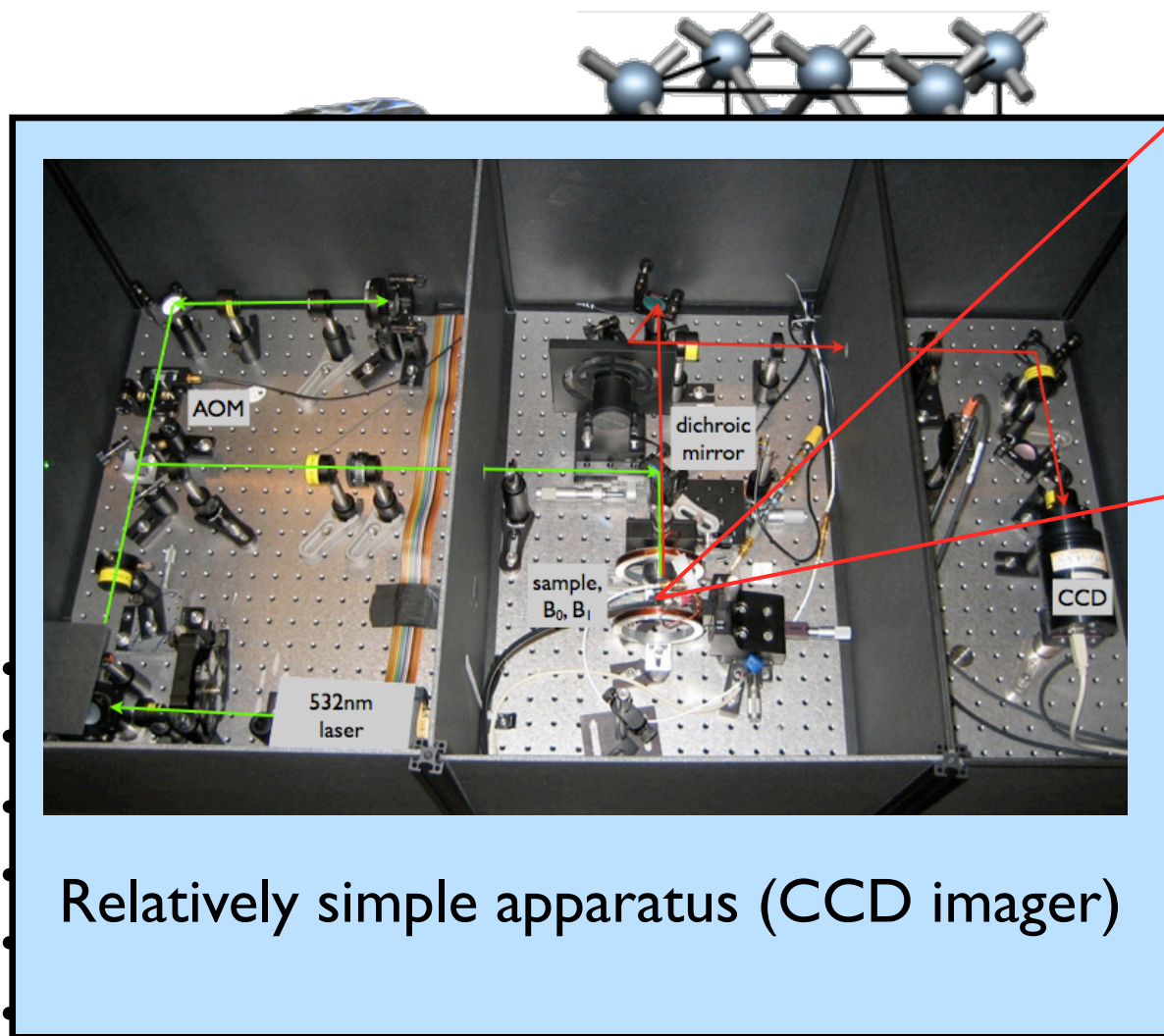


Confocal: individual NVs in diamond

ms, large Zeeman shift

l spin-orbit

# Nitrogen Vacancy (NV) color centers in diamond

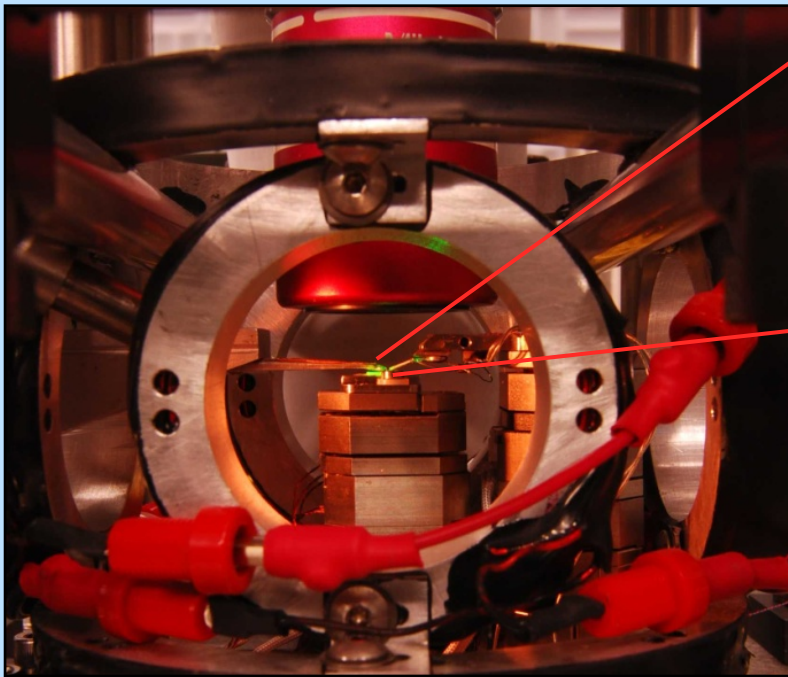
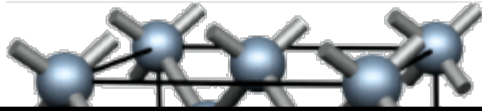


Confocal: individual NVs in diamond

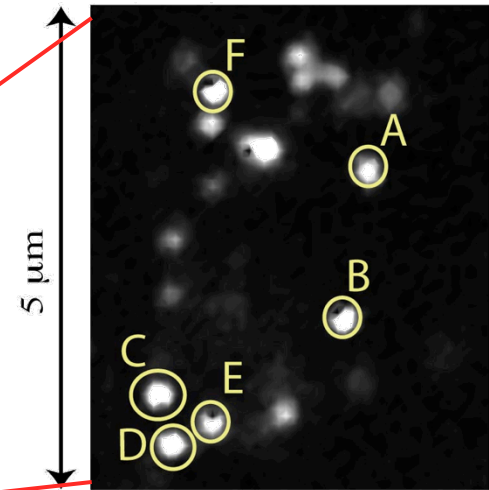
ms, large Zeeman shift

l spin-orbit

# Nitrogen Vacancy (NV) color centers in diamond



Less simple... (confocal + AFM)

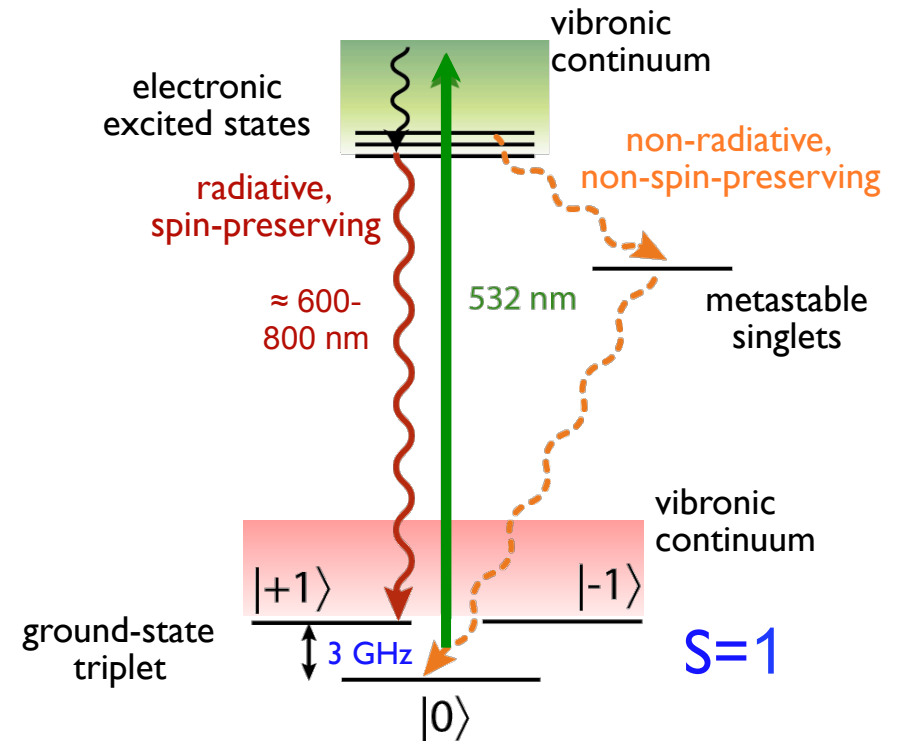


Confocal: individual NVs in diamond

ms, large Zeeman shift

l spin-orbit

# Coherent spin manipulation in NV-diamond

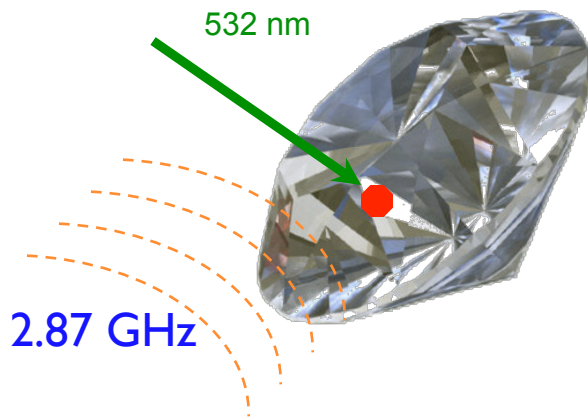
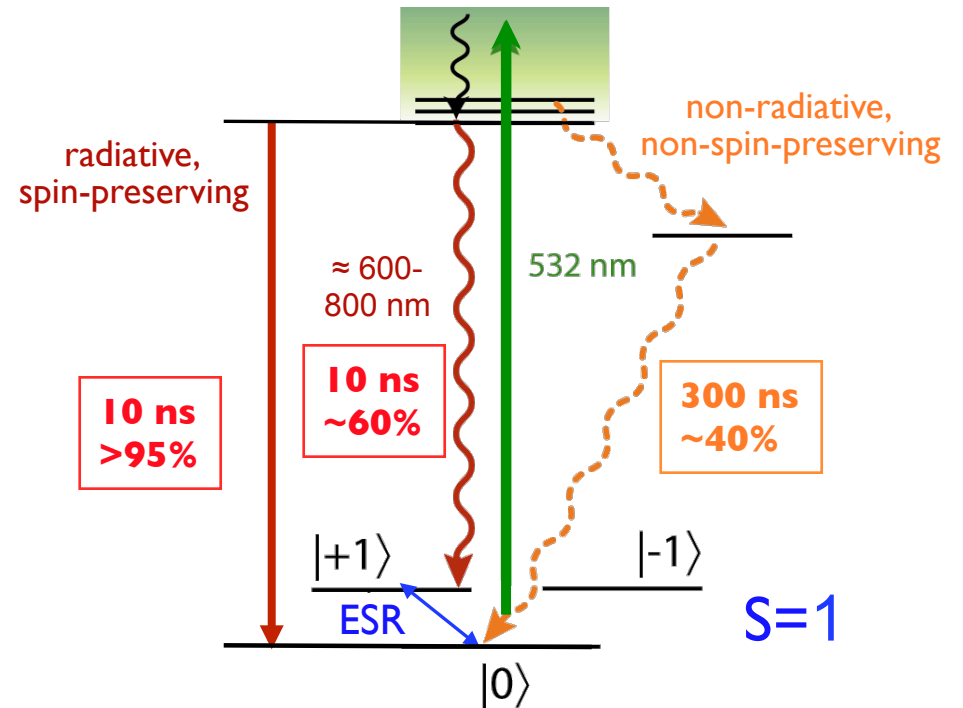


- Atom-like energy levels coupled by strong optical transitions
- Ground-state hyperfine & Zeeman structure with long ESR coherence times ( $T_2 \sim 1$  ms)
- Spin-state dependent fluorescence & decay



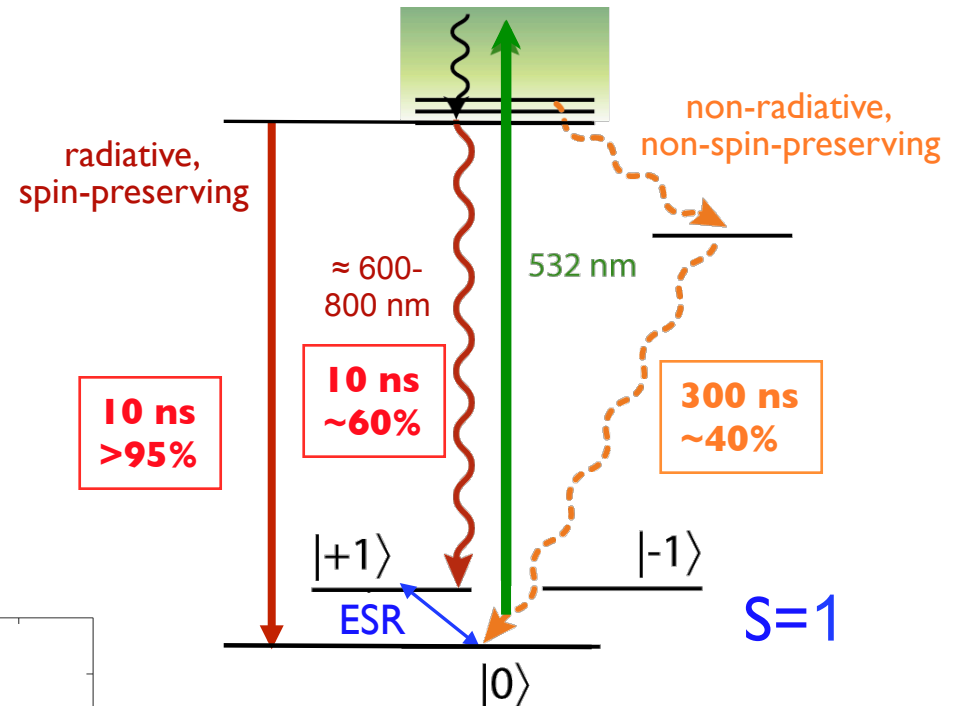
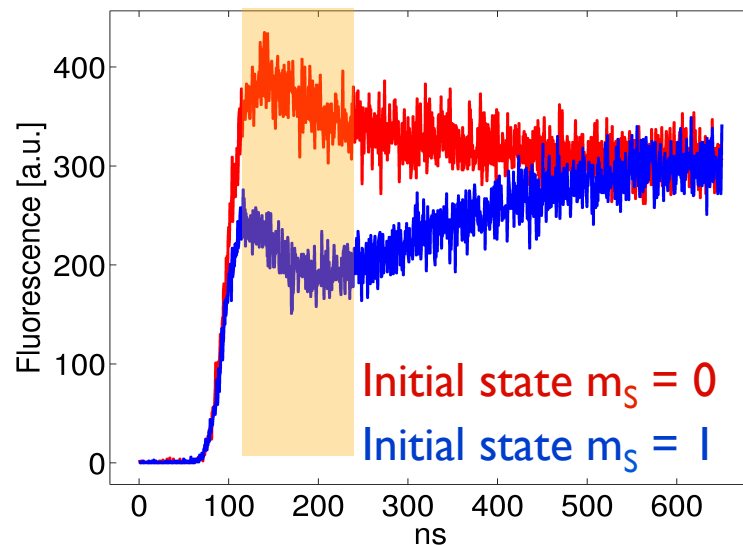
# Coherent spin manipulation in NV-diamond

- 1) Spin-polarization (into  $m_S=0$ ) by optical pumping (>95%)
- 2) Spin coherence control with ESR
- 3) Spin-state optical read-out



# Coherent spin manipulation in NV-diamond

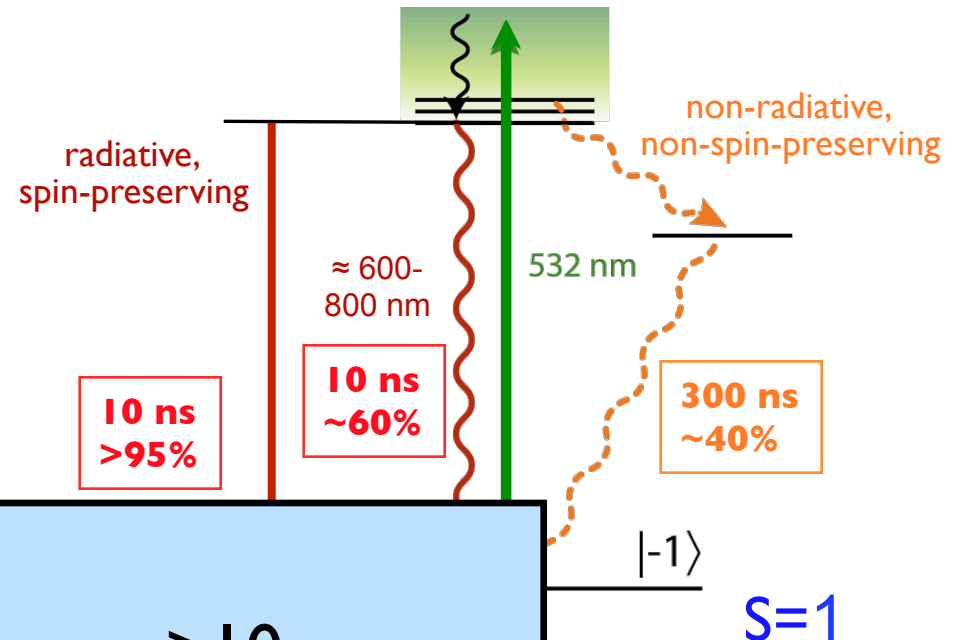
- 1) Spin-polarization (into  $m_S=0$ ) by optical pumping (>95%)
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Typical single NV fluorescence data

# Coherent spin manipulation in NV-diamond

- 1) Spin-polarization (into  $m_S=0$ ) by optical pumping (>95%)
- 2) Spin coherence control with ESR
- 3) Spin state optical read-out



Large amount of work over >10 years:

N. Manson

T. Kennedy

S. Rand

P. Grangier

F. Jelezko

J.F. Roch

J. Wrachtrup

S. Praver

P. Hemmer

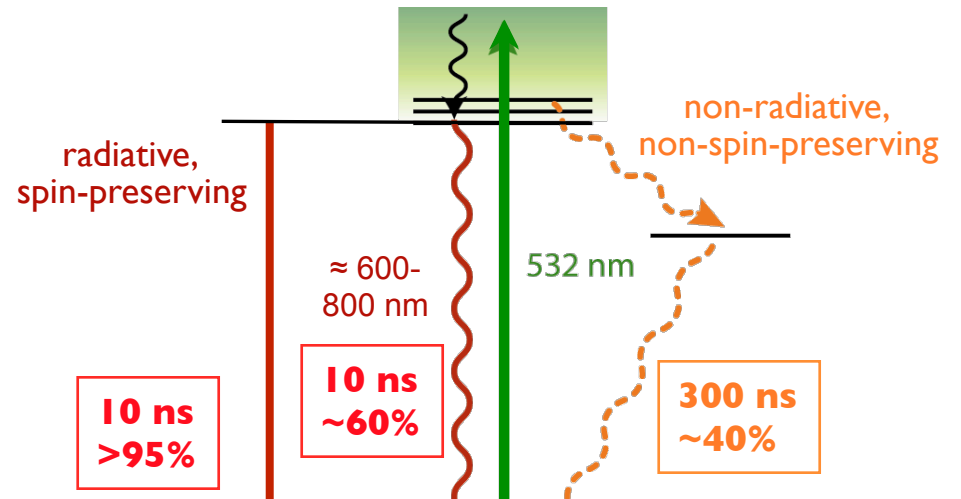
D. Awschalom

# Coherent spin manipulation in NV-diamond

1) Spin-polarization (into  $m_s=0$ )  
by optical pumping (>95%)

2) Spin coherence control  
with ESR

2) Spin state optical read-out



## Other important issues:

Composite spin environment: N,  $^{13}\text{C}$   
=> NV decoherence

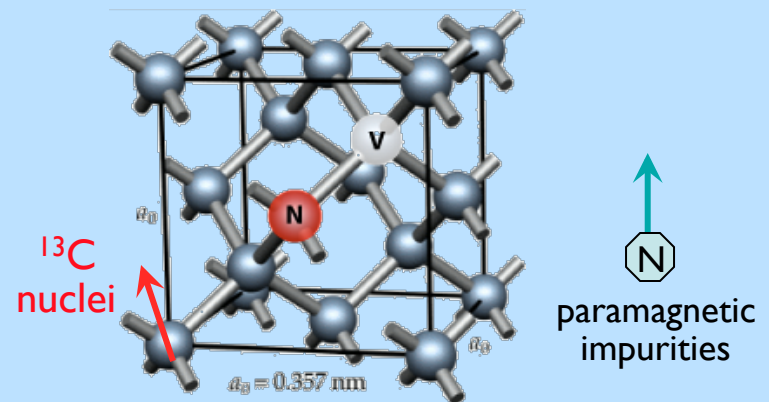
Environment-enhanced metrology?

4 crystal axes

Optical ionization to  $\text{NV}^0$

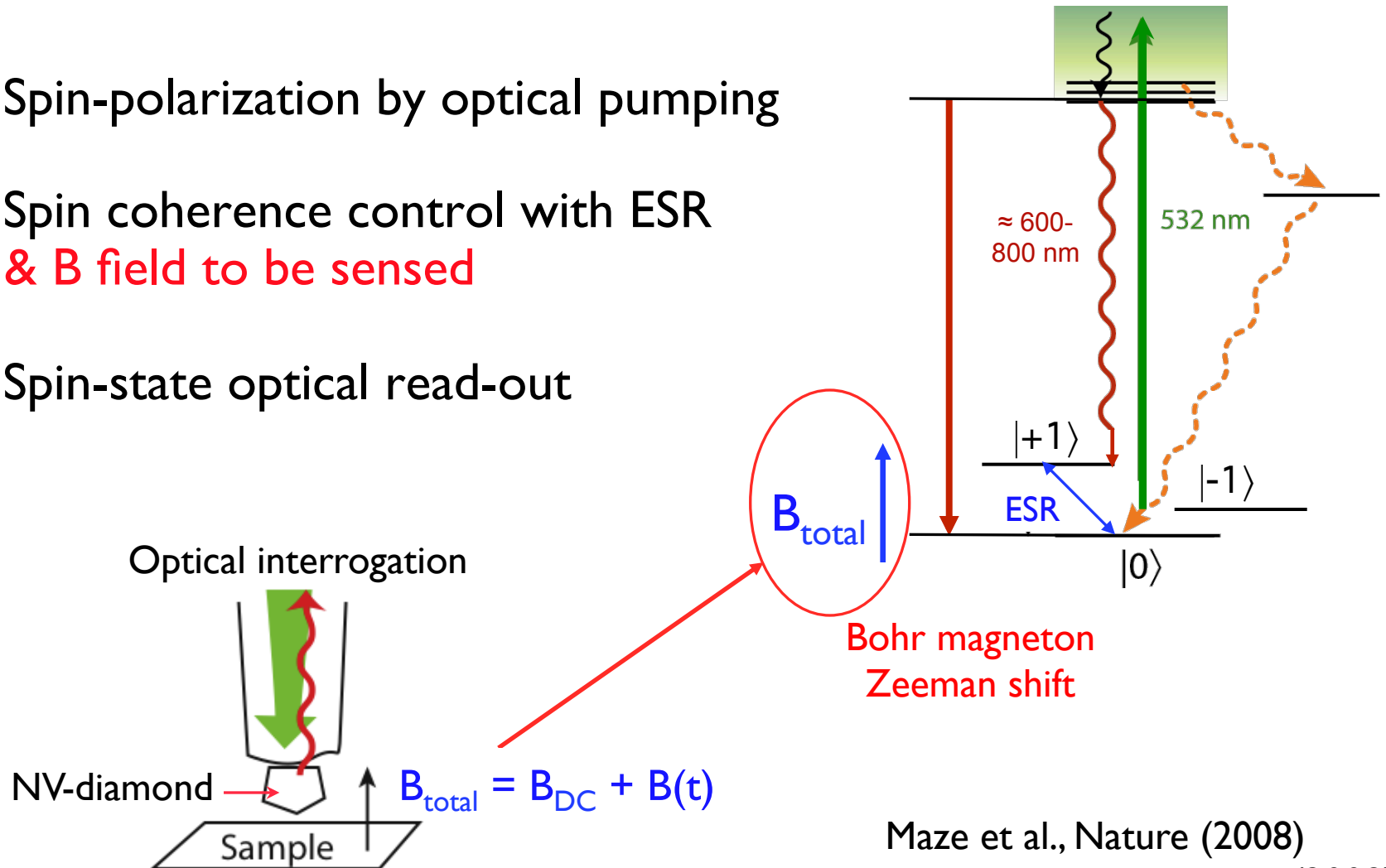
Sample geometry & fabrication

Surface chemistry



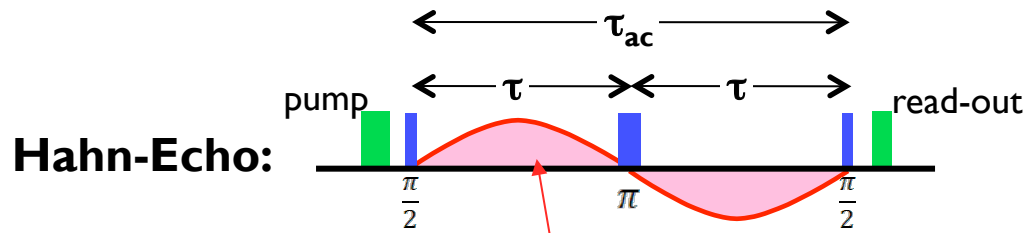
# NV-diamond magnetometry

- 1) Spin-polarization by optical pumping
- 2) Spin coherence control with ESR  
& B field to be sensed
- 3) Spin-state optical read-out



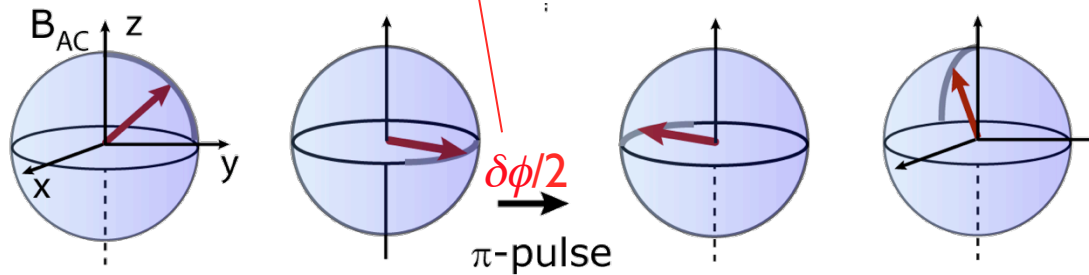
Maze et al., Nature (2008)  
Balasubramanian et al., Nature (2008)  
Taylor et al., Nature Physics (2008)

# Echo-based NV magnetometry



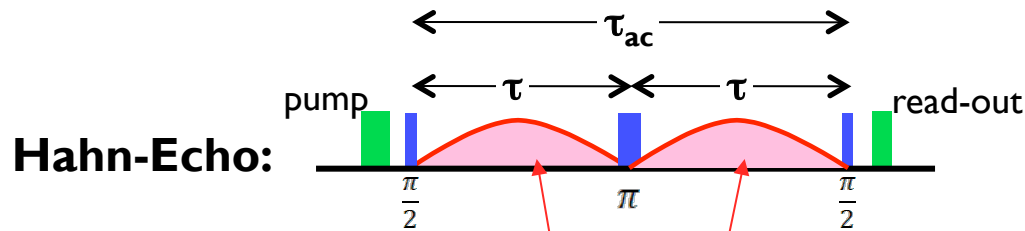
Target ac magnetic field:  

$$B(t) = B_{ac} \sin[(2\pi f_{ac})t + \phi]$$

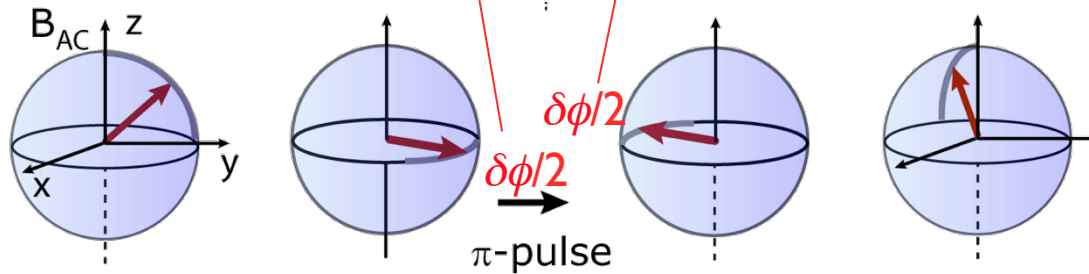


Echo also decouples NV spin from static inhomogeneities in local environment  $\Rightarrow T_2$  limit

# Echo-based NV magnetometry



Target ac magnetic field:  
 $B(t) = B_{ac} \sin[(2\pi f_{ac})t + \phi]$



**NV fluorescence signal  $\sim \delta\phi \approx \gamma_{NV} B_{ac} \tau$**

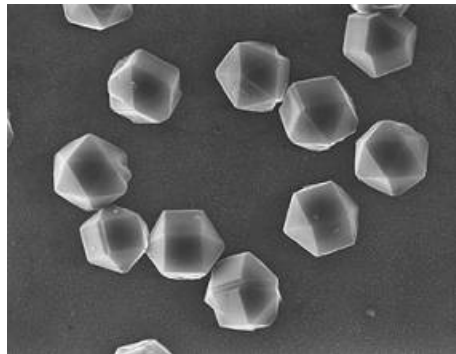
Echo also decouples NV spin from static inhomogeneities in local environment  $\Rightarrow T_2$  limit

Shot-noise sensitivity

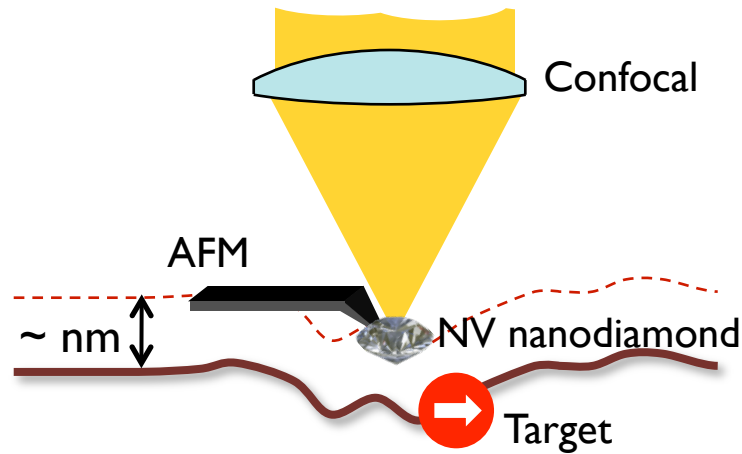
$$\frac{\delta B_{ac}}{\sqrt{\text{Hz}}} \sim \frac{1}{\gamma_e \sqrt{T_2 n}}$$

# NVs in sensor volume

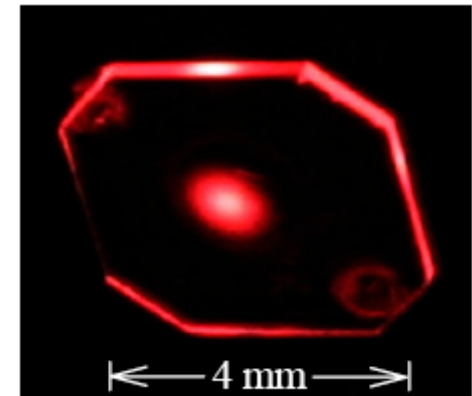
# NV-diamond magnetometry: complementary modalities



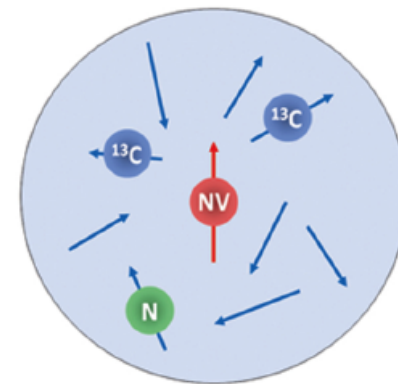
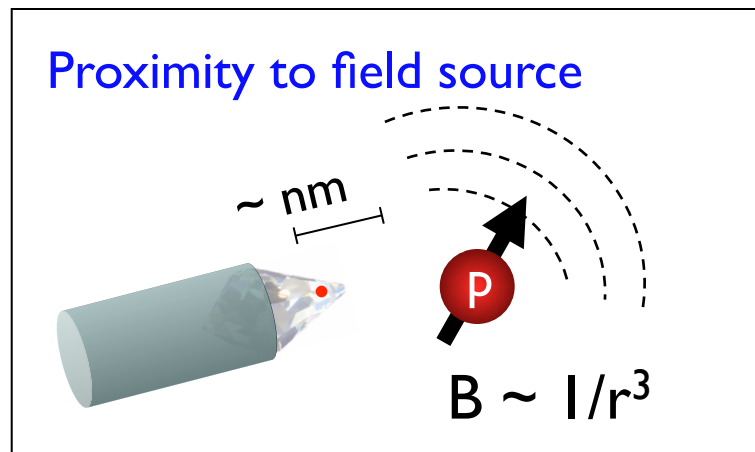
NV nanodiamonds



Scanning diamond-AFM



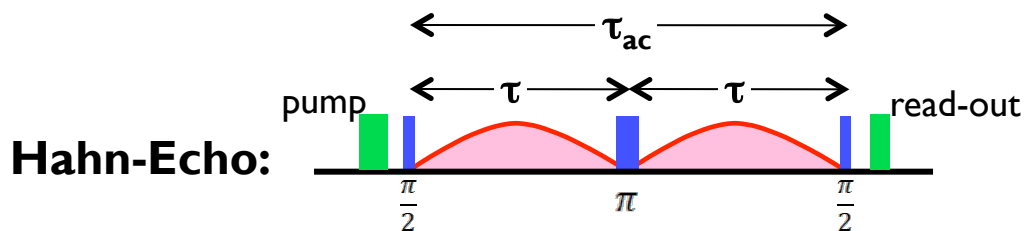
Bulk diamond



Composite spin environment

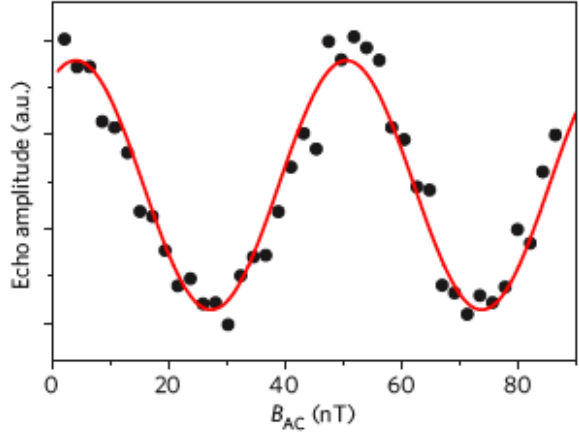
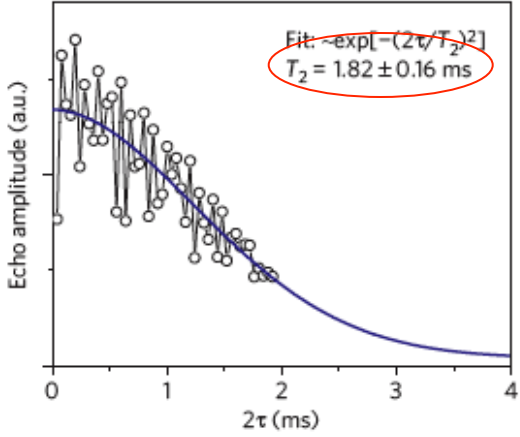
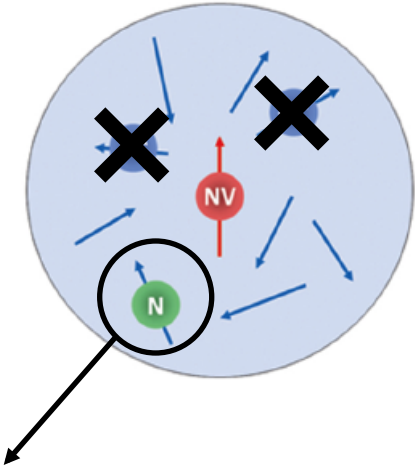


# Best single-NV magnetometry: 4 nT/Hz<sup>1/2</sup>

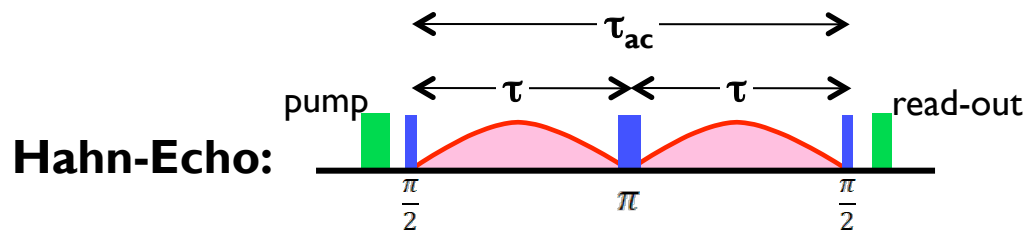


Target ac magnetic field:  
 $B(t) = B_{ac} \sin[(2\pi f_{ac})t + \phi]$

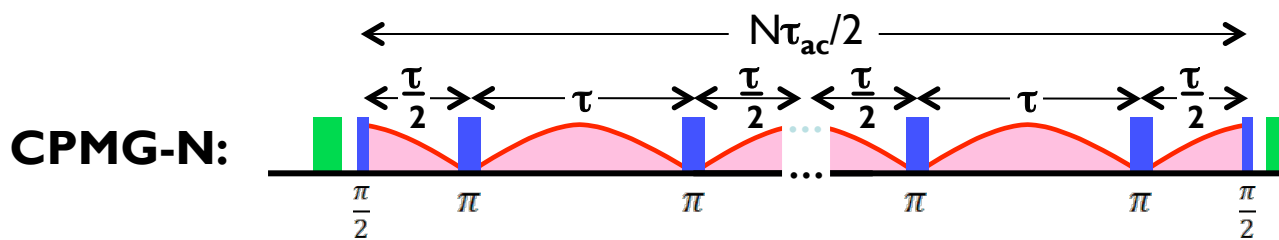
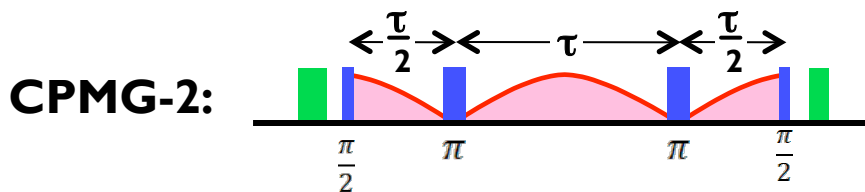
Isotopically purified CVD diamond: <sup>13</sup>C ≈ 0.3%  
 [N] < 10<sup>13</sup> cm<sup>-3</sup>  
 Balasubramanian et al., Nature Materials (2009)



# Progress toward NV magnetometry $< 1 \text{ nT/Hz}^{1/2}$



Target ac magnetic field:  
 $B(t) = B_{ac} \sin[(2\pi f_{ac})t + \phi]$

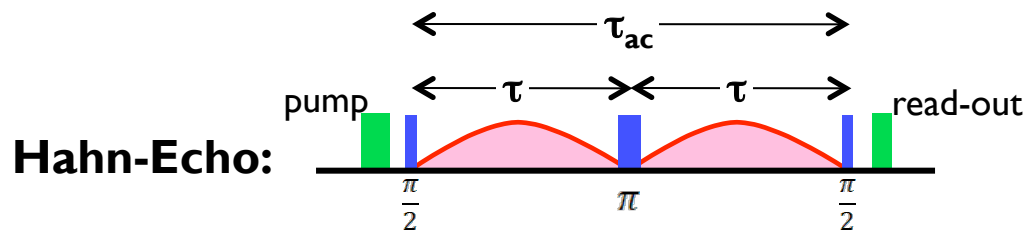


1) Use N-pulse dynamic decoupling sequence:  
 $T_2 \Rightarrow T_2 N^s$   
 $\tau_{ac} \Rightarrow N\tau_{ac} / 2$

Shot-noise sensitivity:

$$\frac{\delta B_{ac}}{\sqrt{\text{Hz}}} \approx \eta_{ac}^{\text{CPMG-N}} \approx \frac{\pi \hbar}{2g\mu_B} \frac{1}{C} \sqrt{\frac{2}{N\tau_{ac}}} \exp \left[ \left( \frac{N^{(1-s)} \tau_{ac}}{2T_2} \right)^p \right]$$

# Progress toward NV magnetometry $< 1 \text{ nT/Hz}^{1/2}$



Target ac magnetic field:  
 $B(t) = B_{ac} \sin[(2\pi f_{ac})t + \phi]$

3) Maximize  $C = \alpha\sqrt{\beta n}$

$\alpha$  = Spin-state-dependent signal contrast

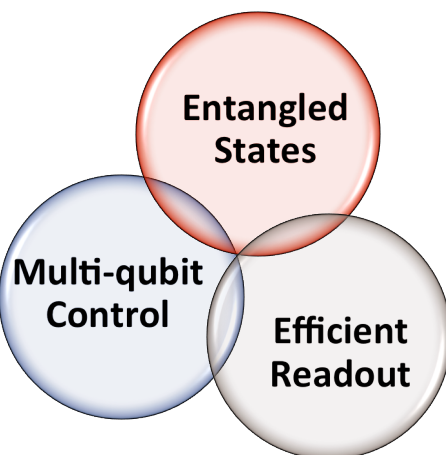
$\beta$  = # photons collected per NV per measurement

$n$  = # sensing NVs (and ancilla spins) in detection volume

1) Use N-pulse dynamic decoupling sequence:

$$T_2 \Rightarrow T_2 N^s$$

$$\tau_{ac} \Rightarrow N\tau_{ac} / 2$$



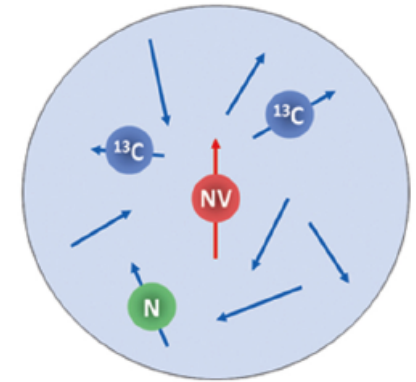
$$\eta_{ac}^{\text{CPMG-N}} \approx \frac{\pi\hbar}{2g\mu_B} \frac{1}{C} \sqrt{\frac{2}{N\tau_{ac}}} \exp \left[ \left( \frac{N^{(1-s)}\tau_{ac}}{2T_2} \right)^p \right]$$

2) Maximize bare  $T_2 \Rightarrow$  control spin environment

# Our role on QuISM team

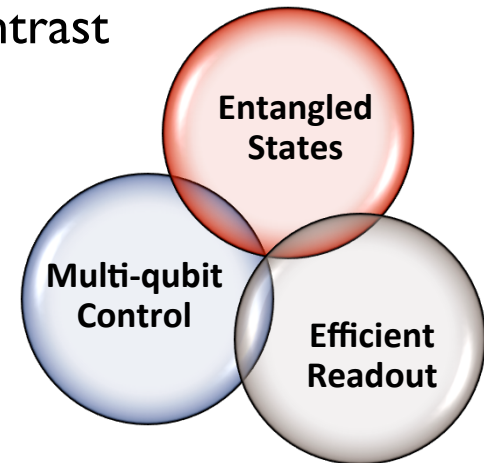
- **NV-diamond as testbed for:**

- Physics of many-body composite-spin systems
- Optimal environmental control (dynamic decoupling)
- Environment-enhanced metrology:
  - e.g., ancilla sensing spins (N) & storage spins ( $^{13}\text{C}$ )
- Creation & application of novel quantum states



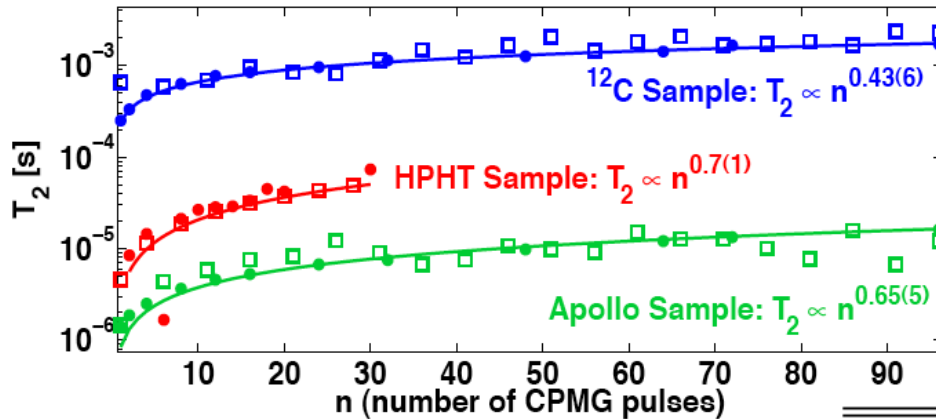
- **Technical optimization of NV-diamond spin measurements:**

- $T_1, T_2, T_2^*$
- Optical collection efficiency & NV spin-state signal contrast
  - => SNR, single-shot NV read-out
- Diamond material properties & control techniques
- Single NV vs. ensembles
- Bulk diamond vs. thin NV layers vs. nanodiamonds
- NV electronically excited, metastable & dark states
- $\text{NV}^-$  vs.  $\text{NV}^0$



- **Physics interactions with other team members**

# Recent results: Dynamic decoupling to increase NV $T_2$



CPMG sequence applied to a wide array of NV-diamond samples  
 $\Rightarrow T_2$  increased by  $\sim 10\times$

$$\delta\phi \propto \frac{1}{\sqrt{n_{\text{NV}} T_2}} \equiv \frac{1}{\sqrt{\text{FOM}}}$$

Spin coherence Figure-of-Merit

Largest FOM in any solid-state spin system  
 $\Rightarrow$  improved NV magnetometry

	$^{12}\text{C}$	HPHT	Apollo
Meas. technique	ensemble	confocal	ensemble
N concentration	$\sim 1$ ppm	$\sim 50$ ppm	$\sim 100$ ppm
NV density	$\sim 10^{14} [\text{cm}^{-3}]$	$\sim 10^{12} [\text{cm}^{-3}]$	$\sim 10^{16} [\text{cm}^{-3}]$
$^{13}\text{C}$ concentration	0.01%	1.1%	1.1%
Echo (1-pulse) $T_2$	240(6) $\mu\text{s}$	5(1) $\mu\text{s}$	2(1) $\mu\text{s}$
$T_2$ scaling	$n^{0.43(6)}$	$n^{0.7(1)}$	$n^{0.65(5)}$
Max. achieved $T_2$	2.2 ms	35 $\mu\text{s}$	20 $\mu\text{s}$
$\Delta$ (expected)	$\approx 60$ kHz	$\approx 3$ MHz	$\approx 6$ MHz
$\Delta$ (measured)	30(10) kHz	1(1) MHz	7(3) MHz
$\tau_c$ (expected)	$\approx 15$ $\mu\text{s}$	$\approx 0.34$ $\mu\text{s}$	$\approx 0.17$ $\mu\text{s}$
$\tau_c$ (measured)	10(5) $\mu\text{s}$	10(5) $\mu\text{s}$	3(2) $\mu\text{s}$
FOM [ $\text{ms}/\text{cm}^3$ ]	$2 \times 10^{14}$	$10^{10}$	$2 \times 10^{14}$

# Recent results: Dynamic decoupling to increase NV $T_2$

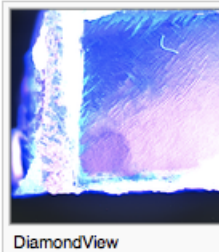
## General Information

ID: 61  
 Substrate #:  
 Run #: 1173101-02  
 Ext Sample #: 1173101-02

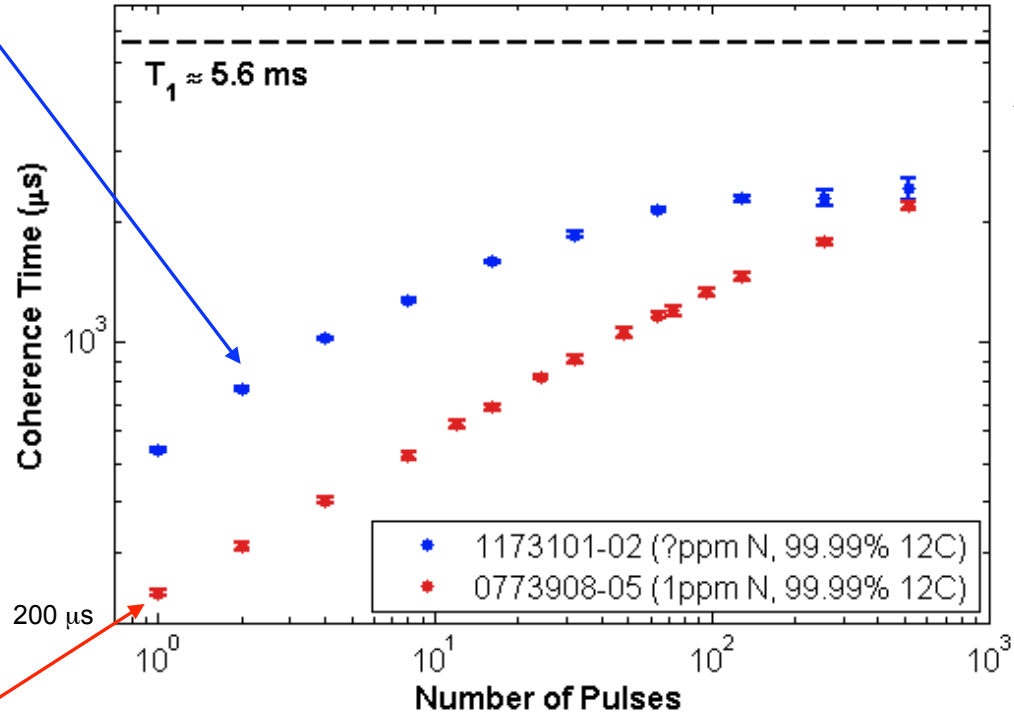
Brief Description: 99.99%  $^{12}\text{C}$  layer on electronic grade diamond.

E6 Comments:

- Sample unpolished, 1
- The sample has a vari do with the way N is i variation is mostly ca will be a thickness de precise numbers on tl that we could eliminat DiamondView (broadt clearly shows the var



Comparative study of CPMG decoupling applied to E6 CVD samples with 0.01%  $^{13}\text{C}$  and variable (low) N content.



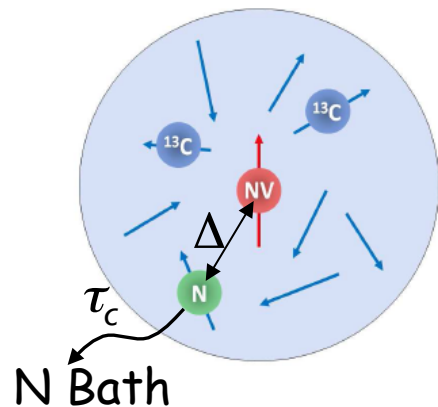
All samples reach  $T_2 \approx 2.5$  ms, with  $T_1 \approx 5.6$  ms

## General Information

ID: 31  
 Substrate #:  
 Run #: 0773908-05  
 Ext Sample #: 0773908-05

Brief Description: 0.6mm growth 99.99%  $^{12}\text{C}$  with 1ppm  $\text{N}_2$  CVD layer on HPHT sample

# Recent results: Probe/control spin-bath spectrum



Spectral decomposition: Measure NV coherence decay as a function of # of CPMG pulses & time

=> fit to determine N spin-bath spectrum parameters

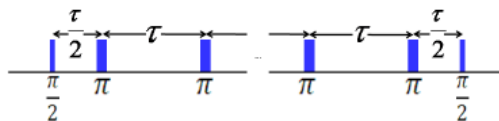
$\Delta$  = NV-N dipolar coupling  $\sim$  N density

$\tau_c$  = N spin-bath correlation (flip-flop) time  $\sim$  (N density)<sup>-1</sup>

Lorentzian N spin-bath

$$S(\omega) = \frac{\Delta^2 \tau_c}{\pi} \frac{1}{1 + (\omega \tau_c)^2}$$

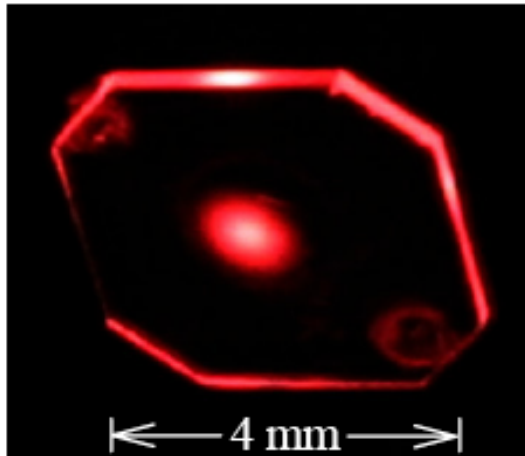
CPMG:



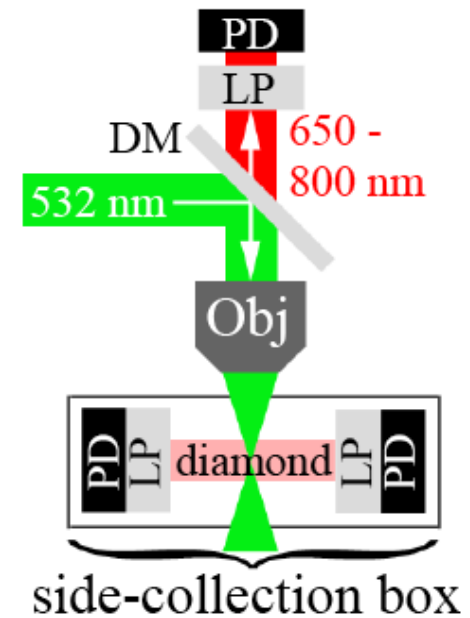
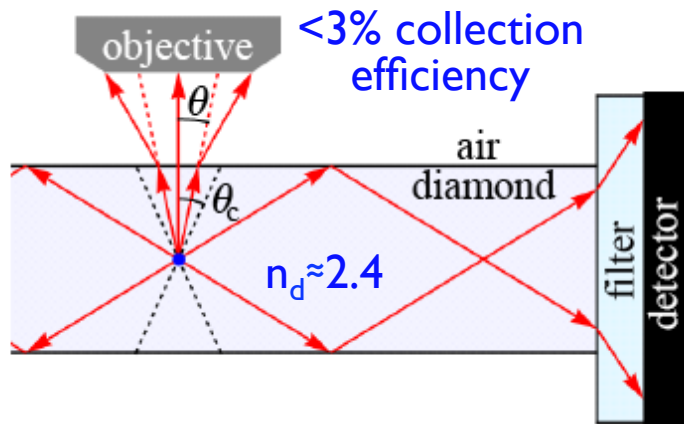
Suppression of N bath flip-flops by random Overhauser field from proximal <sup>13</sup>C spins

	<sup>12</sup> C	HPHT	Apollo
Meas. technique	ensemble	confocal	ensemble
N concentration	$\sim$ 1 ppm	$\sim$ 50 ppm	$\sim$ 100 ppm
NV density	$\sim$ 10 <sup>14</sup> [cm <sup>-3</sup> ]	$\sim$ 10 <sup>12</sup> [cm <sup>-3</sup> ]	$\sim$ 10 <sup>16</sup> [cm <sup>-3</sup> ]
<sup>13</sup> C concentration	0.01%	1.1%	1.1%
Echo (1-pulse) T <sub>2</sub>	240(6) $\mu$ s	5(1) $\mu$ s	2(1) $\mu$ s
T <sub>2</sub> scaling	n <sup>0.43(6)</sup>	n <sup>0.7(1)</sup>	n <sup>0.65(5)</sup>
Max. achieved T <sub>2</sub>	2.2 ms	35 $\mu$ s	20 $\mu$ s
$\Delta$ (expected)	$\approx$ 60 kHz	$\approx$ 3 MHz	$\approx$ 6 MHz
$\Delta$ (measured)	30(10) kHz	1(1) MHz	7(3) MHz
$\tau_c$ (expected)	$\approx$ 15 $\mu$ s	$\approx$ 0.34 $\mu$ s	$\approx$ 0.17 $\mu$ s
$\tau_c$ (measured)	10(5) $\mu$ s	10(5) $\mu$ s	3(2) $\mu$ s
FOM [ms/cm <sup>3</sup> ]	$2 \times 10^{14}$	10 <sup>10</sup>	$2 \times 10^{14}$

## Recent results: Side-collection technique

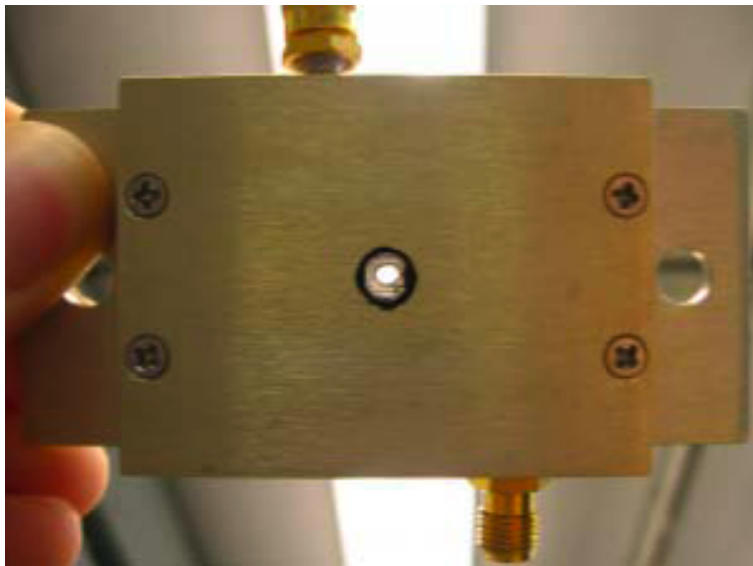
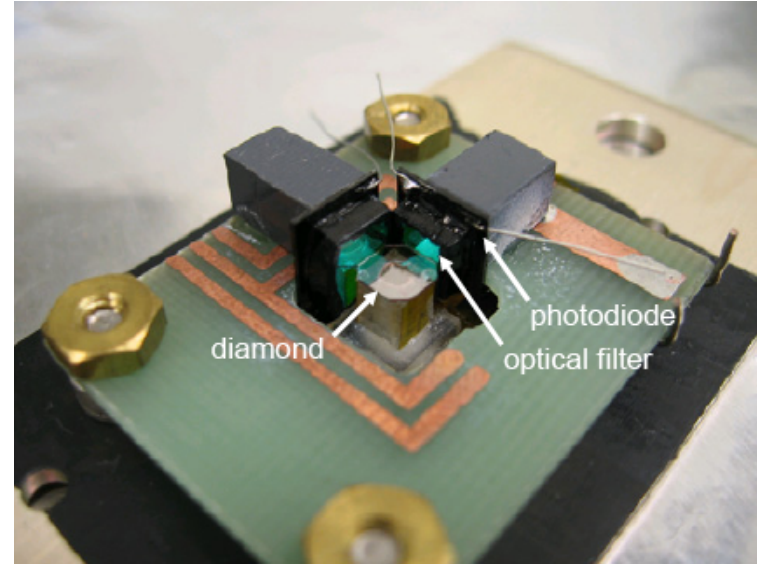
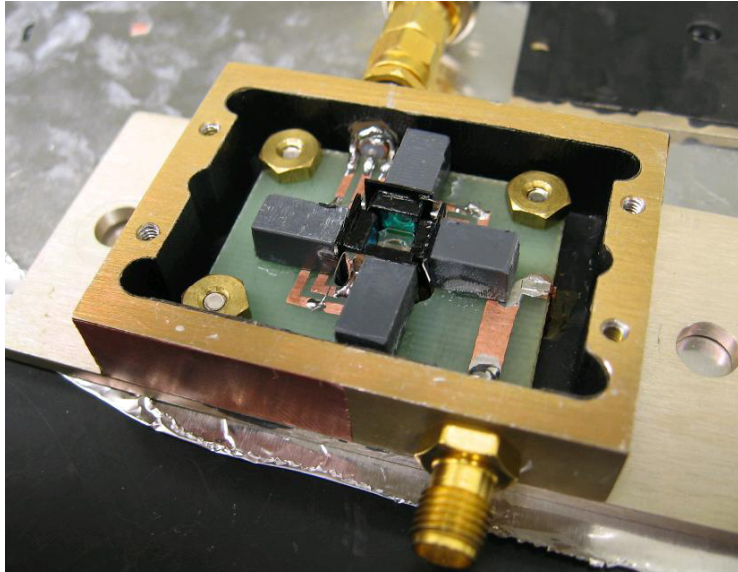


>90% of NV fluorescence leaves via the side (rim) of a diamond chip

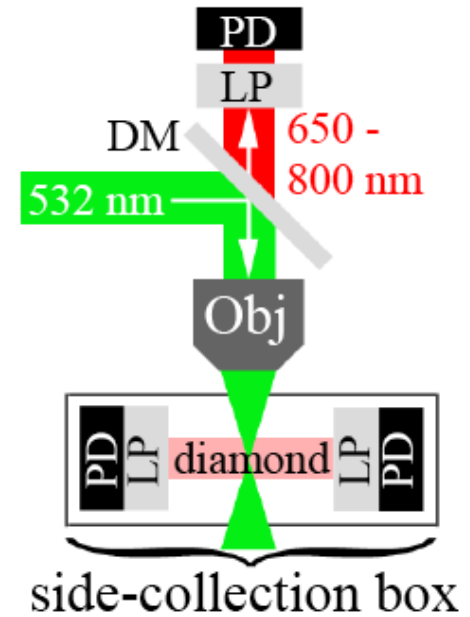




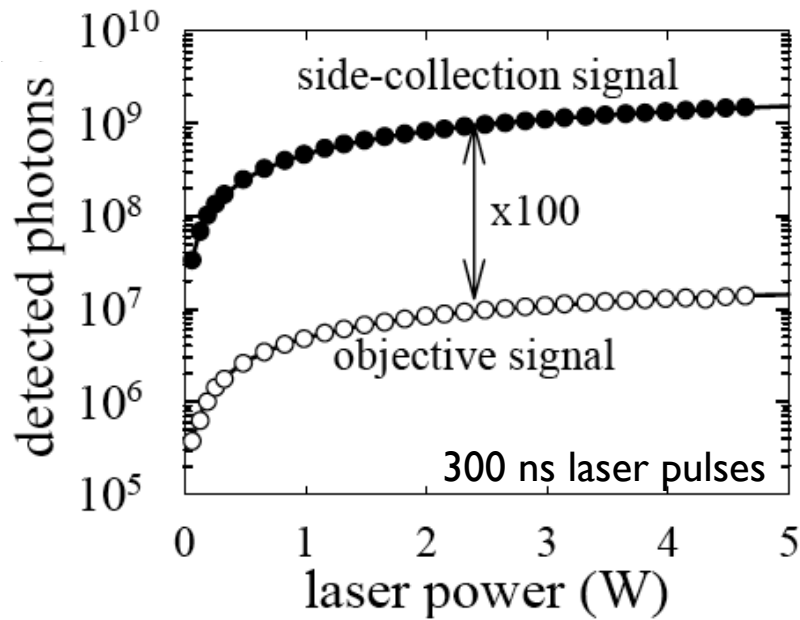
# Recent results: Side-collection technique



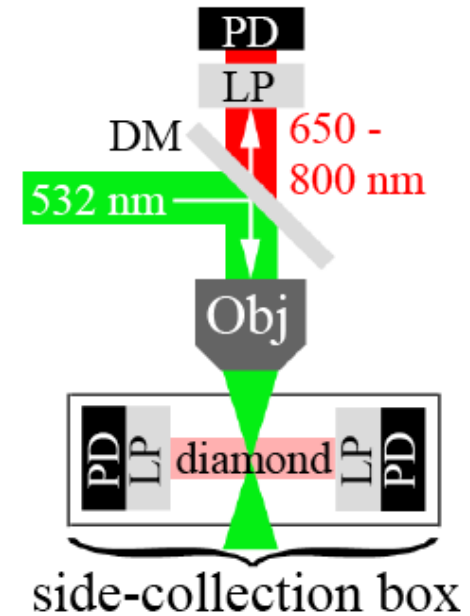
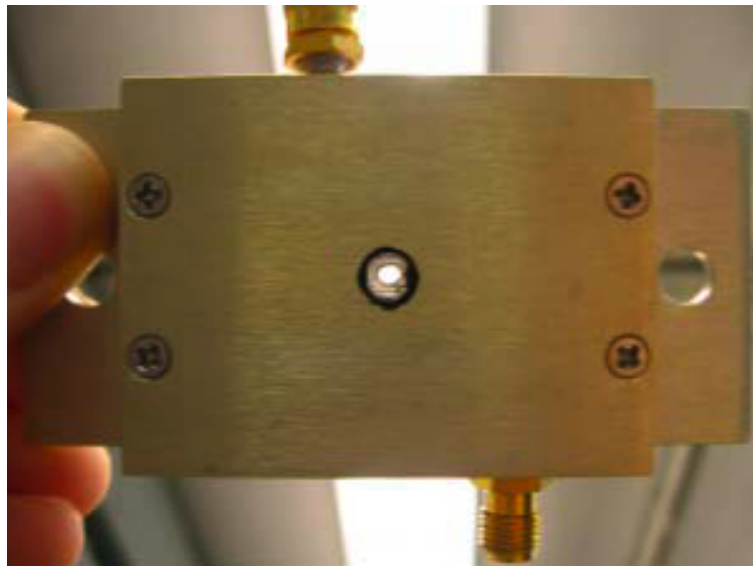
David Le Sage



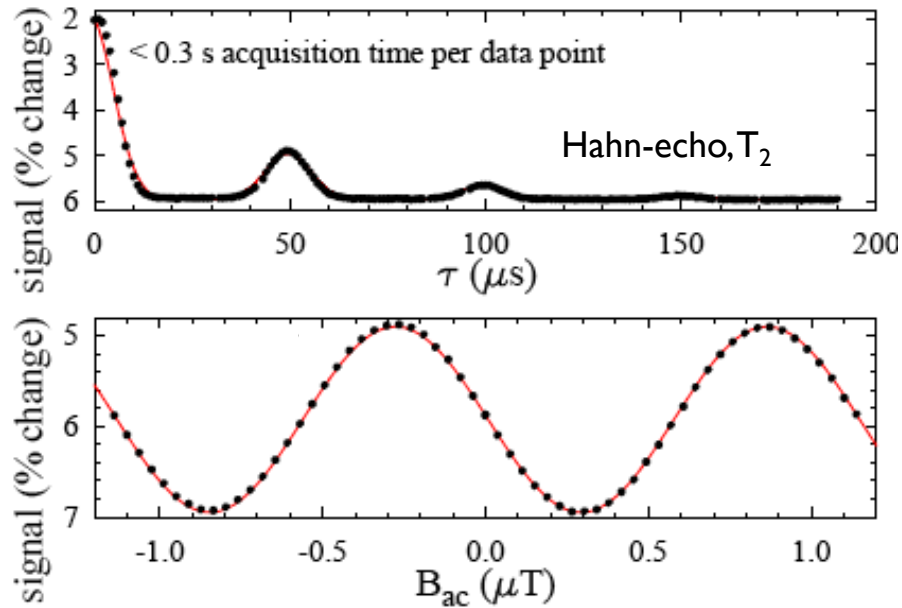
## Recent results: Side-collection technique



≈50% optical side-collection efficiency  
>60% expected with optimization  
=> single-shot NV readout?

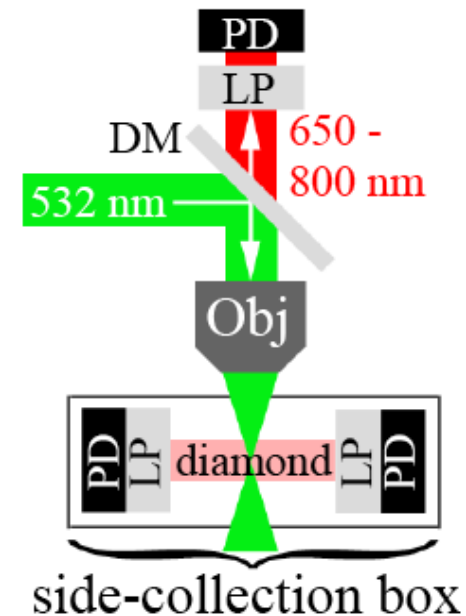
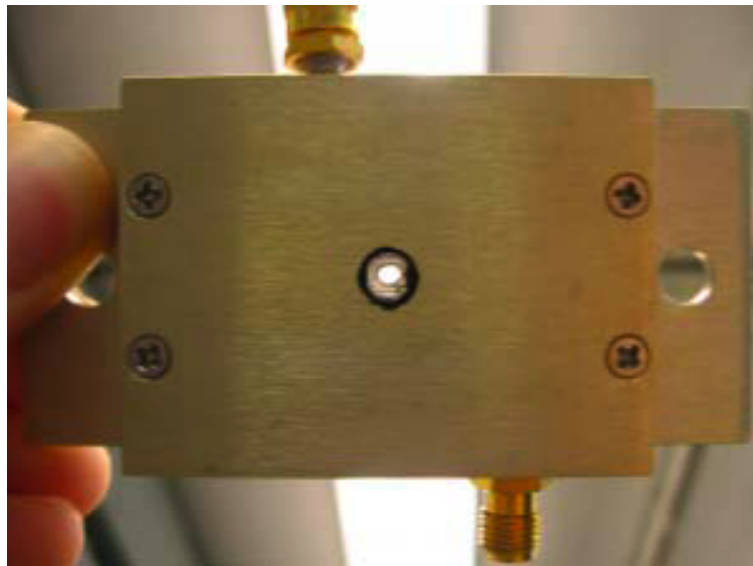


# Recent results: Side-collection technique



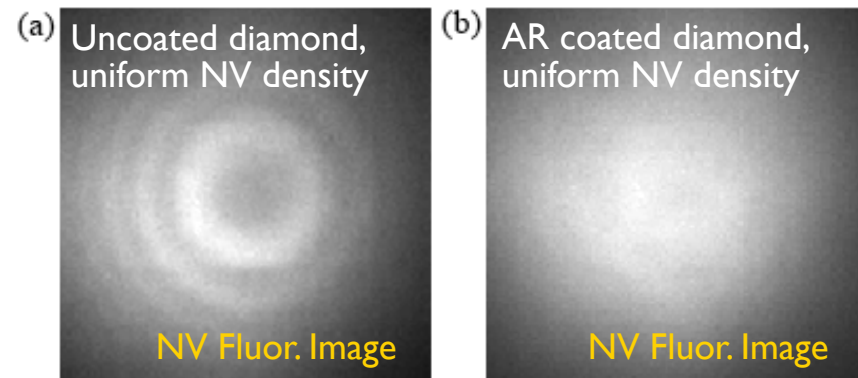
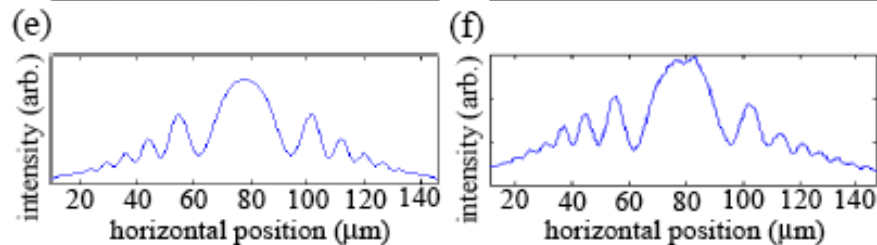
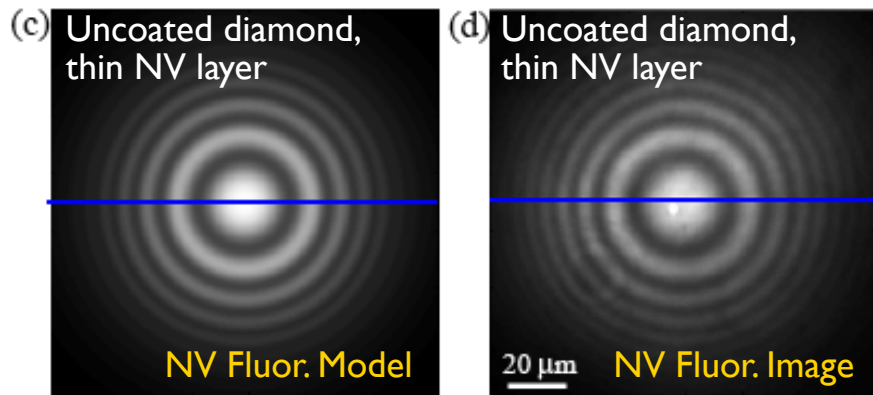
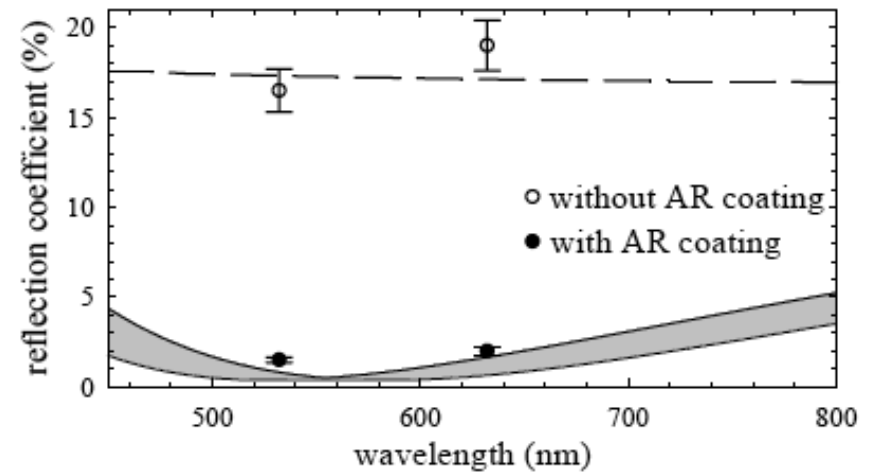
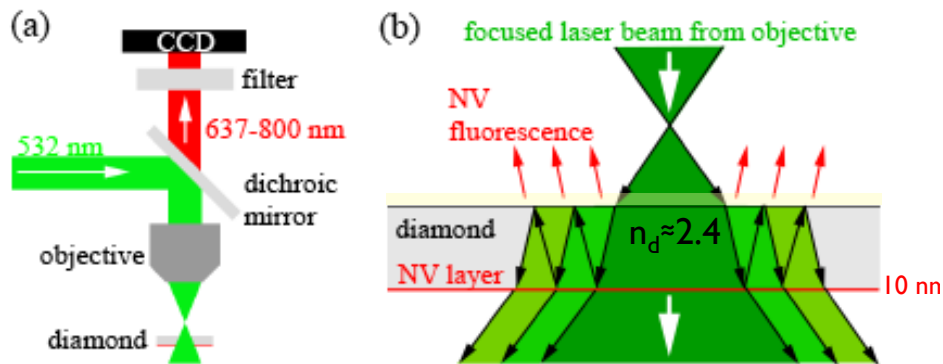
$\approx 50\%$  optical side-collection efficiency  
 $> 60\%$  expected with optimization  
 $\Rightarrow$  single-shot NV readout?

In progress:  $< 100$  pT  $\text{Hz}^{-1/2}$  in  $\sim 1$   $\mu\text{m}^3$ ,  
 $[\text{NV}] \sim 10^{16}$   $\text{cm}^{-3}$ ,  $T_2 \approx 20$   $\mu\text{s}$ ,  
 $\text{FOM} \approx 2 \times 10^{14}$   $\text{ms}\cdot\text{cm}^{-3}$



# Recent results: AR-coating of diamond surface

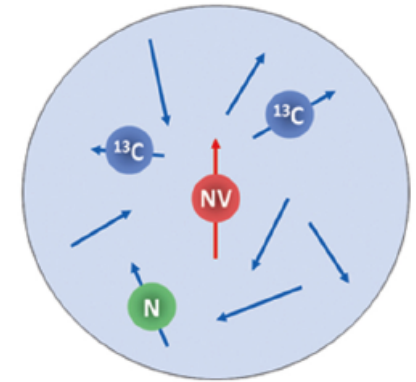
Reduce etalon effect with  $\lambda/4$  layer of silica ( $n=1.46$ )



# Our role on QuISM team

- **NV-diamond as testbed for:**

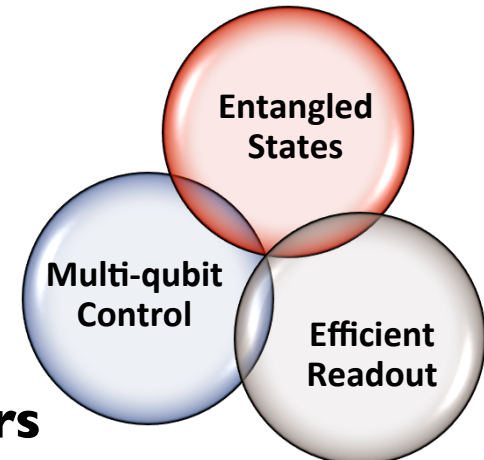
- Physics of many-body composite-spin systems
- Optimal environmental control (dynamic decoupling)
- Environment-enhanced metrology:  
e.g., ancilla sensing spins (N) & storage spins ( $^{13}\text{C}$ )
- Creation & application of novel quantum states





- **Technical optimization of NV-diamond spin measurements:**

- $T_1, T_2, T_2^*$
- Optical collection efficiency & NV spin-state signal contrast  
=> SNR, single-shot NV read-out
- Diamond material properties & control techniques
- Single NV vs. ensembles
- Bulk diamond vs. thin NV layers vs. nanodiamonds
- NV electronically excited, metastable & dark states
- $\text{NV}^-$  vs.  $\text{NV}^0$

- **Physics interactions with other team members**



# Posters



## Application of Dynamical Decoupling to Enhance NV Ensemble Magnetometry



L.M. Pham<sup>1</sup>, N. Bar-Gill<sup>2</sup>, C. Belthangady<sup>2</sup>, D. Le Sage<sup>2</sup>, K. Arai<sup>3</sup>, P. Cappellaro<sup>4</sup>, R.L. Walsworth<sup>5,2</sup>

<sup>1</sup>School of Engineering and Applied Sciences, Harvard, Cambridge, MA 02138. <sup>2</sup>Harvard-Smithsonian Center for Astrophysics, Cambridge, MA 02138  
<sup>3</sup>Physics Department, MIT, Cambridge, MA 02133. <sup>4</sup>Nuclear Science and Engineering Department, MIT, Cambridge, MA 02139. <sup>5</sup>Physics Department, Harvard, Cambridge, MA 02138

## Spectral Decomposition of Composite Solid-State Spin Environments for Improved Ensemble Metrology

Nir Bar-Gill<sup>1,5</sup>, My Linh Pham<sup>2</sup>, Chinmay Belthangady<sup>1</sup>, David Le Sage<sup>1</sup>, Paola Cappellaro<sup>3</sup>, Jeronimo Maze<sup>4</sup>, Mikhail Lukin<sup>5</sup>, Amir Yacoby<sup>5</sup> and Ronald L. Walsworth<sup>1,5</sup>



<sup>1</sup>Harvard-Smithsonian Center for Astrophysics, Cambridge, MA, USA. <sup>2</sup>School of Engineering and Applied Sciences, Harvard University, Cambridge, MA, USA. <sup>3</sup>Department of Nuclear Science and Engineering, Massachusetts Institute of Technology, Cambridge, MA, USA. <sup>4</sup>Pontificia Universidad Catolica, Santiago, Chile. <sup>5</sup>Department of Physics, Harvard University, Cambridge, MA, USA.



## Improved NV-Diamond Fluorescence Detection Technique

D. Le Sage<sup>1</sup>, L.M. Pham<sup>2</sup>, D. Glenn<sup>1</sup>, A. Trifonov<sup>3</sup>, N. Bar-Gill<sup>1</sup>, C. Belthangady<sup>1</sup>, K.Arai<sup>4</sup>, H. Zhang<sup>1</sup>, R.L. Walsworth<sup>1,3</sup>



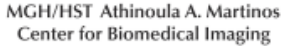
<sup>1</sup>Harvard-Smithsonian Center for Astrophysics, Cambridge, MA 02138,  
<sup>2</sup>School of Engineering and Applied Sciences, Harvard University, Cambridge, MA 02138,  
<sup>3</sup>Department of Physics, Harvard University, Cambridge, MA 02138,  
<sup>4</sup>Department of Physics, MIT, Cambridge, MA 02139.



## Room-temperature solid-state quantum memory using pairs of nuclear spins in diamond

Nir Bar-Gill<sup>1,4</sup>, Stephen DeVience<sup>2</sup>, David Le Sage<sup>1</sup>, Chinmay Belthangady<sup>1</sup>, Linh M. Pham<sup>3</sup> and Ronald L. Walsworth<sup>1,4</sup>

<sup>1</sup>Harvard-Smithsonian Center for Astrophysics, Cambridge, MA, USA. <sup>2</sup>Department of Chemistry and Chemical Biology, Harvard University, Cambridge MA, USA. <sup>3</sup>School of Engineering and Applied Sciences, Harvard University, Cambridge, MA, USA. <sup>4</sup>Department of Physics, Harvard University, Cambridge, MA, USA.



## Nuclear Spin Singlet States in NMR

Stephen DeVience<sup>1</sup>, Ronald Walsworth<sup>2,3</sup>, Matthew Rosen<sup>3,4,5</sup>

1. Harvard University Department of Chemistry and Chemical Biology, 2. Harvard-Smithsonian Center for Astrophysics, 3. Harvard University Department of Physics, 4. Martinos Center for Biomedical Imaging, 5. Harvard Medical School