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Liquid droplet whispering-gallery-mode optical resonators

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2nd International Conference and Exhibition on
Lasers, Optics & Photonics, Philadelphia 8-10 Sept, 2014

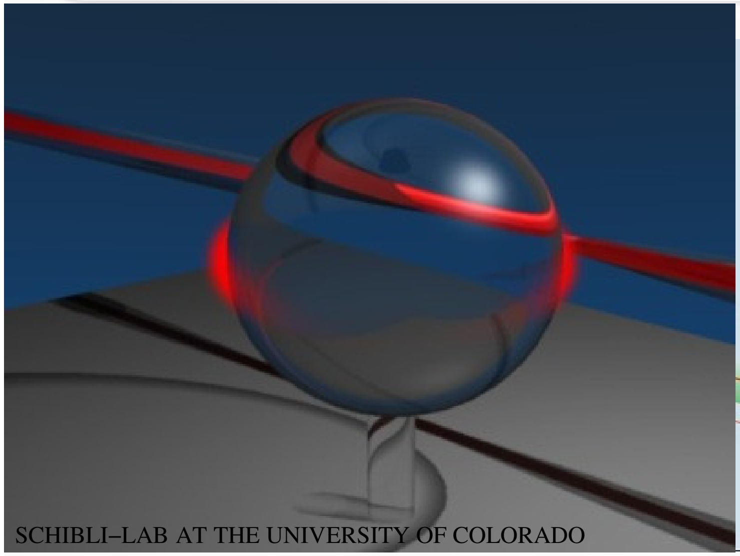
Gianluca Gagliardi

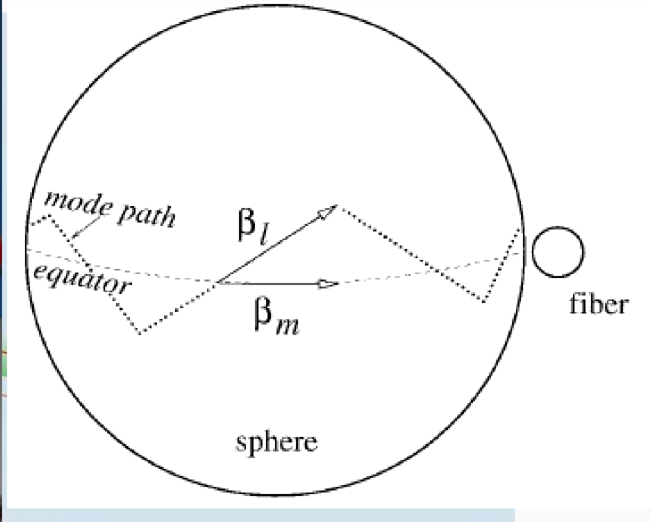
National Institute of Optics (INO),
Italian Research Council
Napoli, ITALY

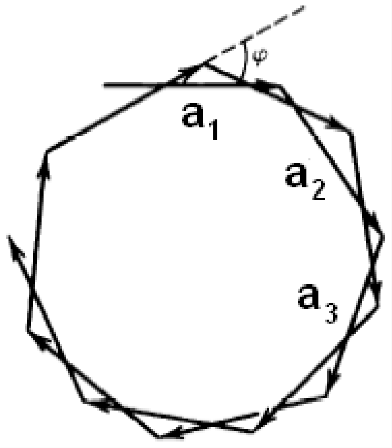
Outline

- Introduction to WGMs
- WGM of liquid droplets in the near IR
- Laser frequency locking to WGMs
- Cavity ring-down spectroscopy in droplet resonators
- Wavelength-shift tracking for dissolved particles
- WGMs detection of NPs in the visible
- Conclusions and outlook

Whispering gallery mode (WGM) resonators


SCHIBLI–LAB AT THE UNIVERSITY OF COLORADO





Total internal reflection

Resonance condition

$$x_{N,l} \cong \frac{2\pi R}{\lambda}$$

$$\Downarrow$$

Fundamental equatorial modes

$$FSR_{WGM} \cong \frac{c/n}{2\pi R}$$

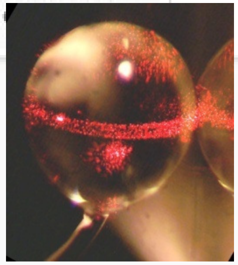
- Solution: spherical Bessel fun *N*, spherical harmonics *l*,*m*
- Modes: angular momentum num *l*, projection *m*

N number of radial maxima

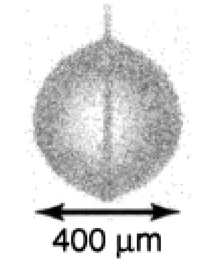
$$\psi \propto Y_{lm}(\theta,\phi)\, j_n(kr)$$

- Analogy between WGMs and acoustic waves
- Smallest volume, lowest loss (n=1, l max, $m=\pm l$)
- Light field confined near the sphere surface
- EW into the external medium

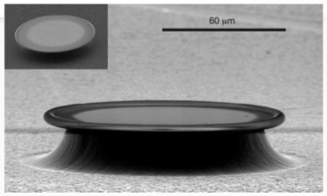
WGM resonators



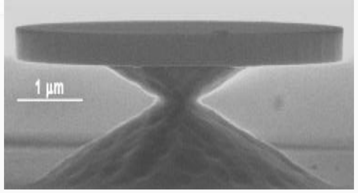
Fused silica: Gorodetsky et al., OL **21**, 453 (1996).



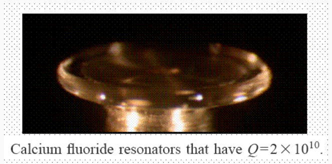
Solid H_2: K. Hakuta et al. OL **27** (2002)



Fused silica: K. Vahala et al., Nature **421**, 925 (2003)

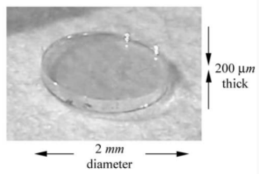


Si: Appl. Phys. Lett. **85** (2004)



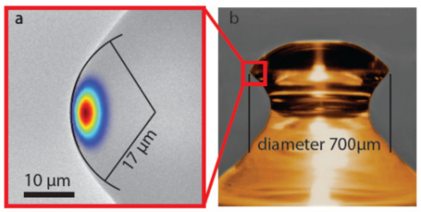
Calcium fluoride resonators that have $Q=2\times10^{10}$.

CaF_2: Savchenkov et al, PRA **70**, 051804 (2004); OE **15**, 6768 (2007).

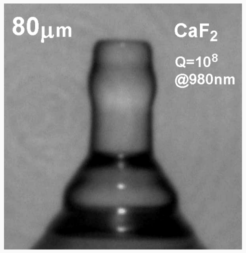


LN WGM: D.A. Cohen and A.F.J. Levi, Electron. Lett. **37** (1) , 2001.

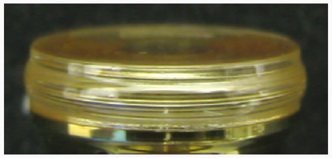
CaF_2 $Q_m>10^5$: J. Hofer, A. Schliesser, P. Del'Haye, and T. Kippenberg (CLEO'09)



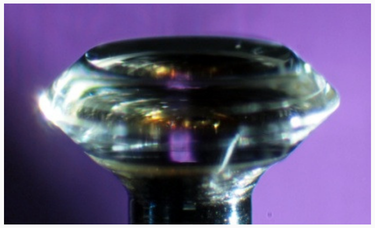
MgF_2 resonator, C. Y. Wang, T. Herr, P. Del'Haye, A. Schliesser, J. Hofer, R. Holzwarth, T. W. Hänsch, N. Picqué, T. J. Kippenberg, arXiv:1109.2716



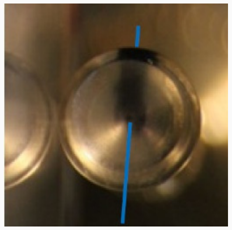
Grudinin et al Opt. Commun. 265, 33-38 (2006).



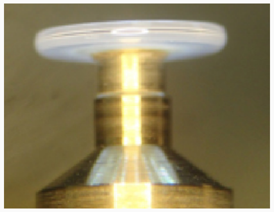
LN WGM resonator, D. Haertle, T. Beckmann, J. Schwesyg, S. Hermann, A. Zimmermann, K. Buse Photonics West 2009



MgF_2 resonator, OEwaves (2011)



BBO resonator, JPL, 2012



SBN resonator, OEwaves (2012)

Fabricating high Q spherical microresonators is very easy…

Spherical micro-resonators as sensors

$$Q=\omega_0\tau=\frac{\omega_0}{\Delta\omega}\cong\frac{\omega_0 L}{c\cdot\gamma}\qquad(>10^9)$$

$$F=\frac{FSR}{\Delta\omega}\cong\frac{\pi}{\gamma}\quad \textit{finesse}$$

Loss mechanisms:

- Radiative loss
- Material absorption
- Surface scattering
- Coupling loss (prism, tapers)

J.A. Barnes, G. Gagliardi, H.-P. Loock, *Optica* 1 (2), 75 (2014)

Whispering-gallery modes in solid cavities

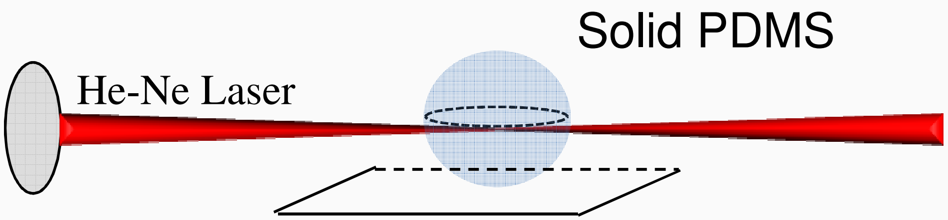
- Long-decay time, high Q (although…)
- Sensitive detection on small volumes
- Silica MRs used mostly for “refractive” sensing
- Optical coupling: prisms, fiber tapers…
- WGM: EW tail outside the sphere
- Few works on gas sensing with CEAS
- Liquid sensing difficult

Liquid droplet resonators: motivations

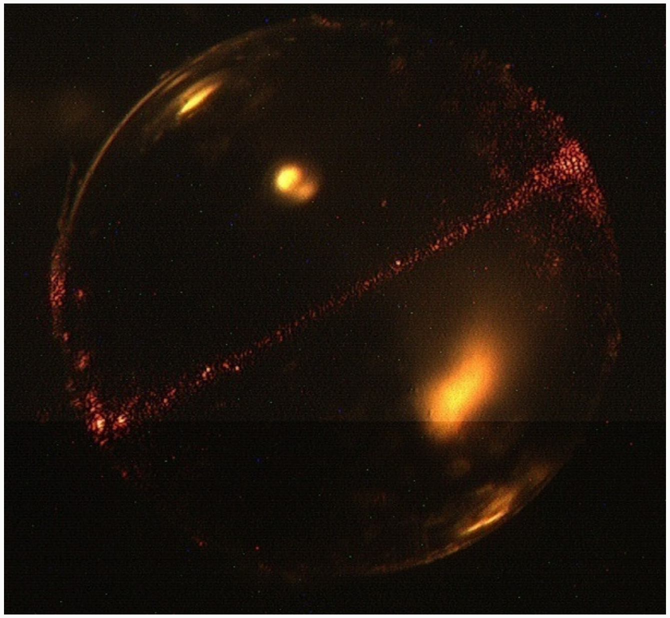
- Optical cavities with relatively-high Q-factor (~ 10^5-10^7)
- Analyte or particles inside resonator
- Full interaction with WGM mode, not only evanescent field
- Ease of fabrication

- How to couple light into a liquid resonator
- How to handle it
- Deal with evaporation if water…

Free-space WGM coupling: preliminary



Ex: Solid PDMS sphere (dia ~ 1.5 mm)

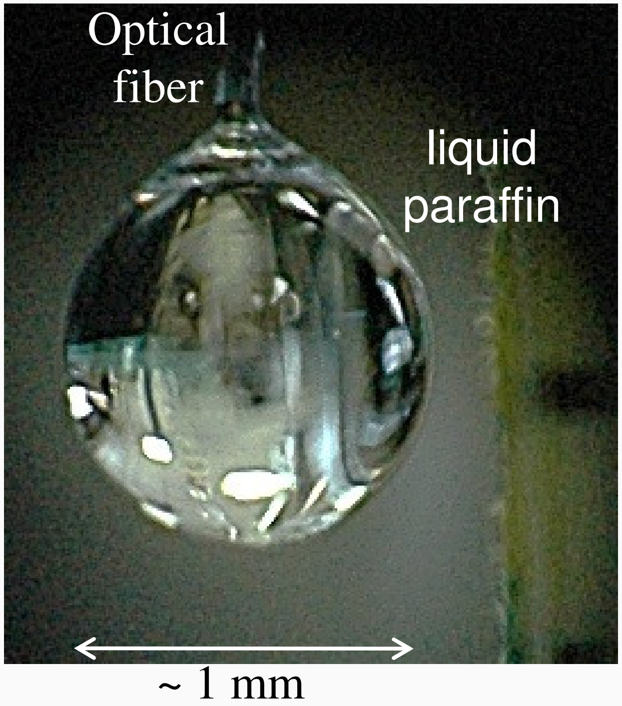


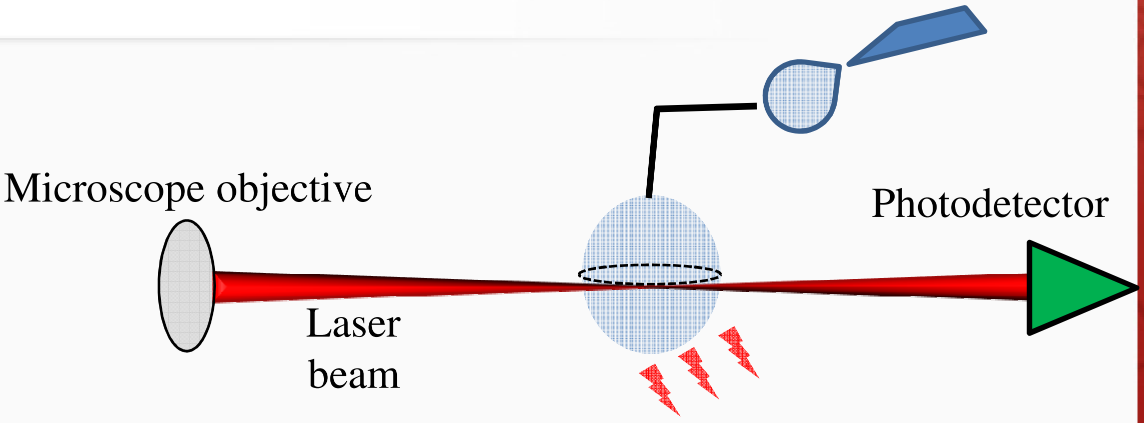
Problems to droplet cavities:

- Alignment
- How to handle the drop:

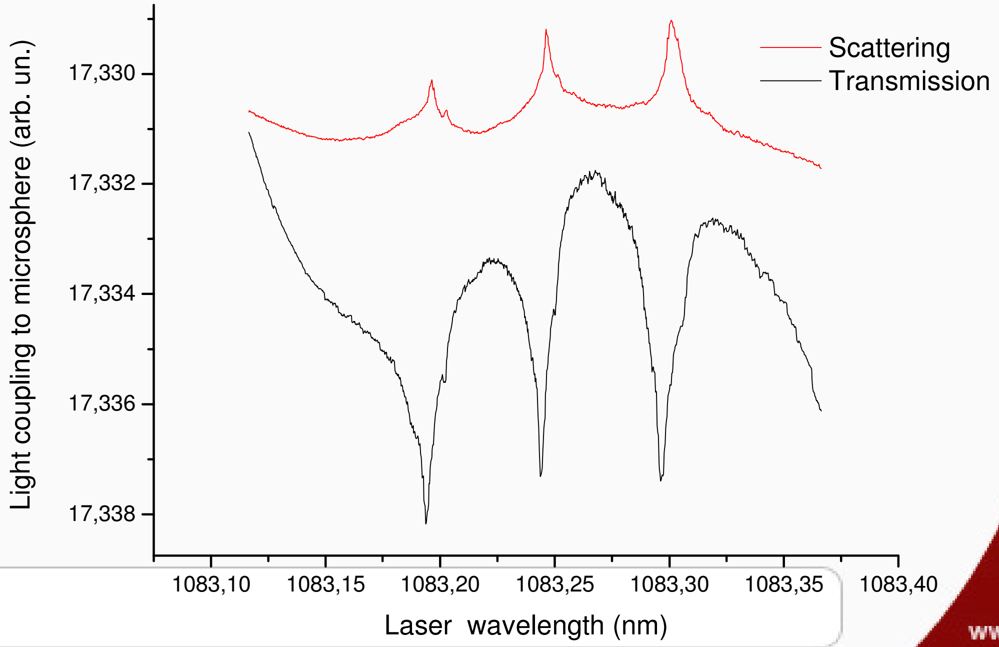
-hydrophobic/hydrophilic surface?

WGMs in liquid drop resonators

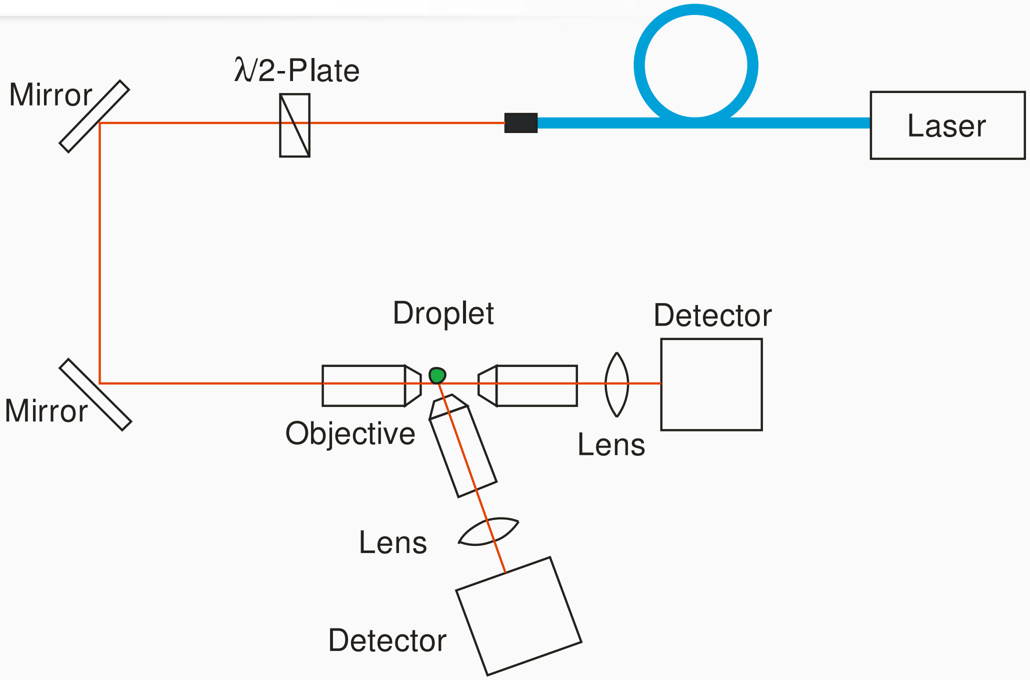




Resonance spectra
observed by a current-modulated
DBR Laser at 1083 nm



Optical configuration for free-space coupling

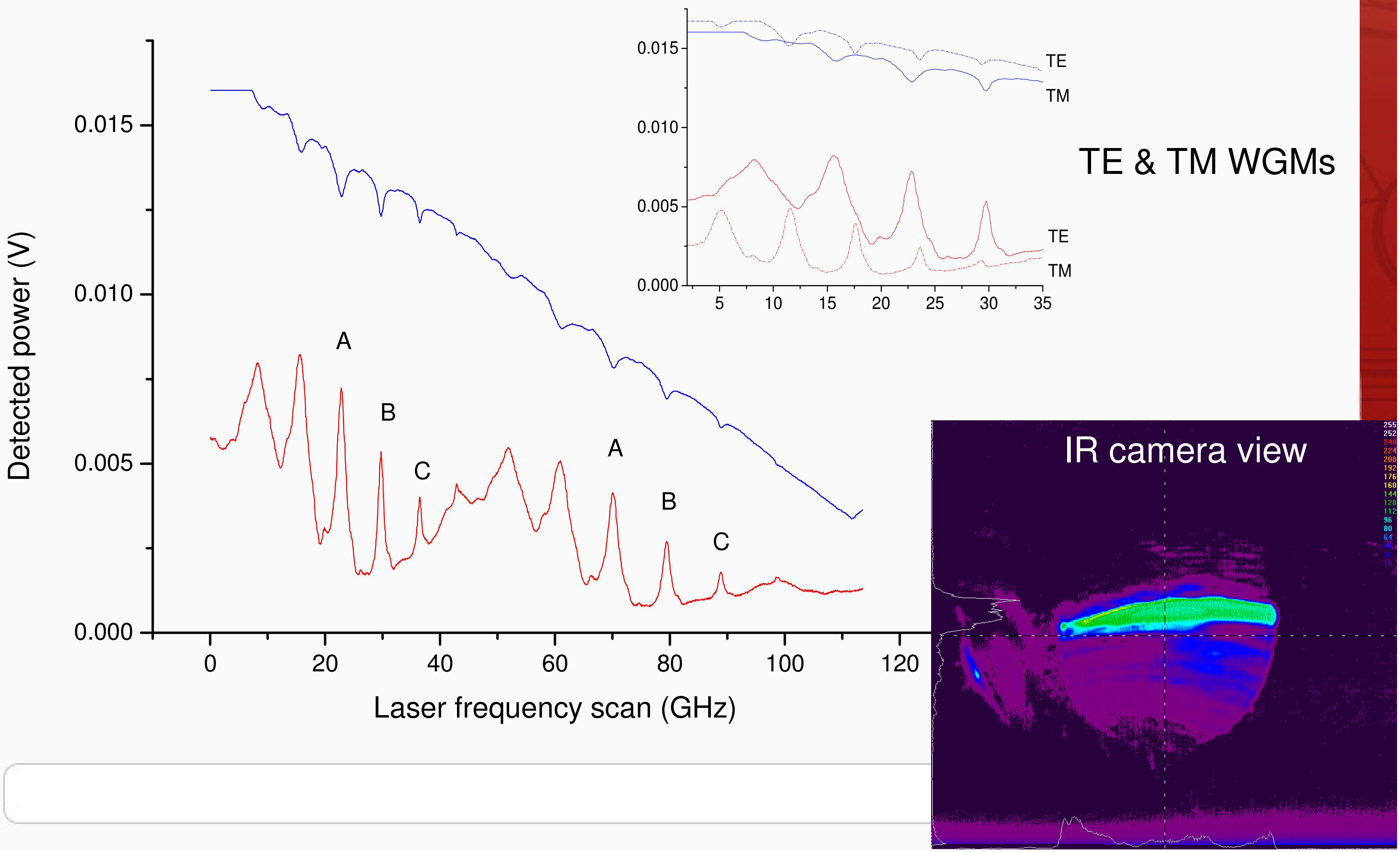


- Q ~ 10^5
- Mechanically robust

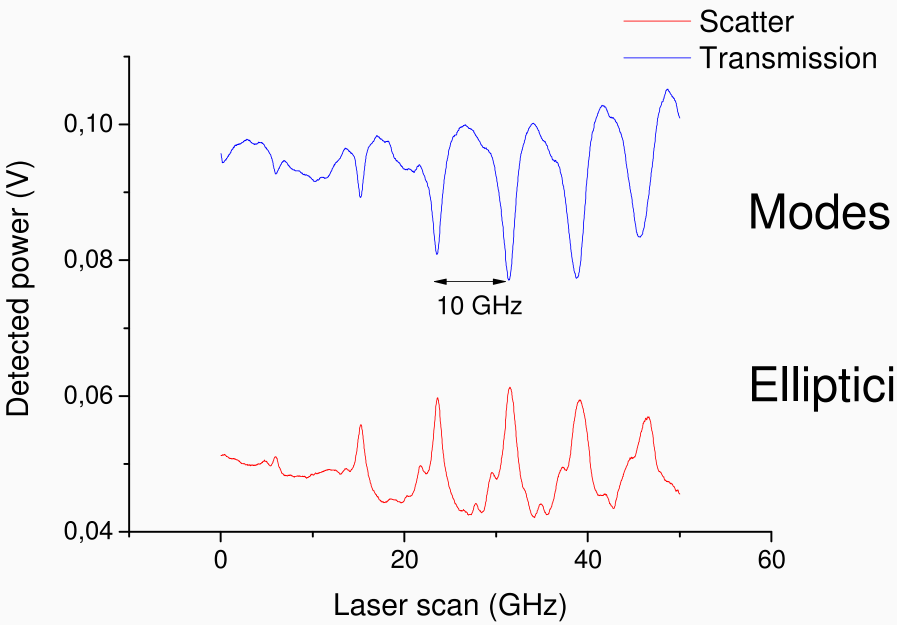
- Highly reproducible

- Diameters from ~ 200 to 1400 µm

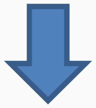
WGMs spectra at 1560 nm



WGMs spectra at 1560 nm



Modes are not degenerated



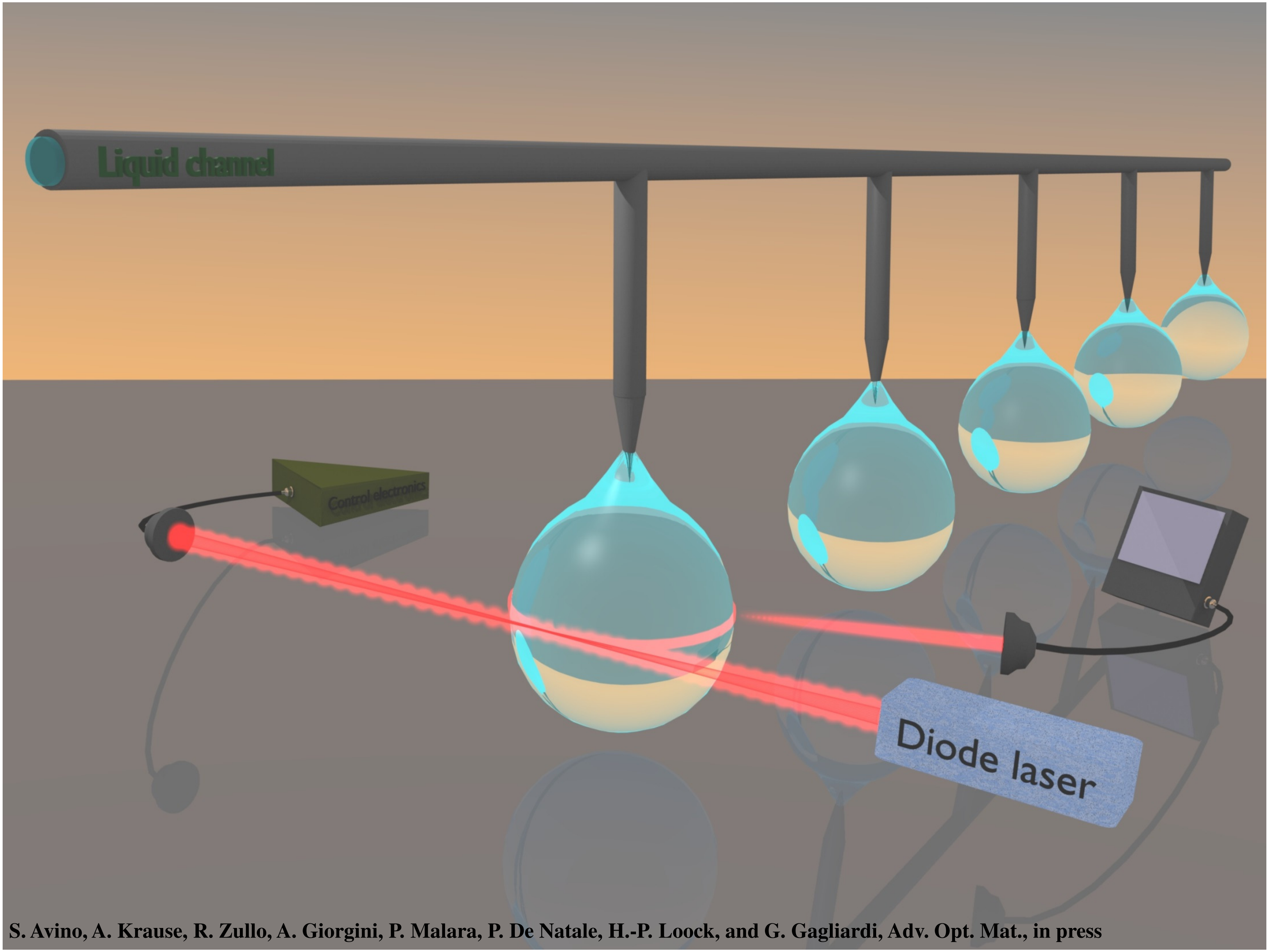
Ellipticity of the droplet

Degeneracy lifting due to ellipticity

$$\nu(m) = \nu_0\left[1 - \frac{e}{6}\left(1 - \frac{3m^2}{l(l+1)}\right)\right]$$

$$e = (r_{pol} - r_{eq})/R,$$

$$\Delta\nu(l) = \nu_0 e / l$$



S. Avino, A. Krause, R. Zullo, A. Giorgini, P. Malara, P. De Natale, H.-P. Loock, and G. Gagliardi, Adv. Opt. Mat., in press

Laser-frequency locking to WGMs

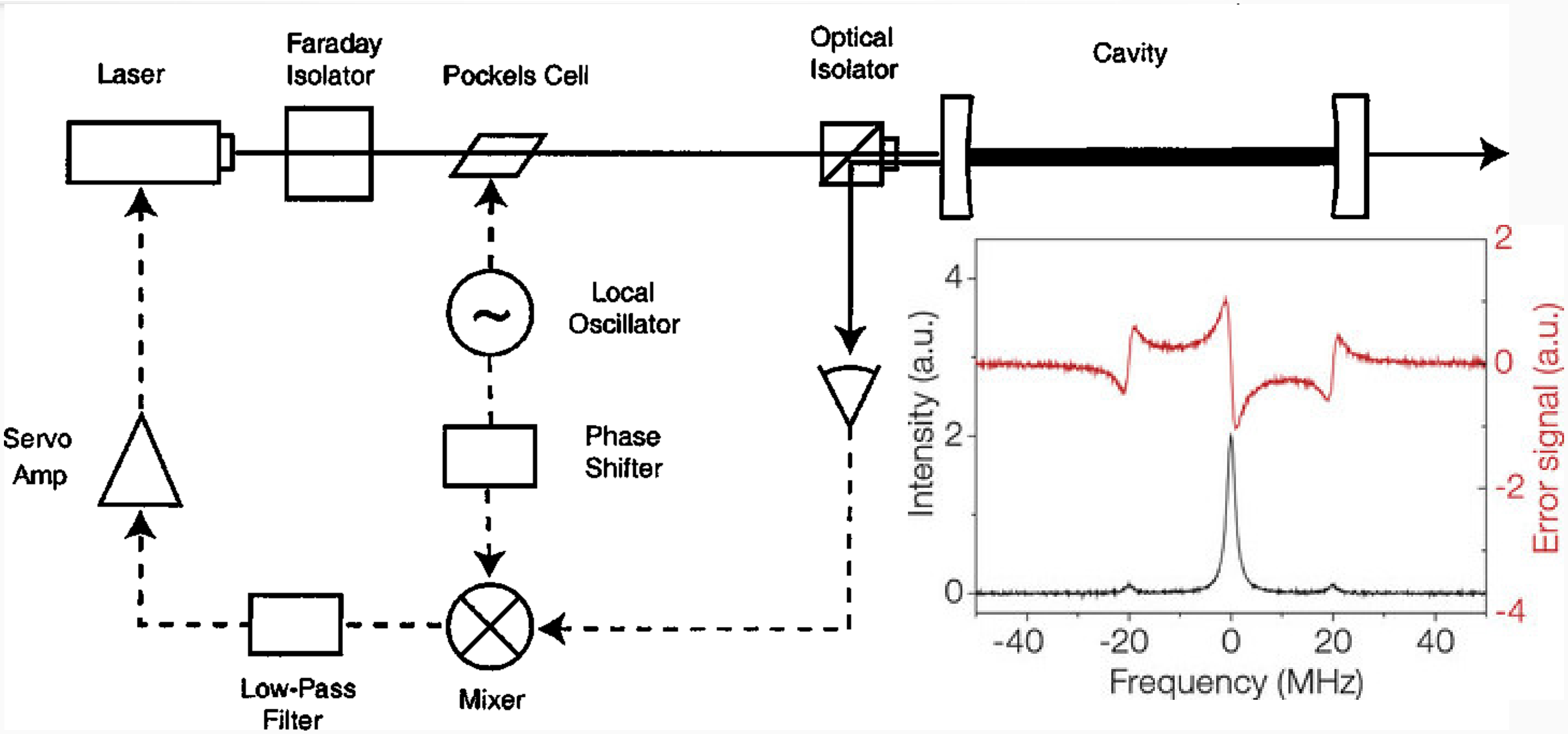
Locking the laser emission frequency to the cavity modes allows

- -Real-time tracking of the resonance
- -No need for laser λ scan
- -Reduced need for fitting or post-processing
- -WGM shift measurements below the HWHM possible
- -static or dynamic measurements possible

Very-low noise techniques can be used to extract a suitable signal for locking & measure tiny effects in the cavity…

(G. Gagliardi e al., Science 330, 1081-1084 (2010))

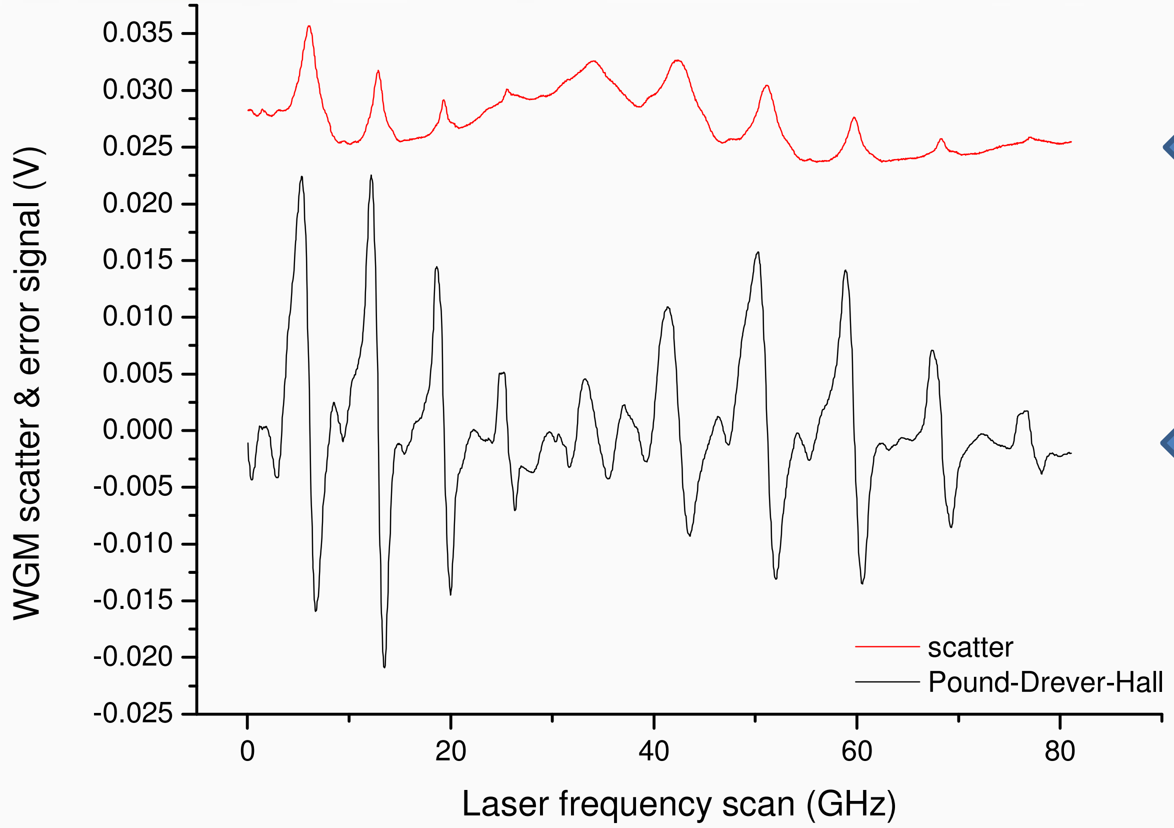
The Pound-Drever-Hall technique



- Laser frequency detuning is measured vs resonance
- The measured signal is fed-back to the laser (or the cavity)
- Null detection – total decoupling from the laser intensity noise
- Intrinsic low-noise measurement - high frequency modulation
- High bandwidth lock

E. D. Black, Am. J. Phys. 69, 79 (2001)

PDH locking signals for WGMs



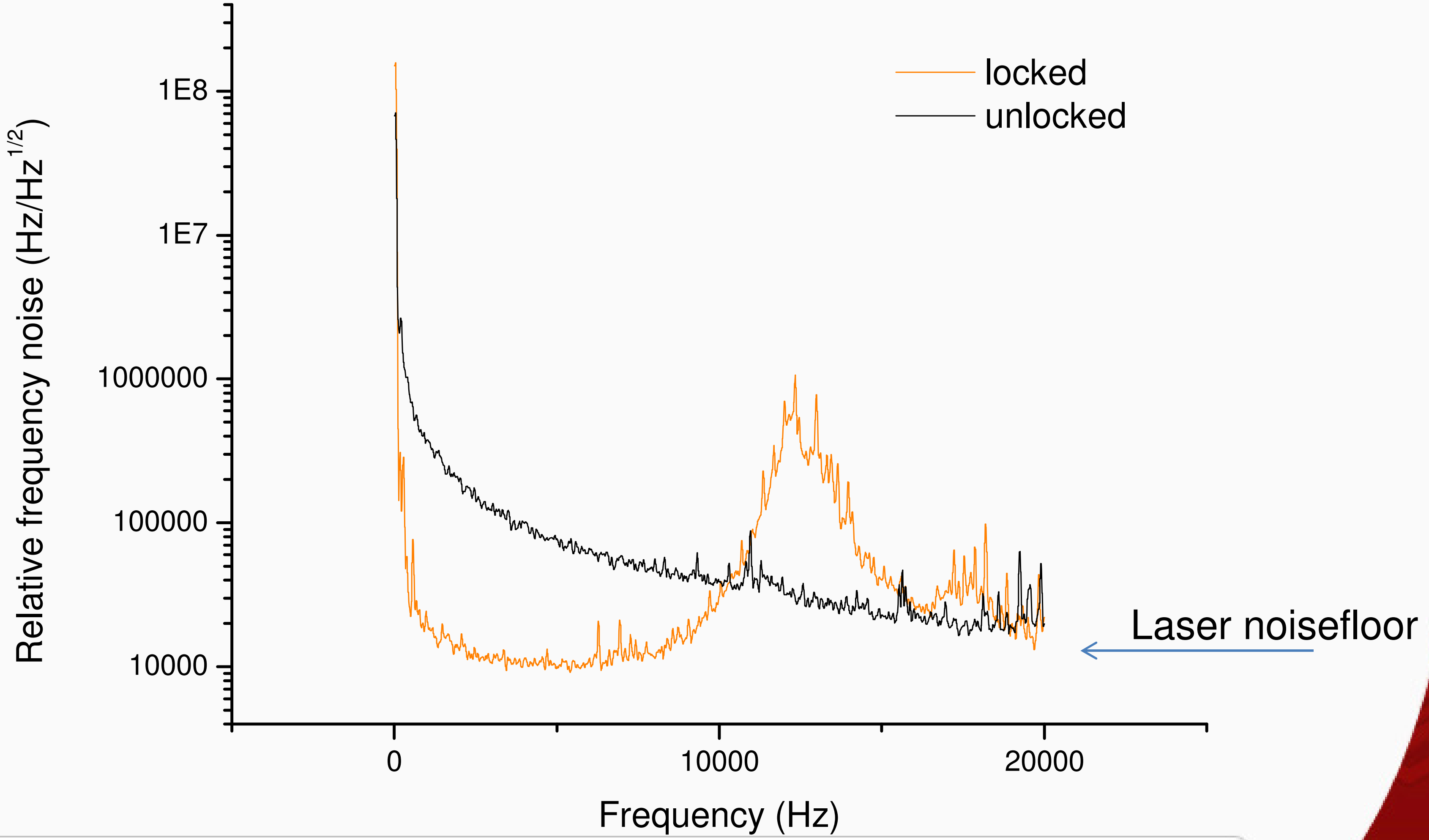
Scatter

PDH from transmission
high S/N

Active stabilization of the laser on a single WGM …even below the mode linewidth

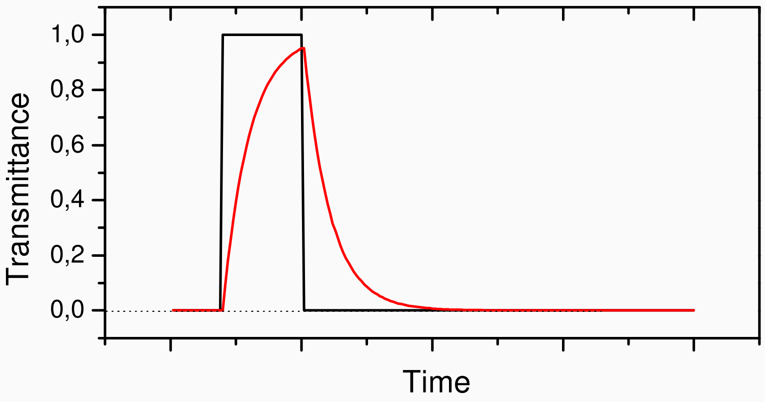
Laser-frequency locking to the resonance

From the FFT of the error signal:

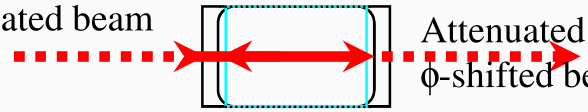


Laser noisefloor

Cavity ring-down spectroscopy (CRDS)

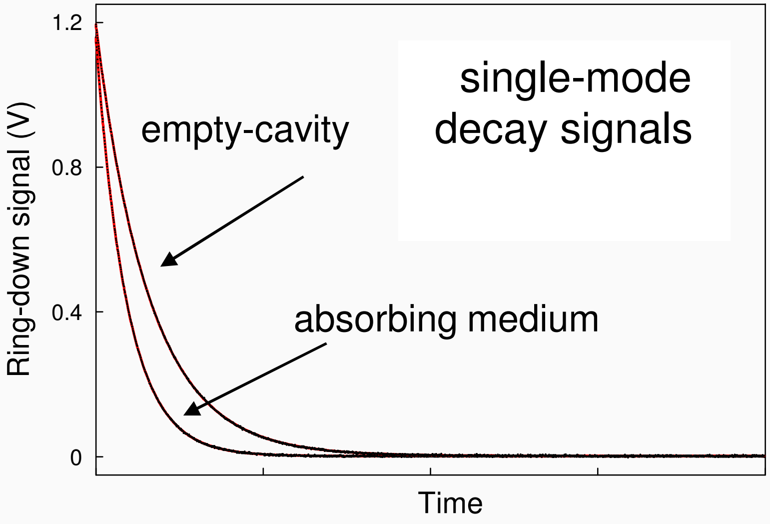


$$\tau = \frac{t_R}{-2\ln R + \varepsilon C d}$$



Amp modulated beam

Attenuated &
ϕ-shifted beam



Absorption coefficient

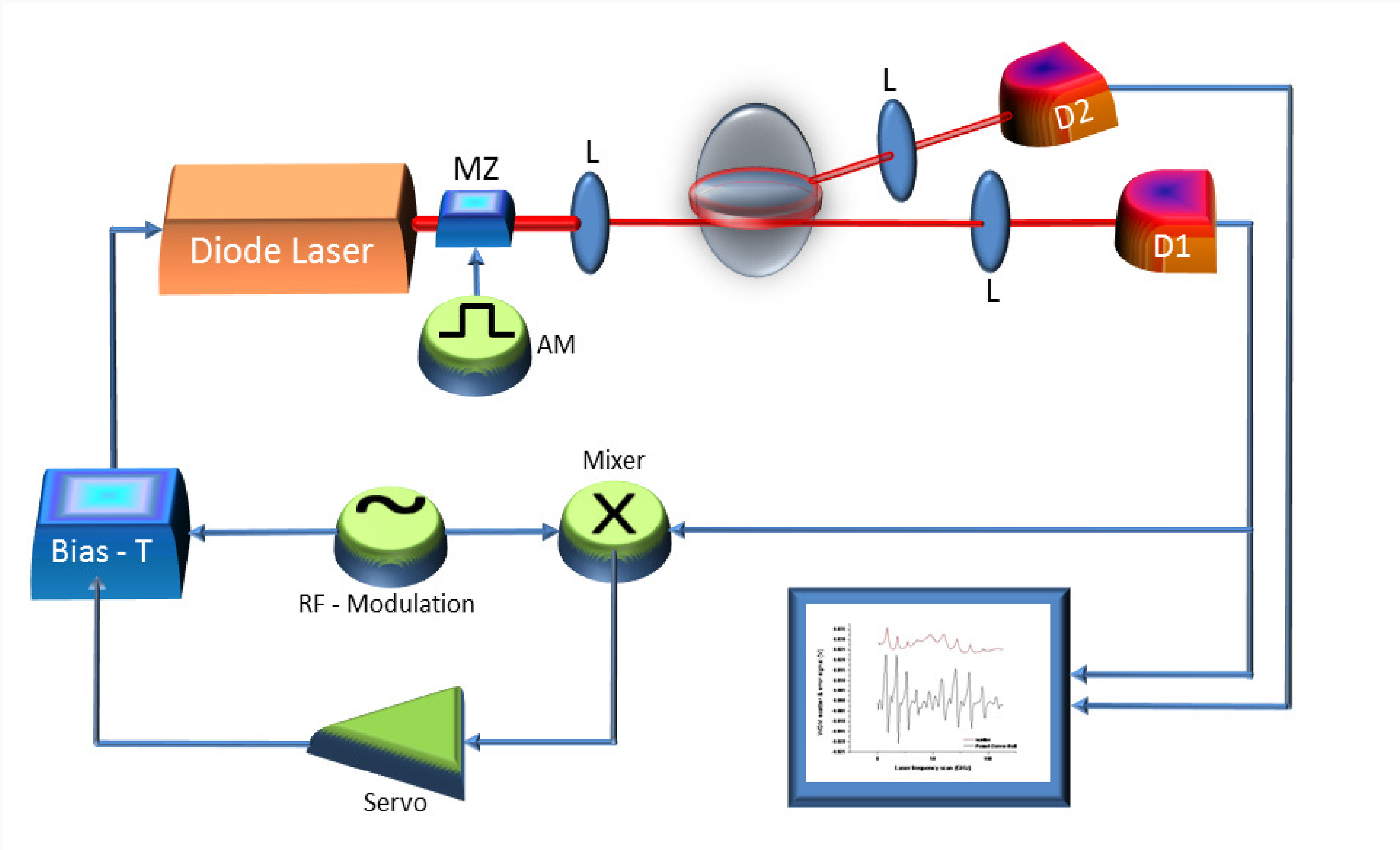
$$\alpha = \frac{n}{c}\left(\frac{1}{\tau} - \frac{1}{\tau_0}\right)$$

$$\alpha_{\min} \cong \frac{(1-T)\Delta\tau_{\min}}{L(\tau_0 - \Delta\tau_{\min})} \cong \frac{1}{c\tau_0}\,\frac{\Delta\tau_{\min}}{\tau_0}$$

Time-domain CRDS:

- Direct absorption coeff
- Fast detect
- Noise immune

Lock & Detection scheme

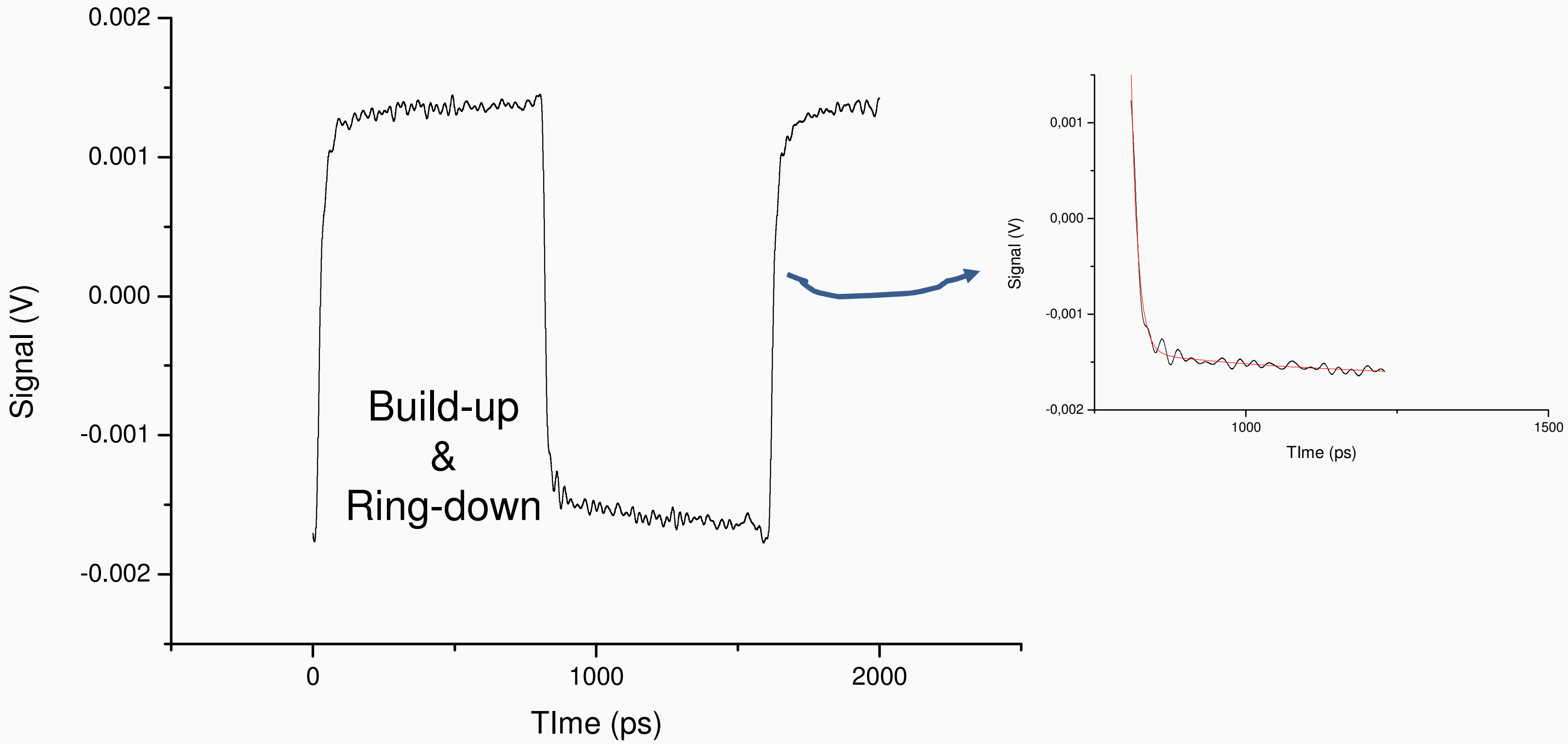


S. Avino, A. Krause, R. Zullo, A. Giorgini, P. Malara, P. De Natale, H.-P. Loock, and G. Gagliardi, Adv. Opt. Mat., in press

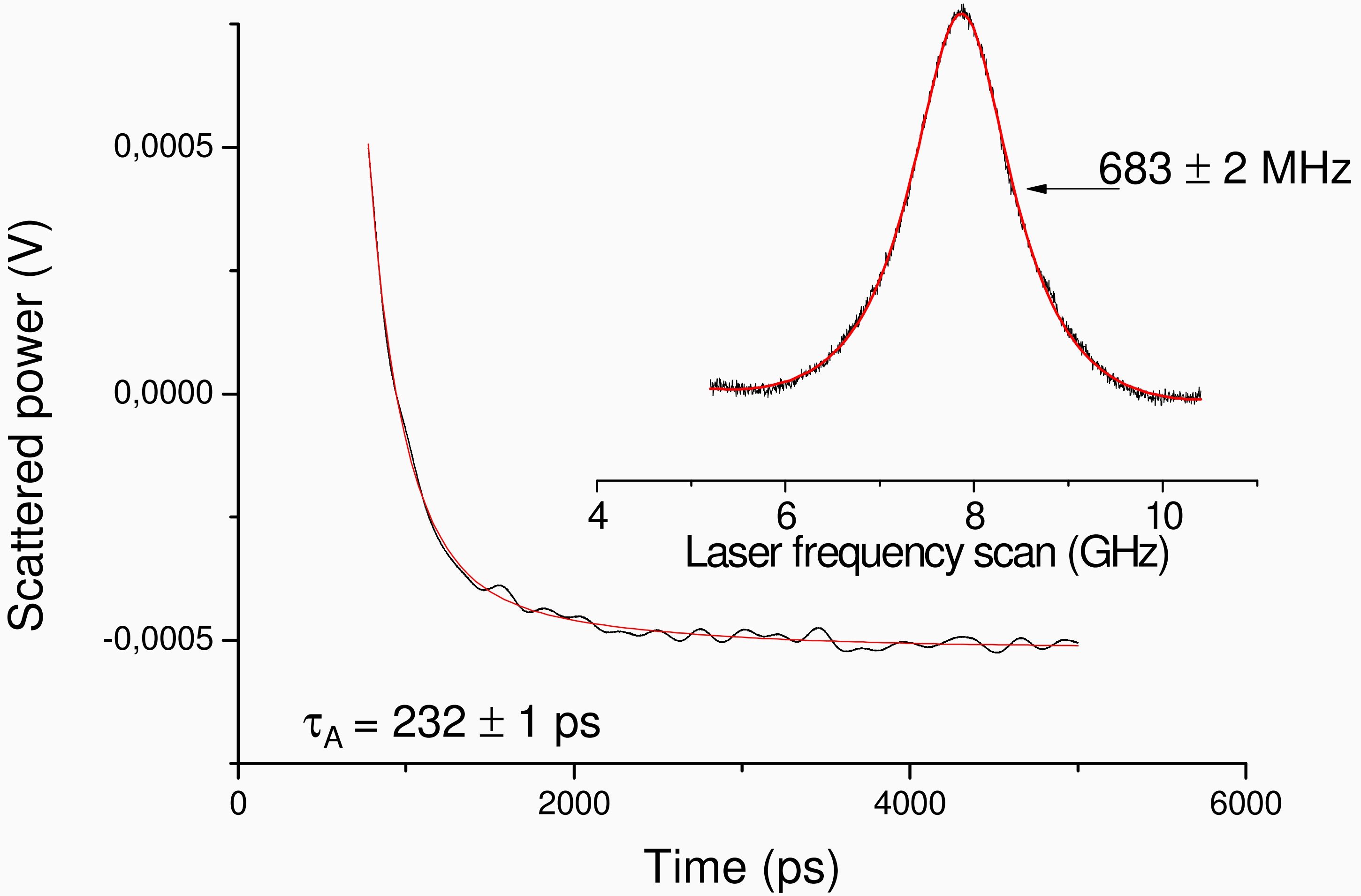
WGM cavity ring-down signals

Once the laser is locked to the cavity mode, a square-wave AM is applied:

➡ The photon lifetime is very short: very fast response/transition times are required

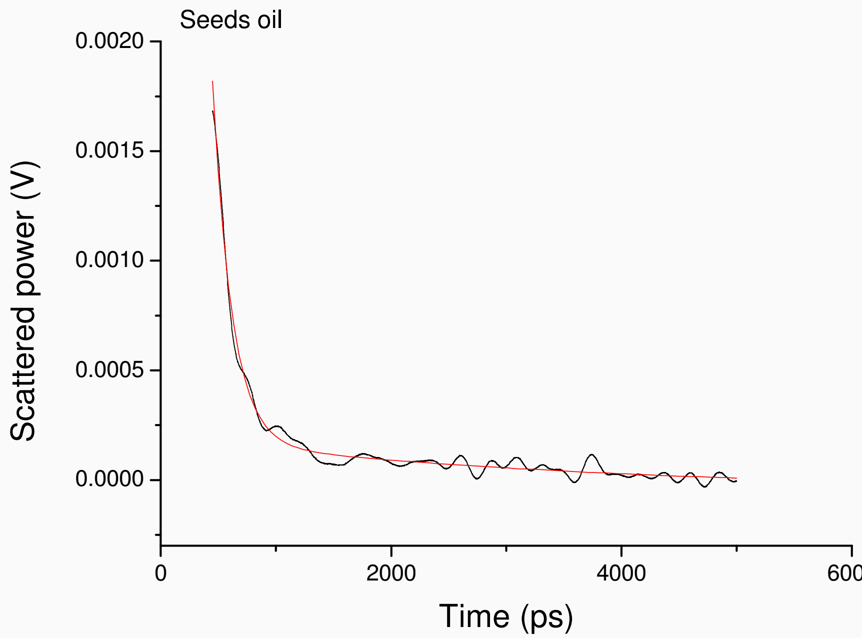


WGM: t-cavity ring-down & linewidth

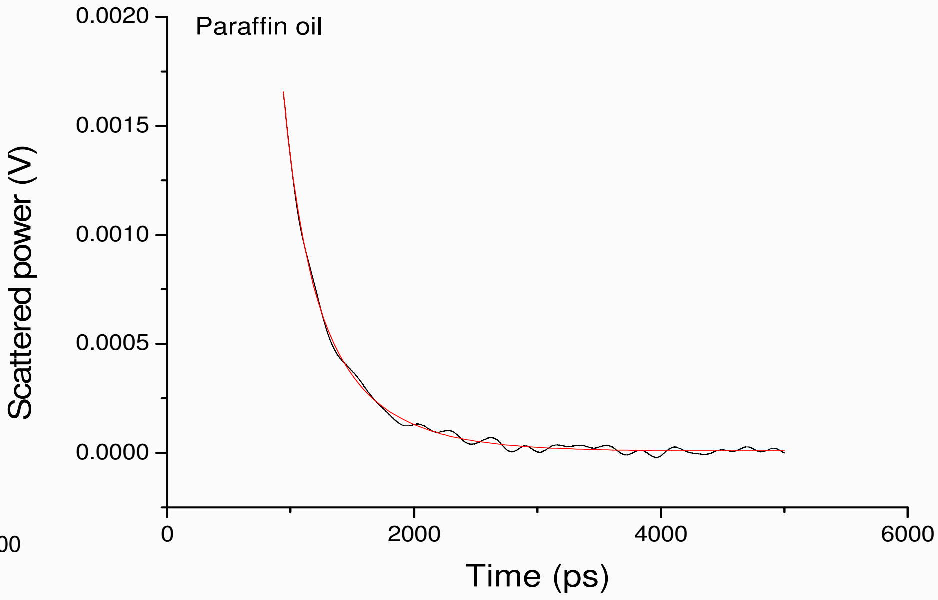


- Good consistency between the linewidth and photon lifetime
- Minimum detectable abs coeff change 7 10^{-5} mm^{-1}

CRDS: decay signals for different oils

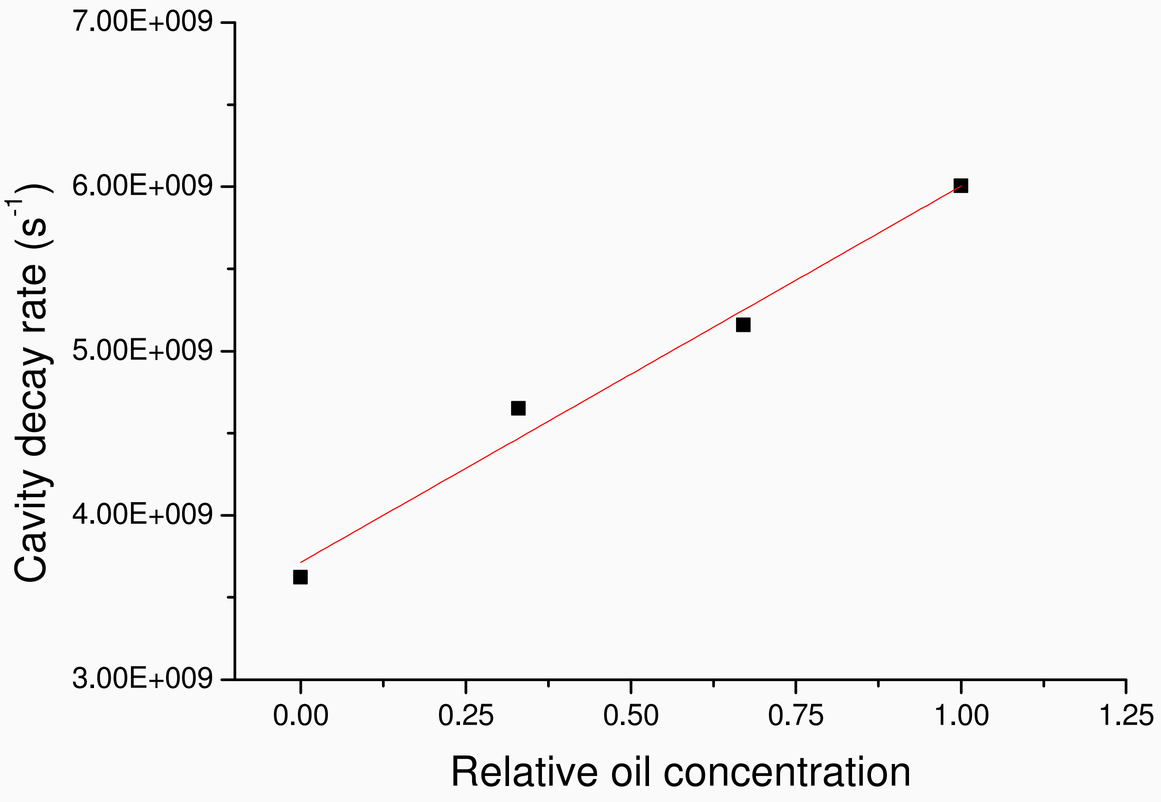


$\tau = 166.5 \pm 0.4\text{ps}$



$\tau = 367.0 \pm 0.4\text{ ps}$

Oil contamination: CRD vs. concentration

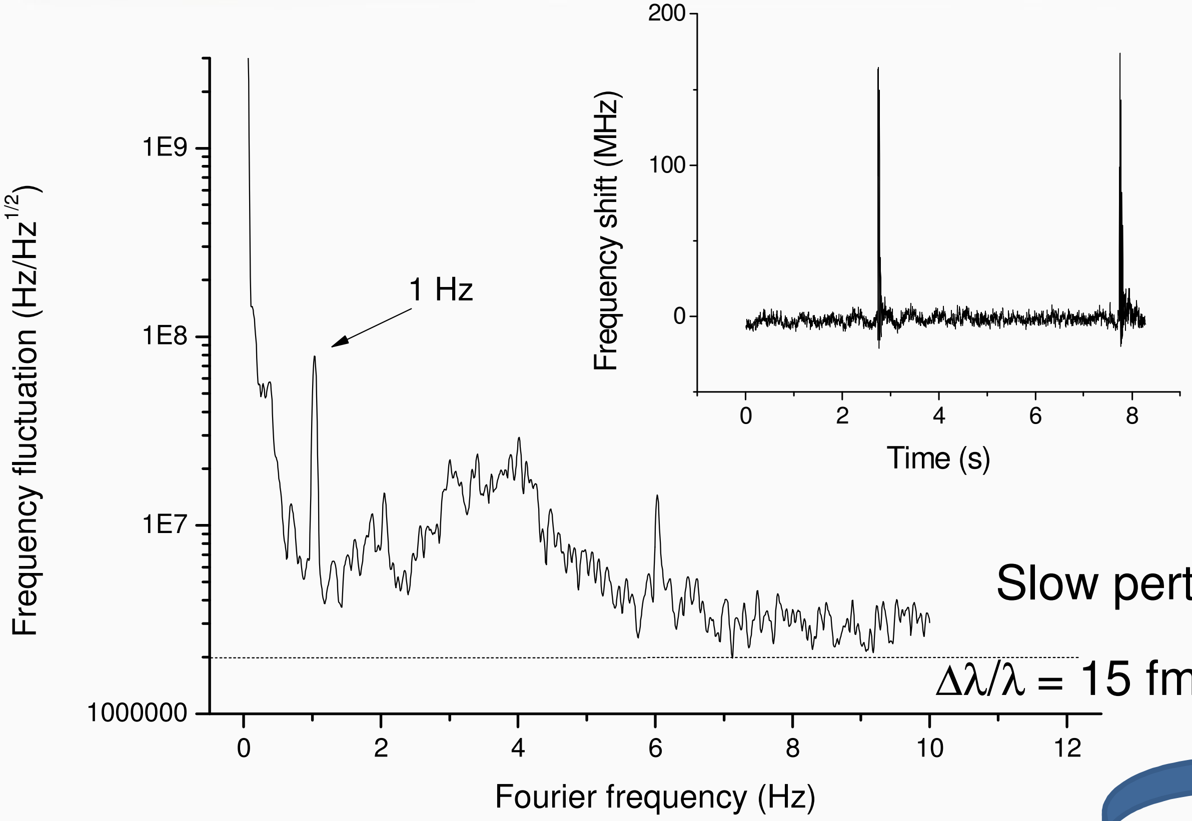


Seeds oil in olive oil
with
0, 33, 66 and 100 %
concentration

$$\frac{1}{\tau} = \frac{c}{n}(\alpha + C\varepsilon)$$

Good agreement with the measured abs cofficients of 2 components

Droplet real-time response to mechanical stimuli



Pulsed excitation

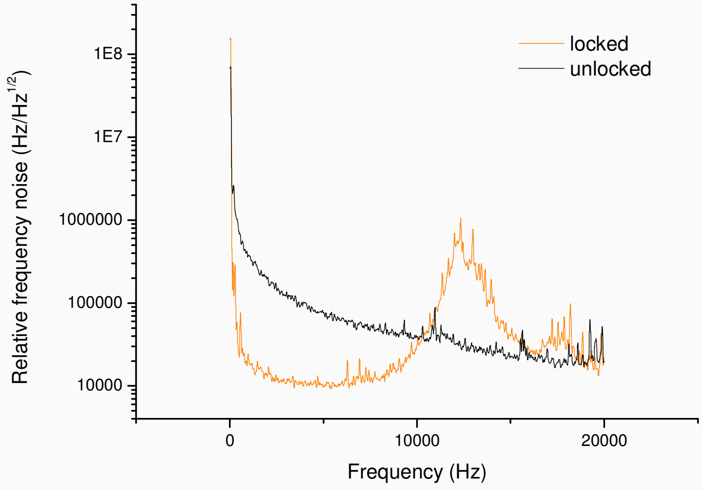
Slow perturbation

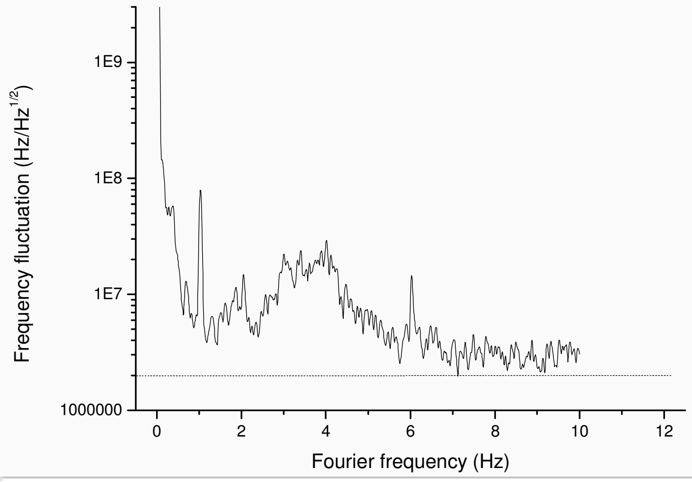
$\Delta\lambda/\lambda$ = 15 fm/Hz^{1}

Calibration of real-time WGM shift

The feedback signal exhibits a fast response
High S/N
Limited by ambient noise

Prospect for NP detection





- Laser frequency noise sets the ultimate limit on short time scales:

  30 kHz/√Hz → $\Delta\lambda \approx$ 0.2 fm/√Hz on a ms scale

  2 kHz/√Hz → $\Delta\lambda <$ 10^{-2} fm/√Hz on a μs scale

- Ambient noise affects the low-frequency range:

  $\Delta\lambda \approx$ 15 fm/√Hz on a s scale

Q-factor considerations

$$\frac{1}{Q} = \frac{1}{Q_{abs}} + \frac{1}{Q_{rad}} + \frac{1}{Q_{scat}} + \frac{1}{Q_{shape}}$$

- In droplet cavities the dominant loss can be due to liquid

Ex. In the NIR, Liquid paraffin

- Ultimate limit: scattering caused by thermally-induced

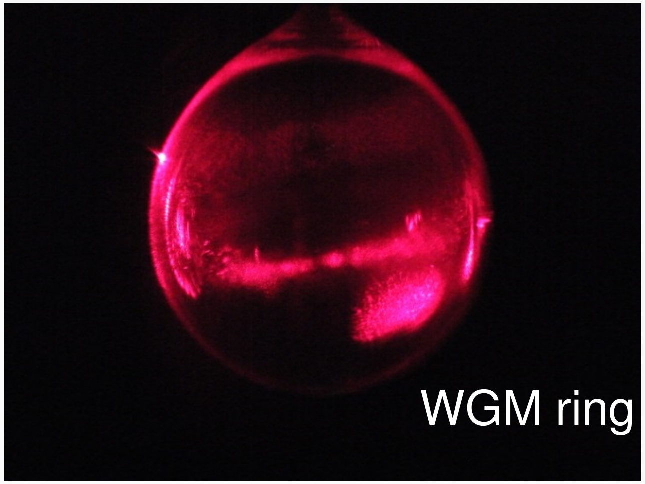
Shape fluctuations

Lai et al. PRA 1990

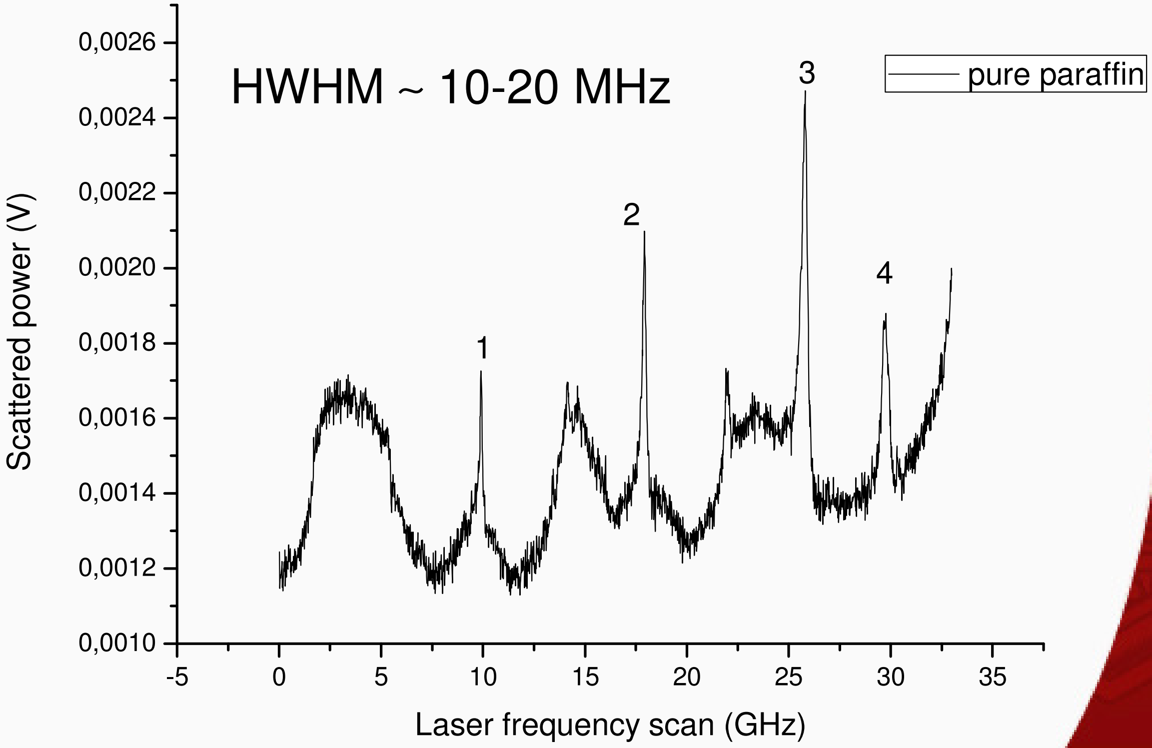
$$Q = \frac{2a}{\Delta} \approx 10^7 \qquad \Delta = \sqrt{\frac{k_B T}{4\pi\gamma}}$$

- We can move to the *visible* to increase Q

WGMs in the visible region (660 nm)



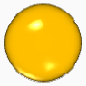
As expected, the WGMs in the visible range exhibit narrower resonances

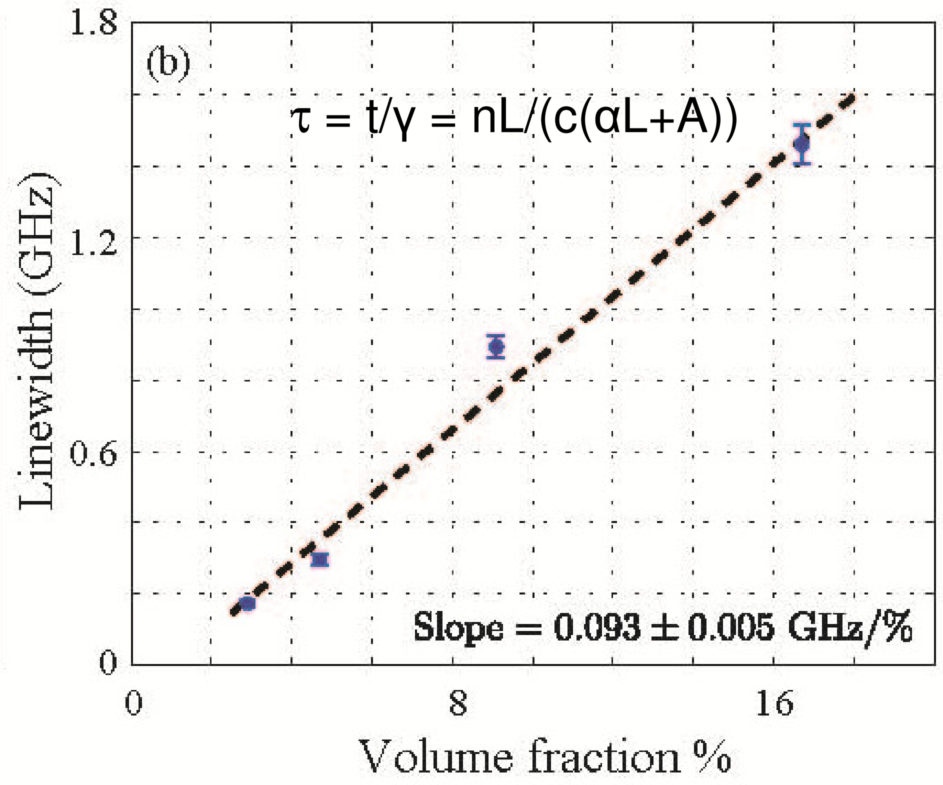


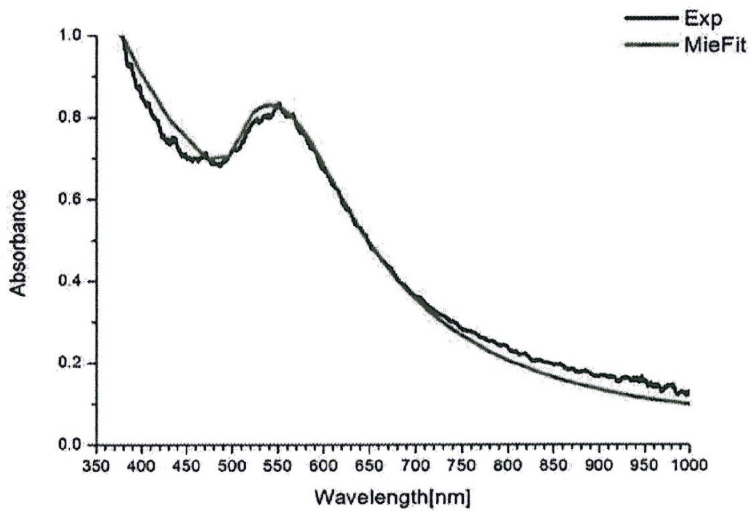
Q-factor ~ 10^7

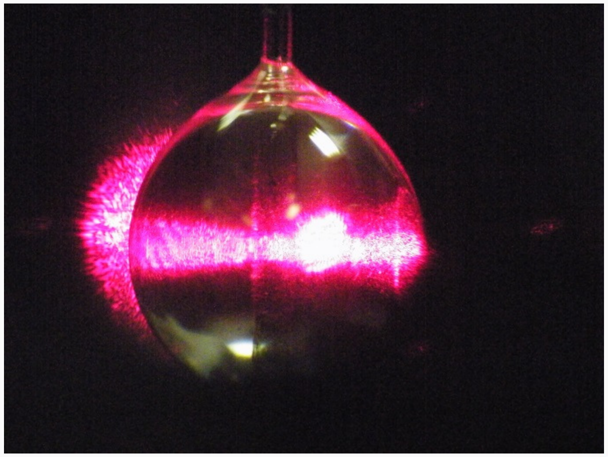
Detection of metallic NPs: preliminary results

Au nanop
10 nm dia









M.R. Foreman, S. Avino, R. Zullo, H.-P. Loock, F. Vollmer, and G. Gagliardi, Eur. Phys. J., in press

Conclusions

Summary

- Liquid droplet as micro-resonator
- Near-IR laser locked to a WGM with PDH
- CRDS with locked laser
- Oil absorption measurements
- NPs detection in the visible by WGM broadening

Outlook

- Use other liquids for droplets, different wavelengths
- Improve method of analyte delivery
- Many other experiments possible…

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Lawrence Berkeley National Laboratory, Molecular Foundry (USA)
Central Glass and Ceramic Research Inst.-CISR, Calcutta (IN)

UNIONE EUROPEA
Fondo europeo di sviluppo regionale

PON Ricerca e Competitività 2007-2013

Ministero dell'Istruzione, dell'Università e della Ricerca

Ministero dello Sviluppo Economico

View of Pozzuoli bay

THANKS
FOR
YOUR
ATTENTION…

….come and visit us
in Napoli!

INO-CNR
Istituto
Nazionale di
Ottica

National Institute of Optics (INO) of CNR: 1918

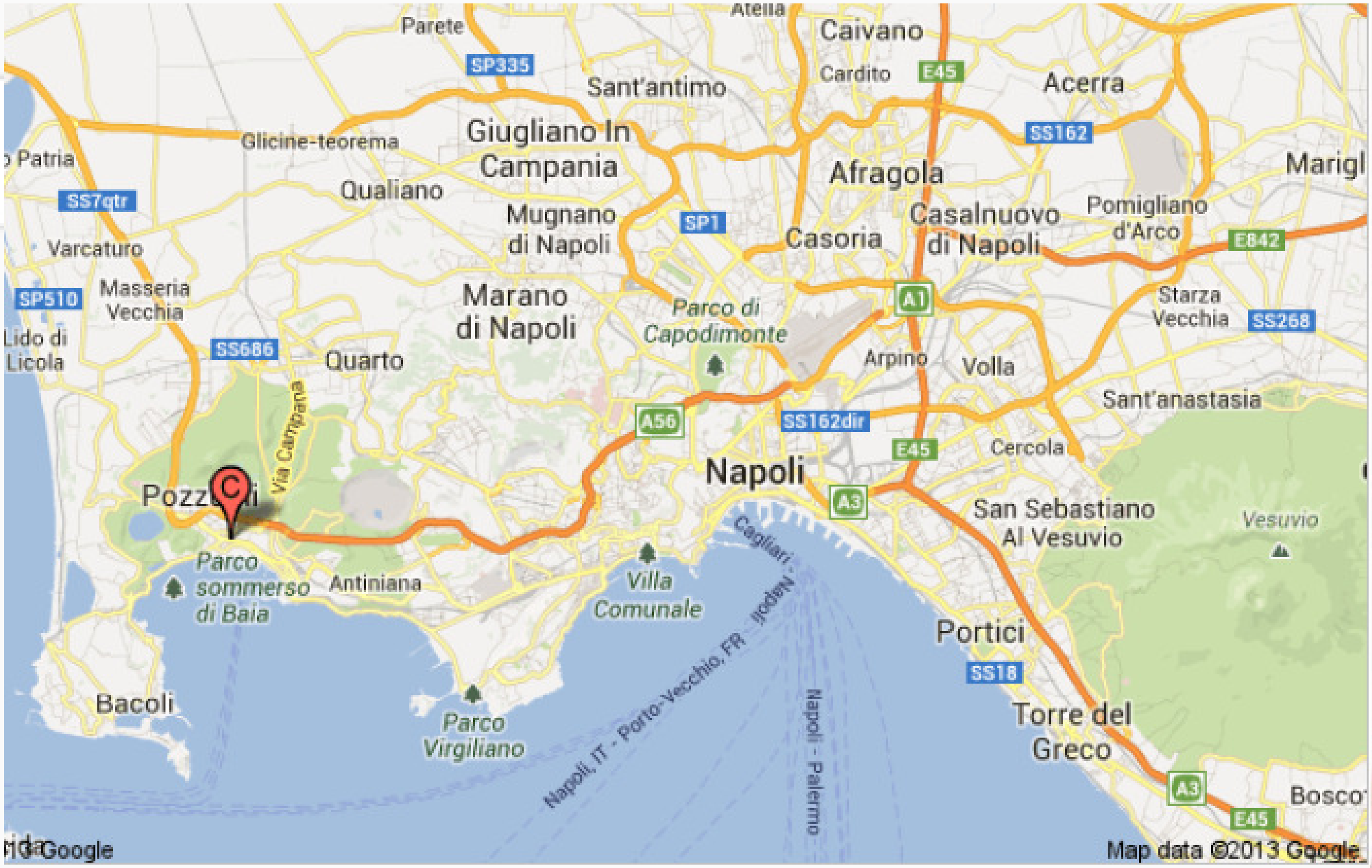
LABORATORIO DI CAPALLE
LABORATORIO C/O OPIFICIO

> …Firenze, la città custode delle tradizioni galileiane e culla e sede fortunata di molte fiorenti istituzioni ottiche, segnerà l'inizio di un periodo nuovo…
>
> Roma, 10 giugno 1936

In the photo, Firenze
Guglielmo Marconi, first CNR president

INO units: Firenze, Napoli, Lecce, Pisa, Trento, Venezia, Brescia

INO-CNR
Naples unit



Staff (admin+research) more than 30 people

Research activities:

- Optical sensing and laser spectroscopy
- Nonlinear optics
- Optical trapping and cooling of molecules
- Interferometry and digital holography
- Optical tweezers

Important milestones

R.D.Richtmyer, J.Appl.Phys., v.10, p.391 (1939) - TIR for high-Q cavity

C.G.B.Garret, W.Kaiser, W.L.Bond, Phys.Rev., v.124,p.1807 (1961) - stimulated emission

A.Ashkin, D.Dziedzic, PRL, v.38,p.1351(1977) - sharp resonances in radiation pressure on levitated droplets

R.K.Chang, J.B.Snow, S.-X.Qian, Opt.Lett.,v.10,p.37(1985) - WGM SRS in individual droplets

V.B.Braginsky, M.L.Gorodetsky, V.S Ilchenko, Phys.Lett.,v.A37, p.393 (1989) - Q>10^8 in solid spheres

L.Collot , V.Lefevre, S.Haroche et al., Europhys.Lett.,v.23,p.327(1993) - Q>10^9

V.Sandoghdar et al, Phys.Rev. A23, p.54, R1777 (1997) - microsphere laser

M.Cai, G.Hunziker, K.J.Vahala, IEEE PTL, v.11,p.686 (1999) - photonic add-drop with µsphere and fiber tapers

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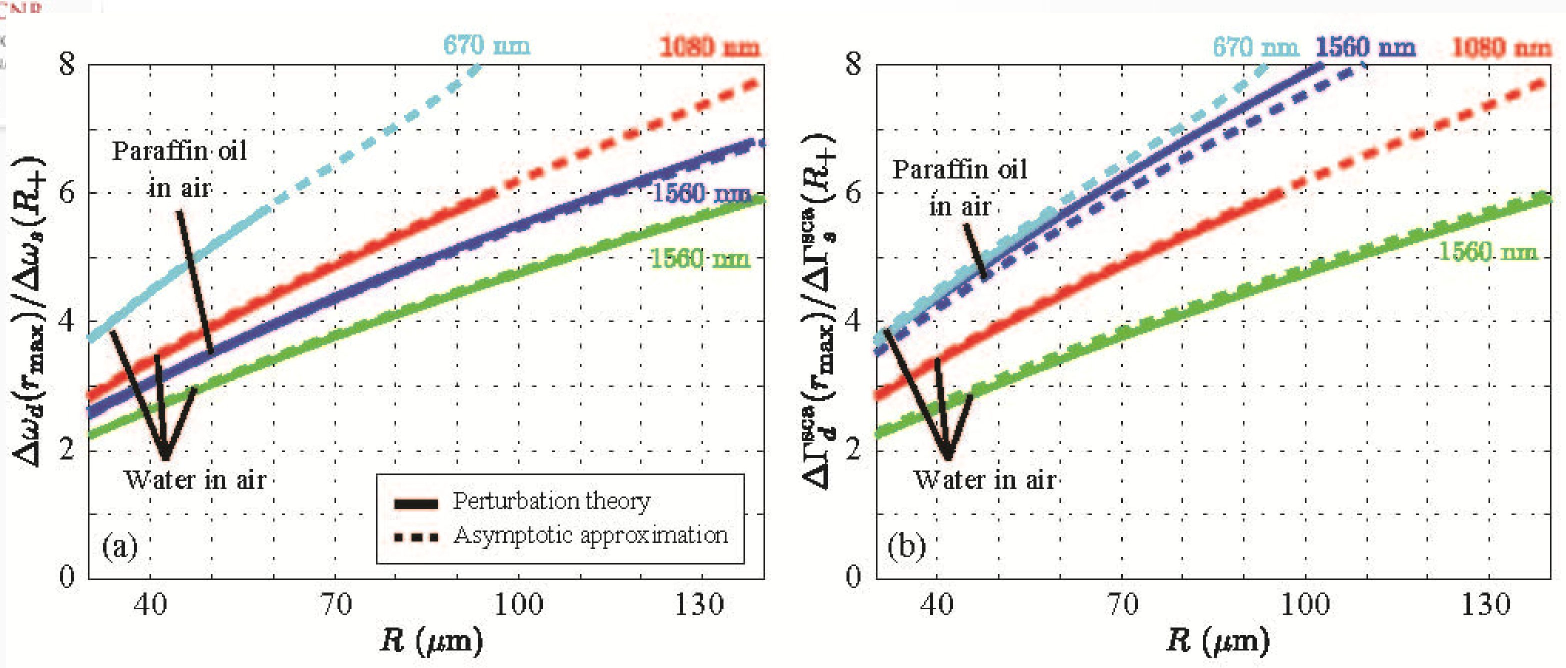
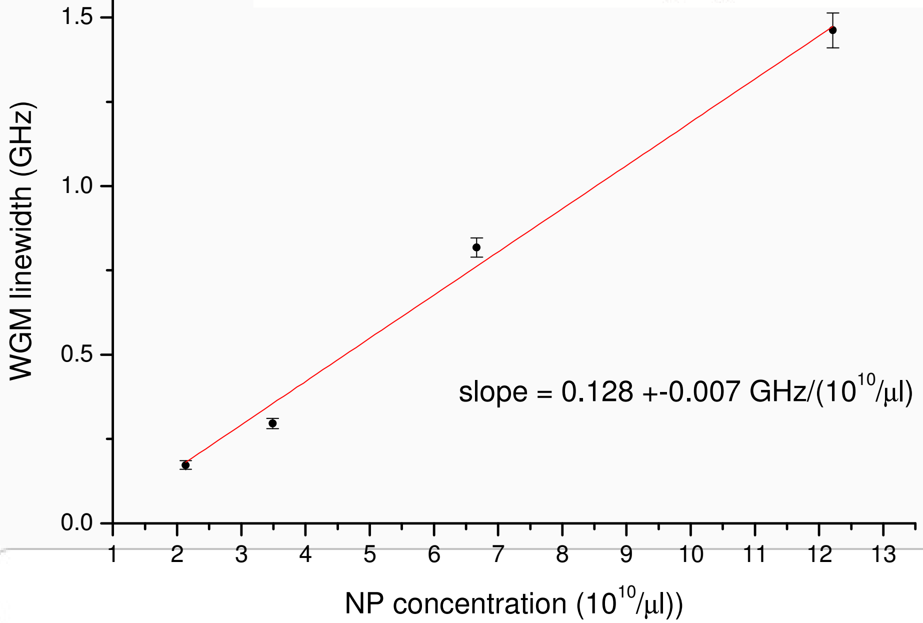

670 nm
1080 nm
1560 nm
1560 nm
Paraffin oil in air
Water in air
(a)
Perturbation theory
Asymptotic approximation
$\Delta\omega_d(r_{\max})/\Delta\omega_s(R_+)$
R (μm)
0
2
4
6
8
40
70
100
130
670 nm
1560 nm
1080 nm
1560 nm
Paraffin oil in air
Water in air
(b)
$\Delta\Gamma_d^{\mathrm{sca}}(r_{\max})/\Delta\Gamma_s^{\mathrm{sca}}(R_+)$


WGM linewidth (GHz)
0.0
0.5
1.0
1.5
1
2
3
4
5
6
7
8
9
10
11
12
13
slope = 0.128 +-0.007 GHz/(10^10/µl)
NP concentration (10^10/µl))

References

•J. Barnes, B. Carver, J. Fraser, G. Gagliardi, H.-P. Loock, Z. Tian, M. Wilson, S.Yam, O. Yastrubshak, "*Loss determination in microsphere resonators by phase-shift cavity ring-down measurements*", Opt. Express **16**, 13158-13167 (2008).

•J.A. Barnes, K. Bescherer, S. Dias, G. Gagliardi, H.P. Loock, N.R. Trefiak, H. Waechter, S. Yam, "*Measurement of Multi-Exponential Decays by Phase-Shift Cavity Ring-Down*", Appl. Phys. B **96**, 193-200 (2009).

•Hans-Peter Loock, Jack A. Barnes, Gianluca Gagliardi, Runkai Li, Richard D. Oleschuk, Helen Wächter, "*Absorption detection using optical waveguide cavities*", Can. J. Chem. (cover article) *88, 401–410 (2010).*

•G. Gagliardi, M. Salza, P. Ferraro, E. Chehura, R. Tatam, T. K. Gangopadhyay, N. Ballard, D. Paz-Soldan, J. A. Barnes, H.-P. Loock, T. T-Y. Lam, J. H. Chow, and P. De Natale, *"Optical-fiber sensing based on reflection laser spectroscopy"*, Sensors (MDPI) 10 (3), 1823-1845 (2010).

•T. T-Y. Lam, M. Salza, G. Gagliardi, J. H. Chow, P. De Natale, "*Optical fiber 3-axis accelerometer based on lasers locked to π phase-shifted Bragg gratings*", Meas. Sci. Technol., **21** 094010 (2010).

•T. T-Y. Lam, J. H. Chow, D. A. Shaddock, M. B. Gray, G. Gagliardi, and D. E. McClelland, "*High resolution absolute frequency referenced fiber optic sensor for quasi-static strain sensing*", Appl. Opt. 49 (21), 4029–4033 (2010).

•G. Gagliardi, M. Salza, S. Avino, P. Ferraro, P. De Natale, **"***Probing the Ultimate Limit of Fiber-optic Strain Sensing*", Science 330, 1081-1084, 2010

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