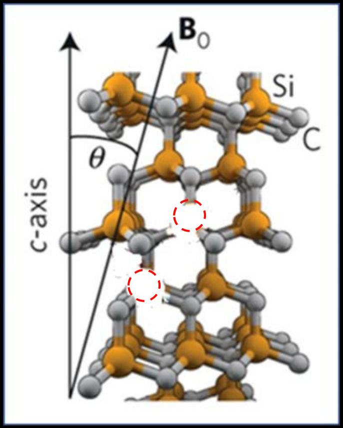
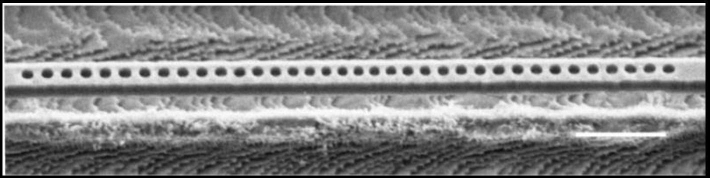
HARVARD
John A. Paulson
School of Engineering
and Applied Sciences

"Inverted Atoms": New Photon Sources

Evelyn L. Hu





Based on Widmann et al., *Nat. Mat.* 2015

June 7, 2019

NSF/DOE Quantum Science Summer School (QS3)

YESTERDAY’S TOPICS

- Semiconductor (GaAs) QUANTUM DOTS
- Photonic Crystal Cavities: structure, fabrication & metrics
- Achieving STRONG COUPLING: new entangled light-matter state

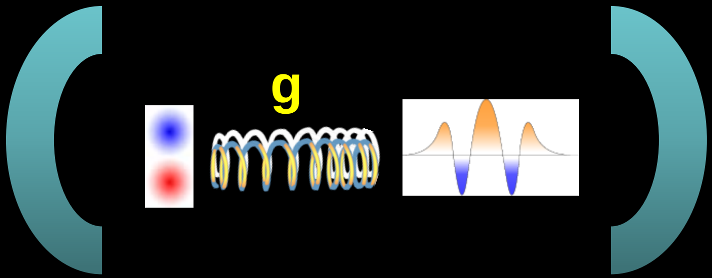
THE TOPICS FOR TODAY

- Atomic-scale “defects” in SiC
- Integrated defect-photonic crystal cavity results
- New challenges and opportunities

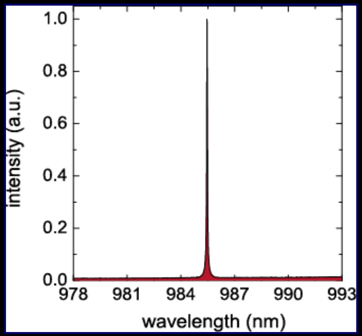
But first…

PUTTING TOGETHER THE QD-CAVITY SYSTEM:

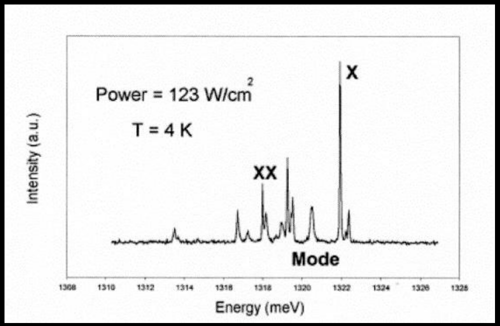
What do you think is important here?



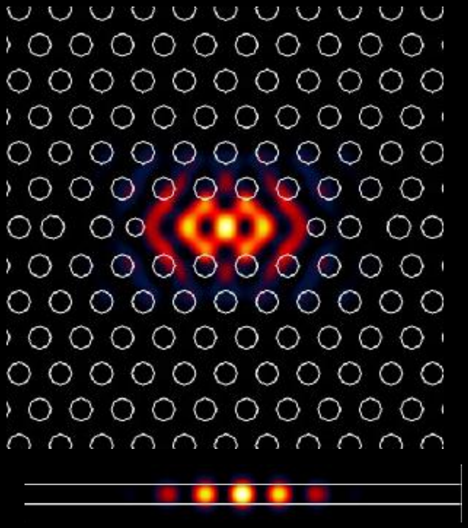
CAVITY



Cavity resonance



Quantum dot spectrum



top

side

PLEASE TAKE 15 MINUTES WITHIN GROUPS TO DISCUSS THIS, NOTE CONCEPTS THAT ARE UNCLEAR

PUTTING TOGETHER THE QD-CAVITY SYSTEM: Your answers

1. Group A
   a. IDEAS

      i. Match resonant energies of QD and cavity: should couple states
      ii. Maybe superposition of exciton and photon in cavity?

   b. QUESTIONS
      i. How to read state?
      ii. What are the two states: photon and exciton, no photon nor exciton, photon OR exciton?
      iii. QD in cavity? Near cavity?
      iv. How to change between states selectively?
2. The “AVENGERS”
      i. QUESTION: what does it mean physically for the cavity and the QD to be coupled?

      ii. IDEA: we shoot broad spectrum laser into the cavity. If we see a sharp peak for the output, we have coupled the QDs to the cavity…we move the photonic crystal on top of the QD membrane and measure the output intensity…

Your answers (2)

3. "DENTISTS" (we like cavities)
   a. QUESTIONS
      i. How close does the QD need to be spatially? Can they be designed to be spatially closer?
      ii. How does the # of QDs matter/affect things?
      iii. How to control lifetimes?
      iv. MATHEMATICALLY, what is g?
      v. How close does the cavity excitation have to be to the cavity?
      vi. How do you measure the light in the cavity? How does the light get out?
      vii. We know the QD can excite the cavity, but can the cavity excite the QD? Does the location of the QD matter more?
      viii. Can you tune the cavity? Tune the QD?
      ix. How do you measure the light in the cavity?
4. Group B
   a. The cavity needs to be in resonance with the QD 
   b. QUESTIONS
      i. How important is the placement of the QD?
      ii. More detail on fabrication

Your answers (3)

5. Antonio's User Group
    a. REPLIES
        i. Dielectric environment tuning of QD with cavity
        ii. Match QD frequency with cavity
        iii. Good selection rules, dipole orientation
        iv. QDs have to where the cavity nodes are

6. Group C
    a. QUESTIONS
        i. How to tune the resonance of frequency of a cavity?
        ii. Are we using the cavity to pick up QD's photon emission?
        iii. What's the purpose of e-beam lithography? (fine spatial features)
        iv. What's the purpose of tuning the emission spectrum of a single QD? (we tuned the cavity, not the QD)
7. Group D
    a. What is important?
        i. What mechanism for coupling
        ii. Excitation of QD
        iii. Whether or not exciton is made
        iv. Whether or not cavity resonates

Your answers (4)

7. Group D
   b. QUESTIONS
      i. Is the QD inside/outside of the cavity?
      ii. What about multiple (QDs)?
8. Group E
   a. QUESTIONS
      i. How many photonic modes could be allowed?
      ii. Can you engineer the defect to support 2 modes?
      iii. How do we send wave into cavity with an intensity not in the cavity?
      iv. How does light interact with QD?
      v. How does coupling work?
      vi. How does full-wave mode interact with photon? (Maxwell's EM vs QED)
      vii. Is the qubit I0> state the system where the resonant mode is present and the QD is in its ground state? Is the |1> where the QD is excited into its mode and photon is not present?

Your answers (5)

9. Group F

   a. RECIPE

Then there should be strong coupling

      i. Find a QD with a good energy level splitting in the wavelength range you want
      ii. You need to make sure it is spatially separated from other quantum dots, on a range determined by the size of cavity your want
      iii. Build a cavity that has a maximum resonance mode E field at the position of the desired QD (overlap spatially)
      iv. Design the resonance frequency of the cavity to be resonant with QD's ΔE
      v. Maximize matrix elements.

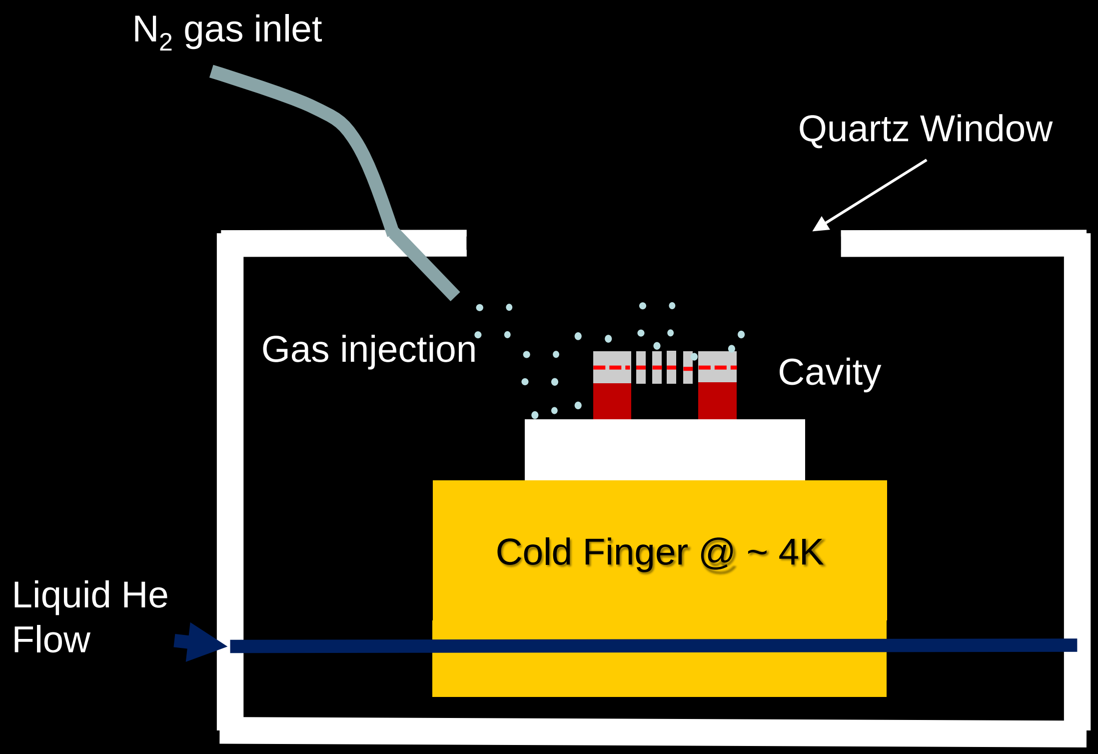
   b. QUESTIONS
      i. Does the parity of the mode matter?
      ii. Which QD excited state do we couple to?
      iii. How does this relate to the shape of the cavity?
      iv. Does the quantum dot have to be in the center?

Your answers (6)

10. Group G
    a. RECIPE
        i. Cavity has a specific resonant mode, establishes a standing EM wave at particular frequency upon excitation
        ii. QD ~ 2 –level system and energy gap ΔE between electronic energy levels set characteristic energy scale for excitation
            – QDs are not mono-disperse
            – QDs have some characteristic spacing in cavity materials
        iii. EM interacts with dipole of QD, depending on if cavity resonance energy coincides with ΔE, may excite excitons.

TUNING THE CAVITY FREQUENCY THROUGH ADSORPTION (reversible)

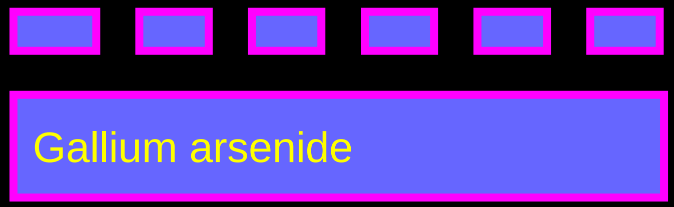
We tune the resonant frequency by changing the dimensions of the cavity at the monolayer level: through successive, calibrated injections of nitrogen



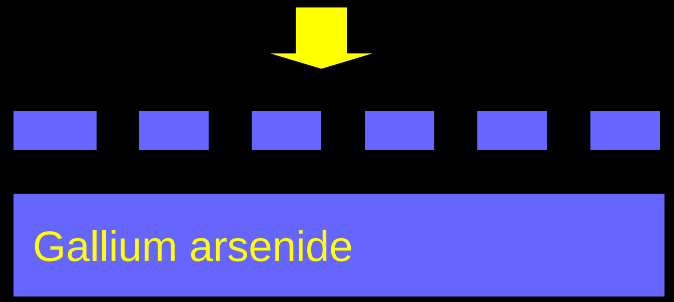
TUNING THE CAVITY FREQUENCY THROUGH "DIGITAL ETCHING" (not reversible)

Mode and QD transitions not quite matched

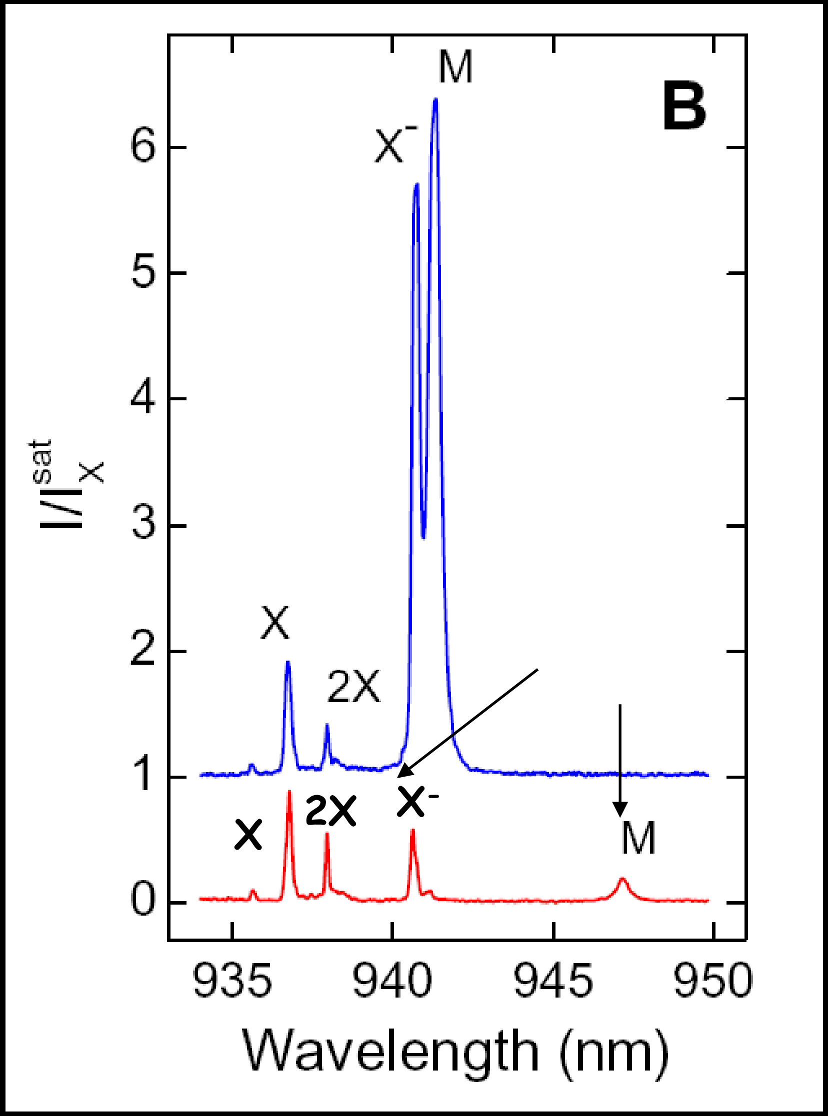
Form thin oxide



Selectively etch oxide

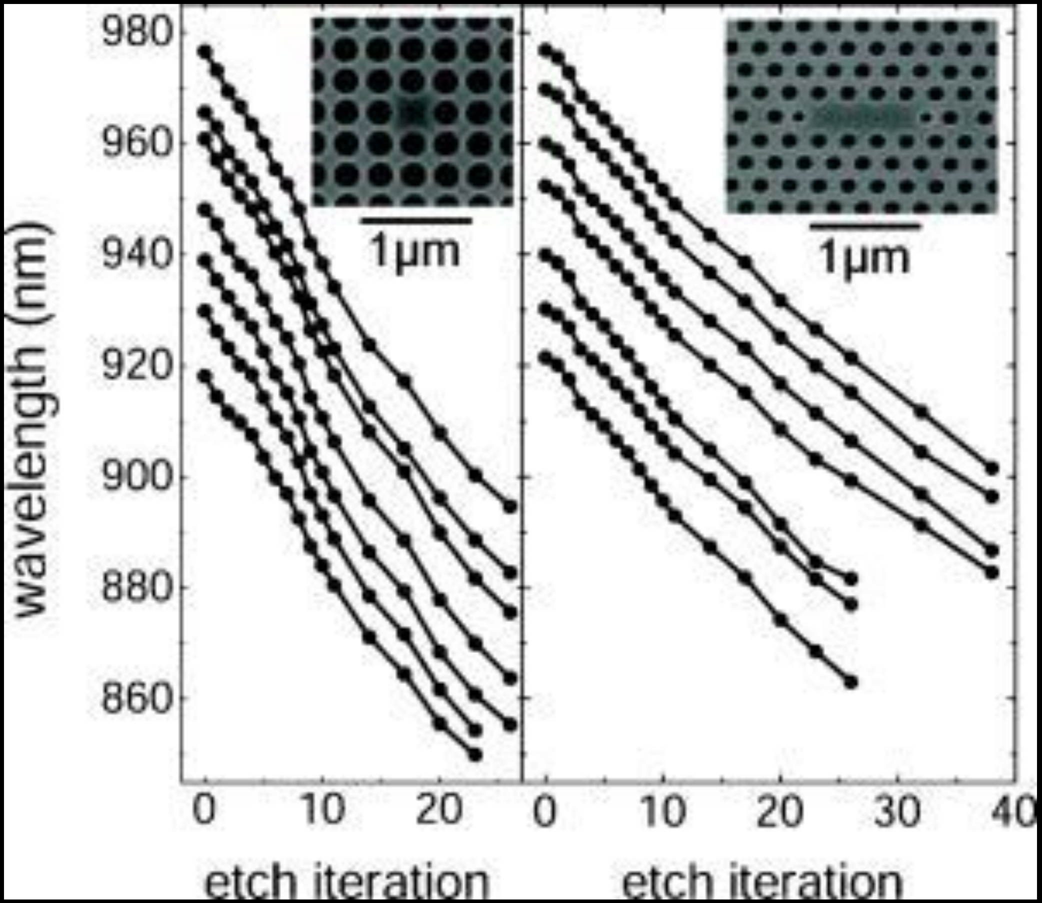


Repeat as needed

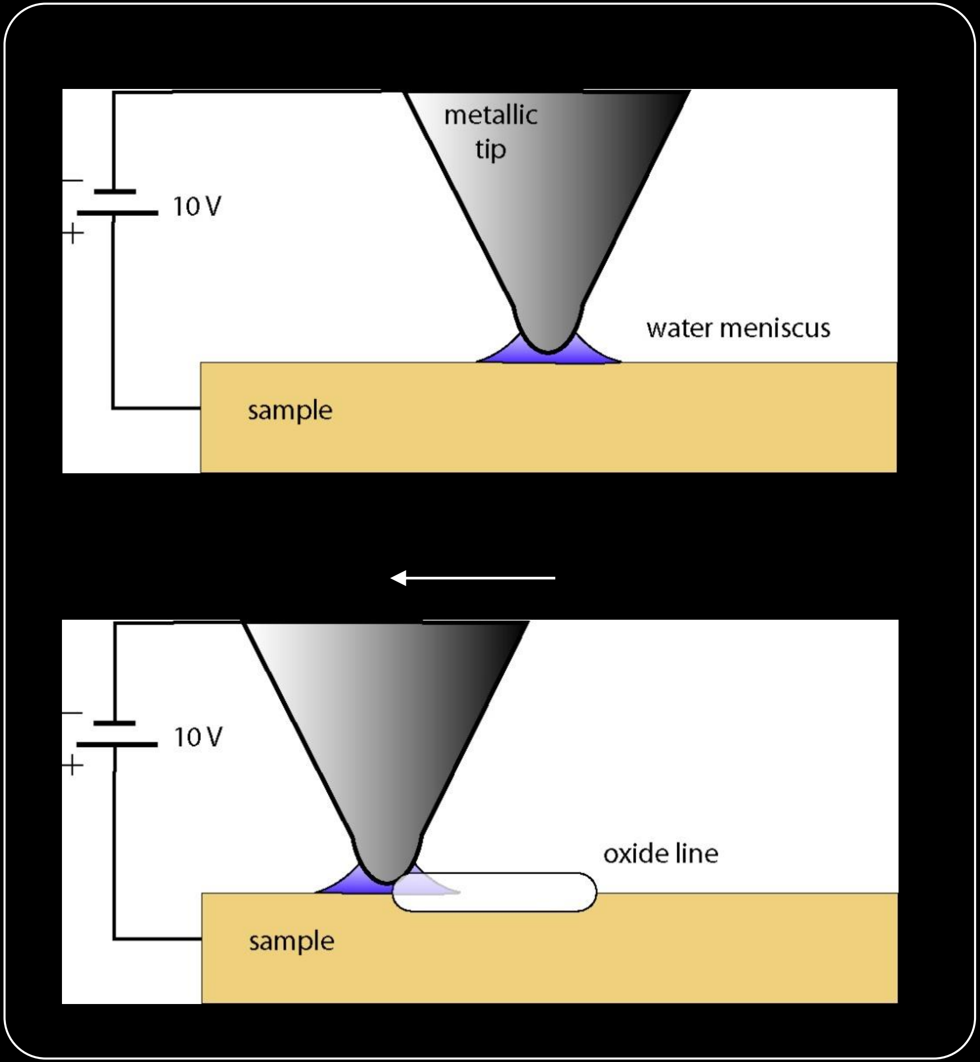


Hennessey at al.,
APL 87, 21108 (2005)

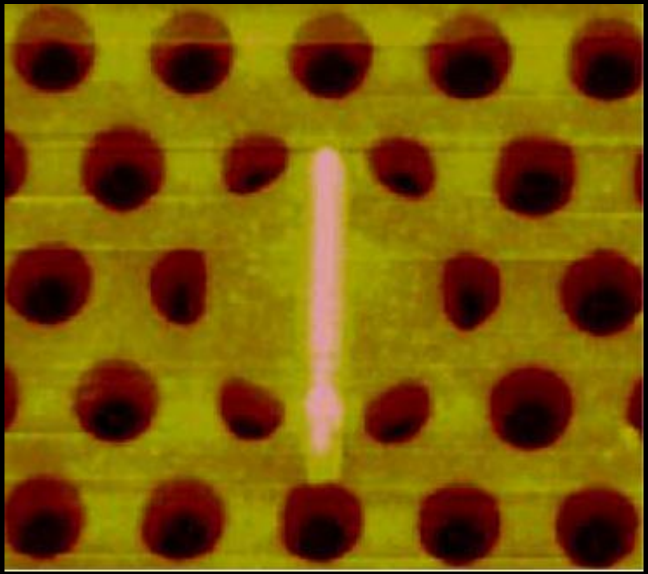
TUNING THE CAVITY FREQUENCY THROUGH "DIGITAL ETCHING" (not reversible)



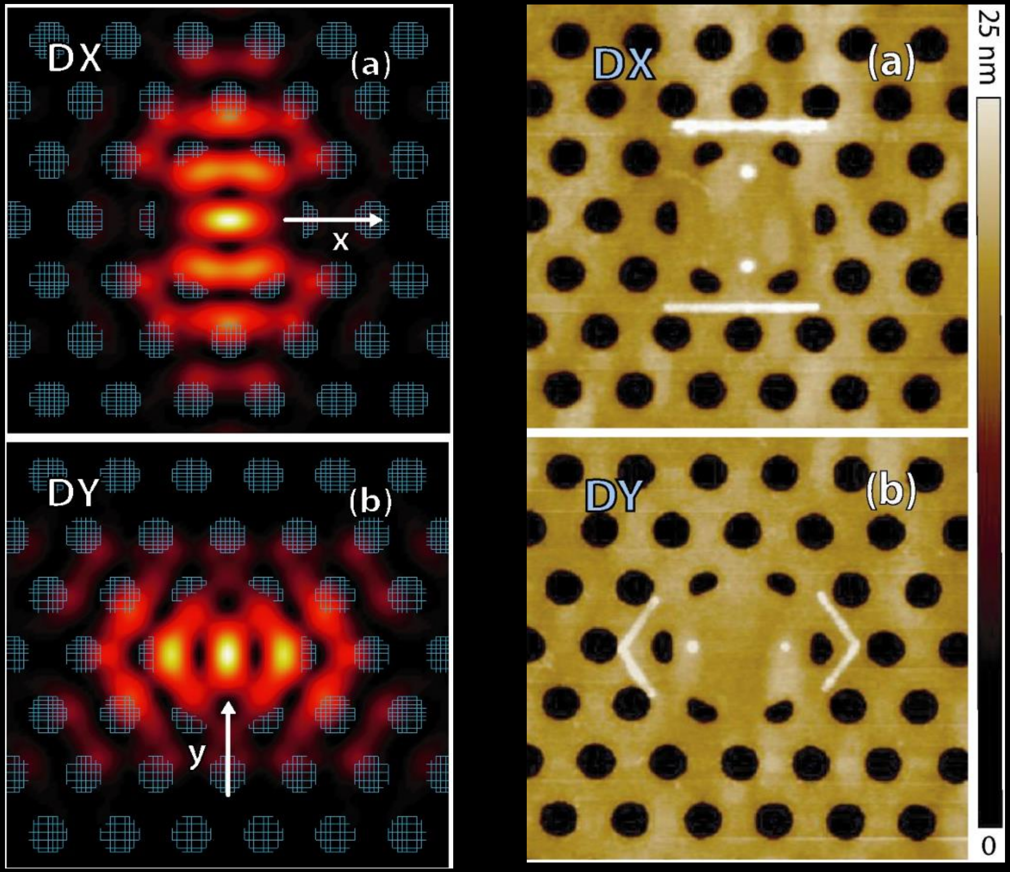
TUNING THE CAVITY FREQUENCY THROUGH LOCAL OXIDATION



- Ni/Pt coated tip
- -10 V bias
- 0.1 micron/s scan speed
- oxide is 50 nm X 4 nm

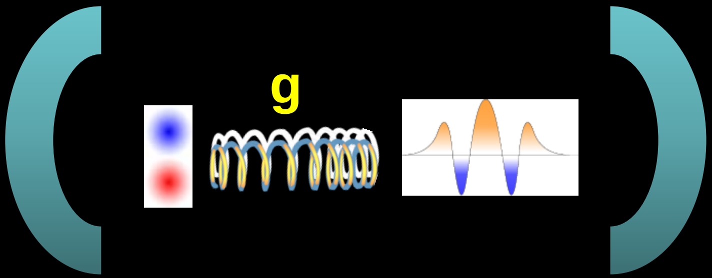


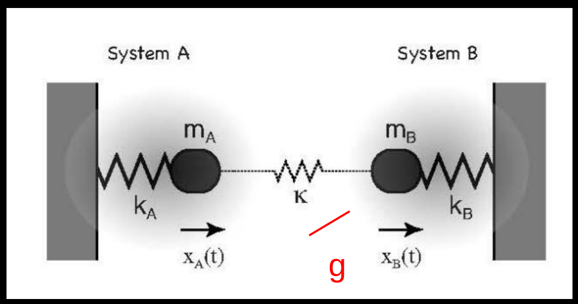
MODIFYING A "DEFECT" TO SUPPORT DIFFERENT PHOTON STATES



WHAT IS THE NATURE OF THE EMITTER-CAVITY COUPLING?

What is g?





Novotny, *Am. J. Phys.* **78** (2010)

Classical analogue: coupling of 2 wave solutions (oscillators) , resulting in a new set of *eigenfunctions, frequencies*

Ph.D. Thesis: “Sol. State Cavity QED in QDs Coupled to Photonic Crystal Cavities,” Arka Majumdar, Stanford (Aug. 2012)

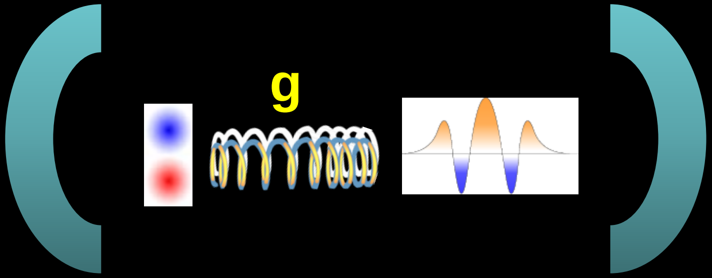
Hamiltonian for atom-cavity system

$$\mathcal{H} = \omega_a \sigma^\dagger \sigma + \omega_c a^\dagger a + ig(a\sigma^\dagger - a^\dagger \sigma)$$

ω_a, ω_c = atom, cavity frequencies
σ, σ^+ operators on atomic states, |g><e| (ground and excited)
a, a^+ annihilation and creation operators for cavity
g couples atomic (electronic) and photonic states

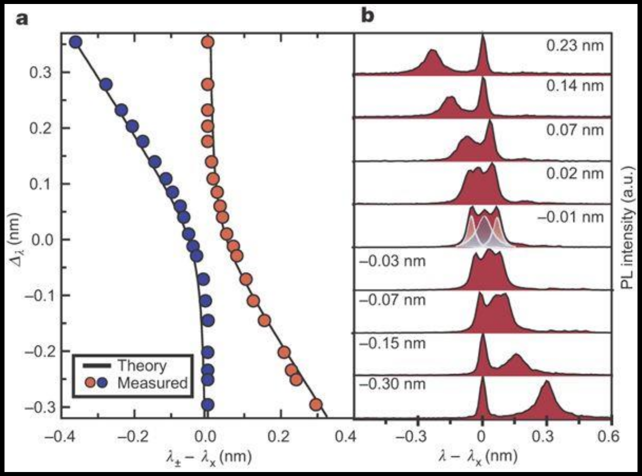
WHAT IS THE NATURE OF THE EMITTER-CAVITY COUPLING?

What is g?



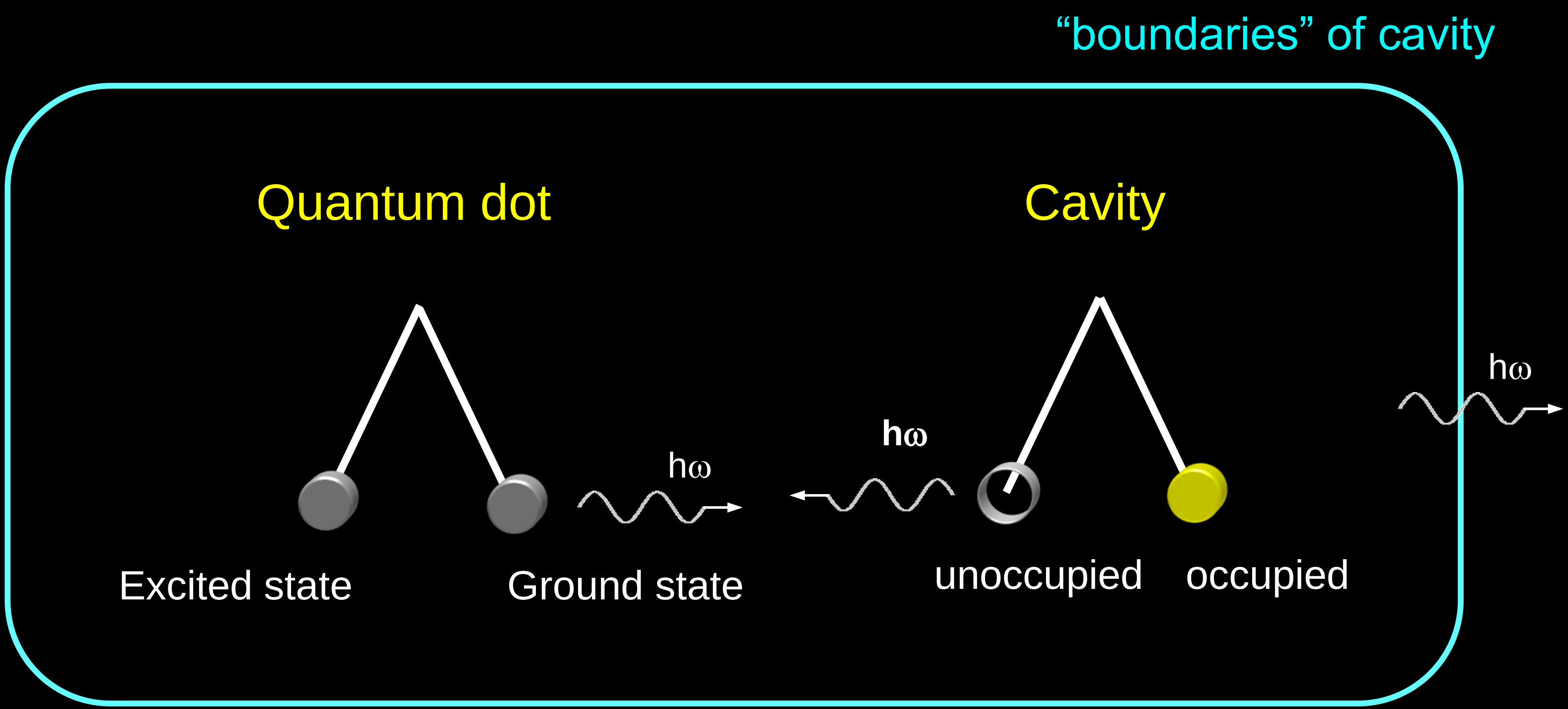
Physically: interaction of atomic dipole with field of the cavity $\vec{\mu} \cdot \vec{E}$

Mathematically, often expressed as a FREQUENCY, or rate

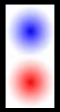


g~ 18.4 GHz

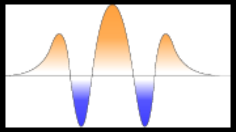
AN "IDEAL MATCH" BETWEEN CAVITY AND QUANTUM DOT



Electronic states =



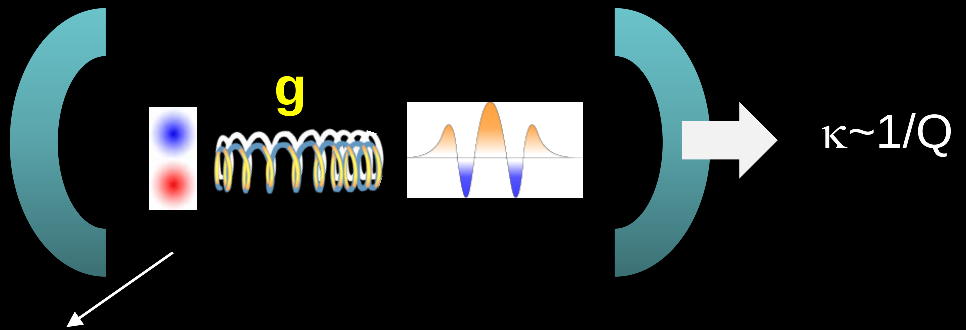
Photon states =



lossless transfer of energy between quantum dot (matter) and cavity (photons)

In fact there IS loss of photons from the cavity

THE METRICS OF A GOOD EMITTER-CAVITY MATCH (with loss)



γ = spontaneous decay rate

g, the coupling constant (rate) should be larger than : g~ 18.4 GHz

- the rate of photon loss from the cavity, κ ~ 1/Q (interaction of cavity field with external environment κ = 24.1 GHz
- The rate of photon loss from the “atom” (QD) γ = 8.5 GHz

Values from Hennessy et al., *Nature* 445, p. 896-9 (2007)

$$\omega_{\pm} = \frac{\omega_c + \omega_a}{2} - i\frac{\kappa + \gamma}{2} \pm \sqrt{g^2 + \frac{1}{4}\left(\delta - i(\kappa - \gamma)\right)^2}$$

δ = QD-cavity detuning

g > κ/2

Ph.D. Thesis: “Sol. State Cavity QED in QDs Coupled to Photonic Crystal Cavities,” Arka Majumdar, Stanford (Aug. 2012)

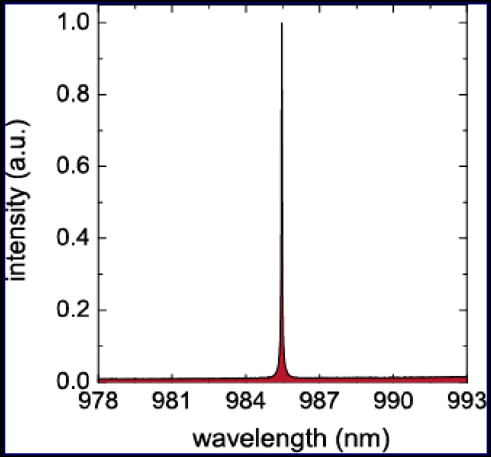
PURCELL FACTOR: A METRIC OF CAVITY-EMITTER MATCHING

Emission rate in cavity

$$F = \frac{\Gamma_{cav}}{\Gamma_0} \sim \frac{Q}{V}\left[\frac{\vec{\mu}\cdot\vec{E}(r)}{|\mu||E_{max}|}\right]^2$$

Emission rate in free space

- ✓ High Q/V = low photon loss, high fields confined to small modal volume
- ✓ High spatial overlap of emitter with cavity field

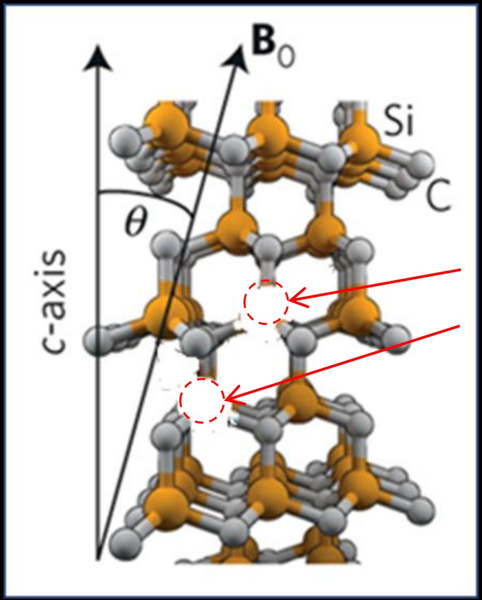


Pragmatically, Q = $\frac{\lambda}{\delta\lambda}$

$$V \sim \left(\frac{\lambda}{n}\right)^3$$

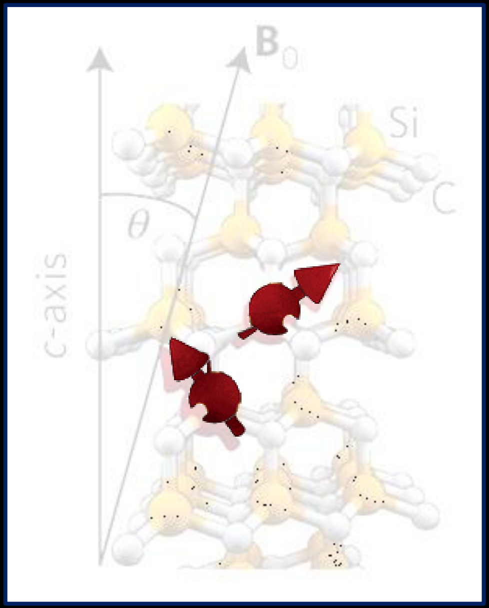
WHAT IF we could create qubits…

- Having long spin coherence times.
- Where spin states could be *initialized* and *read out* by photons.
- Where we could create the qubits in their own protective, isolating environment.



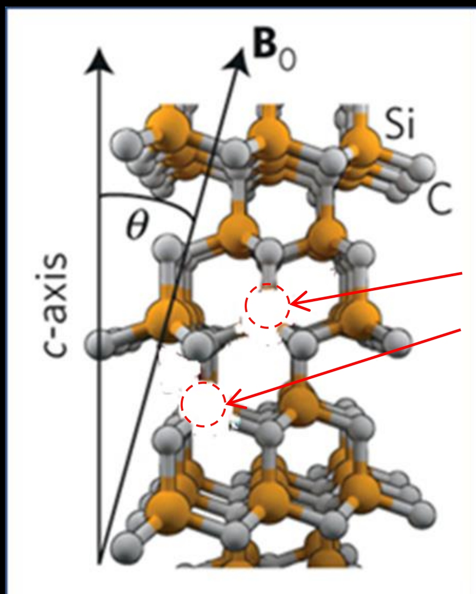
Based on Widmann et al., *Nat. Mat.* 2015

Si vacancies



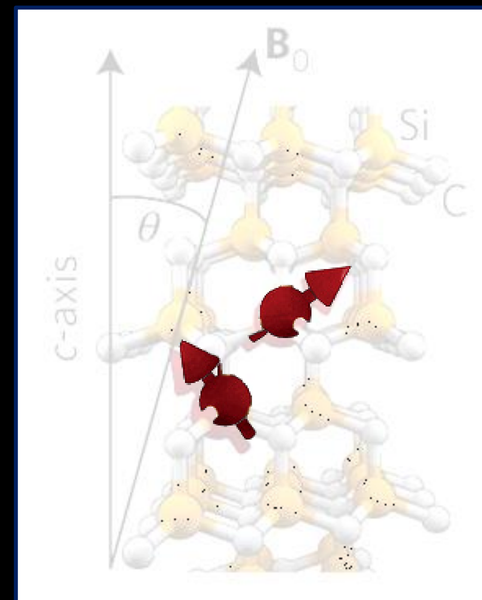
"inverted atoms"

Silicon Vacancies (SiVs) in 4H-SiC



Si vacancies

Based on Widmann et al., *Nat. Mat.* 2015



2 inequivalent Si vacancy sites,
h (V1); k (V2)

Atomic-scale entities with distinctive spin and electronic states…
within a wide bandgap material, isolating the defects from electronic interactions

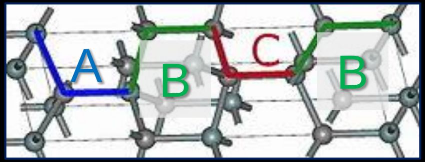
The rich possibilities of SiC

Multiple polytypes (3C, 4H, 6H ~ 250 total): different "stacking sequences" of the Si-C tetrahedron.

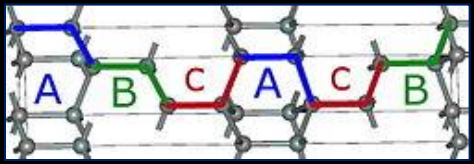
- Proven value for high-power transistors, LEDs
  ➡ material readily available with high quality crystalline quality.
- Materials processing approaches fairly well understood.

SiC wafers, CREE

4H



6H

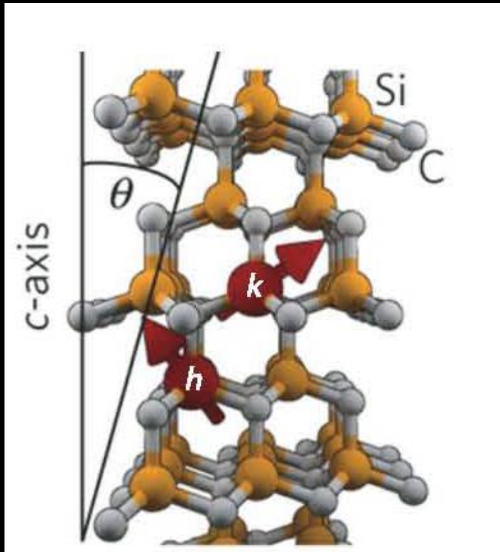


New photonic-spin source:

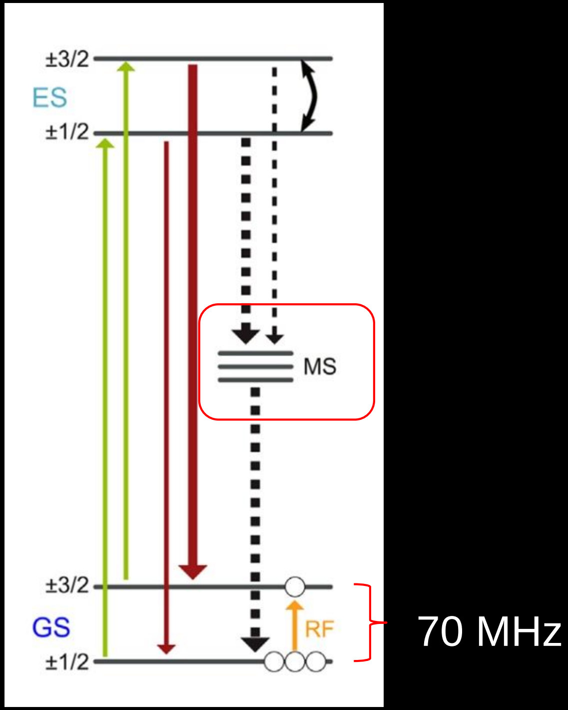
- Spin-coherent defects have been observed at room temperature in all major polytypes (e.g. millisec coherence times for divacancies in 4H SiC*).
- A treasure trove of optically- and spin-active defects that extend the spin possibilities and spectral range of devices.

* Christle et al., *Nat. Mat.* (2015)

Coupled Electronic-Spin Energy Levels



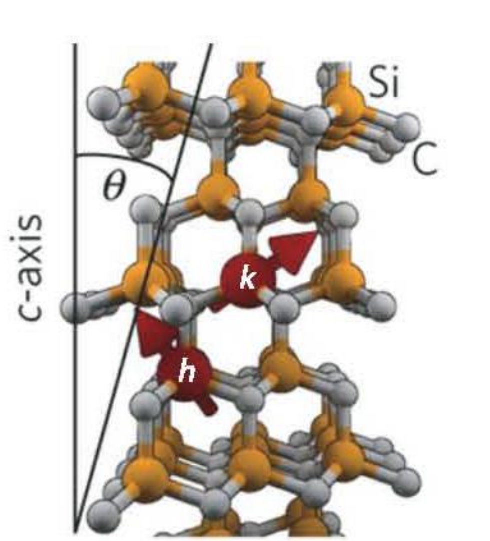
M. Widmann, et al.
Nature Materials, 2015.



Tarasenko et al.,
Phys. Stat. Sol. B (2018)

- Negatively-charged vacancies have S = 3/2
- Metastable state (MS) and selection rules for electronic transitions -> correlation between photon intensity and spin state
- Optically Detected Magnetic Resonance (ODMR) to prepare or read-out spin states.

Coupled Electronic-Spin Energy Levels



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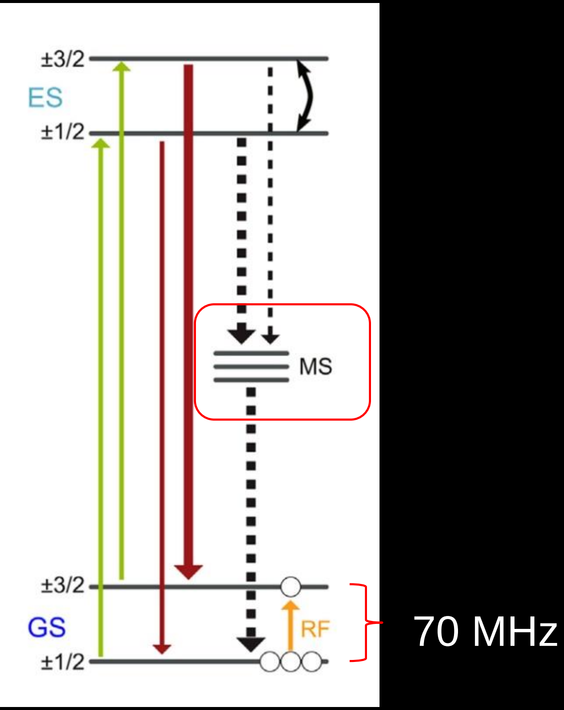


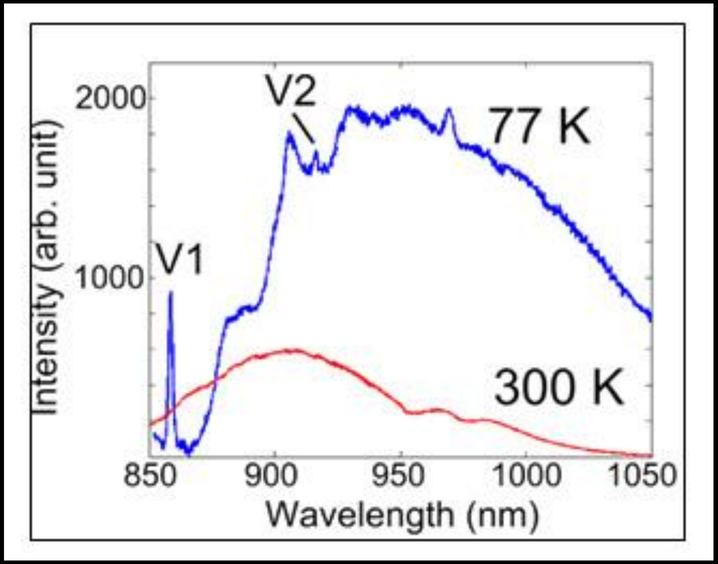
Tarasenko et al.,
Phys. Stat. Sol. B (2018)

Intimate coupling of spin and photon

- Preparation and read-out of spin states
- A “flying” qubit that can carry qubit signals across long distances

We’ll focus on optical properties

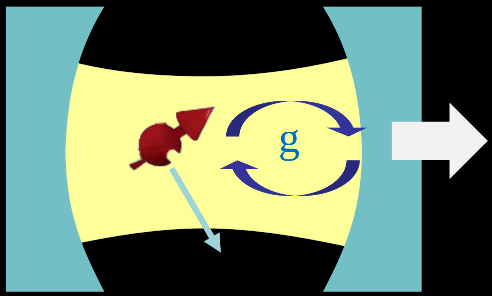
The Optical Spectrum of the SiVs



- Photoluminescence spectrum shows broad phonon sideband at 300 K
- 77 K, two *zero phonon lines* (ZPLs) are visible (labelled V_1 and V_2) corresponding to the two sites
- The fraction of light emitted into the ZPLs is ~ just a few percent

We use the integration of an optical cavity to bring out the full potential of the SiV

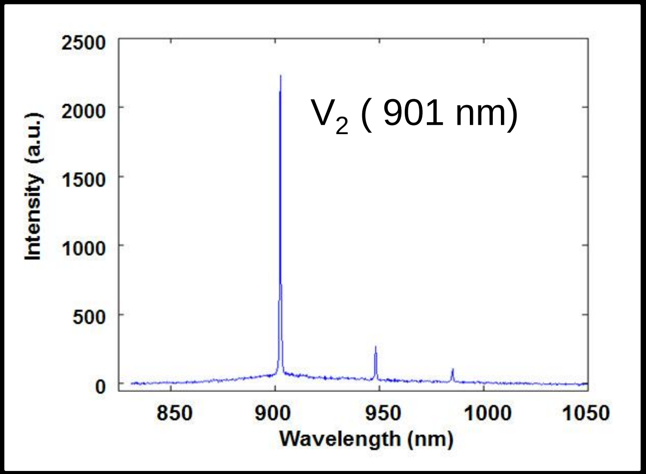
The Power of a Matched, High Quality Optical Cavity



Photon loss
$\kappa \sim 1/Q$

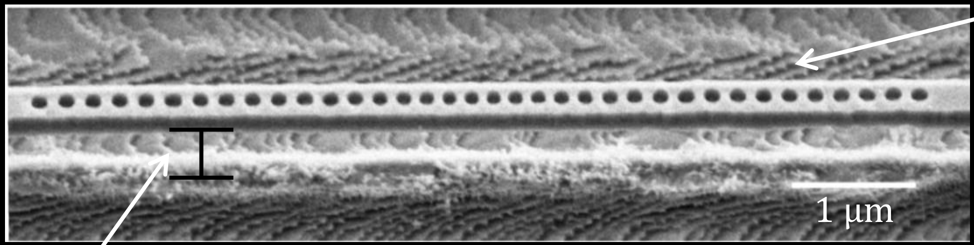
γ = spontaneous decay

$Q \sim \lambda/\delta\lambda \sim 5000$



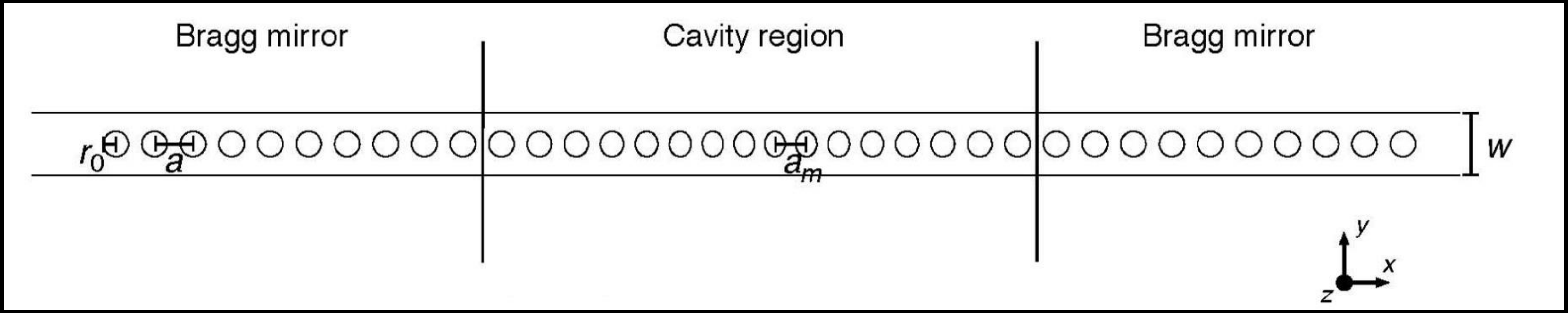
V_2 within untuned cavity at room temperature

The Design of the Cavity

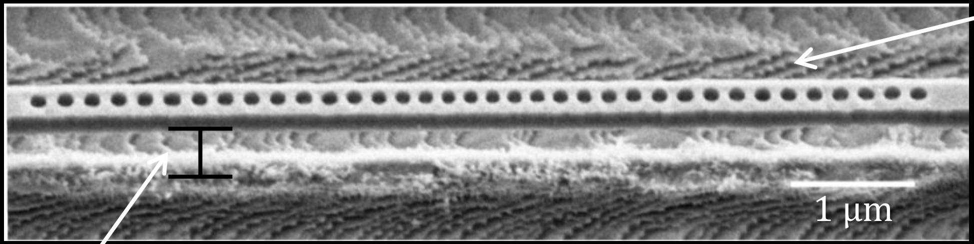


Etched holes modulate index of refraction & set up standing waves (modes) of light at particular wavelengths

Optical isolation prevents loss of light to substrate



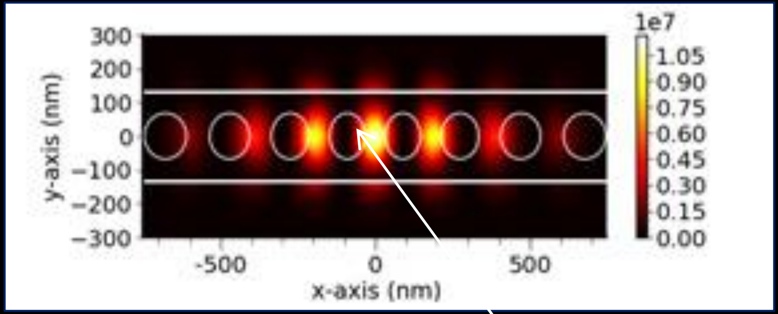
The Design of the Cavity



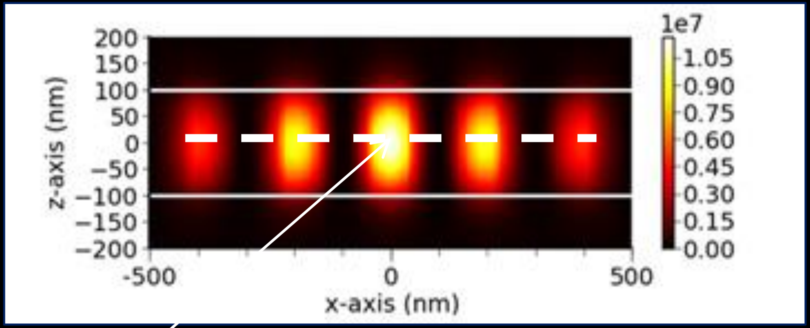
Etched holes modulate index of refraction & set up standing waves (modes) of light at particular wavelengths

Optical isolation prevents loss of light to substrate

Simulated field intensity profile (Fundamental TE mode)



Top-down



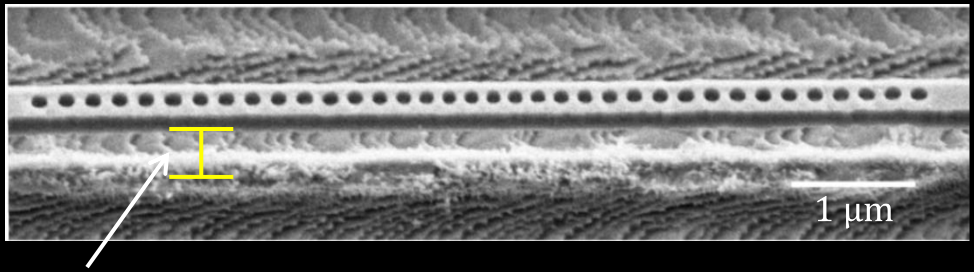
Side view

SiV placed here

The Fabrication Challenges

Beginning with the bulk materials, how do we sculpt an optical nanocavity…

- With good optical isolation? (undercut structures)
- High fabrication fidelity at the nanoscale?
- Correct spatial overlap of defects to modes?
- Ultra-low fabrication-related damage to preserve the defect centers? (avoiding energetic ion beam processes)



Optical isolation prevents loss of light to substrate

Begin with homo-epitaxial SiC, incorporate a wet chemical selective etch

➡ mitigate strain and defects from lattice mismatched heterostructures

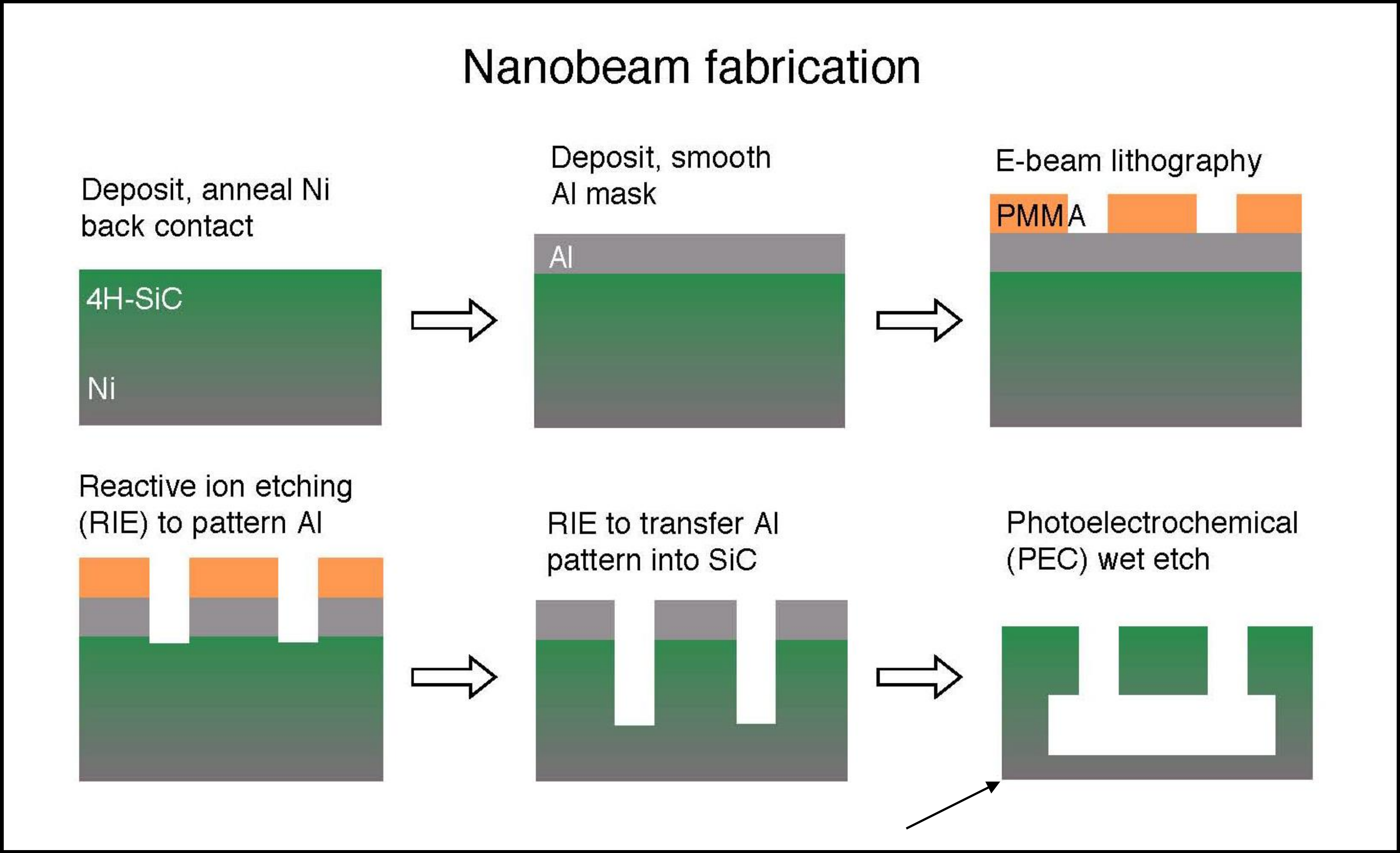
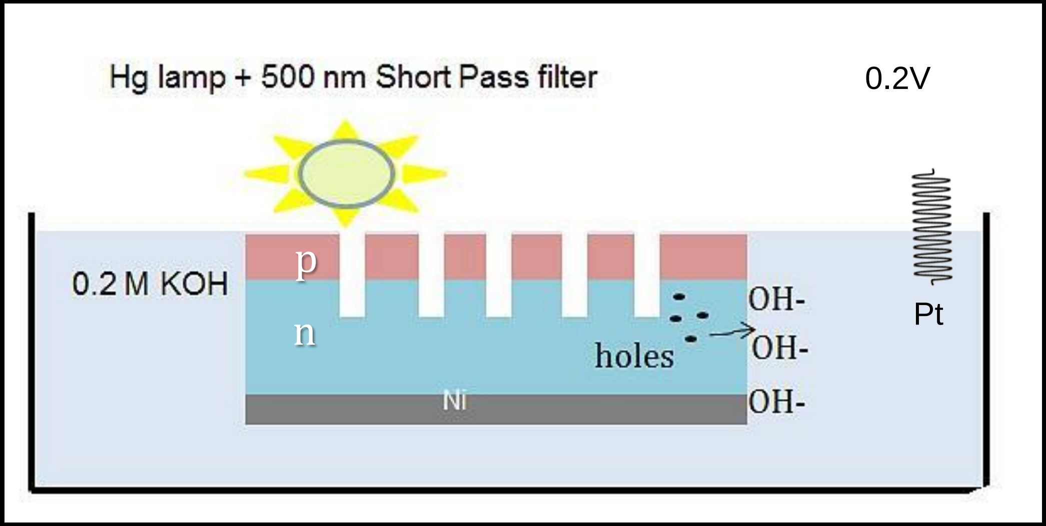


Photo-driven, dopant-selective wet etch for optical isolation

Dopant-selective Photo-Electrochemical Etch

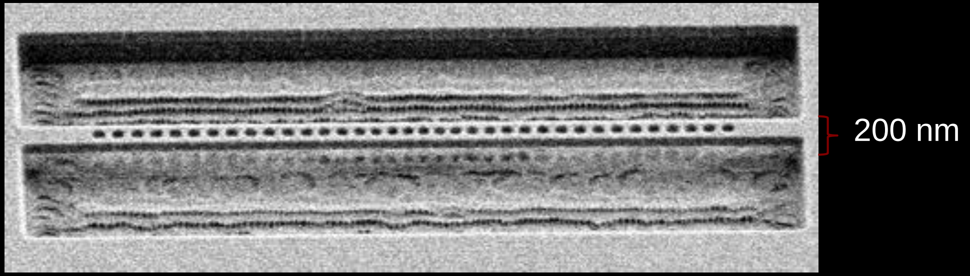


Bracher & Hu, *Nano Letters* (2015)

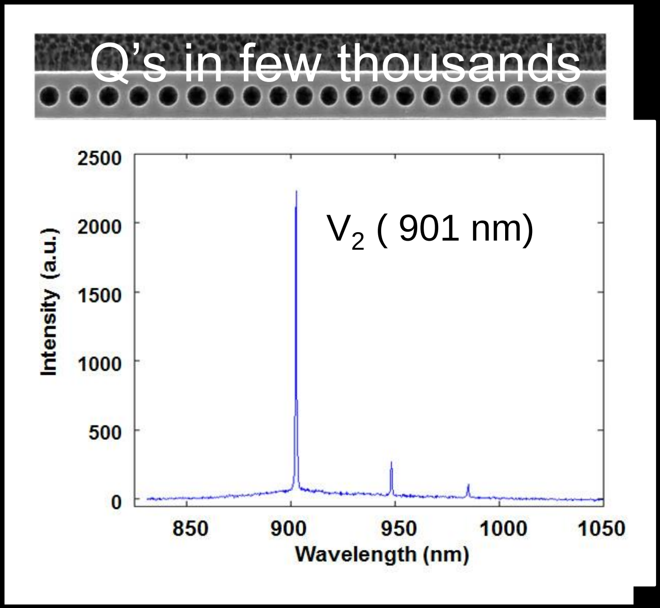
Undercut-isolate nanocavity through selective Photo-Electrochemical Etch of the n-SiC: photo-generated holes drive chemical etch process

$$SiC + 8OH^{-} + 6h^{+} \rightarrow \left[Si\,(OH)_2 O_2\right]^{2-} + CO + 3\,H_2O$$

Shor et al, *APL* **60** (1992);
Dorp et al., *J. Micromechanics & Microeng.*.**17** (2007)



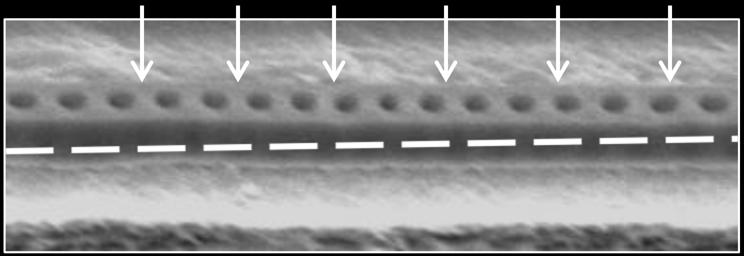
Final Implantation/Creation of SiVs in Completed Cavities



V$_2$ within untuned cavity at room temperature

Fabricated Q's ~ 2 x 10 ^{4}

Implantation of C_{12} ions at 70 keV

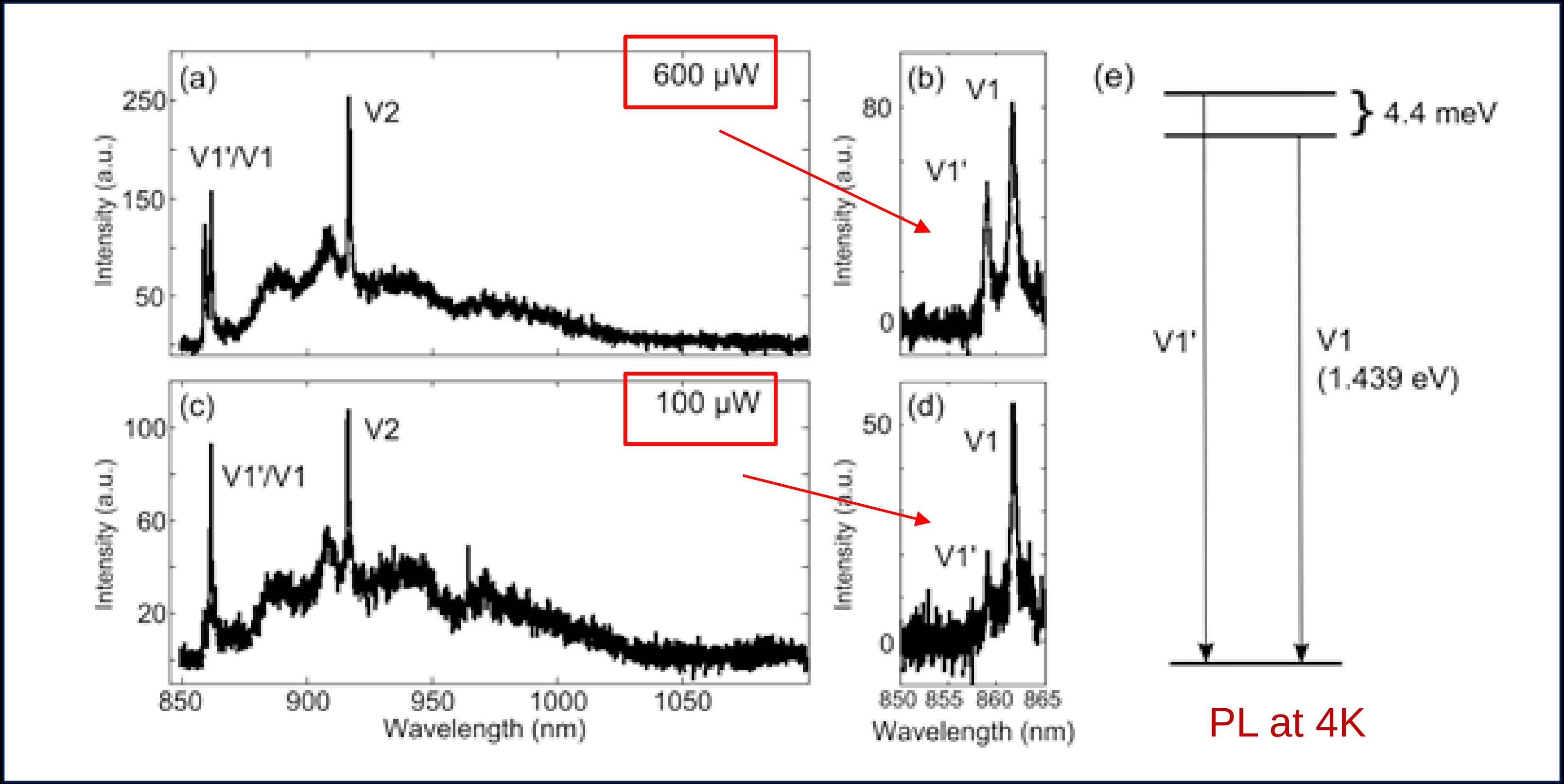


- Ion implantation & electron irradiation to create SiV's
- Ion dose: $10^{12}/cm^2$, $10^{13}/cm^2$
- Initial results without post-implant anneal

What insights can we gain?

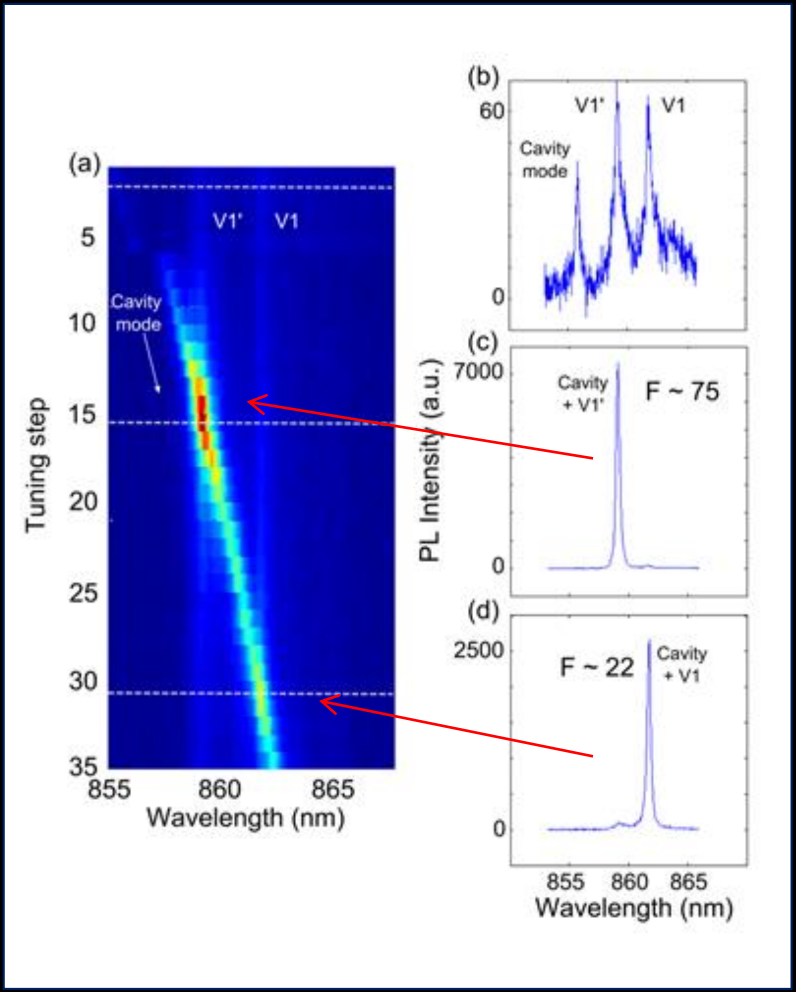
The Untuned (non-resonant) Cavity Shows V1 to have two peaks

Bracher et al., *PNAS* 2017



- Relative fraction of V1', V1 is <u>temperature</u> (power)-dependent
- Higher temperature (power) populates more of V1' excited state
- Learn more by TUNING the cavity into resonance with V1', V1

Tuning the cavity into resonance with V1’, V1

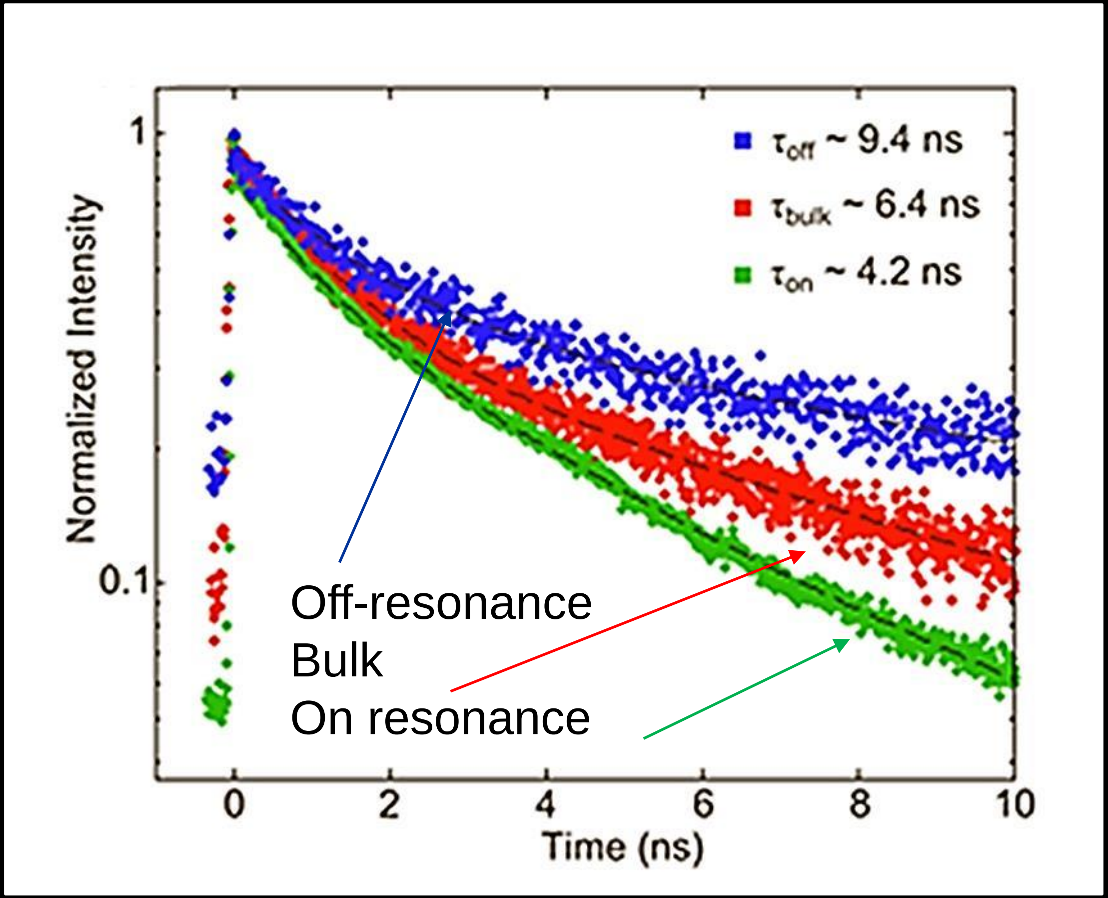


Q ~2200

600 μW excitation:

- Successive tuning of mode into resonance with V1’, then V1
- Tuning carried out through calibrated adsorption of N_2 onto cavity
- Purcell enhancement as high as 75 for V1’; 22 for V1
- Maximum of ~ 50% emission into ZPL on resonance (compared to ~1%)

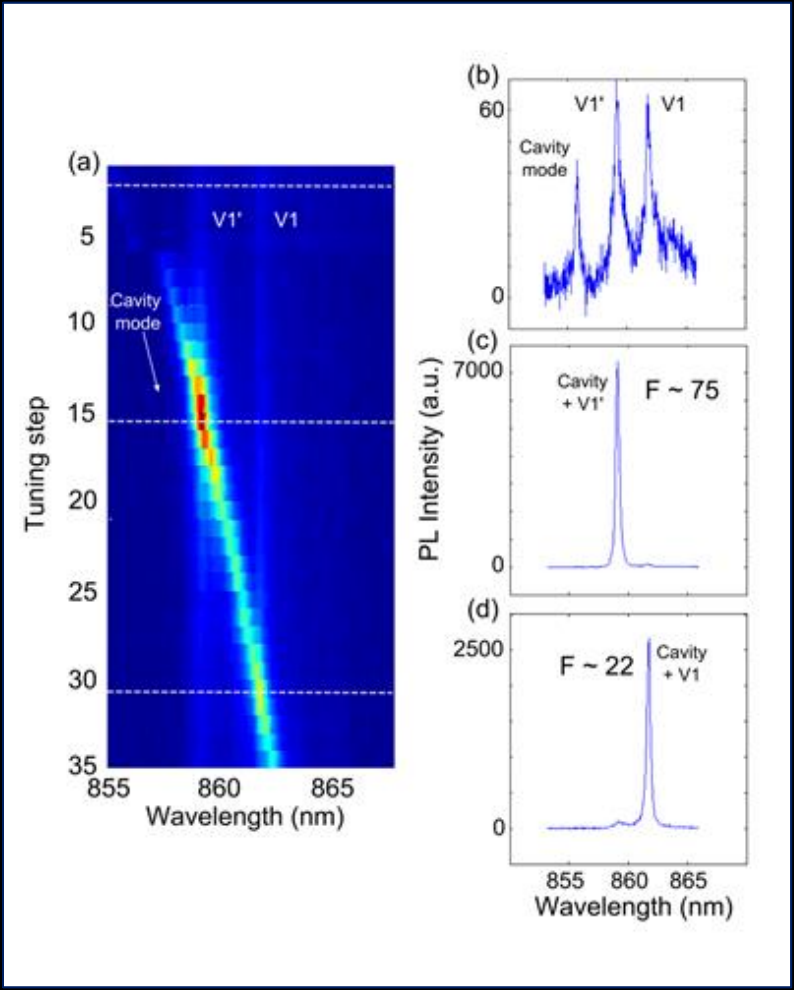
Cavity Modification of Spontaneous Emission Lifetime



Hoped for modification of lifetimes

But wait…there are some important questions remaining.

Differences in Enhancement, V1', V1



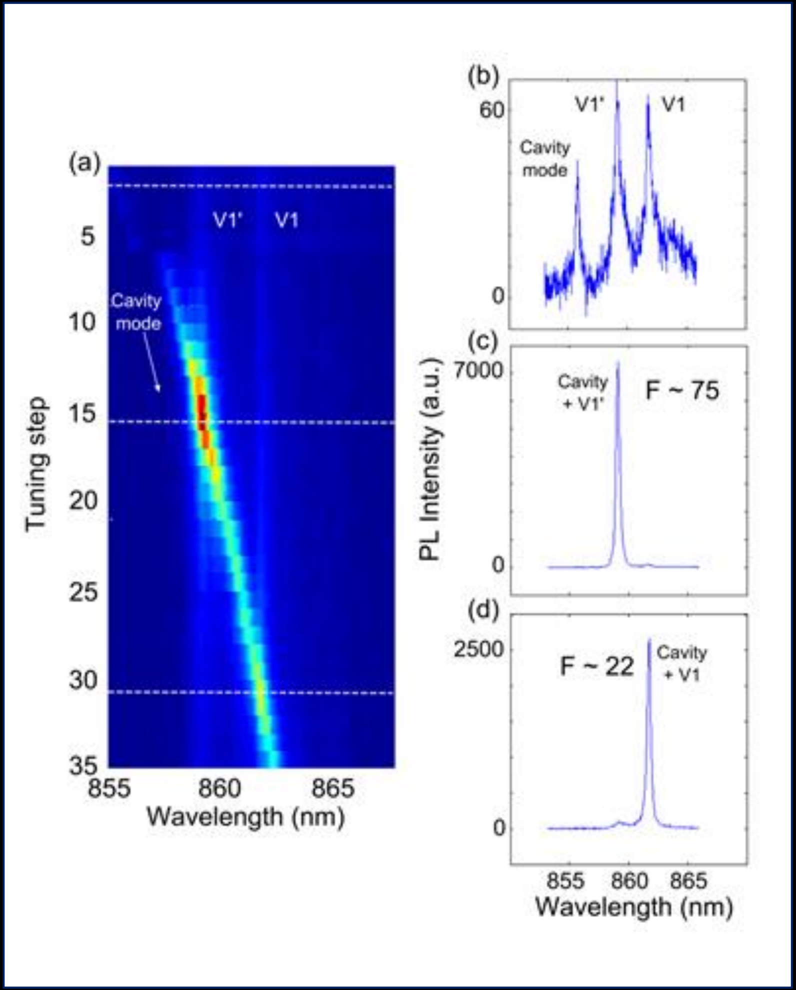
WHY the differences in ENHANCEMENT for V1', V1?

- Assuming same SiV defect, with 2 different excited states…
- Interaction of defect with cavity must be the same for both states….

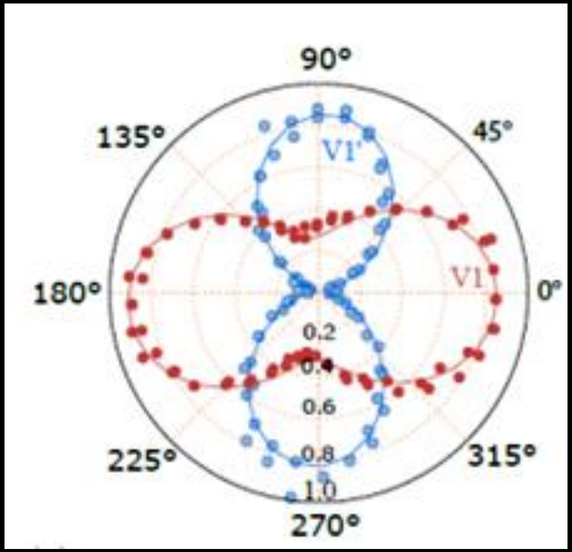
$$F = \frac{\Gamma_{cav}}{\Gamma_0} \sim \frac{Q}{V}\left[\frac{\vec{\mu}\cdot\vec{E}(r)}{|\mu||E_{max}|}\right]^2$$

- Perhaps there is a different orientation of V1, compared to V1'

Differences in Enhancement, V1', V1



Measurement of field polarization



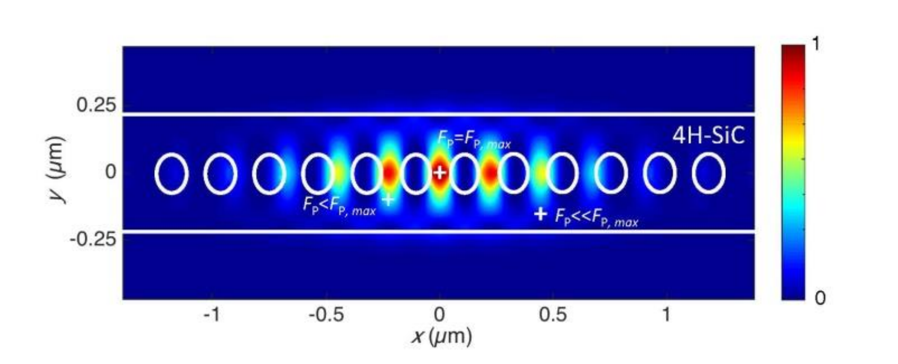
- V1' aligned perpendicular to c-axis; V1 weakly aligned parallel to c-axis*
- We can deduce V1 30º to axis

Cavity provides information about atomic environment

* Wagner et al., *PRB* 2000

What are the next challenges?

Deterministic placement of defects at maximum of cavity field

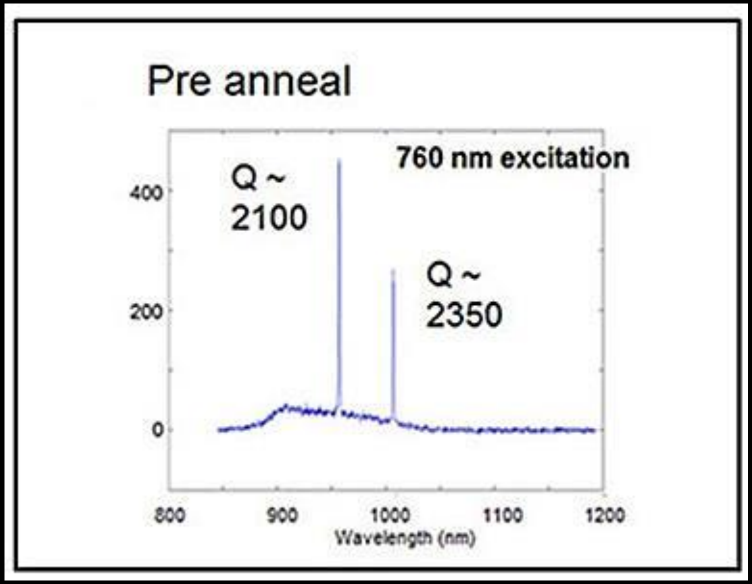


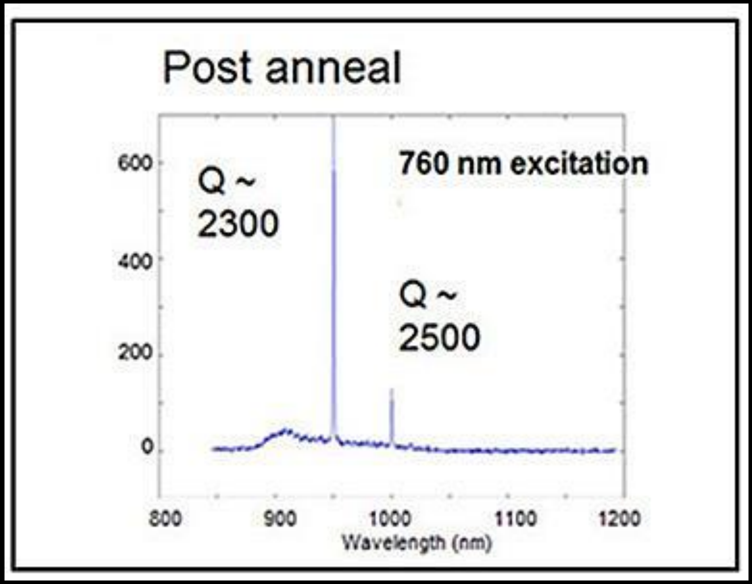
For cavity with Q ~ 2200, if defects were "perfectly positioned", should obtain F ~ 340 (rather than 75)

- Local introduction of defects by FIB, at correct energy and dose
  - Stochastic scattering
  - Difficulty in obtaining single defects at field maximum
- An unusual possibility:

What Happens When We ANNEAL the Cavities?

C implantation, 10^{12} /cm^2 , 70 keV





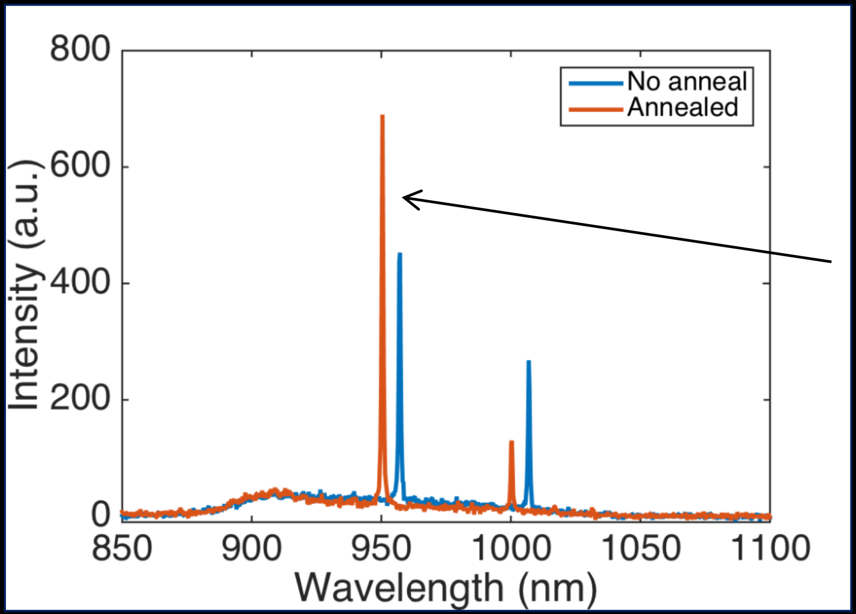
- No anneal: clean spectrum, distinct modes, reasonable Q

AFTER ANNEAL (750C, half hour)

- Slightly higher Q's: anneal removes residual damage
- Change in INTENSITY (~1.5 x) , indicates change in OVERLAP of SiVs with modes

  (therefore, change in POSITION of defect)

Diffusive Motion of Defects Within 4H-SiC Cavities



“blue-shift” due to small dimensional changes in cavity, producing shifts in the modes

(different cavity)

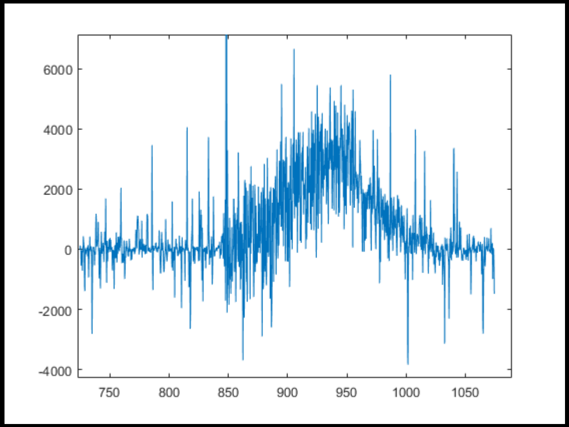
A stochastic process, analyzed for scores of cavities:

- Q of ~ all modes increased, indicating a general improvement in material quality.
- The background luminescence decreases 2x – 3x, indicating some removal of defects
- Modes typically change in brightness with respect to the background: <u>increase or decrease</u> by 10-30%
- CAN WE CONTROL THE MOTION OF THE DEFECTS& “NUDGE THEM” INTO PLACE?

SUMMARY

Presented a case study of “defect” emitters in 4H-SiC

- Showed substantial enhancement of optical signature
- Tuned cavity revealed some details of the V1 excited state and its dipole orientation
- Still coupling to ensembles of SiVs, but there may be innovative ways of achieving more deterministic placement
- A vast frontier of such photon-spin coupled structures are waiting to be characterized.



ESR signature for V1 in bulk 4h SiC

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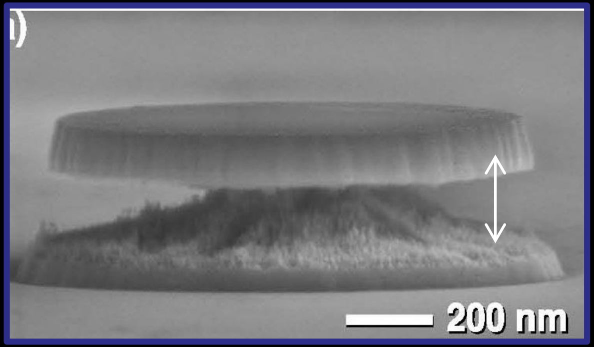
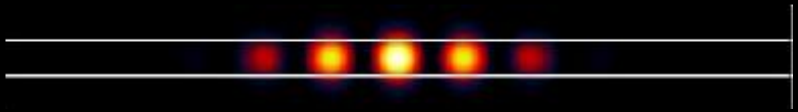
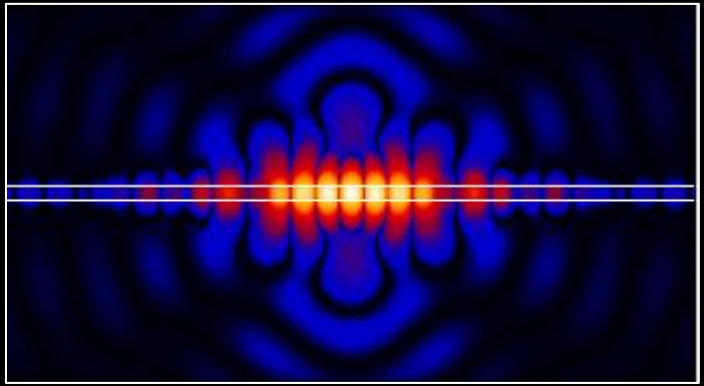
The Importance of Optical Isolation



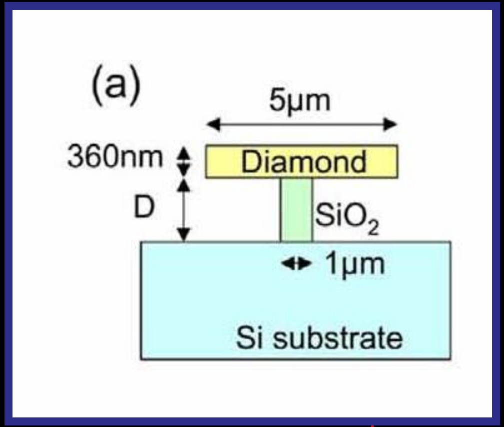
side view

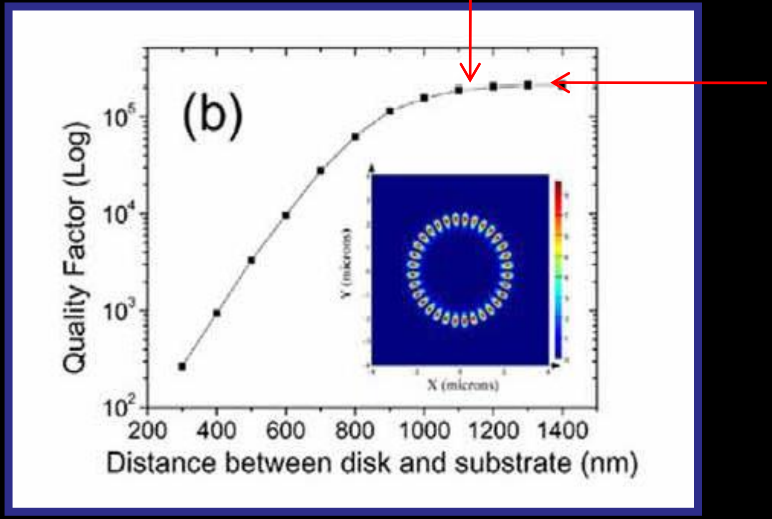




Log scale

Simulation for diamond microdisk (n = ~2.4) on Si substrate (n ~4)





C.F. Wang et al. Appl. Phys. Lett. (2007)