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Influence of Exciton Dimensionality on Spectral Diffusion of Carbon Nanotubes

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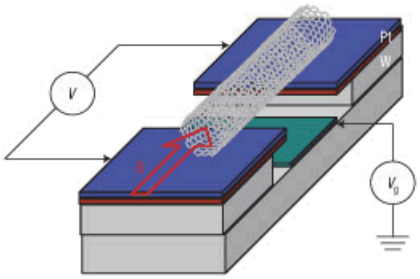
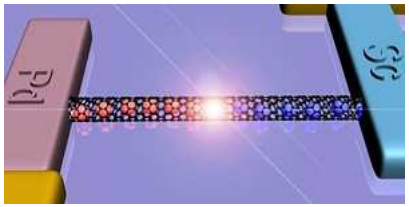
Photoluminescence (PL) of SWCNTs: Application



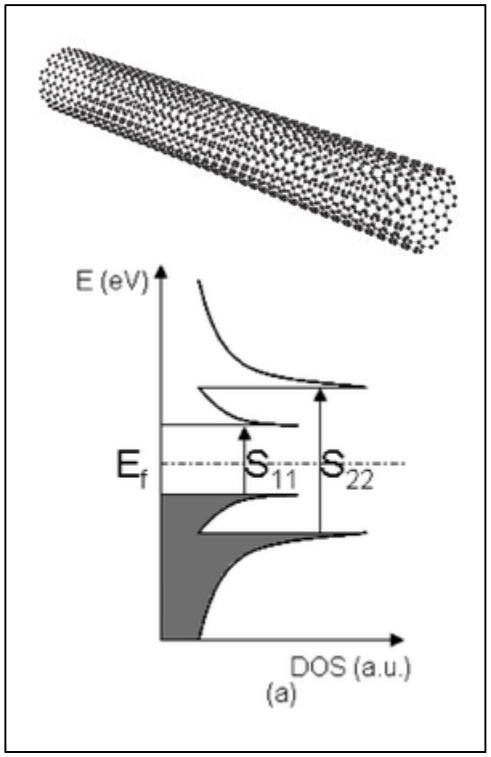
field-effect transistor

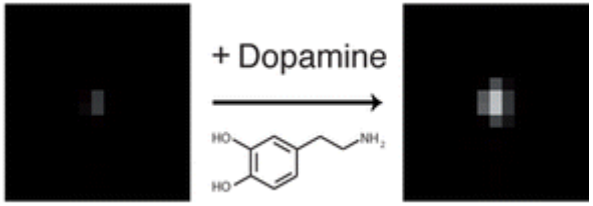
Cao et al. Nature Mater. 2005, 4



light emitting diode

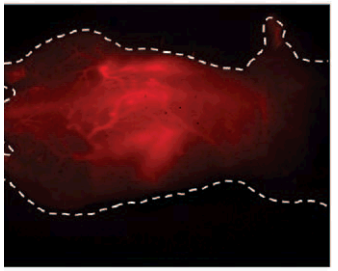
Wang et al. Nano Lett. 2011, 11





sensor

Kruss et al. J. Am. Chem. Soc., 2014



biological imaging

Welsher et al. Nature Nanotech. 2009

Photoluminescence of SWCNTs: Limitation

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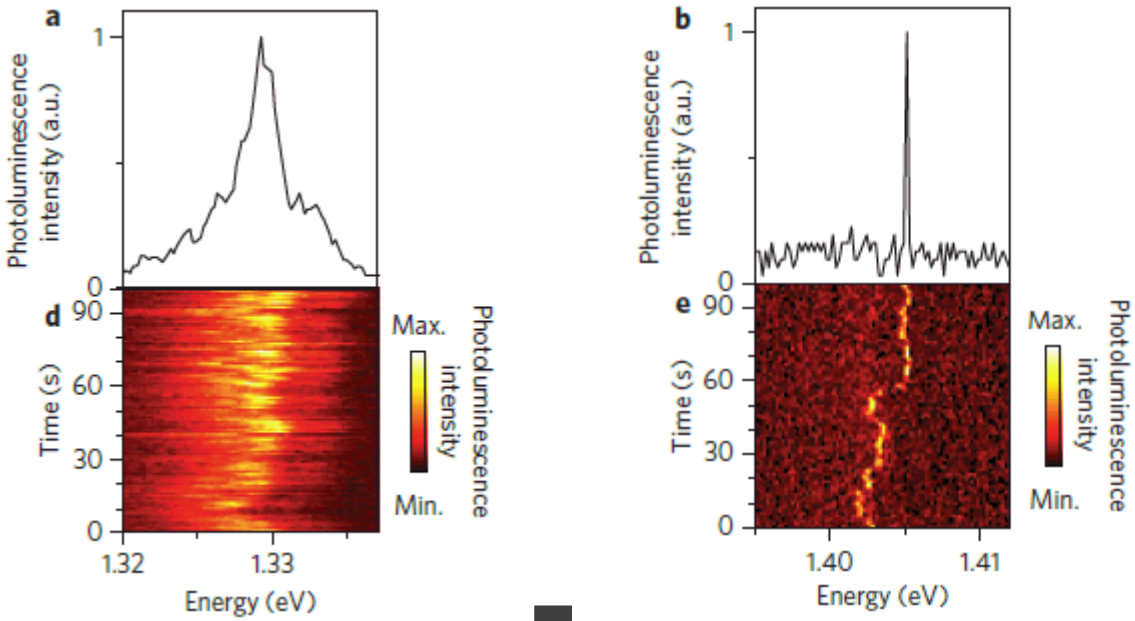
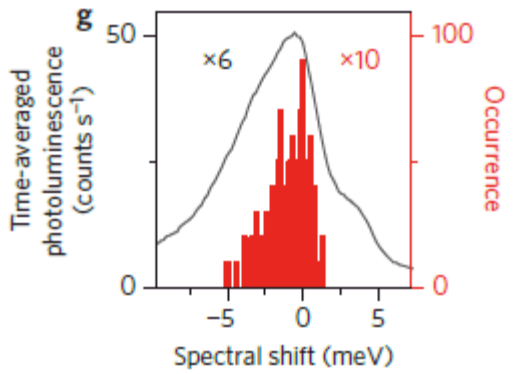
inhomogeneous broadening

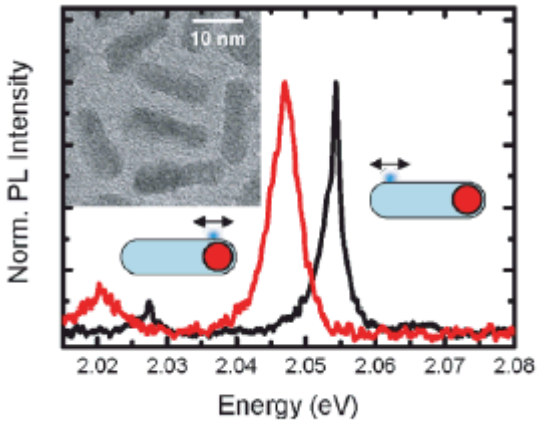


spectral line wondering

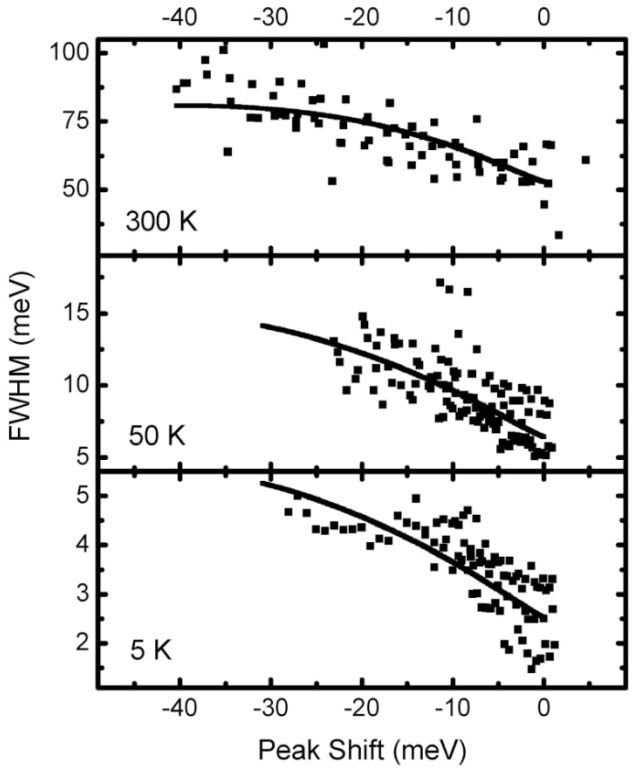
time average



Quantum-confined Stark effect (QCSE)



CdSe/CdS nanorods
(Weller, PRL 2004)



- Charges on nanostructures can create a local electric field.
- Sublinear correlation between peak shift and peak width.

Temperature dependent exciton dimensionality of SWCNTs

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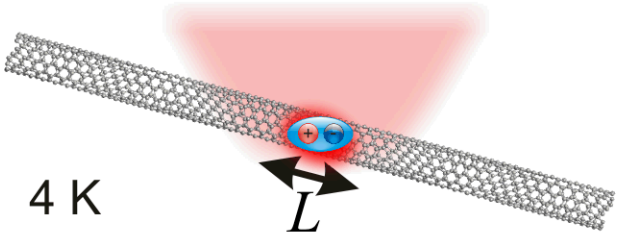
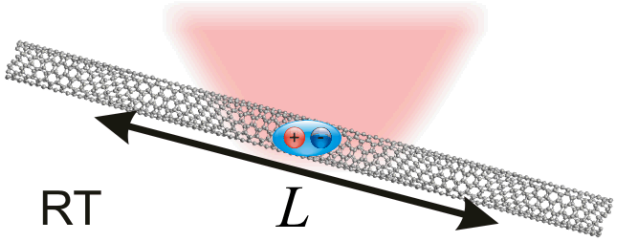
Cryogenic temperature



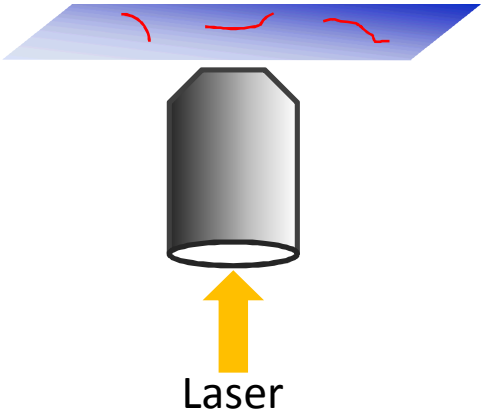
Room temperature



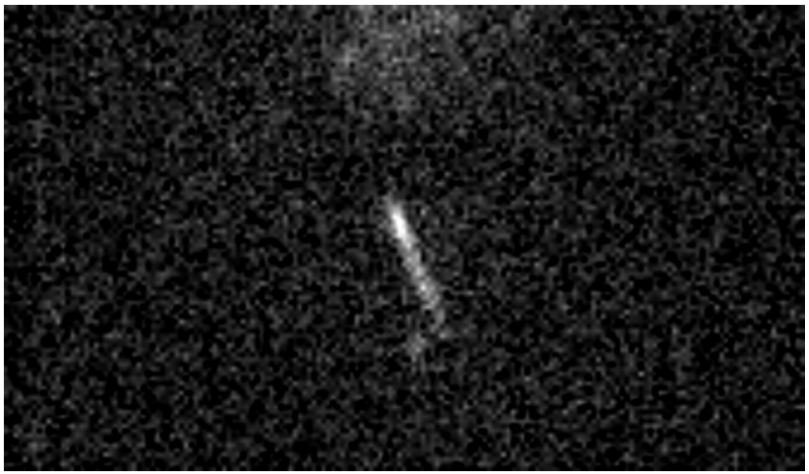
0D quantum dot (QD)-like excitonic states due to local potential barriers

1D diffusive excitonic states

Individual SWCNT PL imaging



Wide field images

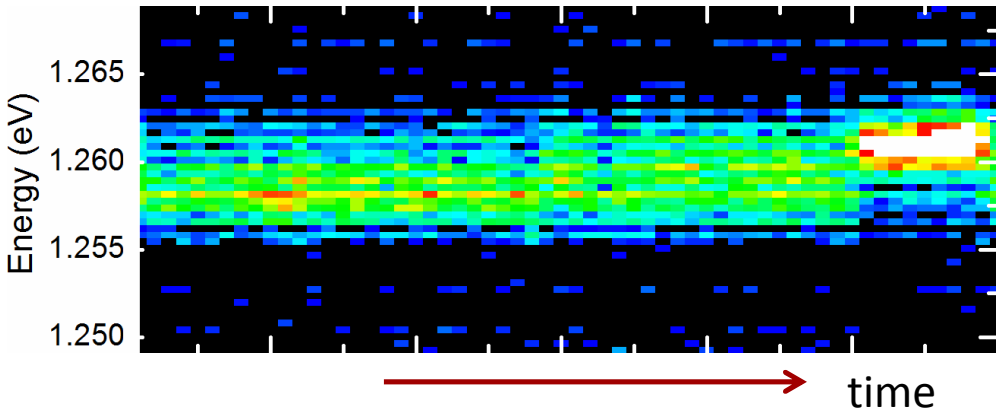


Stable PL

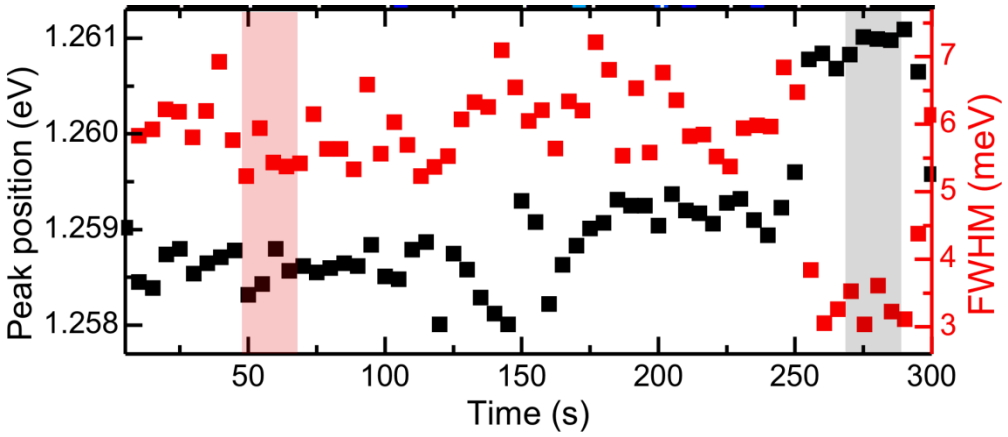
PL blinking/fluctuation

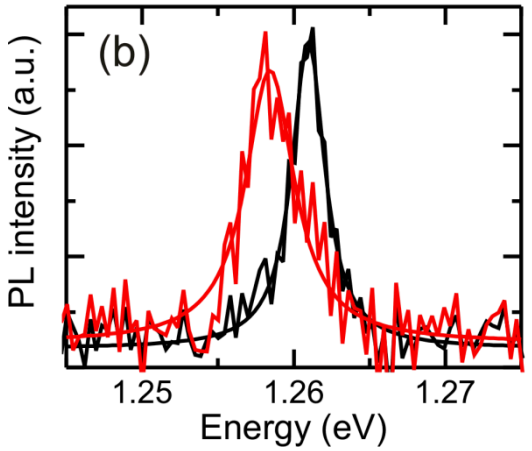
- Homogeneous illumination of the whole SWNTs with expanded laser beam
- Beam size: ~60 x 60 um
- SWCNT length: <15 um

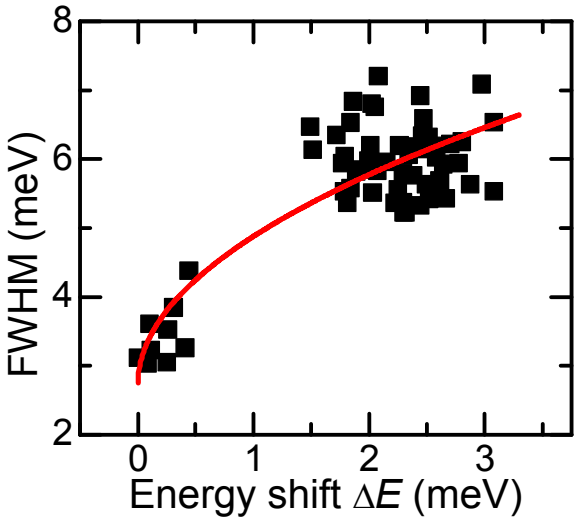
PL spectra at 4K



Gaussian fitting

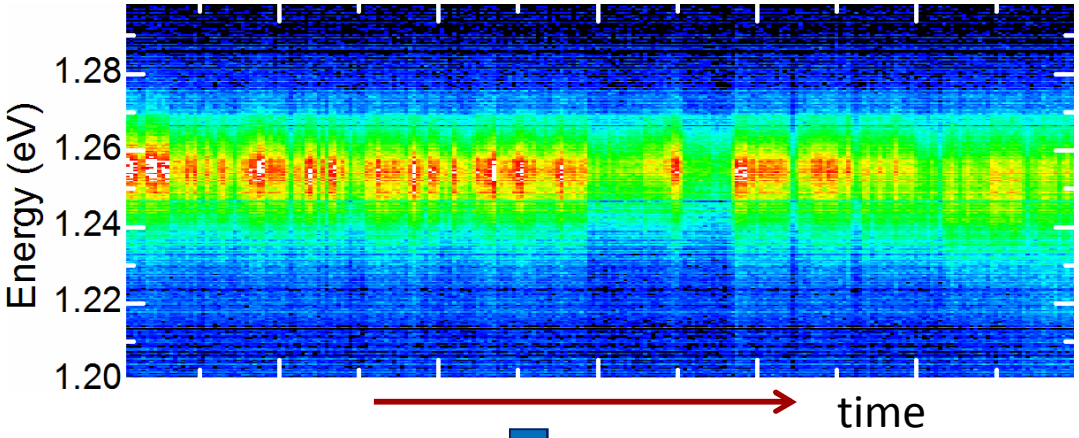




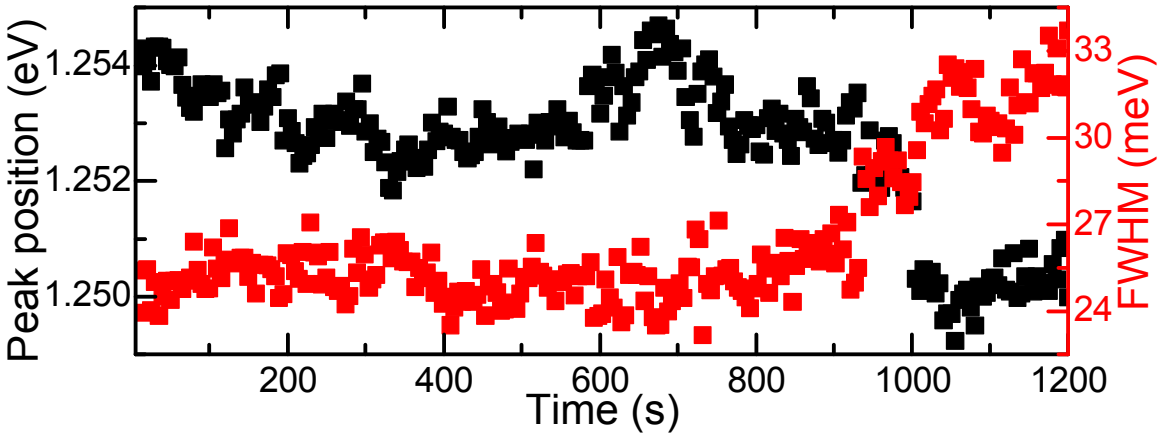


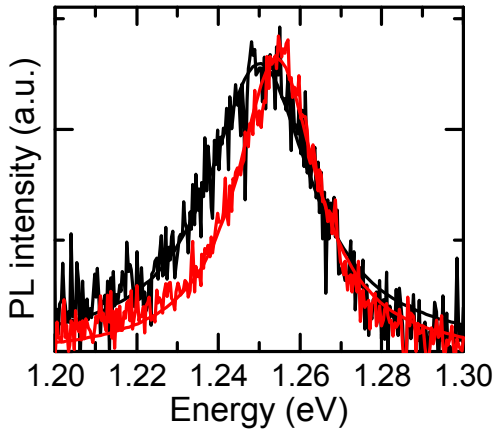
Sublinear correlation: similar to QDs

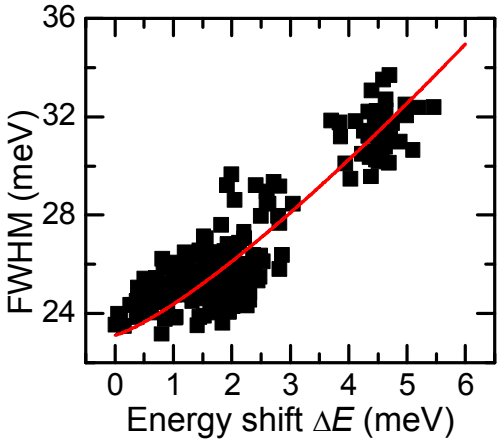
PL spectra at room temperature



Gaussian fitting







Superlinear correlation

QCSE of 0D and 1D excitons

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$$\Delta E = k_b \frac{(edF)^2}{E}$$

ΔE : energy shift of the binding energy caused by QCSE
e : electron charge
d : tube diameter
E : tube binding energy at zero field
F : electric field induced by surface charges

Spectral diffusion described by a a Gaussian stochastic model:

$$\langle \delta(\Delta E)(t)\delta(\Delta E)(0)\rangle = \sigma^2 e^{-t/t_c}$$

σ : spectral broadening in meV
t_c : correlation time

At t_c = 0, we have $\langle \delta^2(\Delta E)(0)\rangle = \sigma^2$

In a simple assumption, we assume that there is an external point charge q located on the surface of a SWCNT that creates a local electric field

$$F = \frac{q}{4\pi\varepsilon r^2}$$

r: exciton-charge distance
ε: permittivity of the surrounding environment

QCSE of 0D and 1D excitons

Cryogenic temperature: 0D QD –like states →

- Exciton-charge distance r and local environment permittivity ε are constants.
- ΔE is mainly induced by fluctuations in surface charges.

→ $\sigma \propto \sqrt{\Delta E}$ sublinear correlation

Room temperature: 1D diffusive states →

- fluctuation in exciton-charge distance r dominates

→ $\sigma \propto \Delta E^{5/4}$ superlinear correlation

QCSE of 0D and 1D excitons

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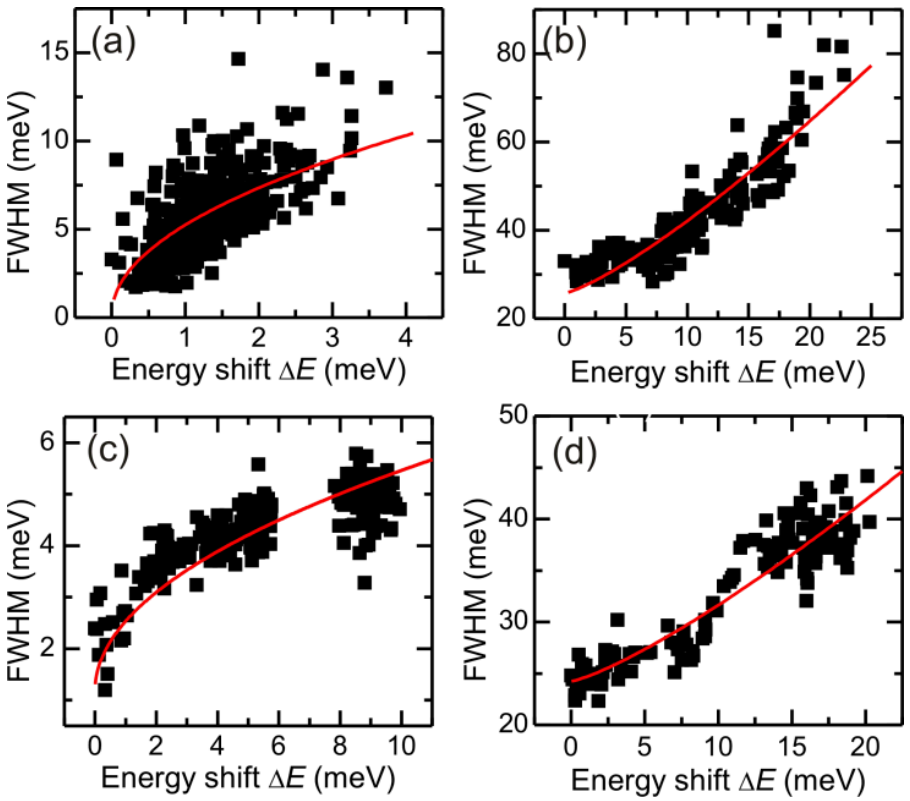
4K

room temperature

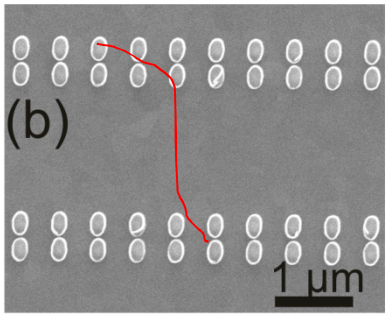


$$\sigma \propto \sqrt{\Delta E}$$

$$\sigma \propto \Delta E^{5/4}$$

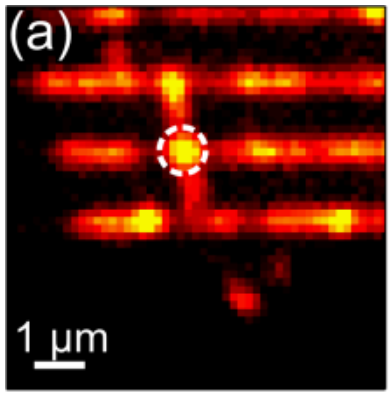
SWCNTs coupled to Au dimers

SEM

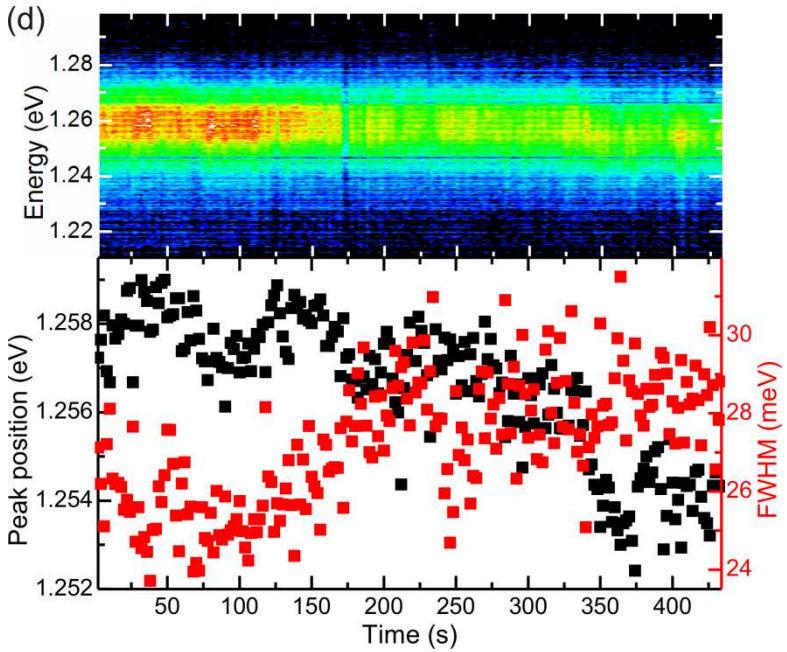


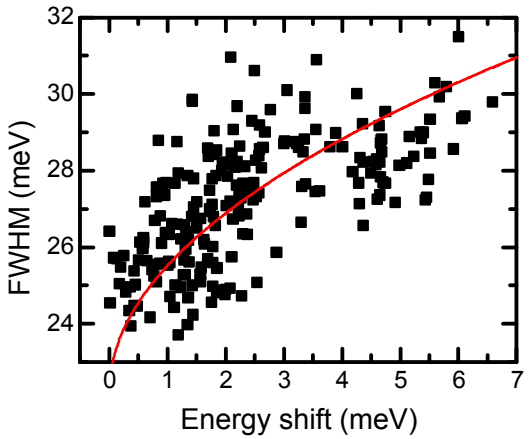
Au dimer nanoantennas

PL image



Localized emission





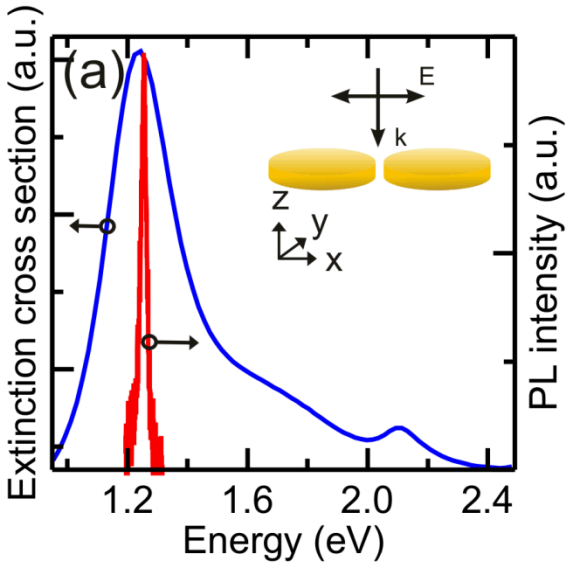
$$\sigma \propto \Delta E^{5/4}$$

superlinear correlation at room temperature!

Numerical Simulations: excitation

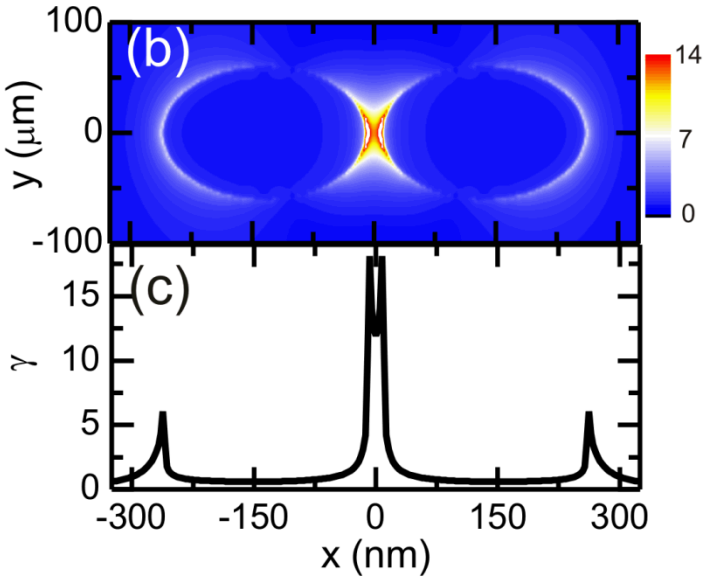
FDTD

Extinction spectrum + PL spectrum



- The longitudinal surface plasmon mode (~ 1.24 eV) from the Au dimer is in good resonance with the PL of the SWCNTs.

Local electric field distribution

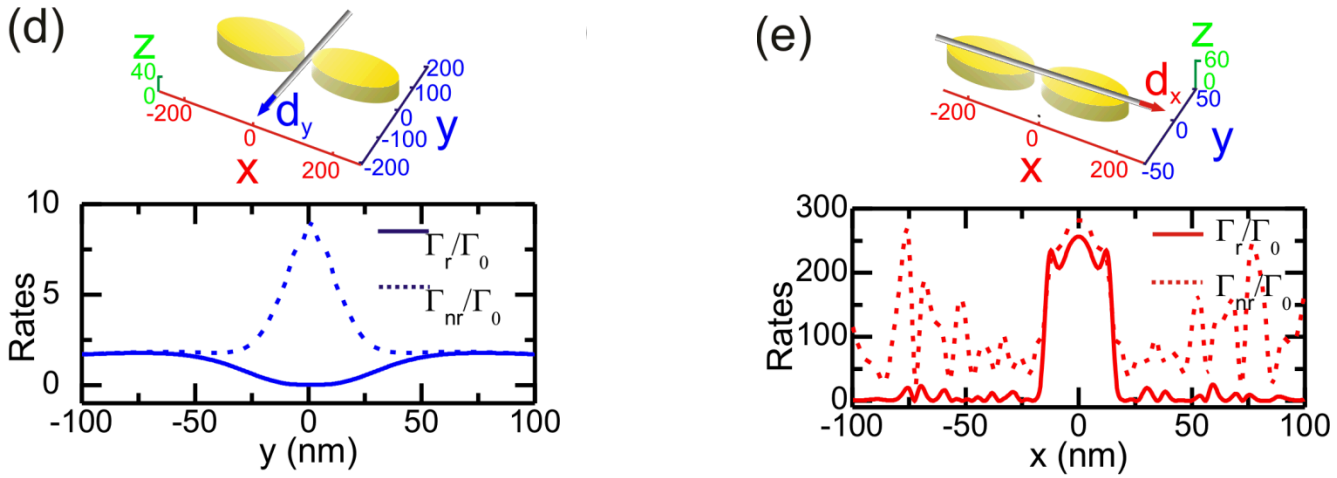


- Electric field is highly confined inbetween the gap.
- Excitation rate is proportional to the local electric field.

→ highly confined excition region

Numerical simulation: decay rates

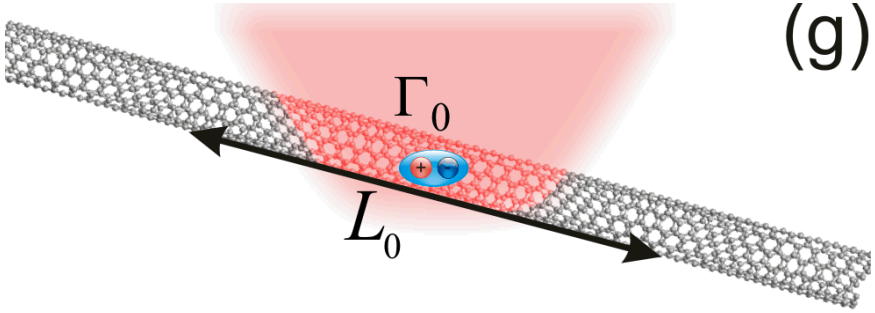
- Based on a general theoretical approach.
- Assume the oscillator strength of the dipole oriented perpendicular to the tube is negligible.



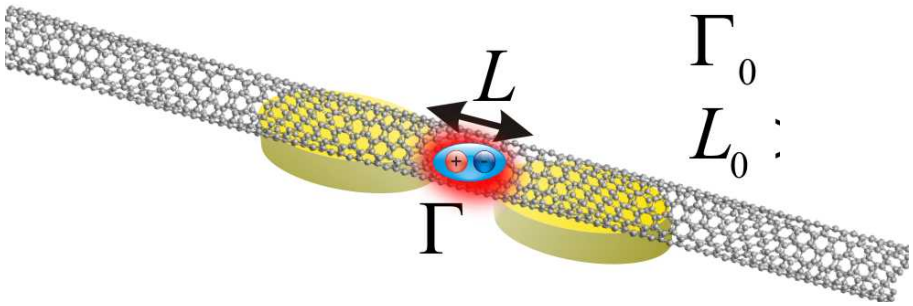
- the total decay rate of SWCNTs coupled to dimer nanoantennas can be enhanced by factors of 10 – 500.

SWCNTs coupled to Au dimers: explanation

room temperature



$$\Gamma_0 \ll \Gamma$$
$$L_0 \gg L$$



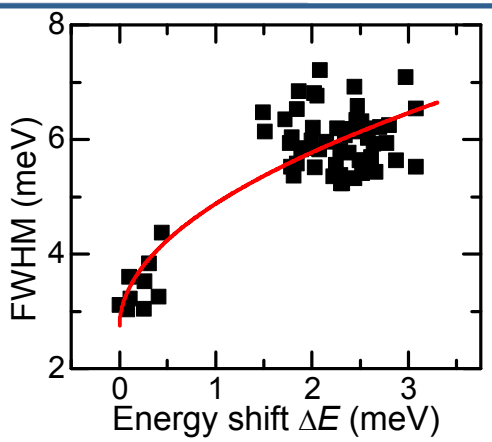
In the Au dimer gap:

- highly confined excitation region
- significantly enhanced decay rate

→ spatial narrowing of the recombination region

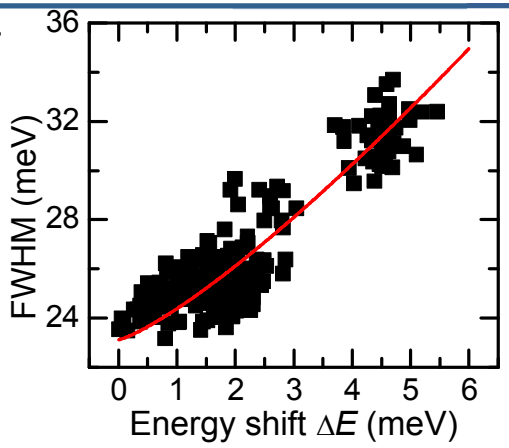
Summary

4K



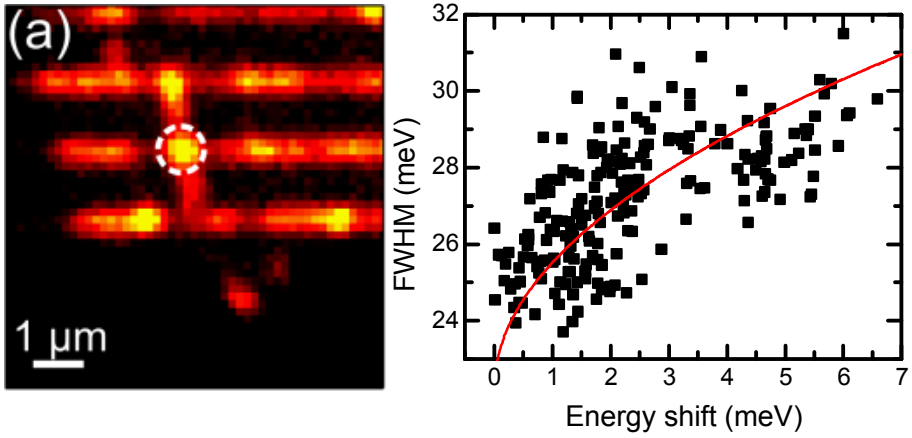
$$\sigma \propto \sqrt{\Delta E}$$

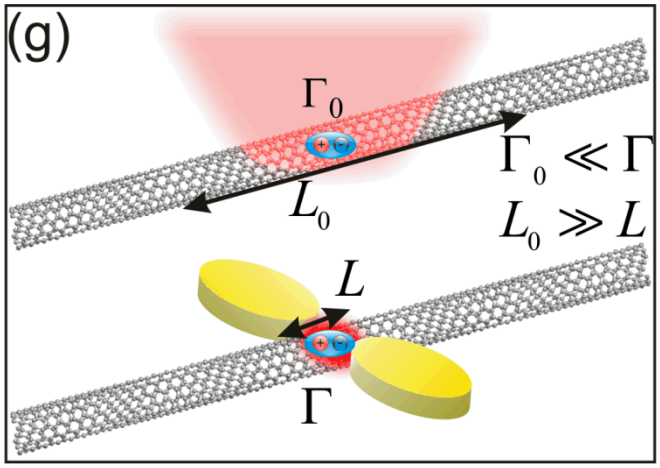
RT



$$\sigma \propto \Delta E^{5/4}$$

RT, coupled to Au dimer





Acknowledgements

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Thank you for your attention!