Optical microresonators as single-particle absorption spectrometers

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About the article

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Optical microresonators as single-particle absorption spectrometers

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Optical measurements of nanoscale objects offer major insights into fundamental biological, material and photonic properties. In absorption spectroscopy, sensitivity limits applications at the nanoscale. Here, we present a new single-particle double-modulation photothermal absorption spectroscopy method that employs on-chip optical whispering-gallery-mode (WGM) microresonators as ultrasensitive thermometers. Optical excitation of a nanoscale object on the microresonator produces increased local temperatures that are proportional to the absorption cross-section of the object. We resolve photothermal shifts in the resonance frequency of the microresonator that are smaller than 100 Hz, orders of magnitude smaller than previous WGM sensing schemes. The application of our new technique to single gold nanorods reveals a dense array of sharp Fano resonances arising from the coupling between the localized surface plasmon of the gold nanorod and the WGMs of the resonator, allowing for the exploration of plasmonic–photonic hybridization. In terms of the wider applicability, our approach adds label-free spectroscopic identification to microresonator-based detection schemes.

Authors

Randall Goldsmith

- Bachelor degree: Cornell University in 2002
- Ph.D : Northwestern University in 2007
- Postdoctoral Researcher at Stanford
- Assistant professor of **Department of chemistry in UW-Madison university**
- Research interest :

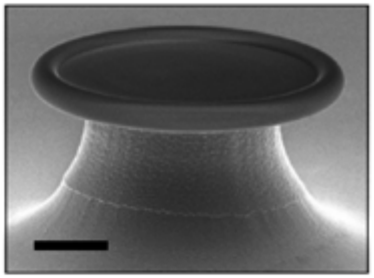
(1) Single-molecule techniques
(2) organic photovoltaics
(3) protein conformational dynamics

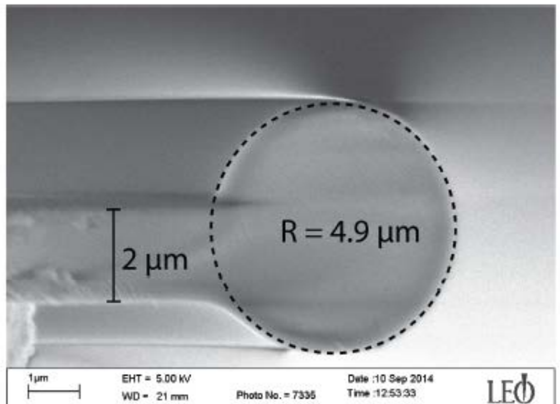
DEPARTMENT OF Chemistry

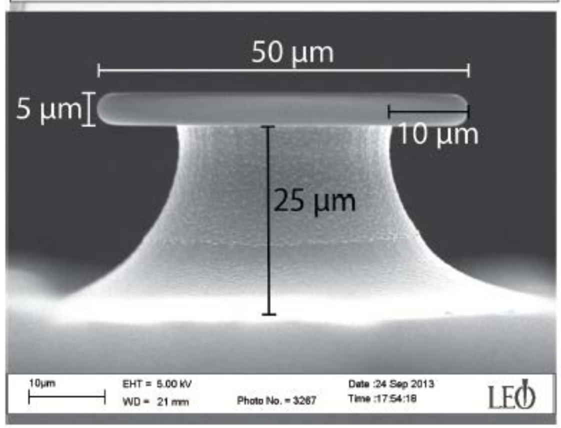
Contents

Setup

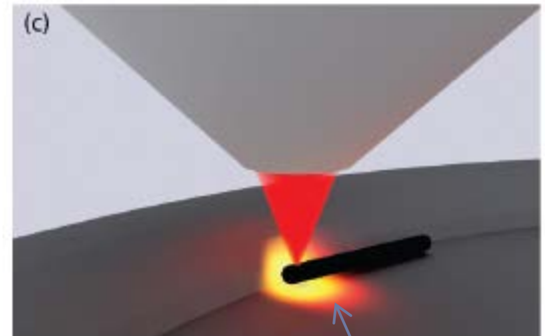
- SEM image of a Toroid microresonator







- AuNR(Gold nanorod)

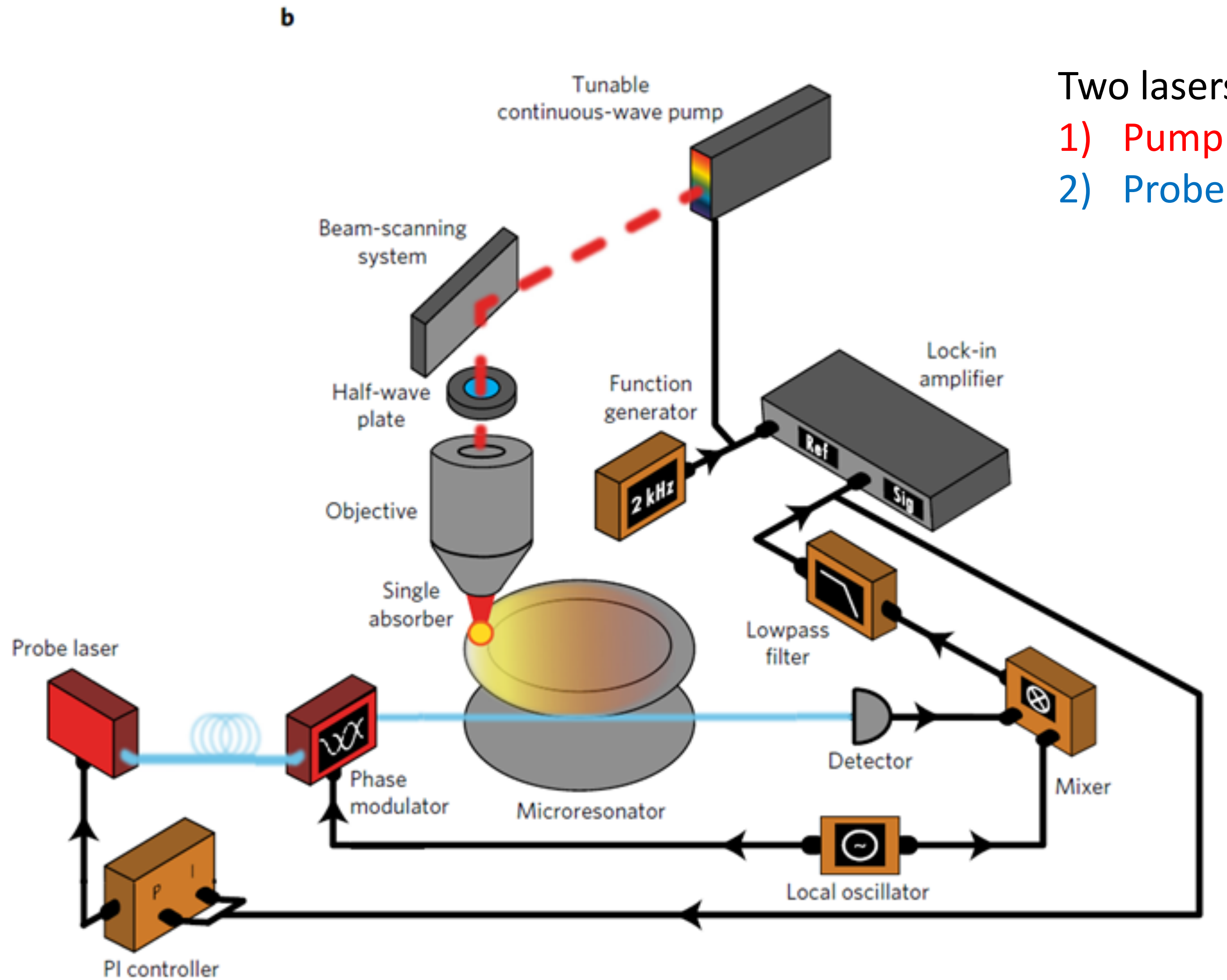


Replaced by Au nanorod

Q factor = 10^7
mode volume= $\sim 300\ \mu\mathrm{m}^3$

Ref. Kevin Heylman’s PhD Thesis

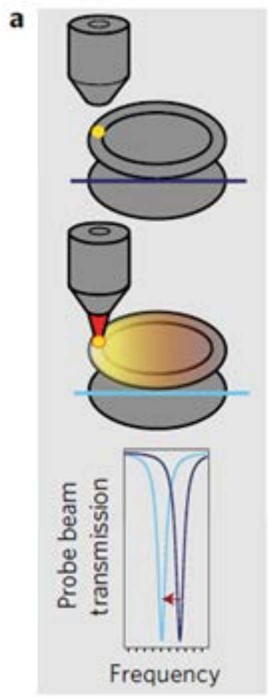
Setup



Two lasers

1) Pump laser
2) Probe laser

Scheme 1. Overall Detection Procedure



Step 1. Pumping AuNR

Step 2. Relaxation->heat dissipation

Step 3. Local temperature increases

Step 4. Photothermal shift of WGM mode freq

Step 5. Detection of shift using probe laser

Step 6. Computing σ_{abs} from shift.

$$\sigma_{\text{abs}} = \frac{q_{\text{heat}}}{I} \longleftarrow$$

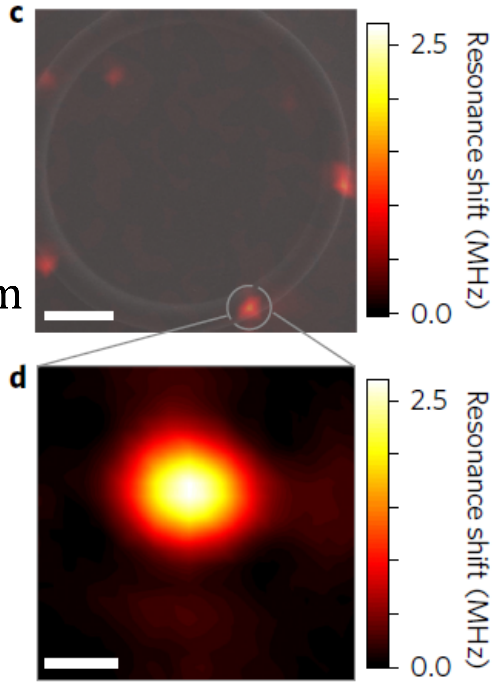
COMSOL FEM simulation

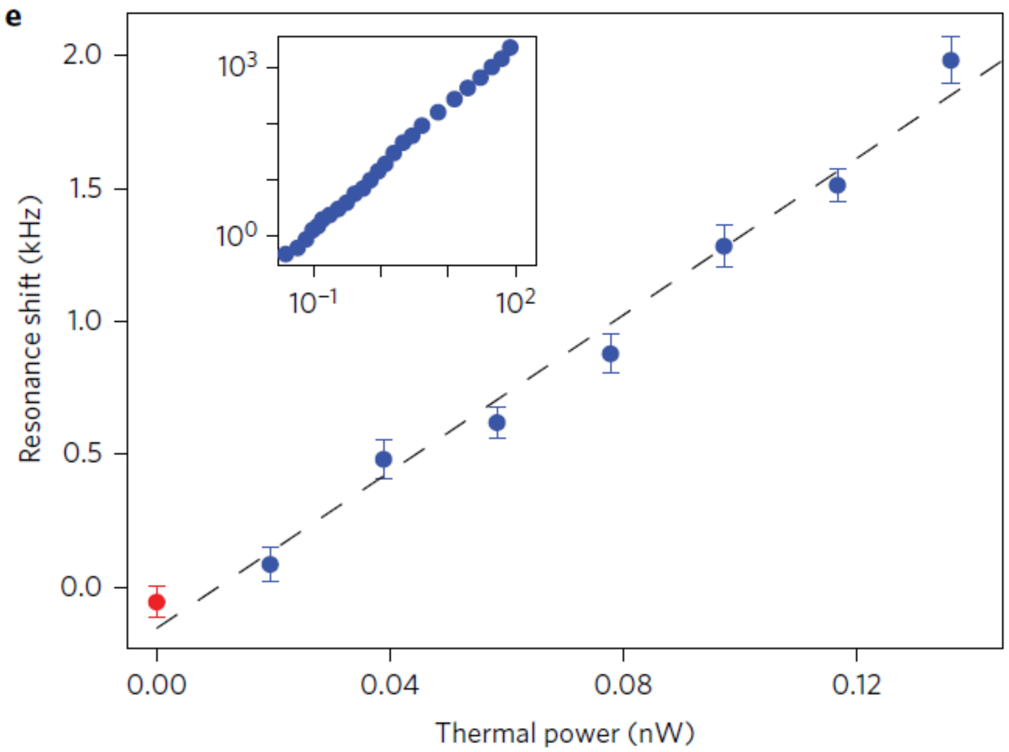
Scheme 1. Overall Detection Procedure

Overlap image of shift and microresonator

10 μm

1 μm





spatial resolution : 1.38 μm

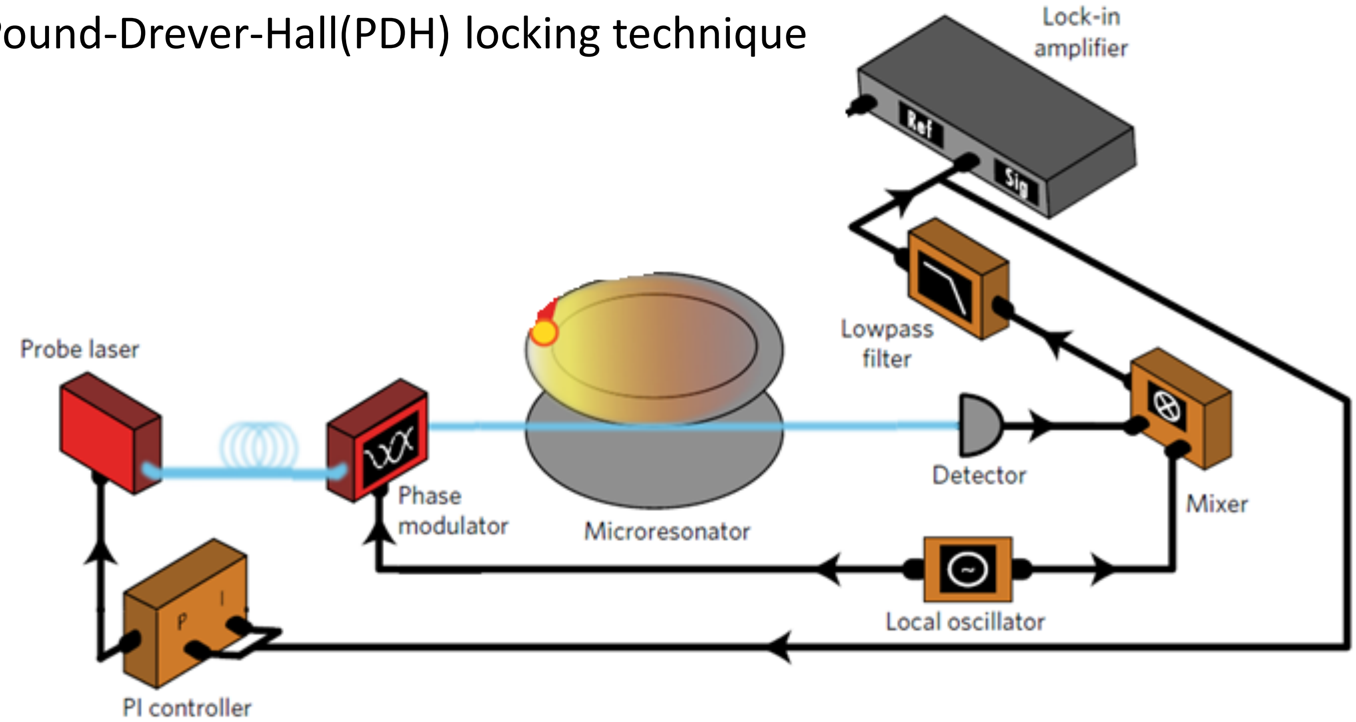
$$\Delta\omega_m = \left(\frac{\omega_m}{n}\frac{dn}{dT}\right)\Delta \mathrm{T}$$

Scheme 2. PDH Locking technique

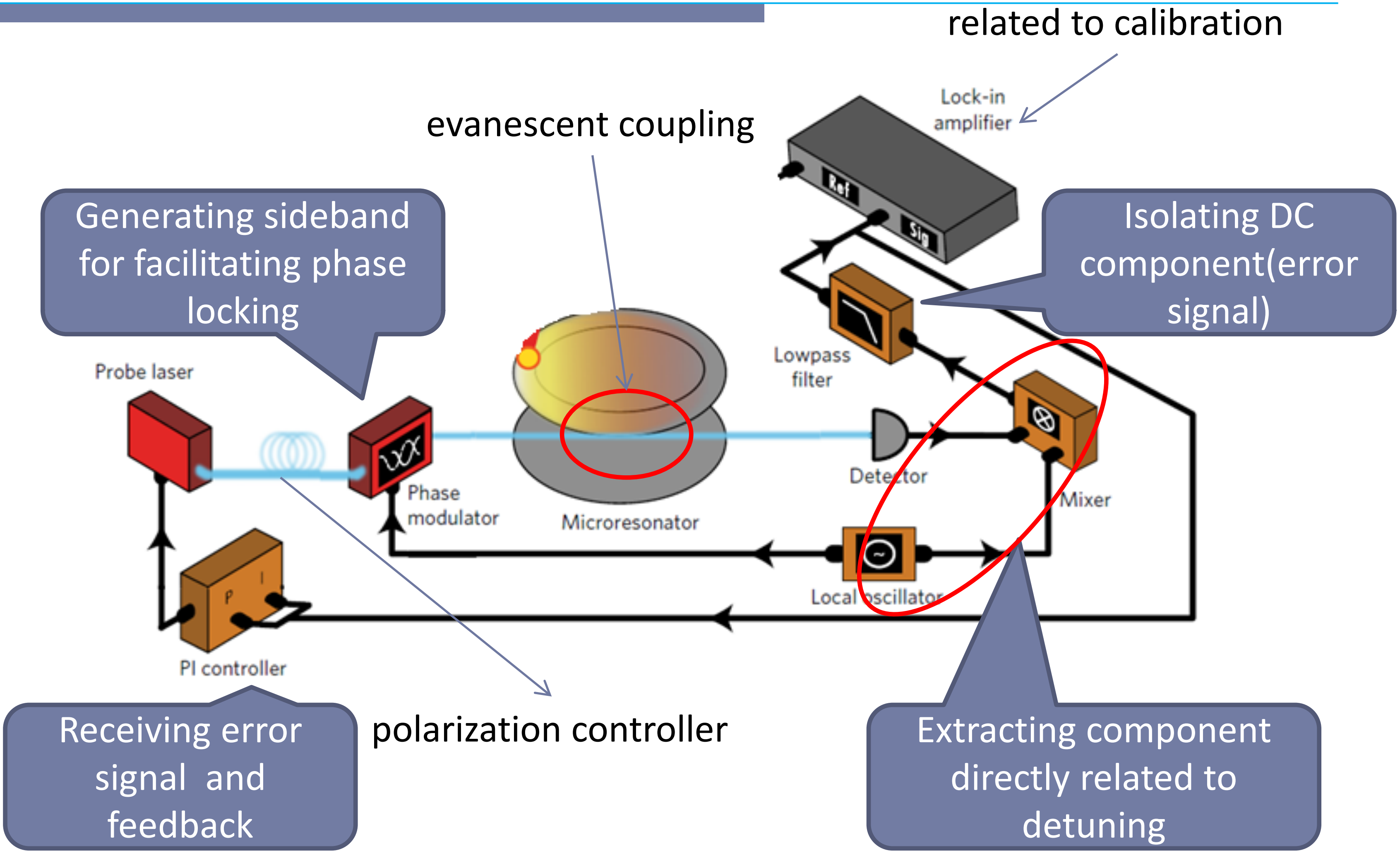
How to measure the small frequency shift

1. Everytime Lorentzian fitting

2. Pound-Drever-Hall(PDH) locking technique



Scheme 2. PDH Locking technique



How sensitive

- Resonance shift : >84 Hz (=> $\Delta T \approx 100\ \mathrm{nK}, \Delta\lambda = 1\ \mathrm{am}$)
- Thermal power detection limit : 20 pW (single dye molecule's : 1 nW)

※Double modulation

acute amp modulation(~0.1 Hz) + PDH locking phase modulation -> very sensitive

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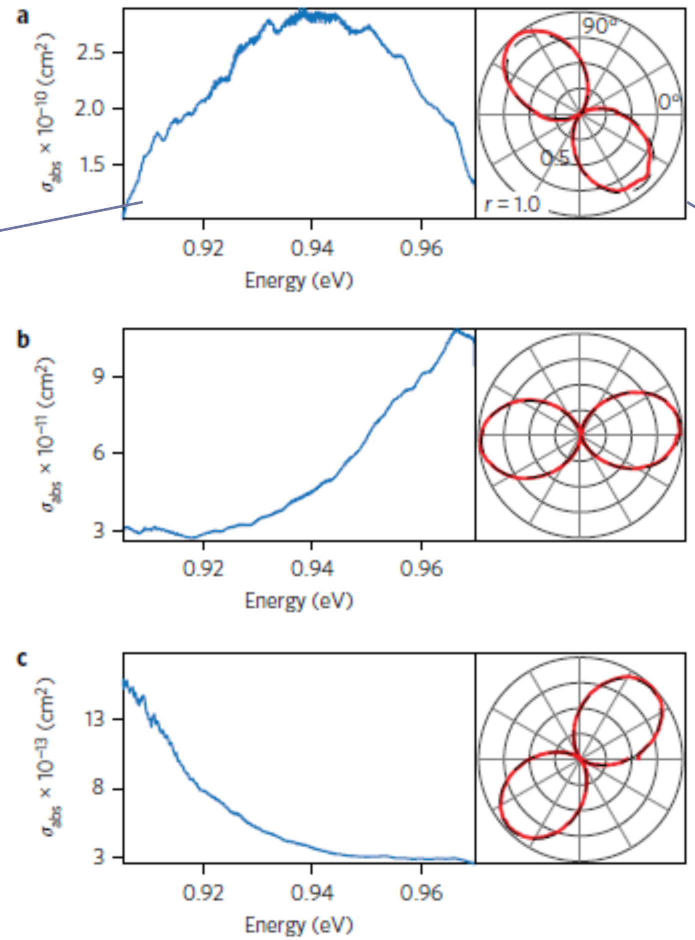
Representative spectroscopic measurement

- Pump and probe are decoupled.->unaffected probing.

LSP peak:
center=0.93 eV
linewidth=68 meV

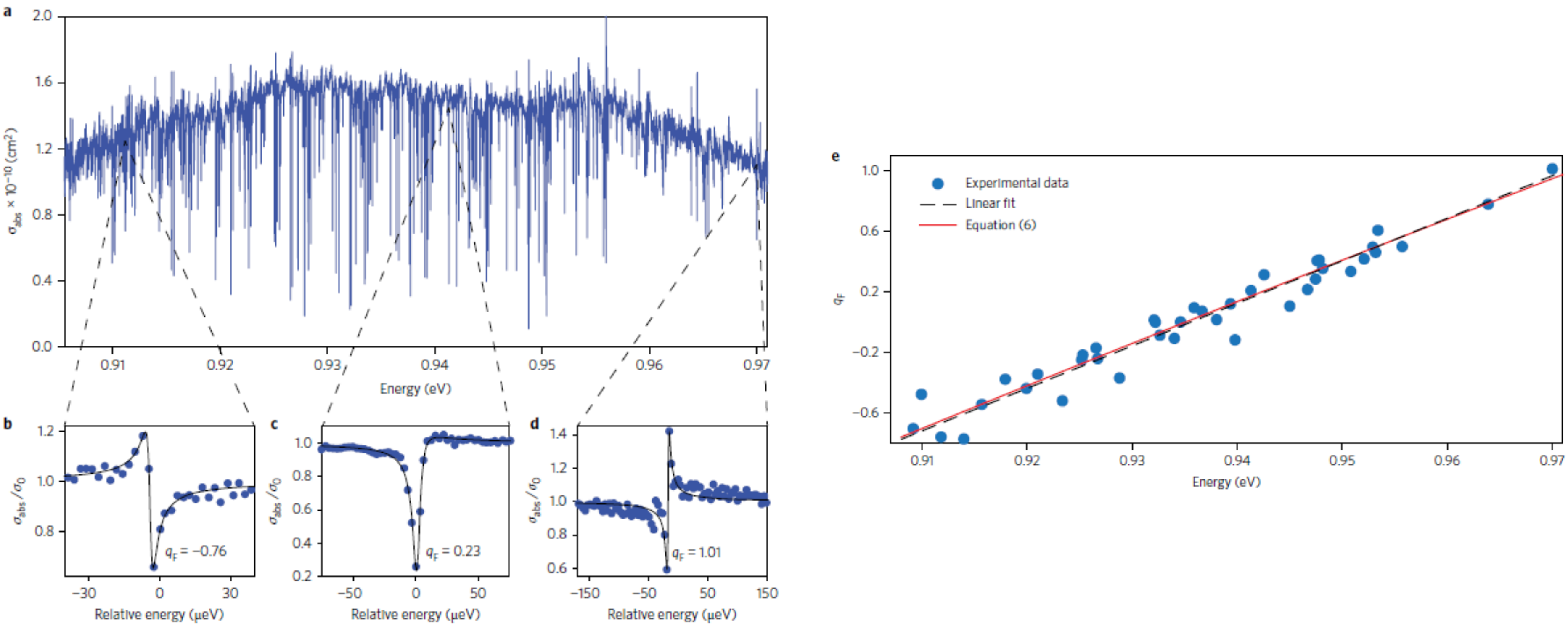


polarization dependence

Asymmetry of Fano resonance

-Signature of WGM-LSP interaction.

-Degree of asymmetry $q_F = \frac{\omega_1}{\gamma_0} - \frac{\omega_0^2}{\omega_1\gamma_0} \approx \frac{2}{\gamma_0}(\omega_1 - \omega_0)$



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Classical coupled oscillator model

1) For LSP in single AuNR(p_0, ω_0, γ_0),

$$\ddot{p_0} + \gamma_0 \dot{p_0} + \omega_0^2 p_0 + \sum_m g_m^2 \omega_0^2 p_m = \omega_0^2 \sqrt{V_0} E_0 e^{-i\omega t}$$

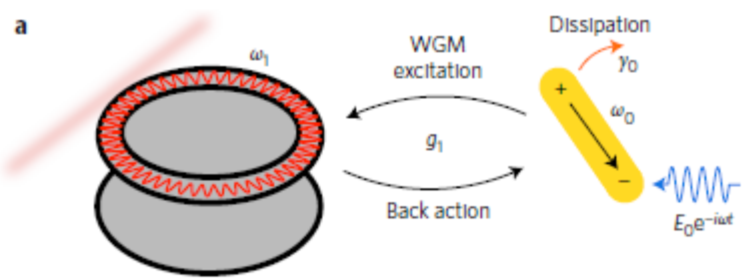
2) For WGM modes(p_m, ω_m)

$$\ddot{p_m} + \omega_m^2 p_m + g_m^2 \omega_m^2 p_0 = 0$$

g_m : coupling between mth WGM and LSP

ω : driving laser frequency

V_0 : mode volume



Classical coupled oscillator model

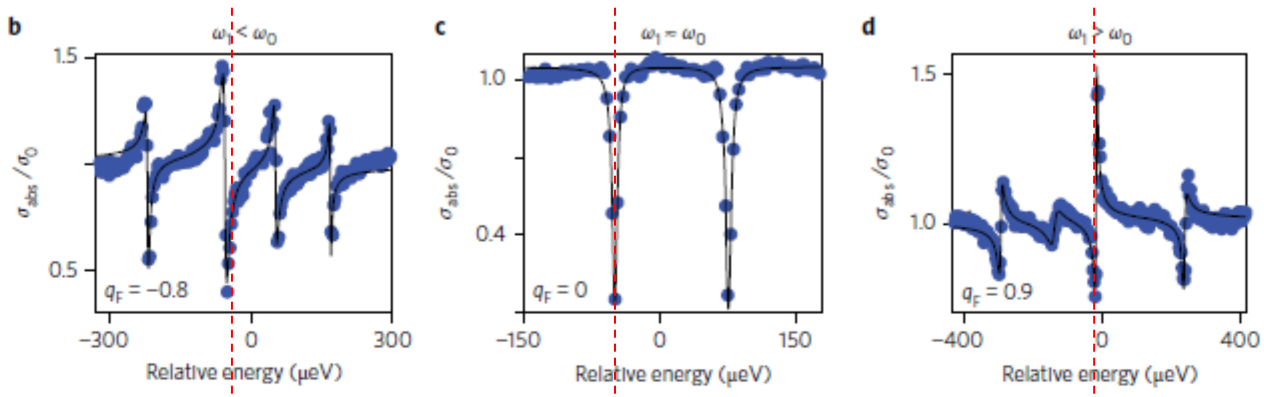
Theoretical absorption cross section

$$\sigma_{\text{abs}}(\omega) = \sigma_0(\omega)\left|\frac{\omega\Gamma q_{\text{F}} + \omega^2 - \Omega^2}{\omega^2 - \Omega^2 + i\omega\Gamma}\right|^2$$

Ω, Γ : Spectral location and linewidth of Fano resonance.

Classical coupled oscillator model

1) Interaction between single AuNR and single WGM mode



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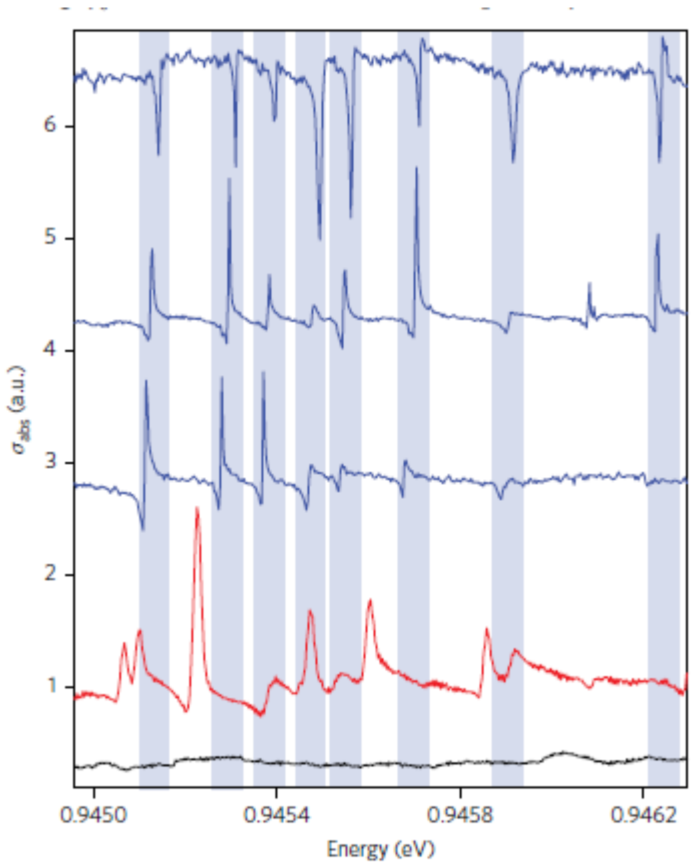
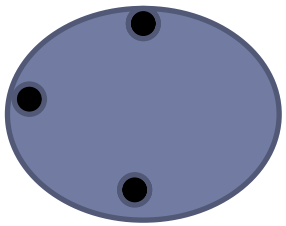
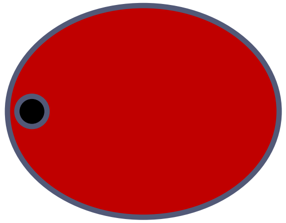
Classical coupled oscillator model

2) Coupling effect on spectrum location





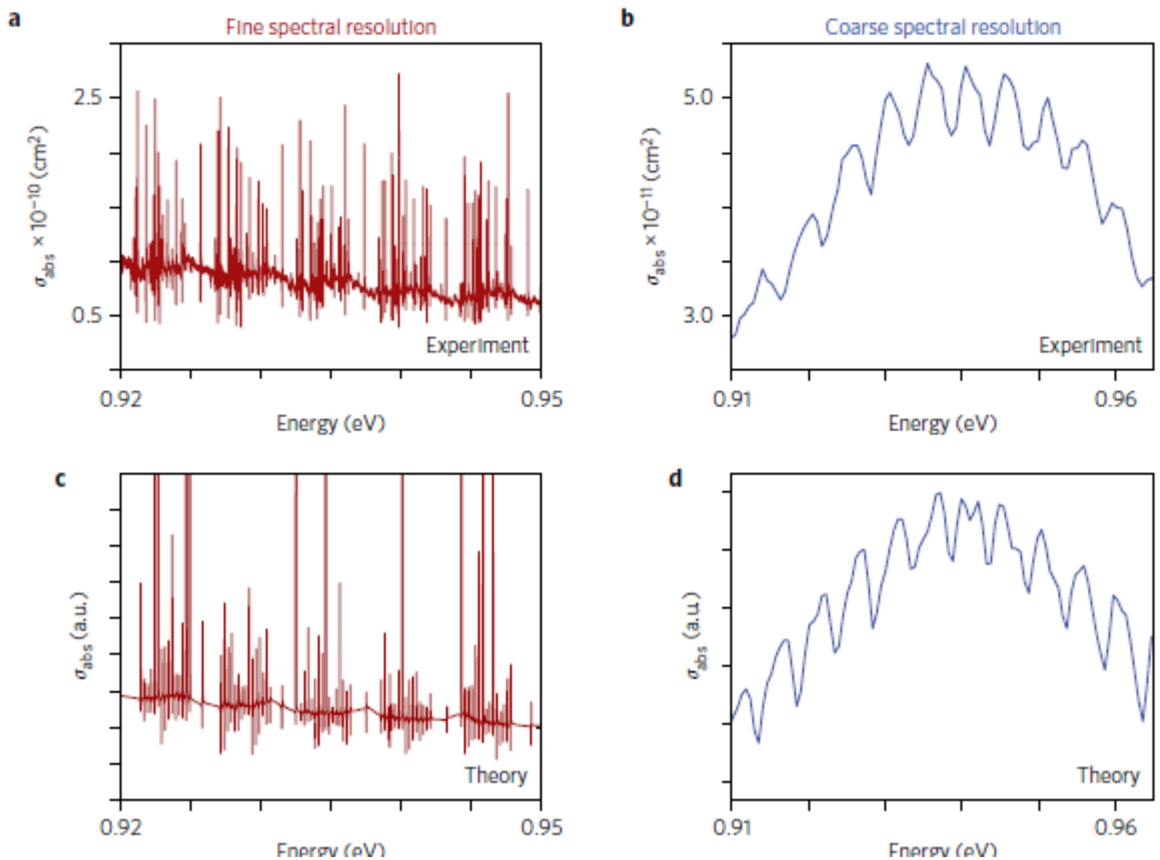
~1 meV shift
-> negligible



dominantly depend on microresonator

Classical coupled oscillator model

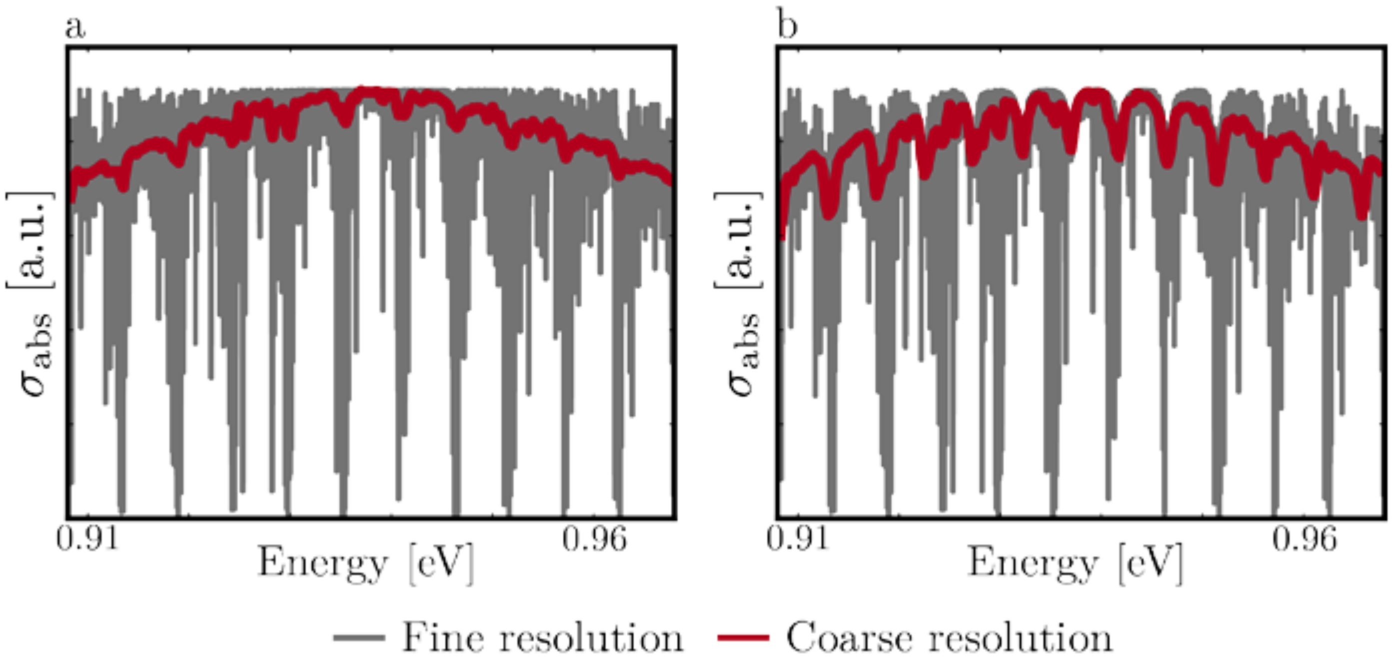
3) Interaction between many AuNR and many WGM modes of same microresonator



Theory for many AuNRs

Classical coupled oscillator model

Theory for single AuNR Theory for two AuNRs



-From this observation, classical coupled oscillator model was verified.

-Unexpected long range interaction?

Question

- Any Question?