Covalent Defects of Carbon Nanotubes: New Class of High Purity, Indistinguishable Quantum Light Sources

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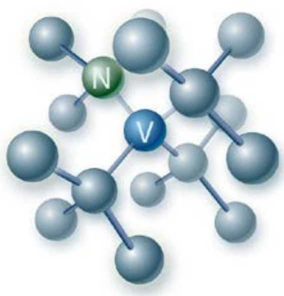
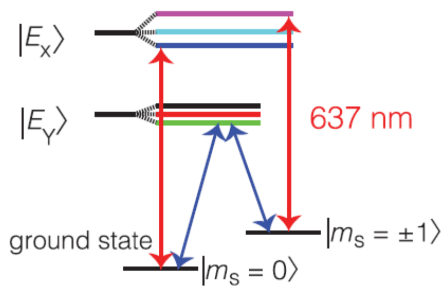
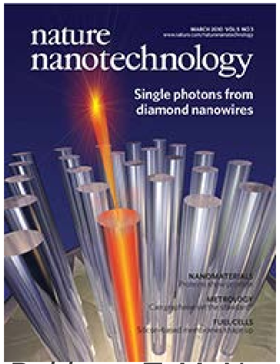
Solitary Dopants in Solid-State Systems

Control incorporation of solitary dopants → Solid state system capable of mimicking a two level atom → Single photon generation, Quantum information processing, Quantum computing with defects Sub-diffraction imaging

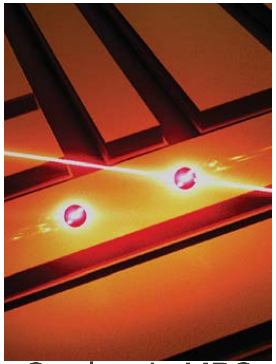
Diamond NV Centers



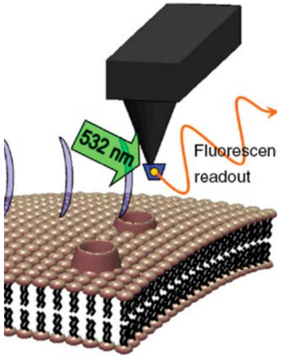




Babinec, T. M. Nat. Nano 5, 195, 2010

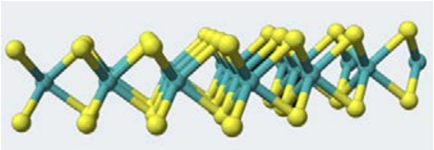


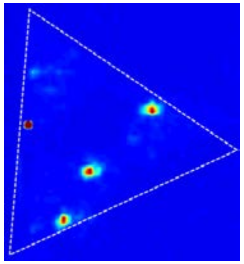
Gordon, L. MRS Bull. 38, 802, 2013

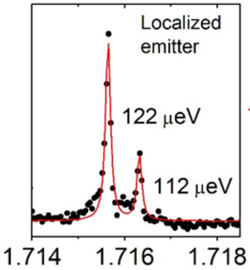


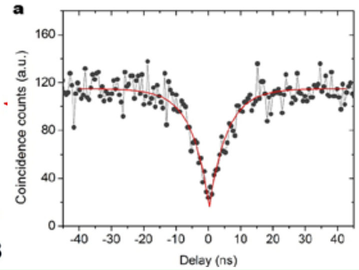
Hall, L. T. PNAS 107, 18777, 2010

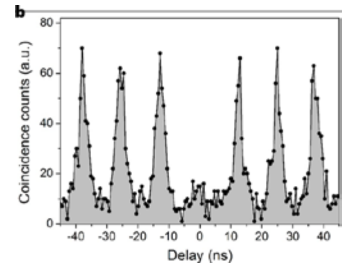
2D TMD (WSe_2)





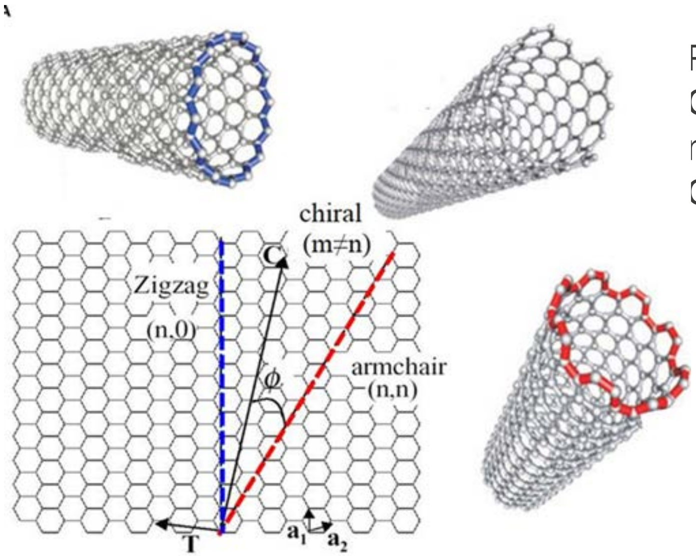




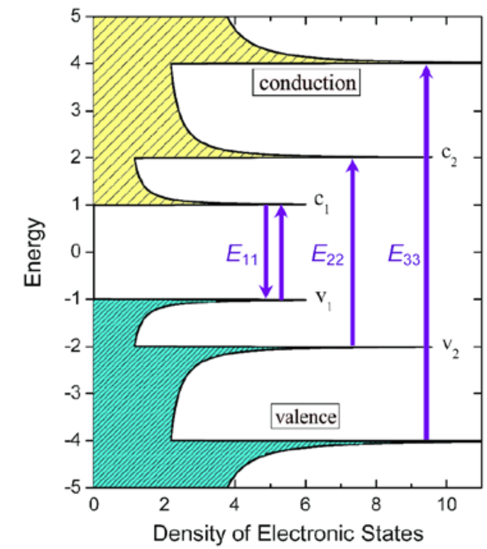


Creating quantum defect in 1D SWCNT with covalent chemistry *5);*

Intrinsic Single Wall Carbon Nanotubes



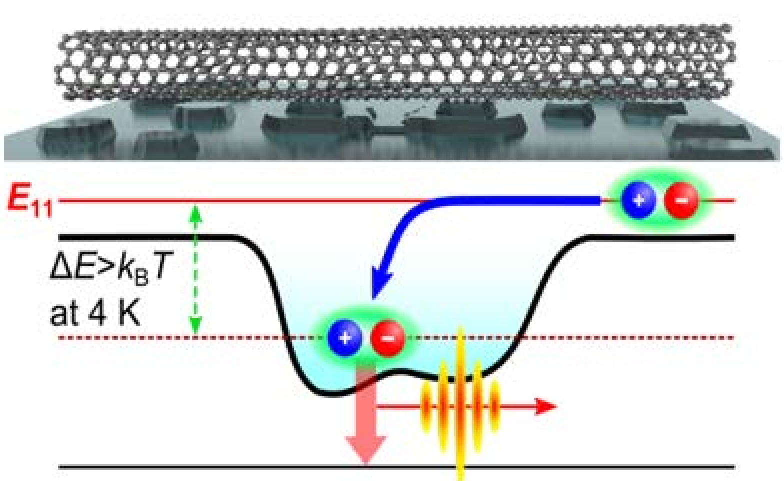
Rolled up a graphene sheet → SWCNT
Chiral indices define a SWCNT.
n=m or (n-m) is multiple of 3 → Metal
Otherwise → Semiconductors



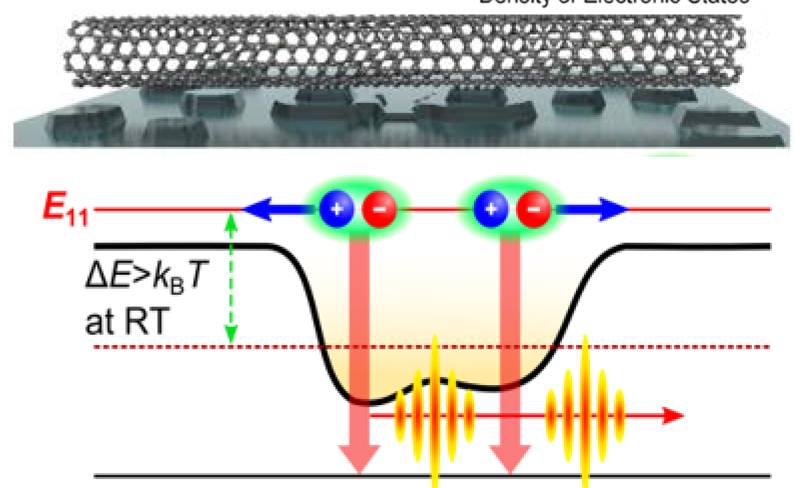
One dimensional density of state

⇩

Cannot imitate quantum mechanical properties of a trap ion.

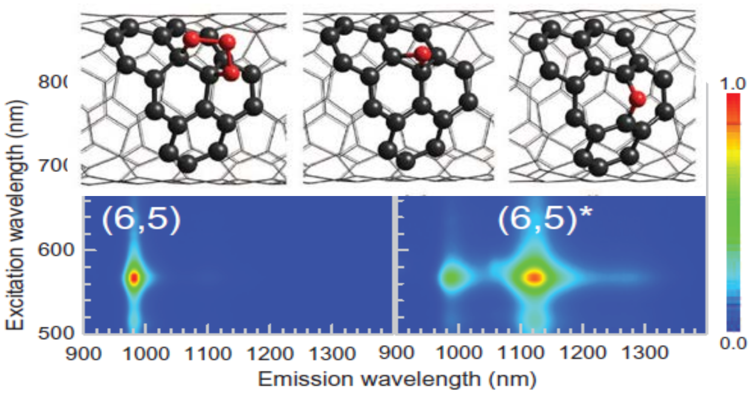


Low T → Exciton localized → quantum dot (artificial atoms)

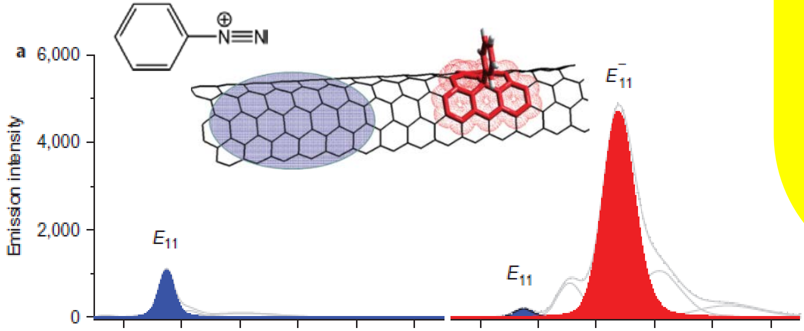


Room T → 1 D system

Defect States of Carbon Nanotubes



Ghosh et al. Science 2010, 330, 1656-1659



Y. M. Piao *et al*. *Nature Chem*. 2013, 5, 840-845.

Outstanding Questions

- Exact chemical nature of dopant.
- Electronic structures of trap-states
- Radiative and non-radiative recombination processes

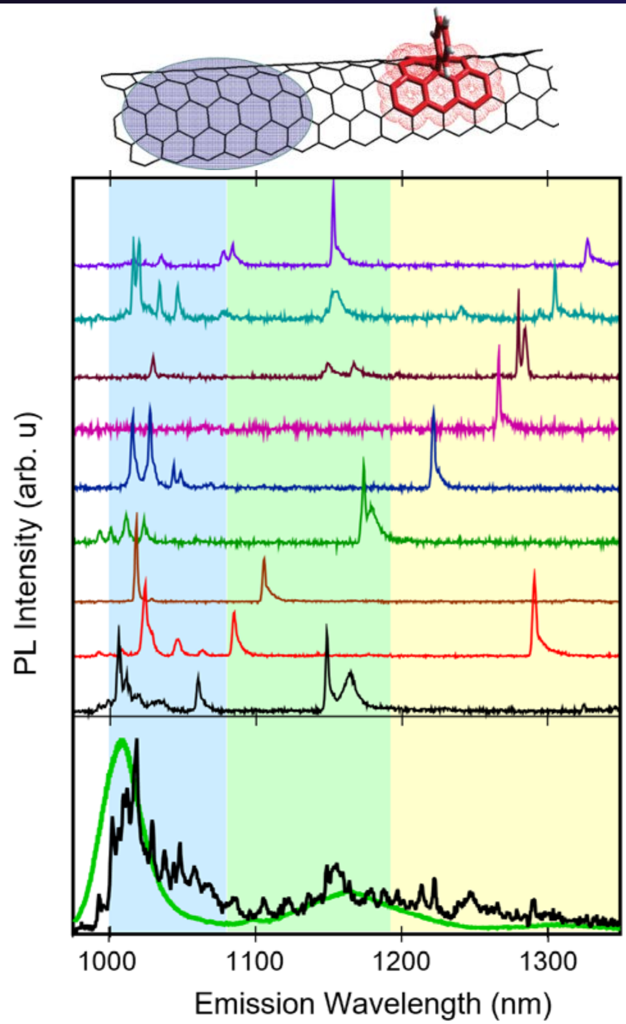
Can solitary dopant states behave as single quantum emitter at RT?

Low temperature PL, time resolve PL, & photon correlation spectroscopy studies on individual doped SWCNTs.

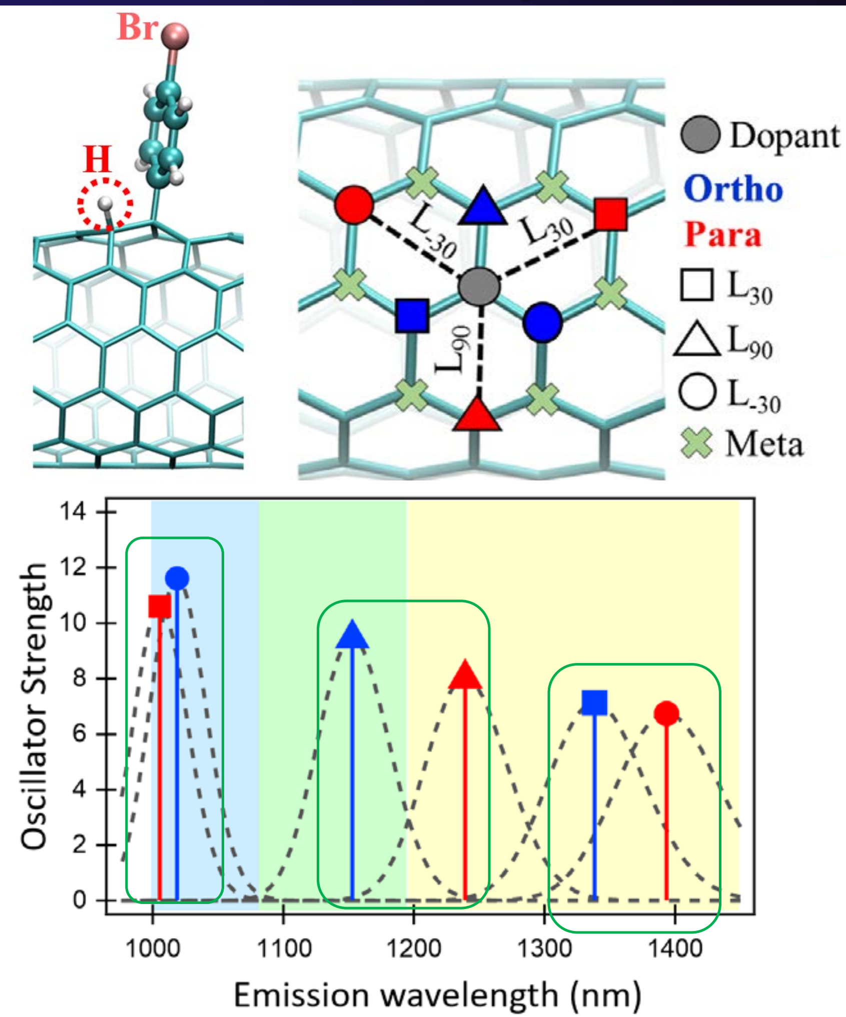
H. Kwon et al JACS 2016, 138 6878–6885

Electronic Structure and Chemical Nature

Low T single defect PL

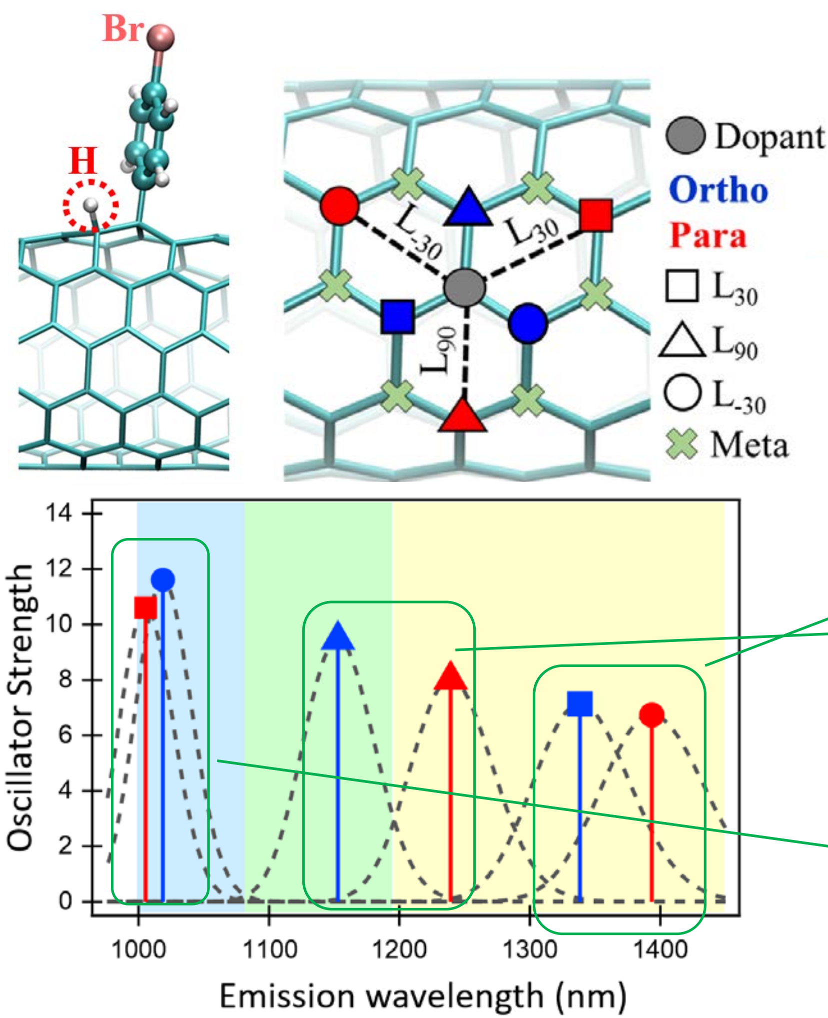


Quantum Chemistry Simulation

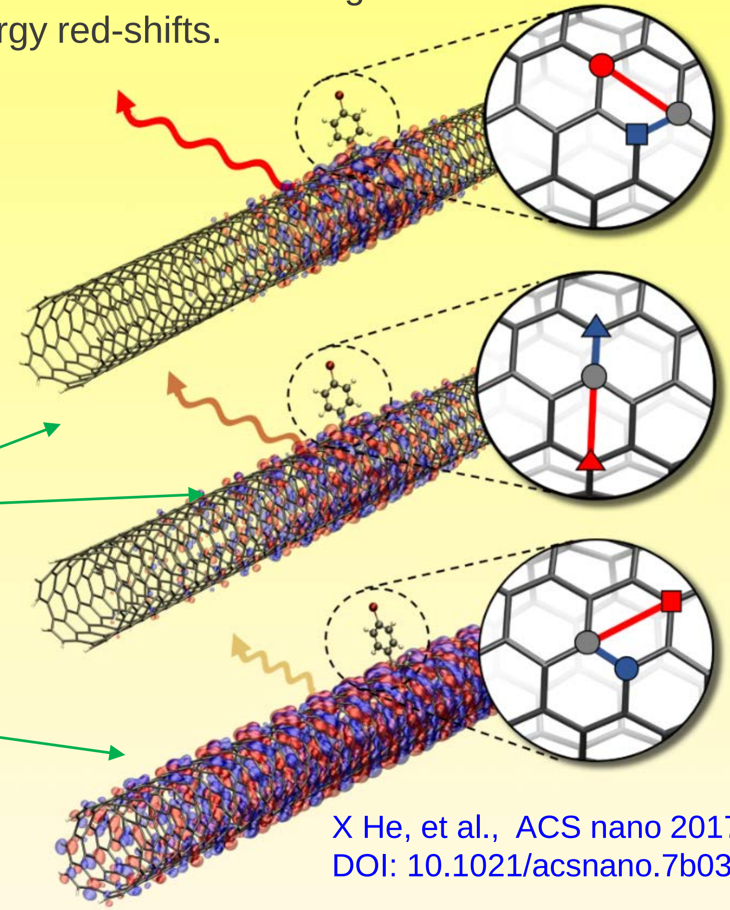


- Low T PL ⇒ Sharp isolated peak spread over 1000-1400 nm range.
- Quantum Chemistry simulation ⇒ Six different chemical configurations

Electronic Structure and Chemical Nature: Quantum Chemistry Simulation

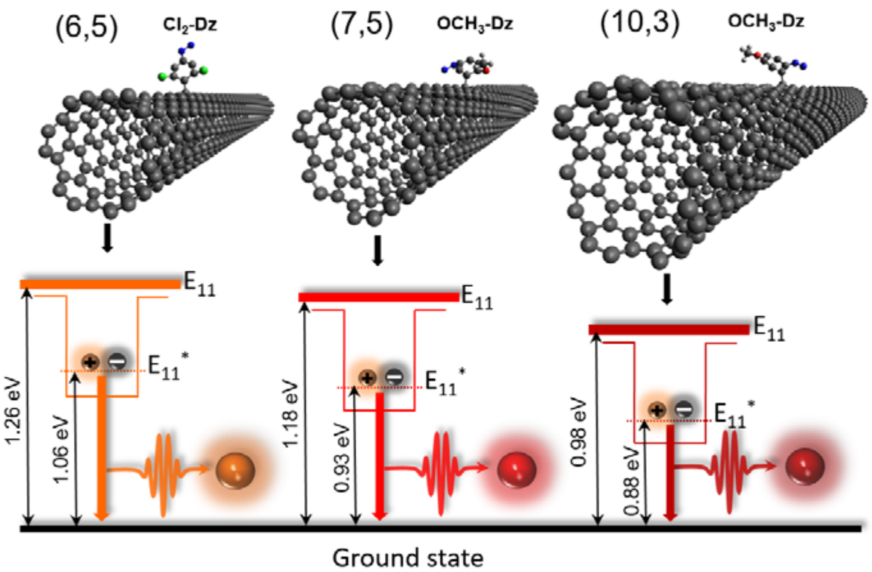


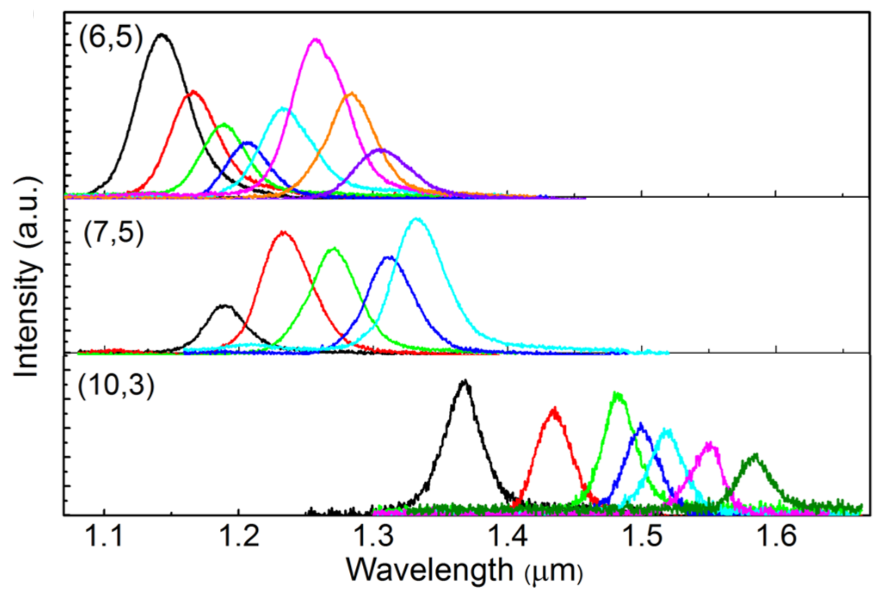
- Six distinct states spreading over 1000-1400 nm
- Exciton localization strengthen with the increase of the energy red-shifts.



X He, et al., ACS nano 2017, DOI: 10.1021/acsnano.7b03022

Tuning defect emission to 1.5 μm

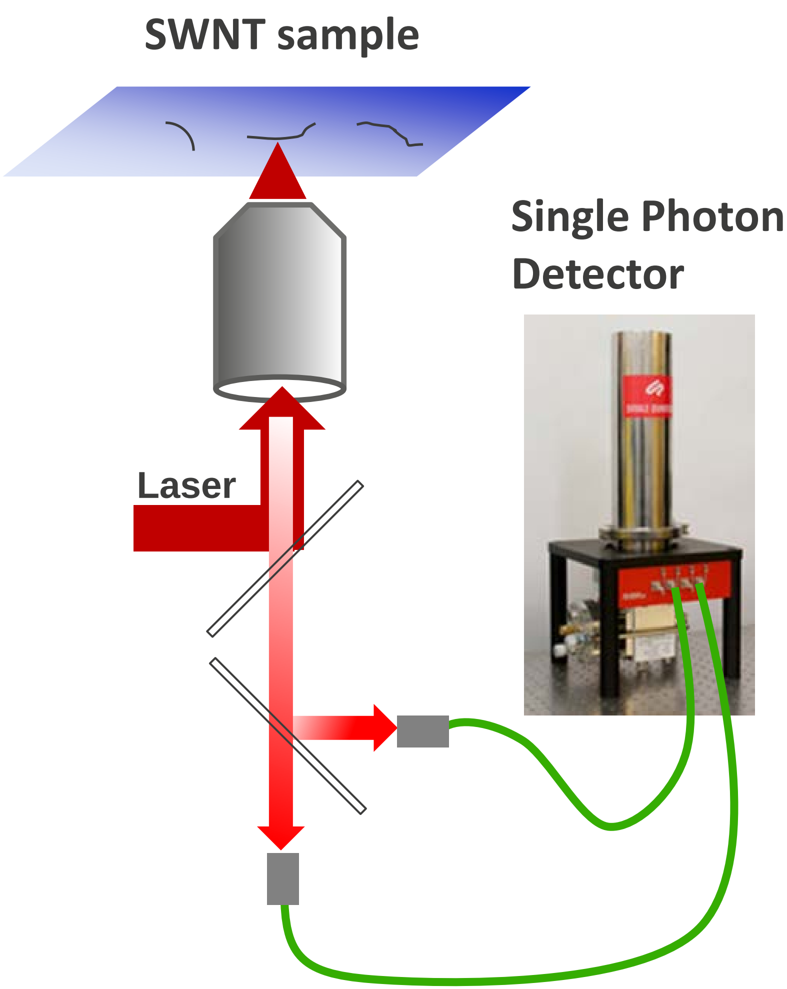




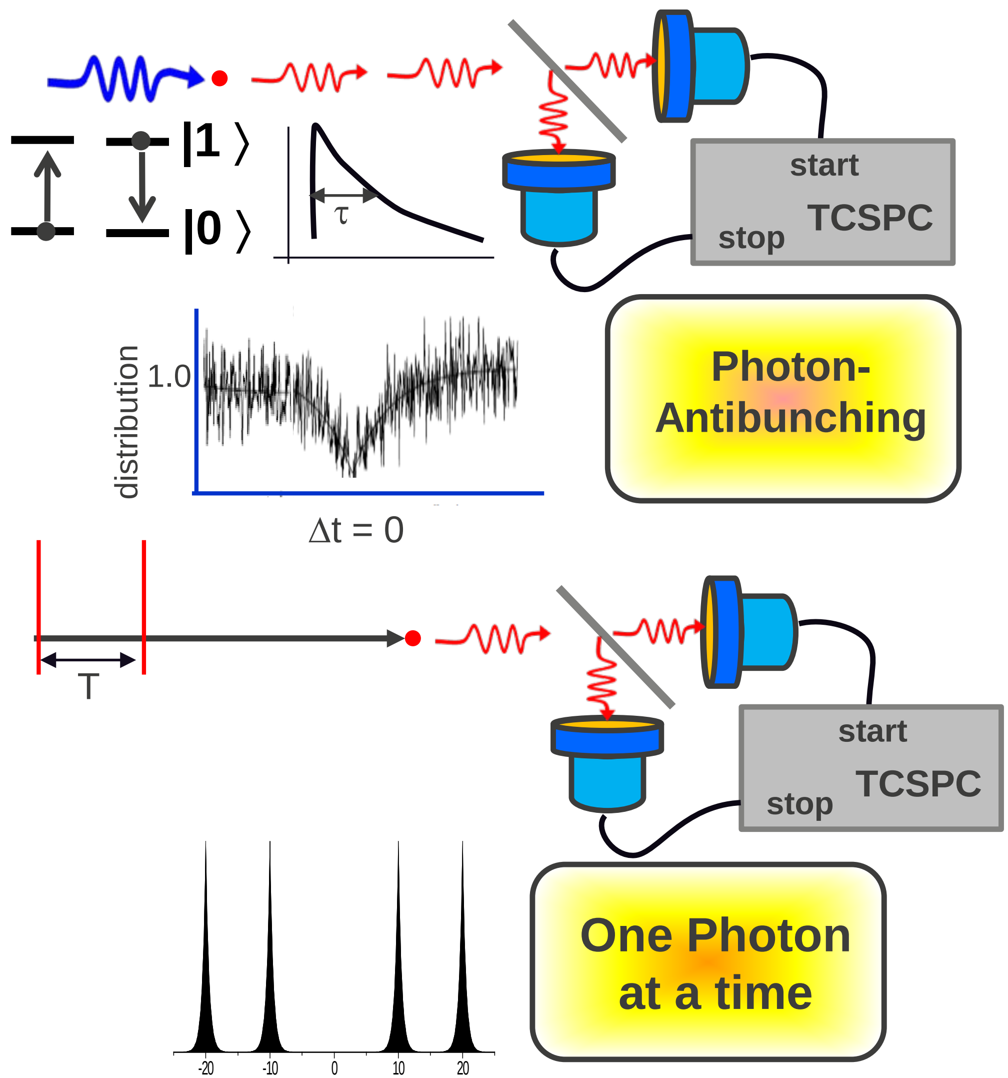
- Flexible diazonium chemistry allow easy attachment of dopants to larger diameter SWCNT
- Quantum defect states are created at 100-300 meV below the band edge

Emission of dopant state can be tuned from 1.0 to 1.6 μm
Cover both 1.3 and 1.55 μm telecommunication bands

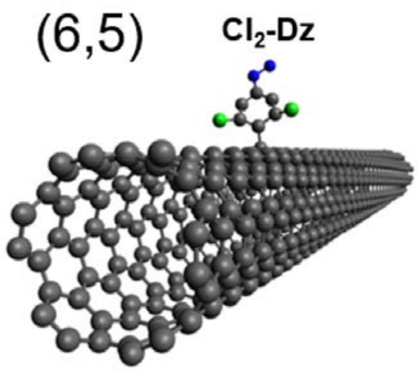
Quantum Optics Experiment: Demonstration of RT Single Photon Generation

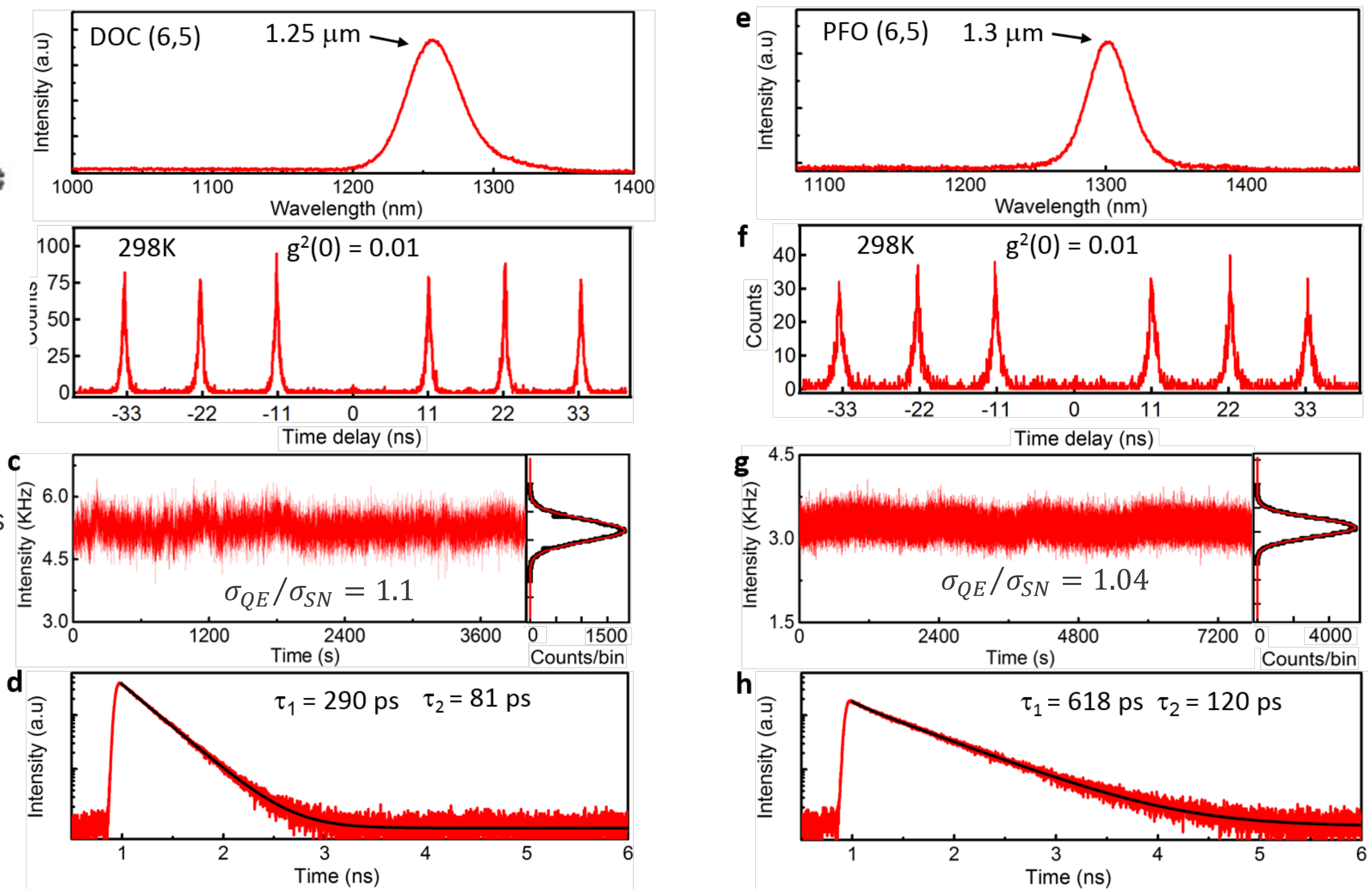


Single two-level atom



Quantum Optics Experiment: Demonstration of RT Single Photon Generation

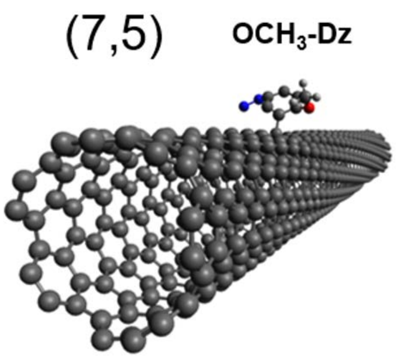


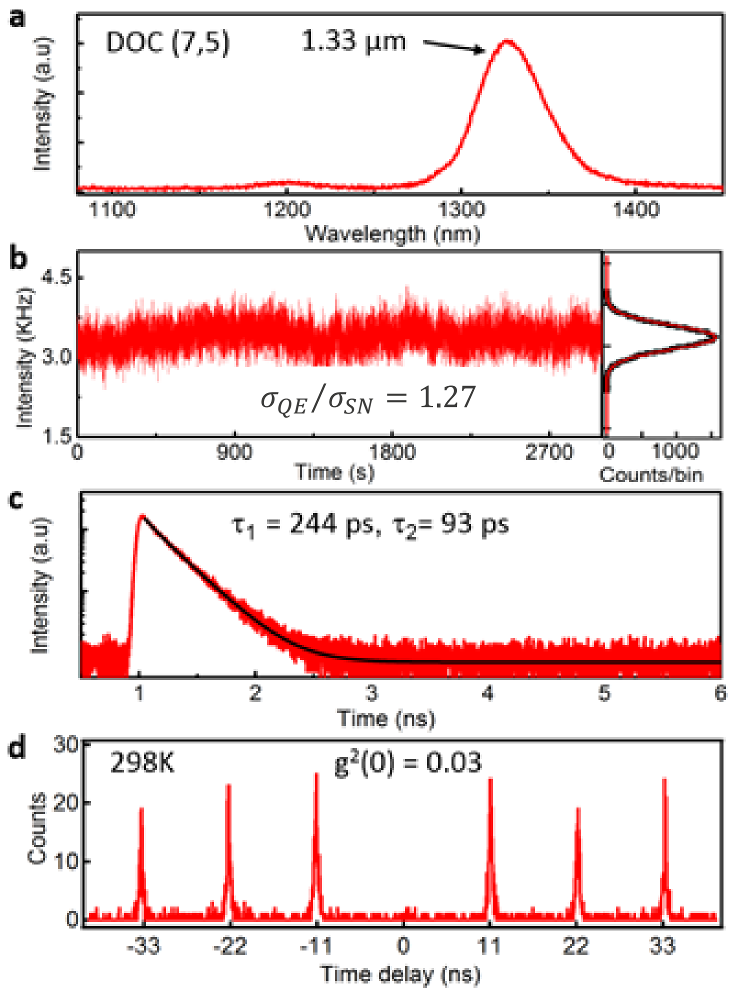


Deviation of count rate distribution of quantum emitters (QE) from the shot noise (SN) limit

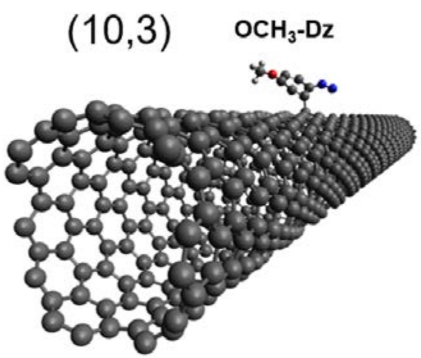
$$\sigma_{QE}/\sigma_{SN} = \sqrt{(\langle n^2\rangle - \langle n\rangle^2)/\langle n\rangle}$$

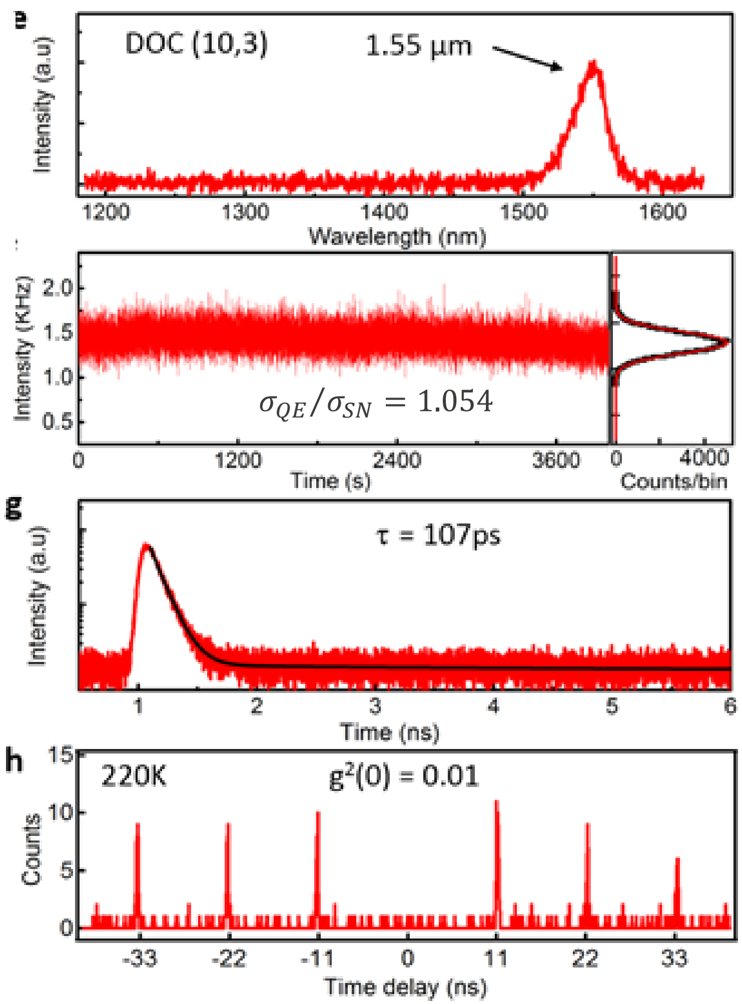
Quantum Optics Experiment: Demonstration of RT Single Photon Generation





Quantum Optics Experiment: Demonstration of RT Single Photon Generation

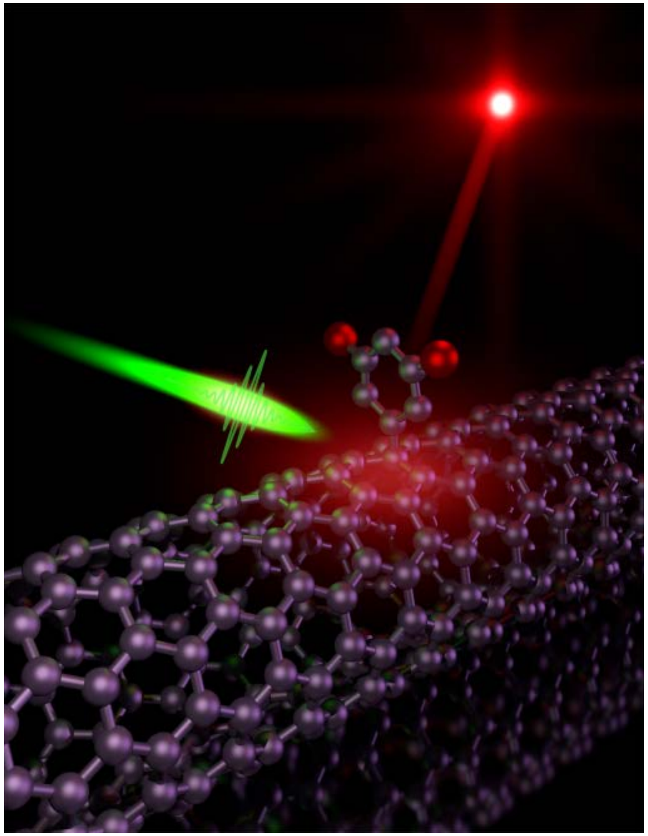




$g^{(2)}$ experiment is performed at 220K to compensate for the decrease of detector efficiency.

sp³ Defects of SWCNTs:

New Building Blocks of Quantum Information Technology



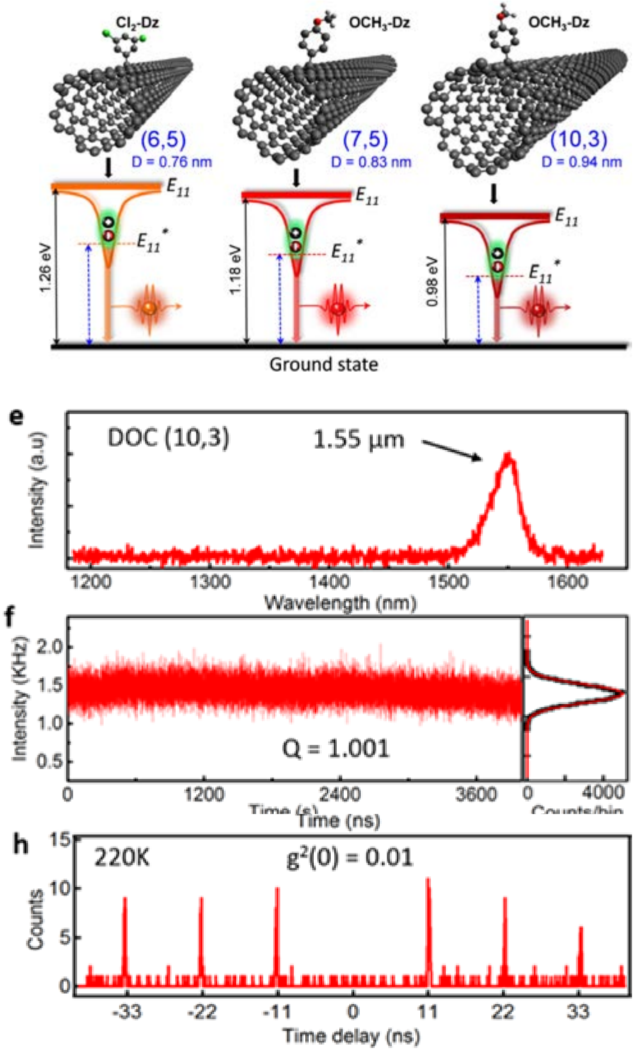
Covalent functionalization

sp³ defects

Deep trap state
130-300 meV
below band gap

An artificial quantum
two level system

Single photon generation
at RT & 1.55 μm

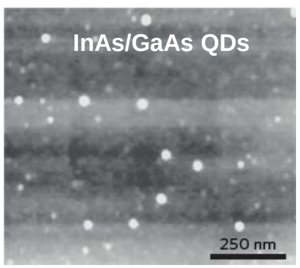


- Xiaowei He, et al., ***Nat. Photonics*. 11**, 577-582 (2017).
- Xiaowei He, et al., ***Nat. Mater*. 17**, 663-670 (2018)
- Avishek Saha et al., ***Nat. Chem***. 10, 1089 (2018)
- Stephen. K, Doorn, H. Htoon, S. Tretiak;, ***Handbook of Carbon Nanomaterials***, pp. 143-189 (2019)

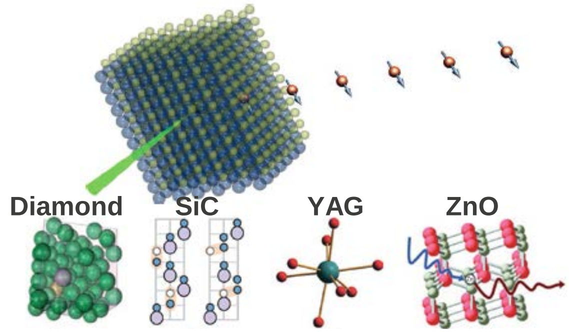
Solid-State Single Photon Emitters (SPE)

Quantum light sources or Single Photon Emitters → A stream of photons with controllable quantum correlations. → Quantum key distribution, quantum computing, quantum metrology
Imaging beyond diffraction limit

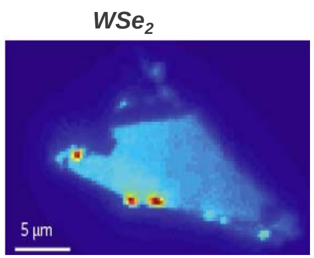
Quantum Dots

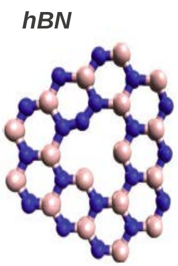


Defects in 3D host and nanocrystals



Defects in 2D host





Igor Aharonovich, Dirk Englund and Milos Toth, Nature Photonic, 10, 631, 2016 and refs. therein.

	InAs Quantum Dot	Diamond NV	Defects in 2D
Count Rate	10^7 Hz	10^6 Hz	10^5 Hz
Temperature	4K	RT	4K
Wavelength	1.3 -1.5 µm	Visible <1.0 µm	Visible <1.0 µm
Integration	Not directly compatible with Si micro-fab		

Indistinguishable Single Photons

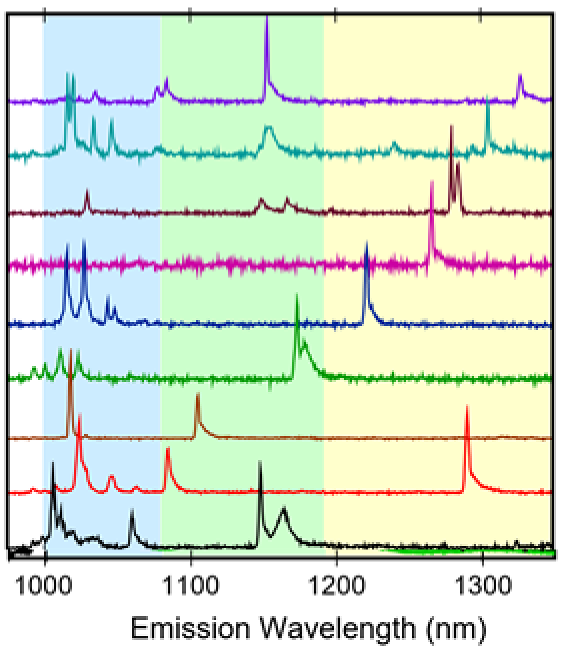
- Spectrally indistinguishable photons are required for quantum computing and quantum repeaters.
- Indistinguishability ⇒Ultra-narrow spectral linewidth of emitters

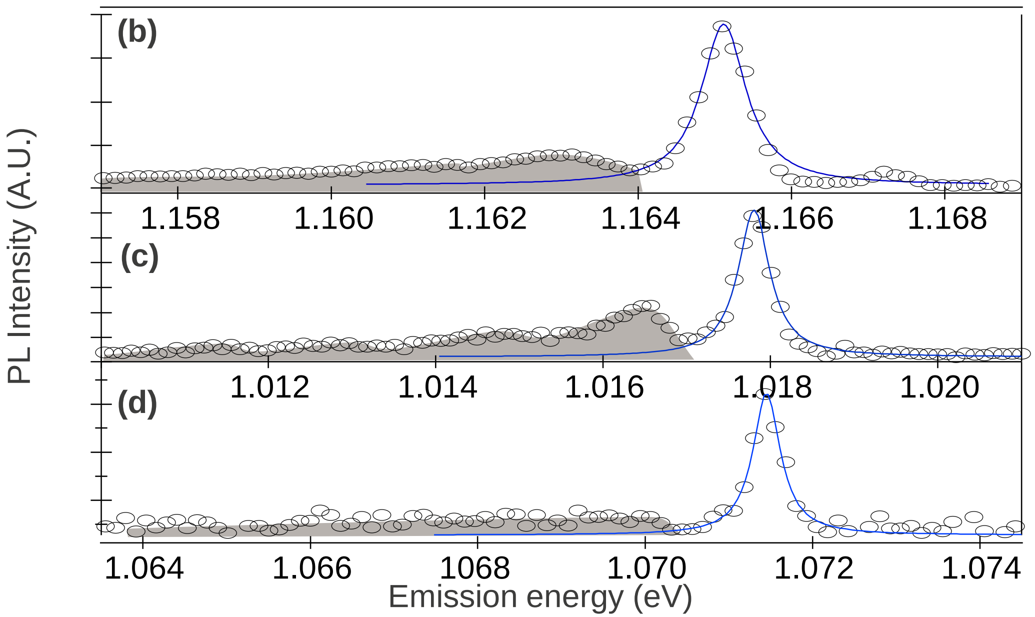
$$\Delta E = \frac{2\hbar}{T_2} = \frac{\hbar}{T_1} + \frac{2\hbar}{T_2^*}$$

T_2: Over all dephasing time; T_1: population decay time; T_2^*: pure dephasing time.

- Traditional approach: Isolate the quantum emitter from environment
⇒ Shut down non-radiative decay channels ⇒ Minimize pure dephasing time

Shape and Width of Spectral Lines



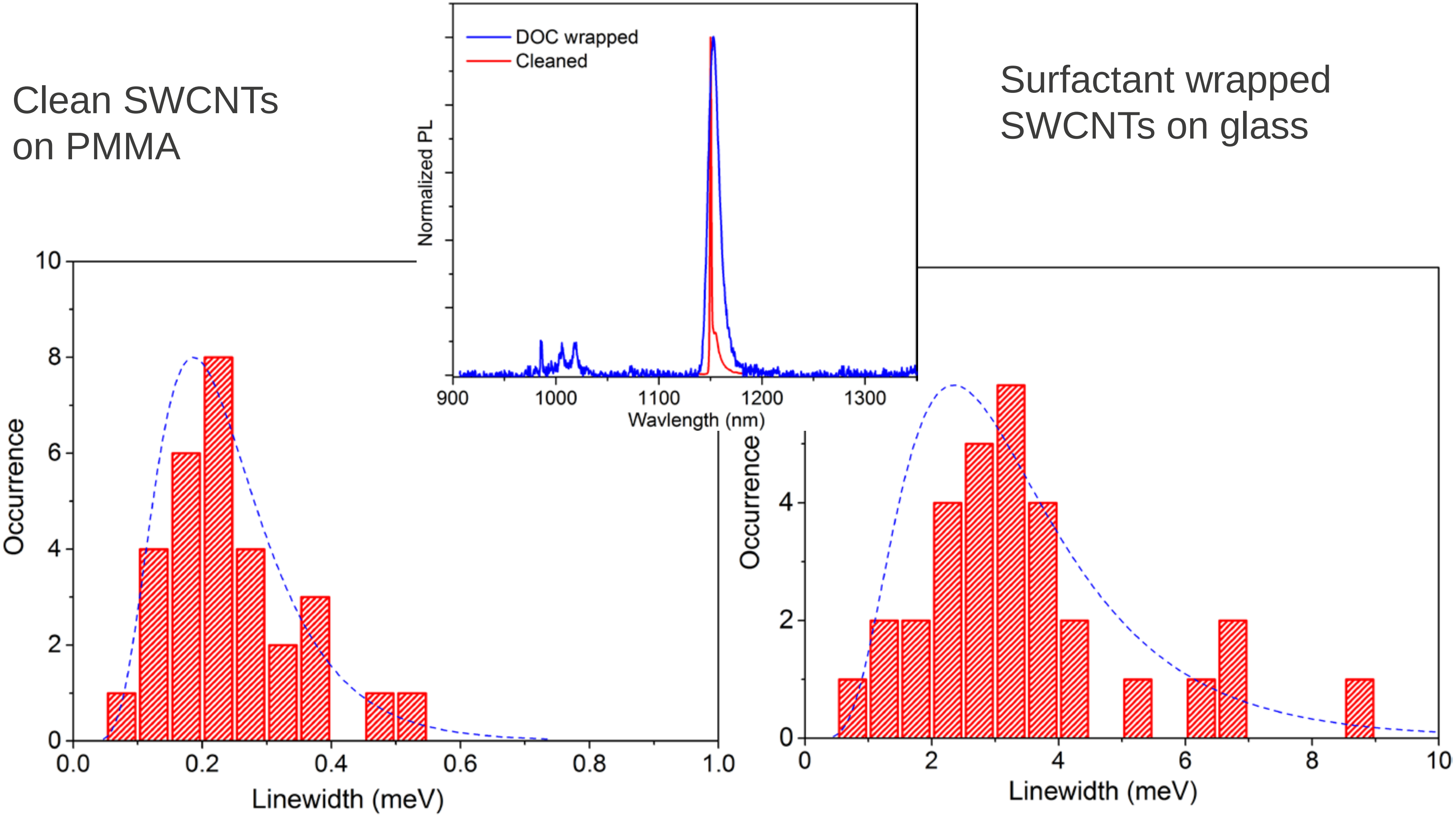


- Low T PL spectra of polymer wrapped SWCNTs
  → Zero phonon line and acoustic phonon side band.
- ZPL Line widths of 200-400 μeV

Shape and Width of Spectral Lines

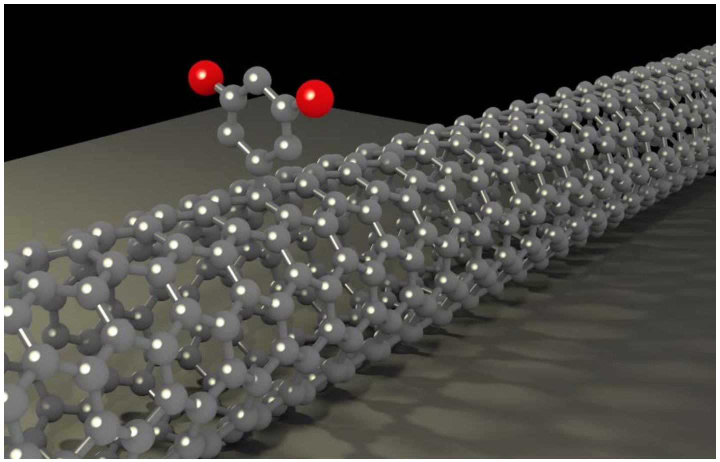
Clean SWCNTs on PMMA

Surfactant wrapped SWCNTs on glass

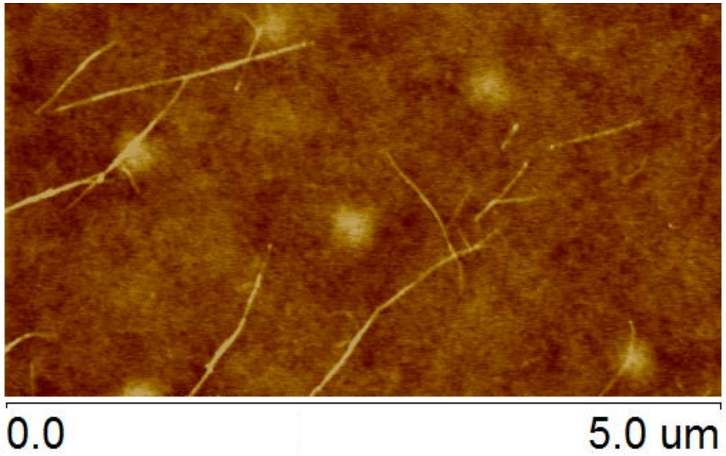


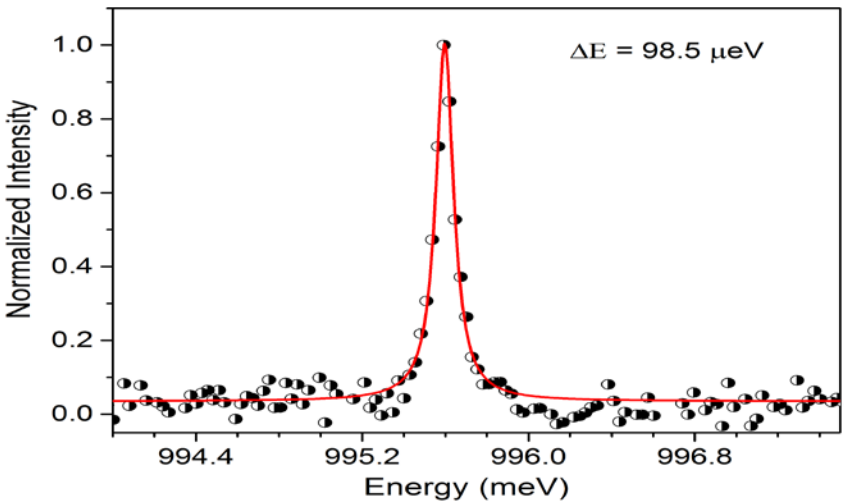
- Environment has strong effect on linewidths

Shape and Width of Spectral Lines



Surfactant (DOC) wrapped (6,5) tubes on Polymer (PS) substrate





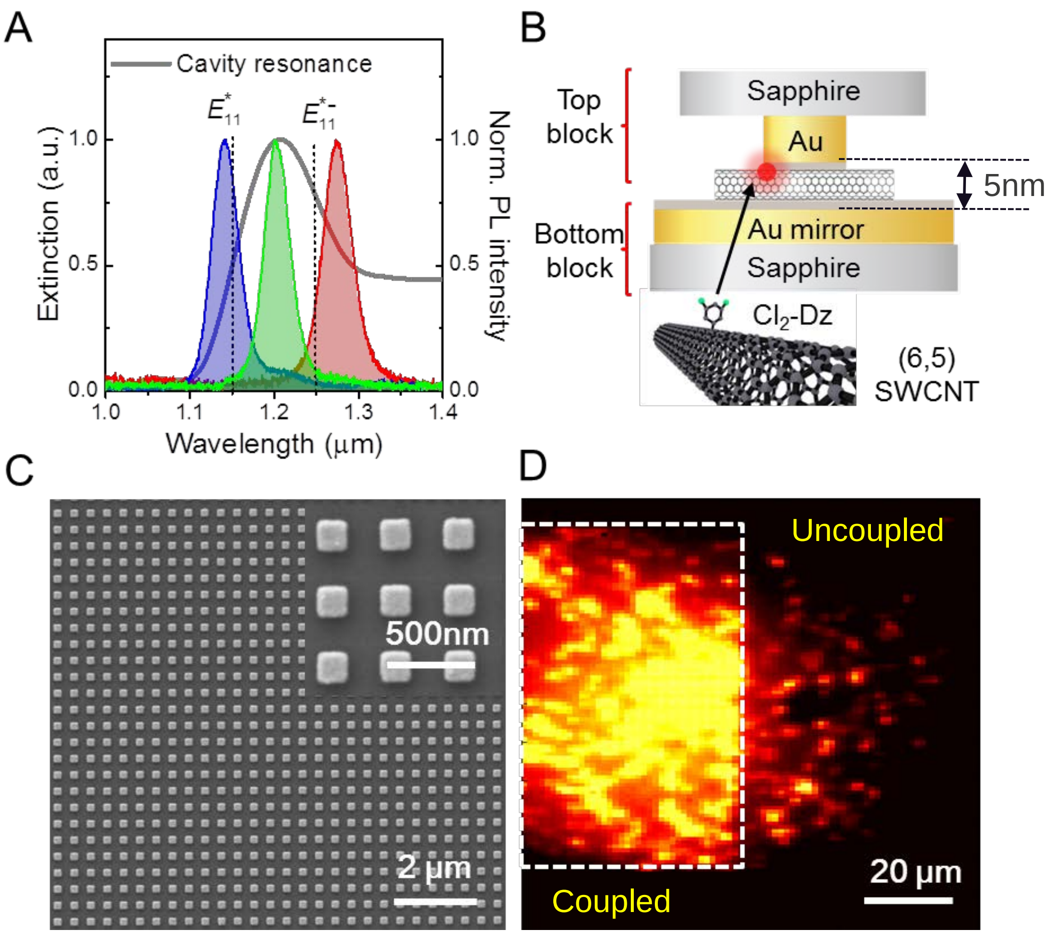
- Narrowest linewidth achieved 98.5 μeV
- PL lifetime T_1 ~ 1.5 ns. Approaching theoretical limit of 3 ns

$$\Delta E = \frac{2\hbar}{T_2} = \frac{\hbar}{T_1} + \frac{2\hbar}{T_2^*}$$

- Linewidth is dominated by pure dephasing T_2^*

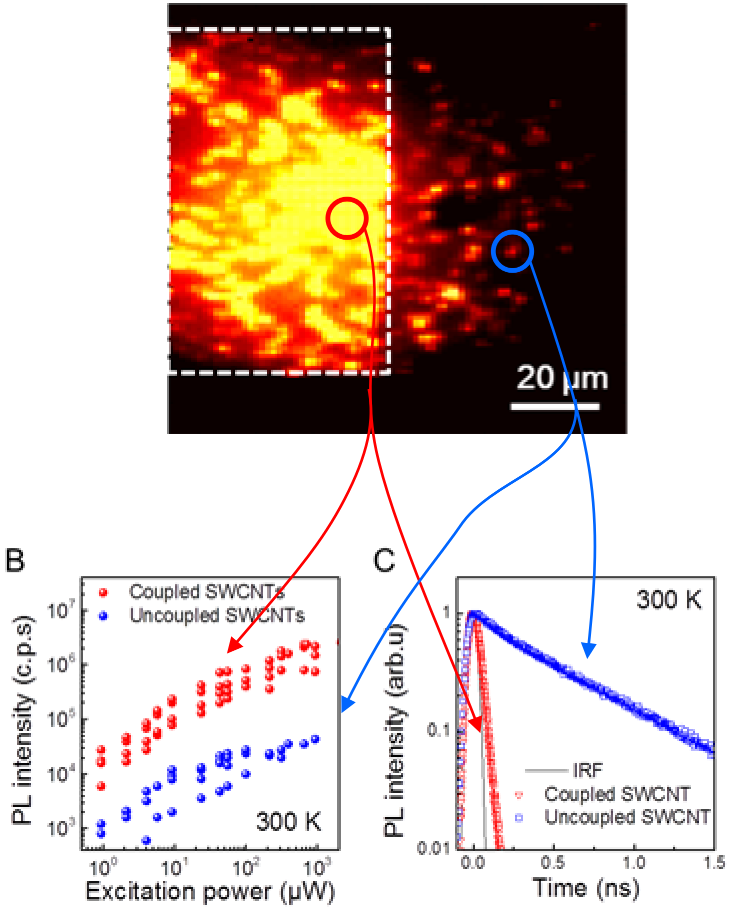
New Paths to Photon Indistinguishability: Plasmonic Enhancement

Exploit Plasmonic Effect to enhance radiative decay rate so that it would dominate over all the other processes.



- Plasmonic cavity
  Au nanocube /Al_2O_3/SWCNT/Al_2O_3/Au Mirror
- Gap size of 5nm ⇒ Field enhancement 800
- Cavity resonance ~ 1200 nm
- SWCNTs are spread randomly over the nanocube array.
- High density nanocube array with filling factor of 65% ensure coupling to the SWCNT.
- PL of SWCNT coupled to nanocube array strongly enhanced compared to that of uncoupled SWCNTs.

Plasmonic Enhancement of PL Emission



Room Temperature Experiment

- PL intensity at saturation is enhanced by 52 fold
- PL lifetime of uncoupled SWCNTs 588 ± 10 ps
- PL lifetime of coupleds SWCNT 11 ± 3 ps
- Decay rate enhancement ~ $\gamma_{on,RT}/\gamma_{off,RT}$ = 53

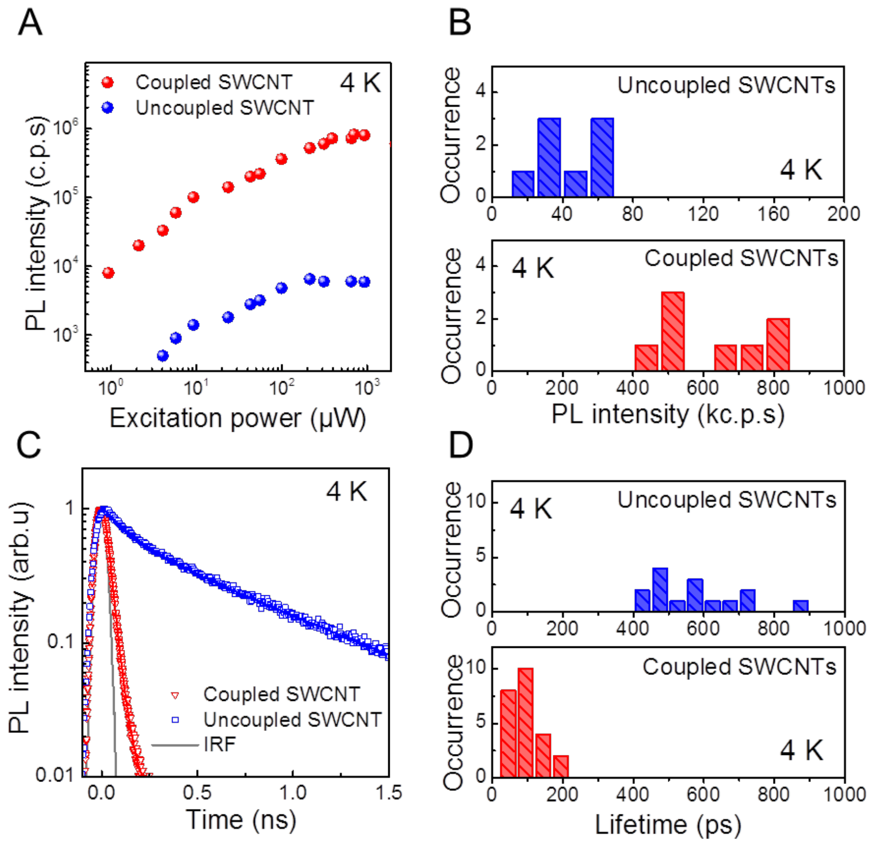
$$F_p = 0.75\left(\frac{\gamma_{on}}{\gamma_{off}} - 1\right)\eta_{off}{}^{-1}$$

$$\eta_{on} = \frac{(Fp+1)\,\gamma_R}{\gamma_{on}} = \frac{(Fp+1)\,\gamma_{off}\,\eta_{off}}{\gamma_{on}} = \frac{(F_p+1)\,\eta_{off}}{\frac{\gamma_{on}}{\gamma_{off}}},$$

Quantum yield of uncoupled emitter: η_{off} = 13%

Purcell Enhancement Factor F_p= 300

Plasmonic Enhancement of PL Emission



Low Temperature Experiment

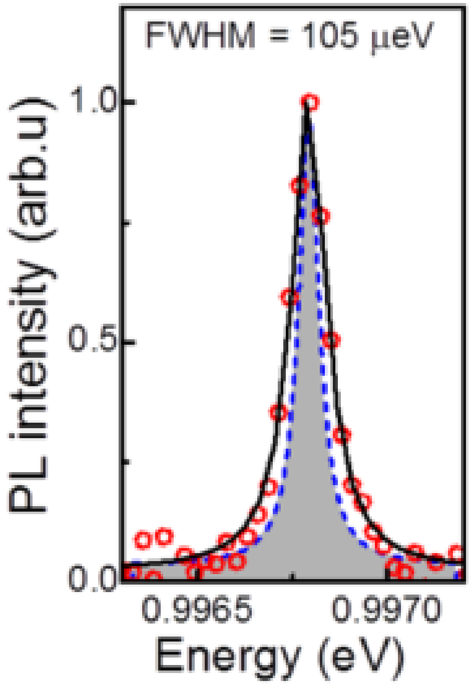
- PL intensity at saturation is enhanced by 133 fold
- PL lifetime of uncoupled SWCNT 726 ± 10 ps
- PL lifetime of coupled SWCNT 10 ± 3 ps
- Decay rate enhancement ~ 73

Strong rate enhancement
1.3 GHz Single photon emission

Purcell Enhancement Factor F_p= 450

Cavity enhanced quantum yield = 74%

Alternative Path to Photon Indistinguishability

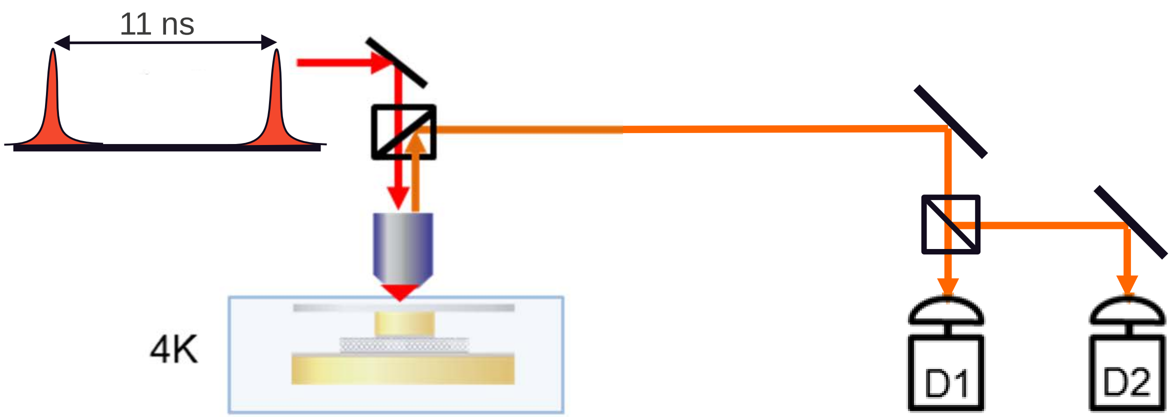


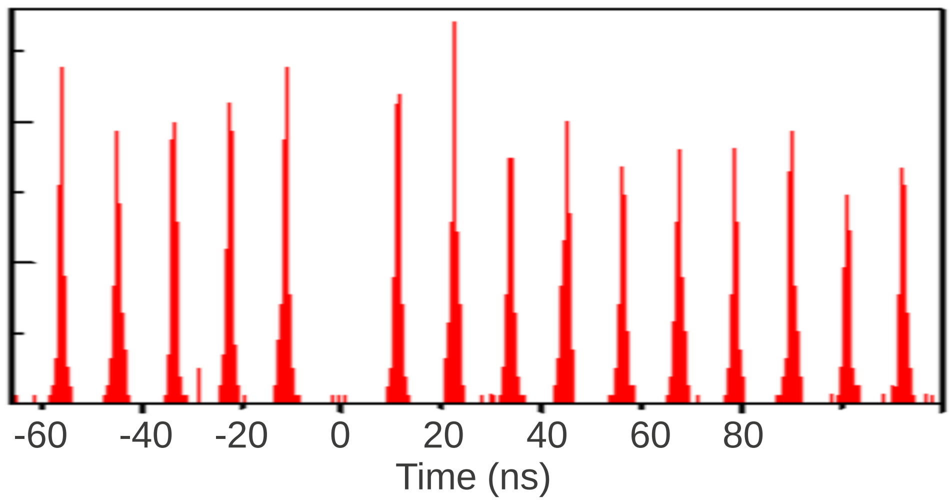
$$\Delta E = \frac{2\hbar}{T_2} = \frac{\hbar}{T_1} + \frac{2\hbar}{T_2^*}$$

- Low T intrinsic linewidths: ΔE = 105 ± 5 μeV
- population decay time: T_1 = 10 ps.
- $\frac{\hbar}{T_1}$ =60 μeV; $\frac{2\hbar}{T_2^*}$ =82 μeV

Single photon indistinguishability $T_2/2T_1$ = 0.63 is achievable !

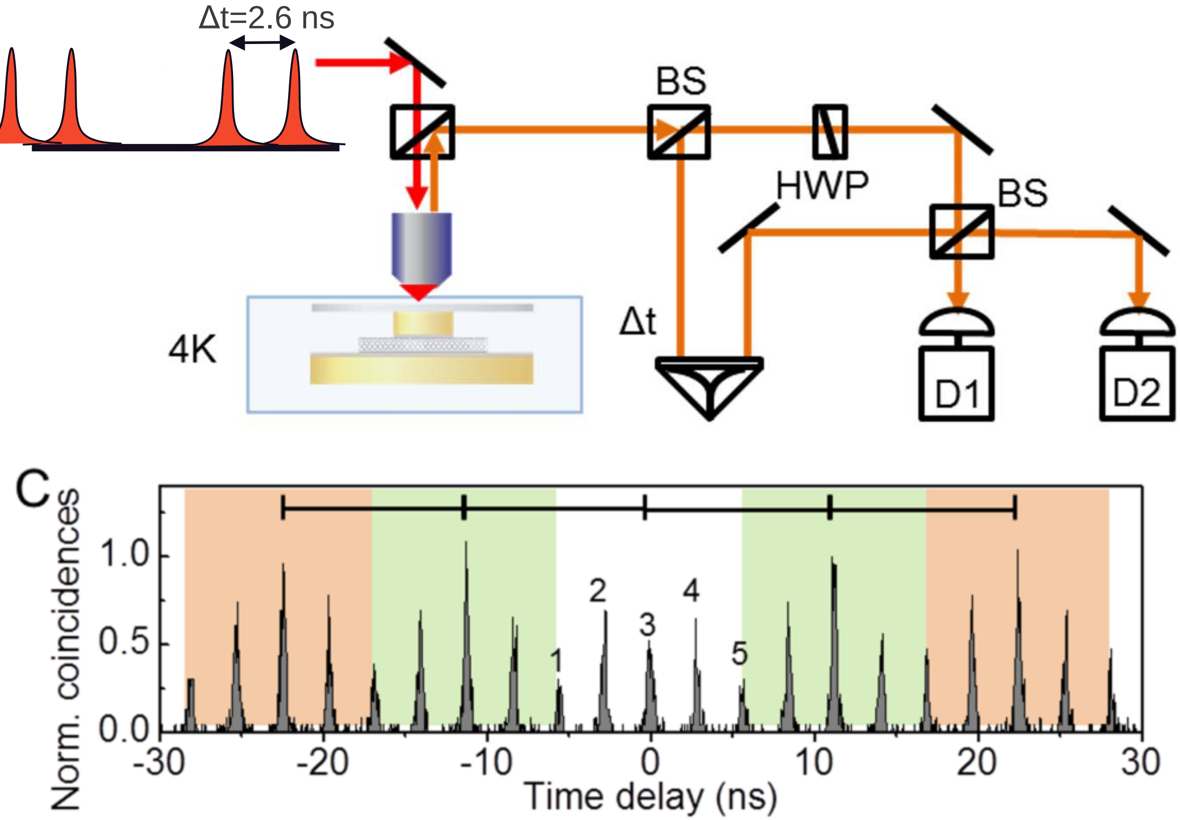
Hanbury-Brown-Twiss (HBT) Experiment





- Second order photon correlation spectra $g^{(2)}$: Histogram of two successive photon detection events.
- Missing center peak ⇒ Antibunching ⇒ One photon / one pulse
- Center peak area < 0.01 ⇒ 99% single photon purity.

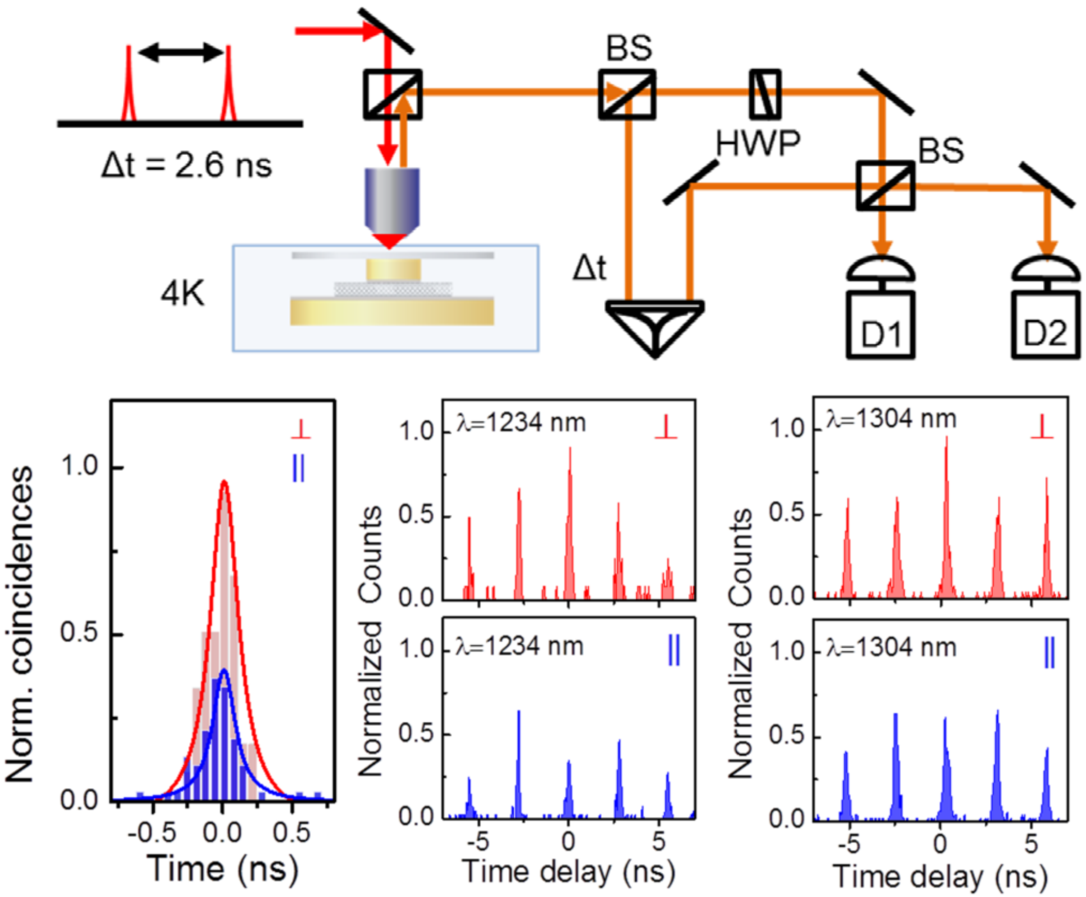
Hong-Ou-Mandel (HOM) Experiment



- Split the excitation pulse into two pulses with $\Delta t = 2.6$ns
- Add a BS and a delay line of the same Δt to the detection channel
- Induce two photon interference at the second BS
- When HWP is turned to rotate polarization of one channel perpendicular to another interference will be destroyed.

$$V = \frac{1}{(1-\varepsilon)^2}\left[2g^{(2)}(0) + \frac{R^2+T^2}{2RT} - \frac{A_3}{A_2+A_4}\left(2 + g^{(2)}(0)\frac{R^2+T^2}{RT}\right)\right]$$

Hong-Ou-Mandel (HOM) Experiment



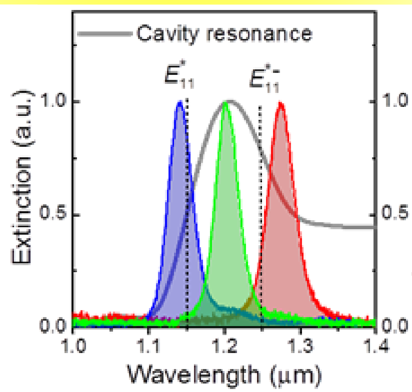
Quantum Defect @ 1234 nm (Resonance with cavity mode) two-photon interference visibility

0.78 ± 0.08

Quantum Defect @ 1304 nm (Off resonance with cavity) Single Photon Indistinguishability

0.40 ± 0.04

$$V = \frac{1}{(1-\varepsilon)^2}\left[2g^{(2)}(0) + \frac{R^2+T^2}{2RT} - \frac{A_3}{A_2+A_4}\left(2 + g^{(2)}(0)\frac{R^2+T^2}{RT}\right)\right]$$



Summary:
Demonstration of Photon Indistinguishability

Quantum defect – plasmonic cavity coupling
On-chip, telecom band, RT quantum light source with 1.3GHz brightness.
Cavity enhanced QY of 74% and Purcell factor of 415
On demand single photon source with
99% single photon purity and
Single photon indistinguishability with
78% fringe visibility

Indistinguishable single photons at telecom O-band from sp^3 functionalized carbon nanotubes coupled to plasmonic nanocavities

Yue Luo[1,2], Xiaowei He[1,2], Younghee Kim[3], Jeffrey L. Blackburn[4], Stephen K. Doorn[3], Han Htoon[3*], Stefan Strauf [1,2*]
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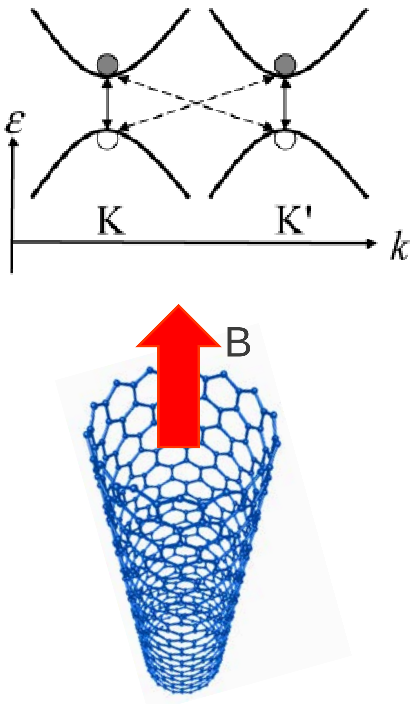
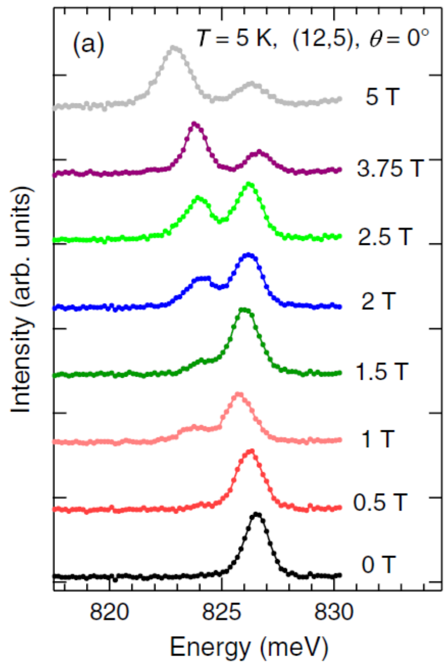
[3] Center for Integrated Nanotechnologies, Materials Physics and Applications Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA
[4] National Renewable Energy Laboratory, Golden, CO 80401, USA

Nano Letter 10.1021/acs.nanolett.9b04069

Probing Excitonic Fine Structure with Single Defect Magneto PL





A. Srivastava et al., *PRL.*, **2008**, 101, 087402

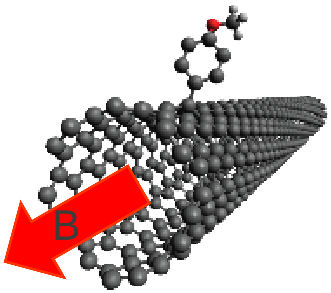
- Band edge exciton (E_{11}) is four fold degenerated.
- Application of magnetic field parallel to the tube axis can lift this degeneracy and brighten the dark exciton state.

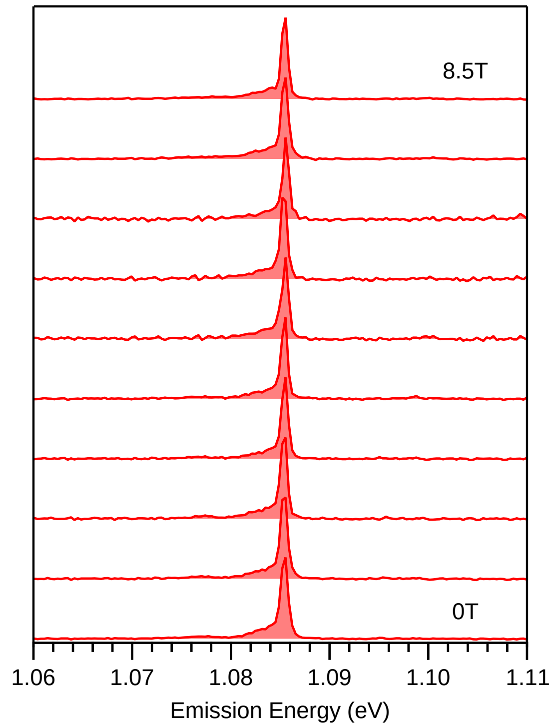
Can similar excitonic fine structure exit in defect states of CNT?

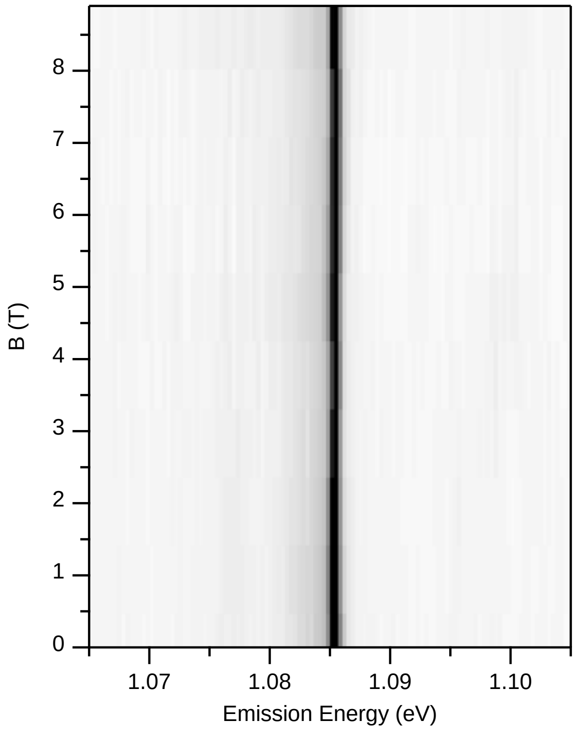
Opportunity of optical manipulation of spin or valley degree of freedom.

Probing Excitonic Fine Structure with Single Defect Magneto PL

4-Methoxybenzene diazonium doped (6,5) CNT



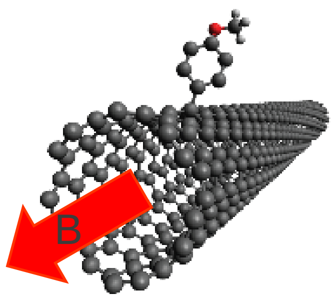


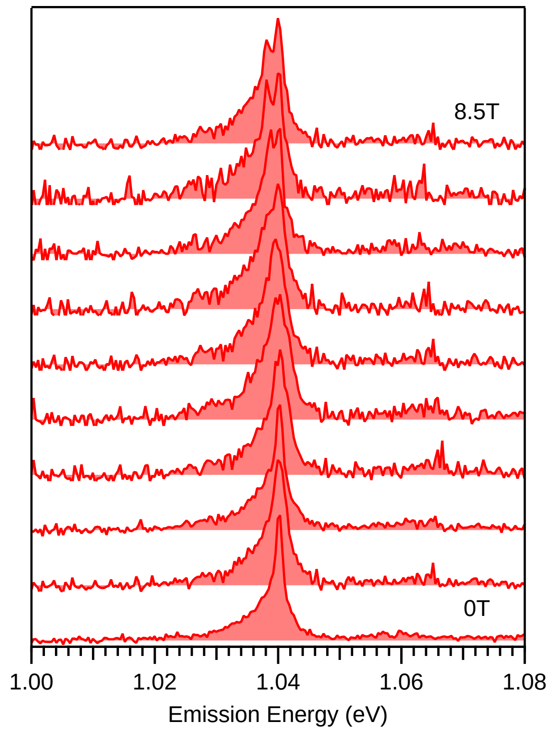


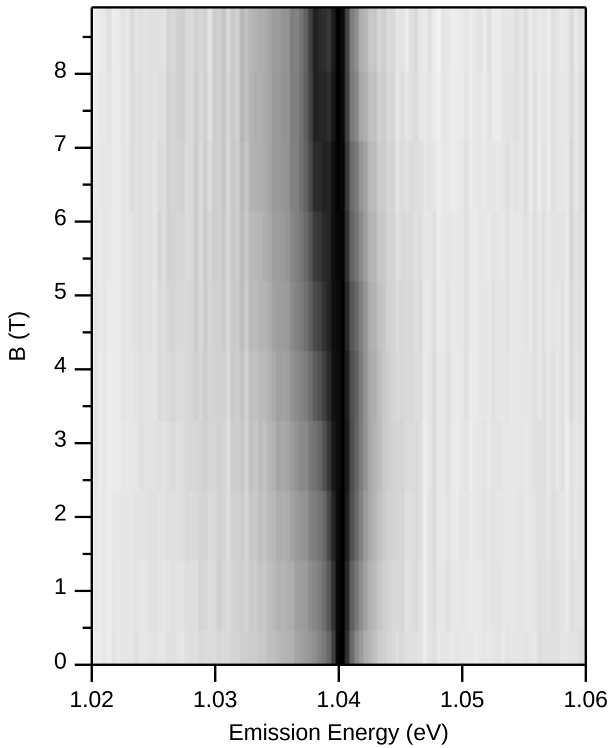
- No splitting was observed in 75% of the defect states

Probing Excitonic Fine Structure with Single Defect Magneto PL

4-Methoxybenzene diazonium doped (6,5) CNT



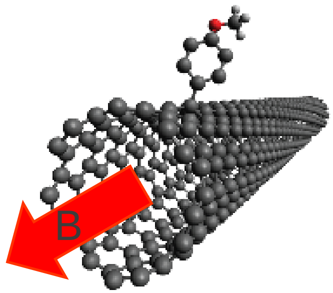


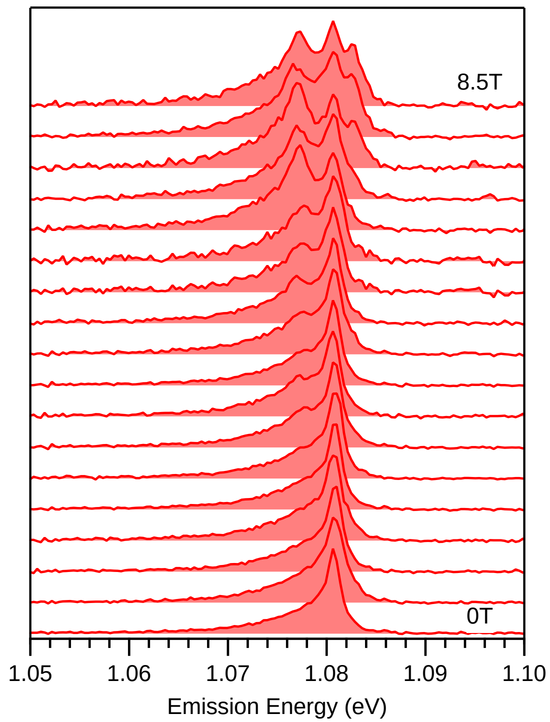


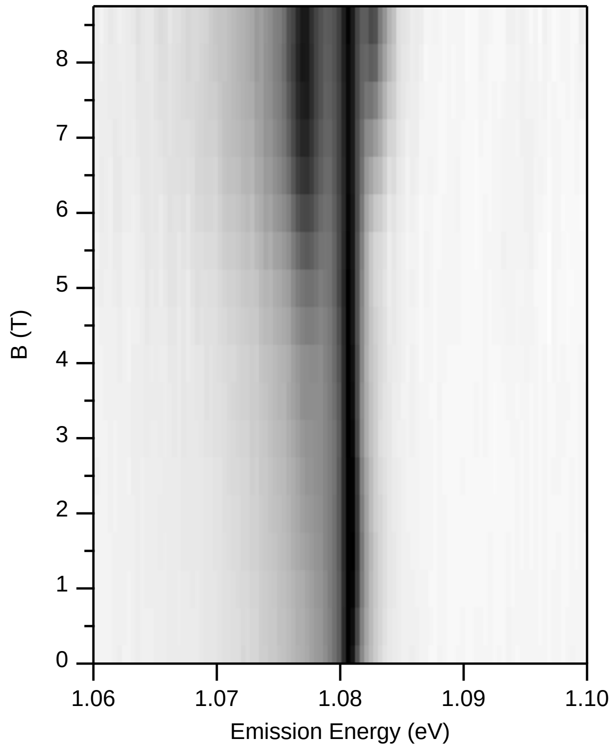
- No splitting was observed in 75% of the defect states
- <5% of the defect emission peaks split into two peaks

Probing Excitonic Fine Structure with Single Defect Magneto PL

4-Methoxybenzene diazonium doped (6,5) CNT

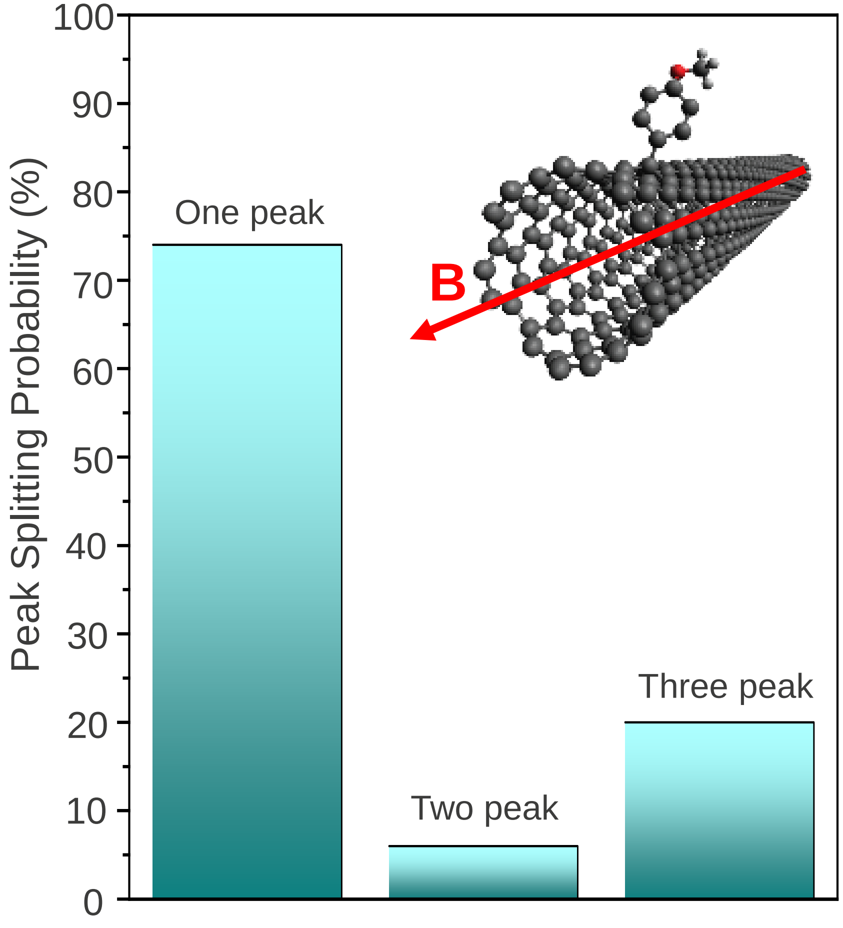


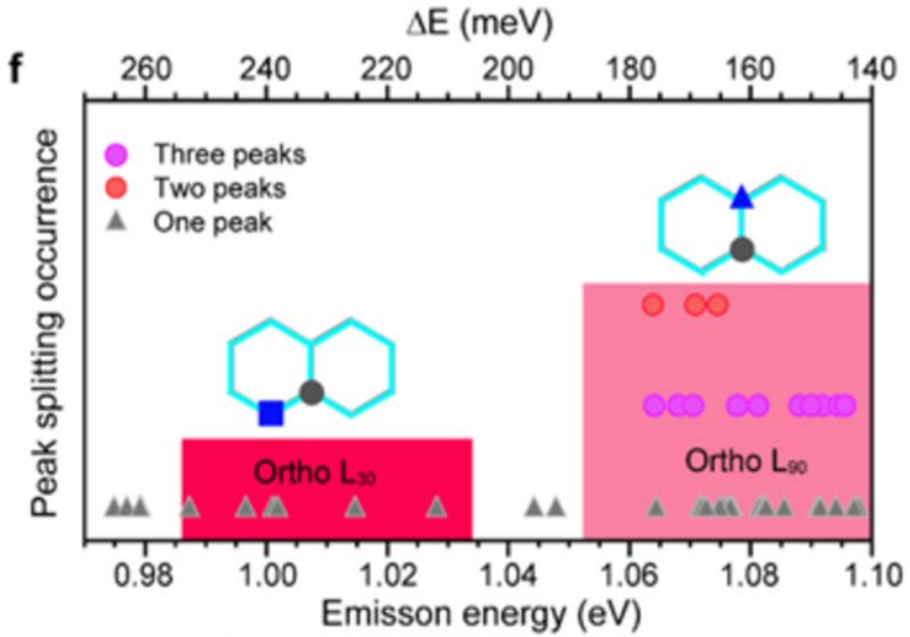




- No splitting was observed in 75% of the defect states
- <5% of the defect emission peaks split into two peaks
- 20% of the defects show 3 or more peaks at 8.5 T

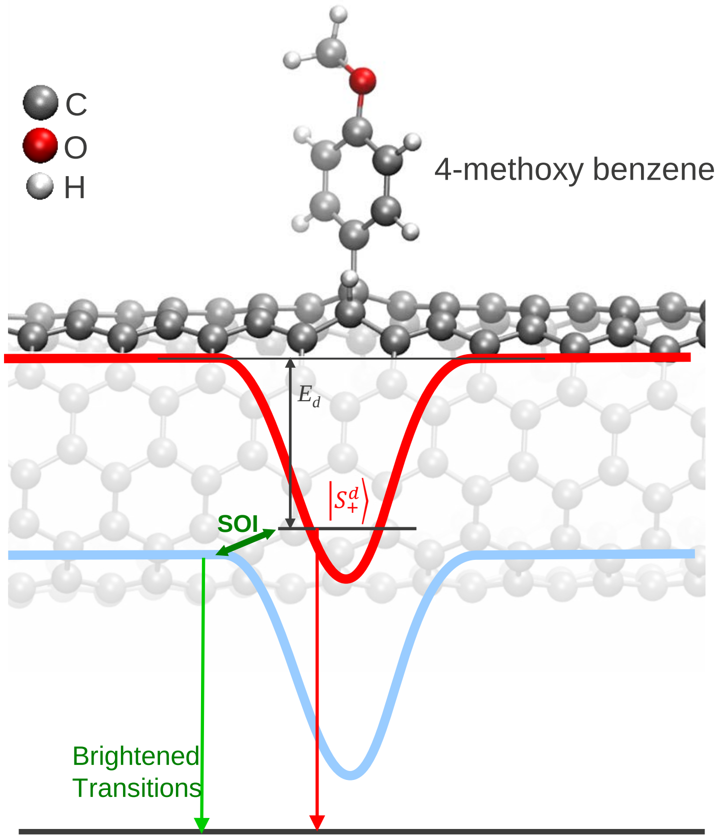
Probing Excitonic Fine Structure with Single Defect Magneto PL



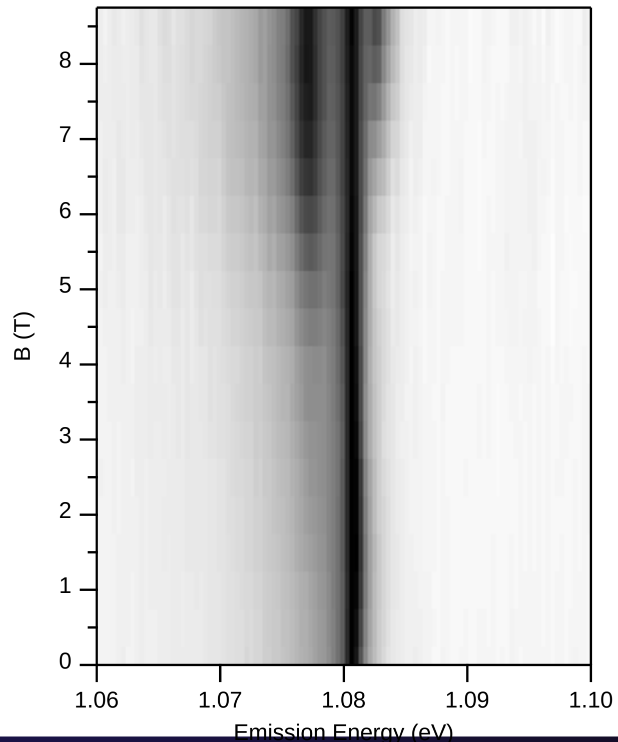


- Emission of defect states distribute over 0.98 to 1.10 eV
- 2 and 3 peak spectral features emerged only from the defects emitting in 1.05-1.1 eV range, corresponding to Ortho L_{90} configuration.

Probing Excitonic Fine Structure with Single Defect Magneto PL



- Defect bound singlet exciton state can get into close vicinity of free 1 D triplet excitons
- Magnetic field can induce the mixing between the two
- Triplet states as a result got brightened.



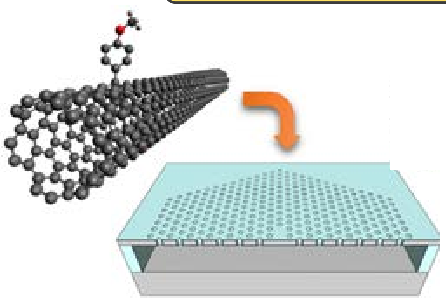
Photonic, Plasmonic, and Electronic Integration

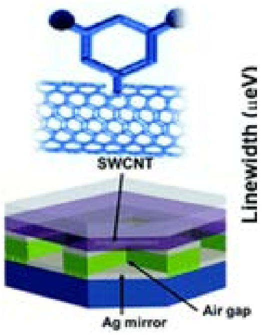
Coupling Quantum emittters with Plasmonic/Photonic Structures

Modification of emission characteristics, e.g. polarization, collimated emission etc.

Modification of emission rate and efficiency

Modification of quantum optical properties.

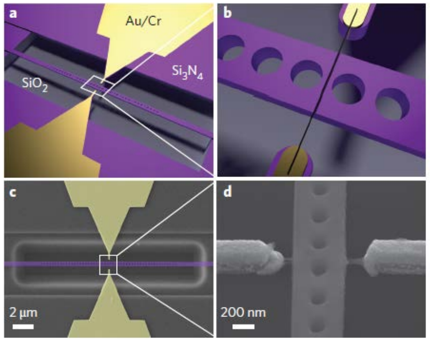




2D Photonic Crytals: Yuichiro. K. Kato, RIKEN, Japan

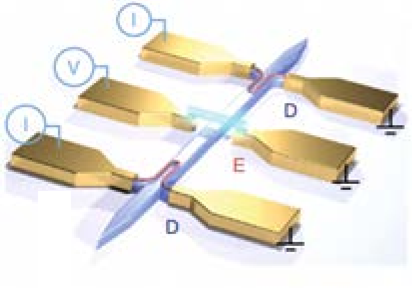
Plasmonic Antenna Array: Stefan Strauf, *Stevens Institute of Technology*

Waveguide Integrated Electrically Driven Single Photon Sources



Ralph Krupke (Karlsrue Institute of Technology)

Electrically driven SPS operating at RT and 1.55 μm
Integrated with wave guide and single photon detectors
for proof of principle quantum photonic experiments



Khasminskaya, S. et al. Fully integrated quantum photonic circuit with an electrically dr en light source. *Nat. Photon.* **10**, 727–732 (2016)

Thanks for your Attention