Single Molecule Detection of NanoMechanical Motion

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Research supported by:

Collaborators

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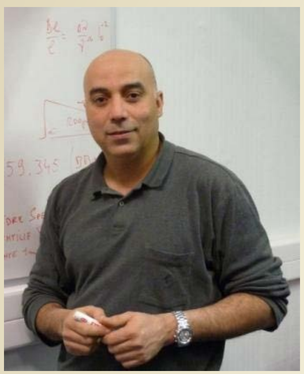
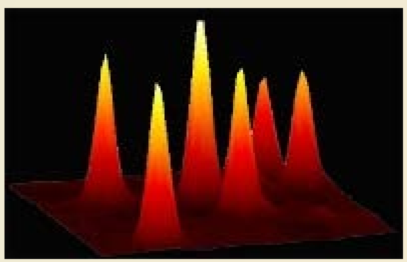


Brahim LOUNIS, Professor at Univ. Bordeaux, LPNN



V. Puller, B. Lounis, F. Pistolesi , Phys. Rev. Lett. **110**, 125501 (2013)

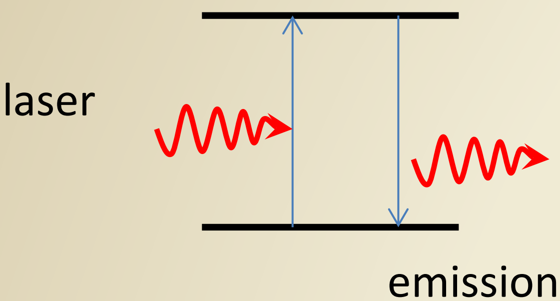
Single Molecule Spectroscopy I

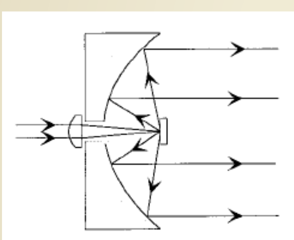
« Detection of laser induced fluorescence in small sample volume in which at most a single molecule can be excited by the incoming laser »

Ten Years of Single-Molecule Spectroscopy

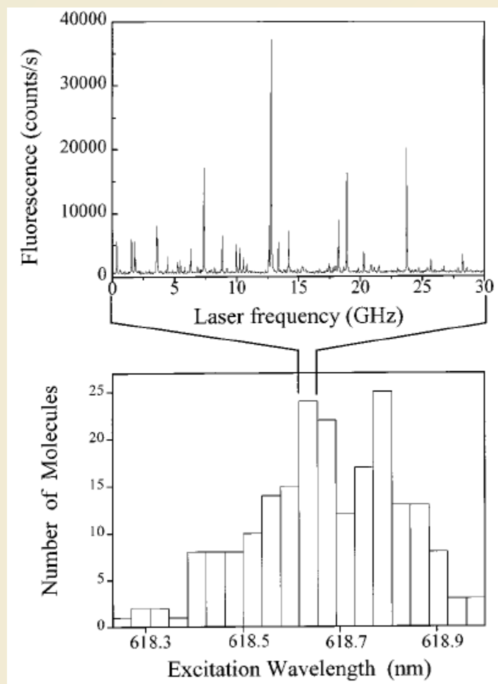
J. Phys. Chem. A, Vol. 104, No. 1, 2000

Ph. Tamarat, A. Maali, B. Lounis, and M. Orrit*

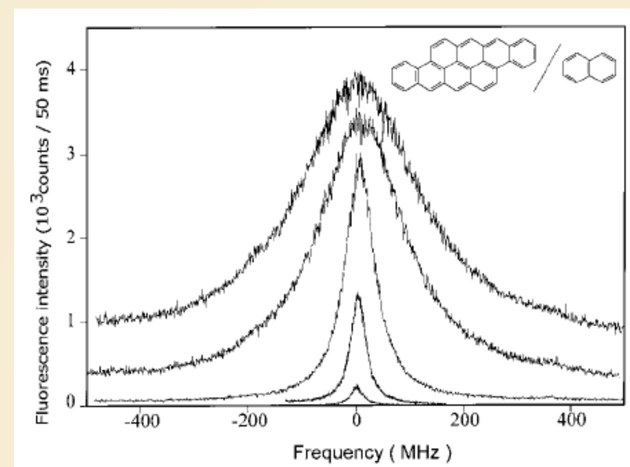




Spectral properties

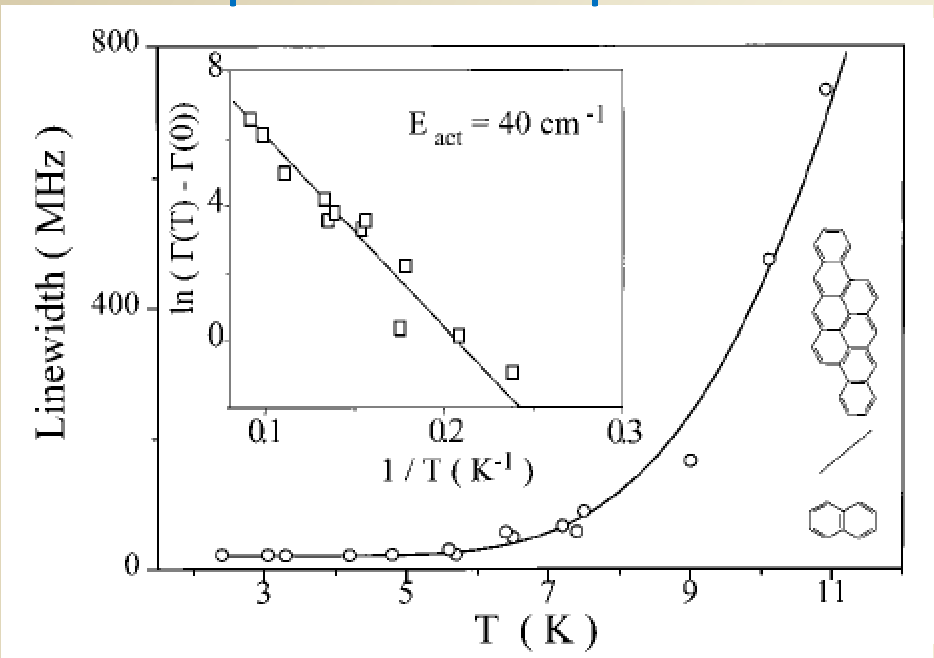


Saturation Diagram

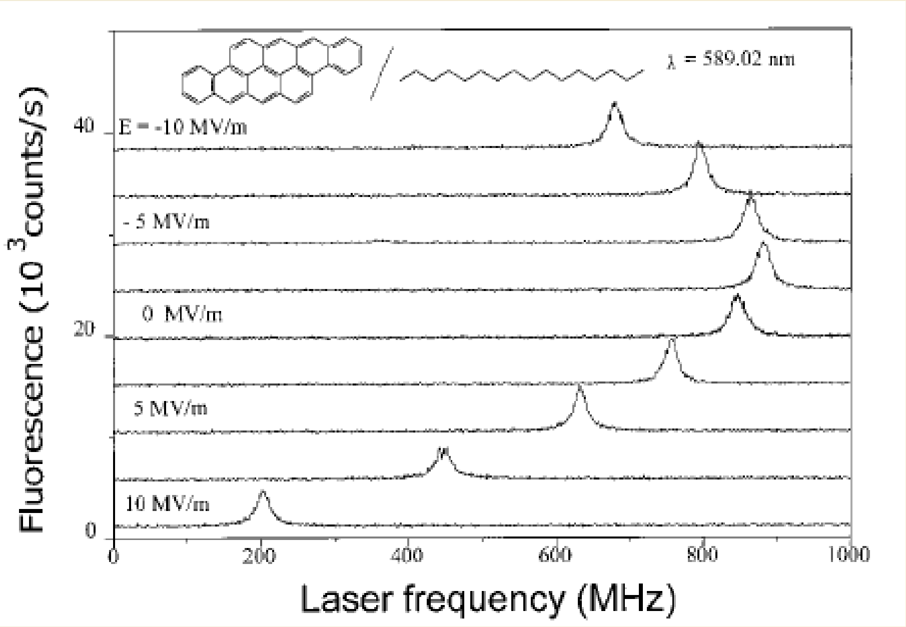


Single Molecule Spectroscopy II

Temperature Dependence



Stark effect



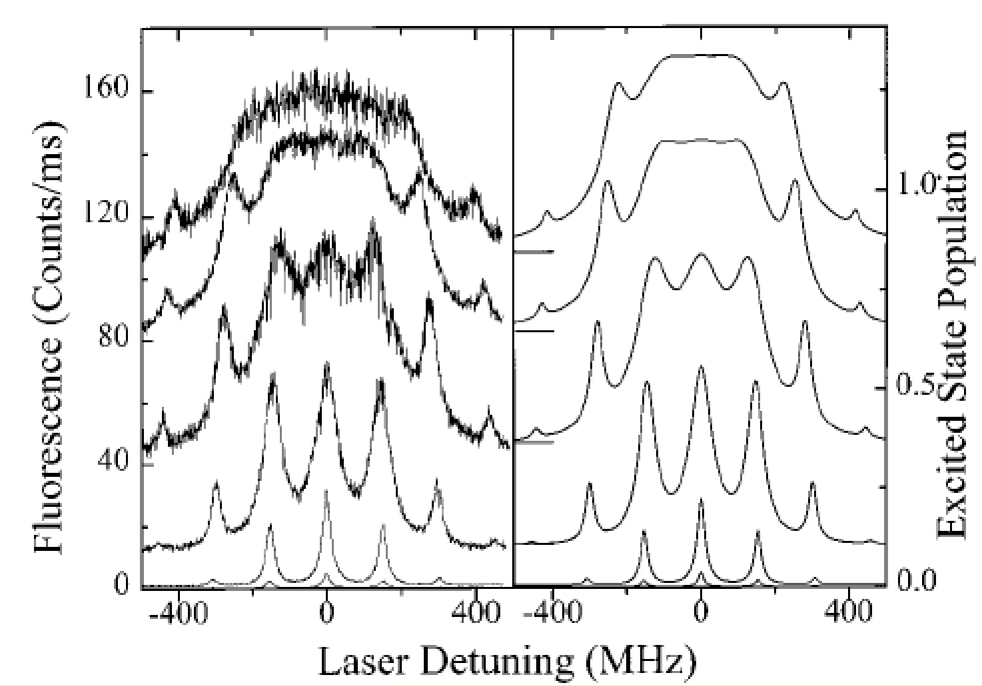
Work at al low temperature

Extremely local and sensitive probe

Typical frequency shift in a solid state matrix
1MHz for a field of 1 kV/cm

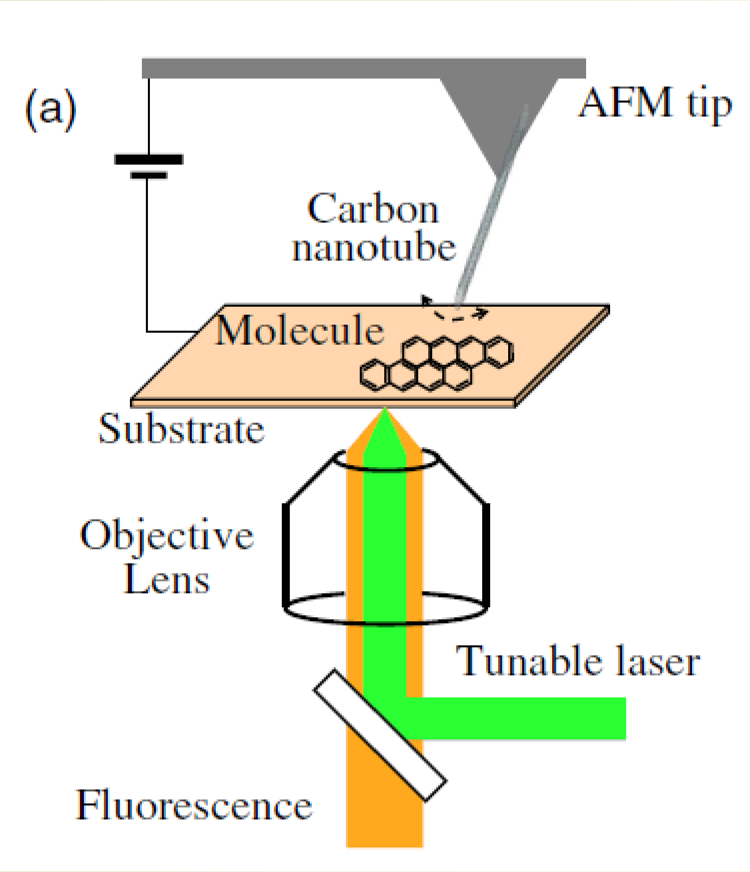
Single Molecule Spectroscopy III

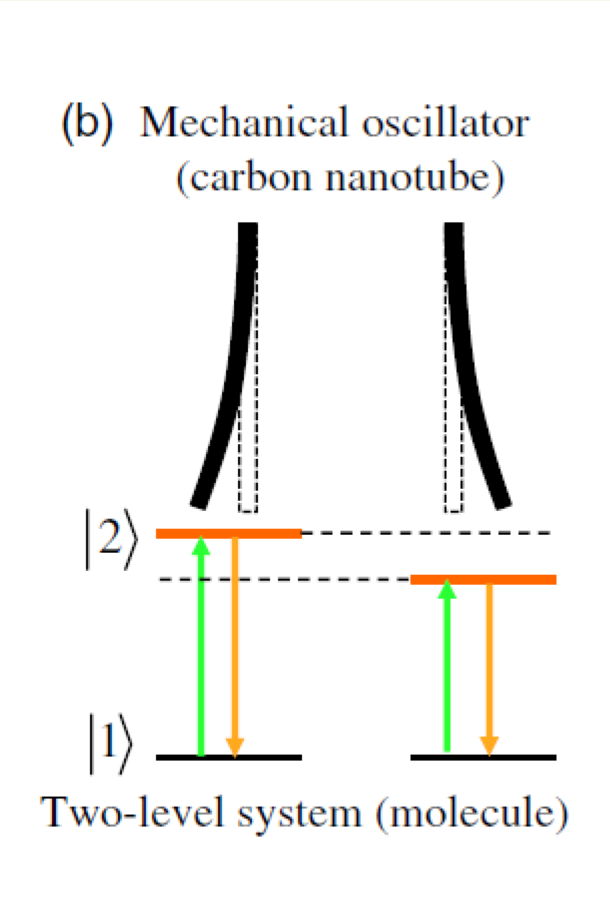
Microwave irradiated molecules



(192) Brunel, Ch.; Lounis, B.; Tamarat, Ph.; Orrit, M. *Phys. Rev. Lett.* **1998**, *81*, 2679.

Detecting Nanotubes Displacement





Using Zeeman splitting of NV centers recent experiments:

O. Arcizet et al., Nature Physics **7**, 879 (2011), S. Kolkowitz et al., Science , **335**, 1603 (2012)
[see also proposal by P. Rabl et al Phys. Rev. B **79**, 041302(R) (2009)]

Coupling constant

Electric field limited by the threshold of field Emission

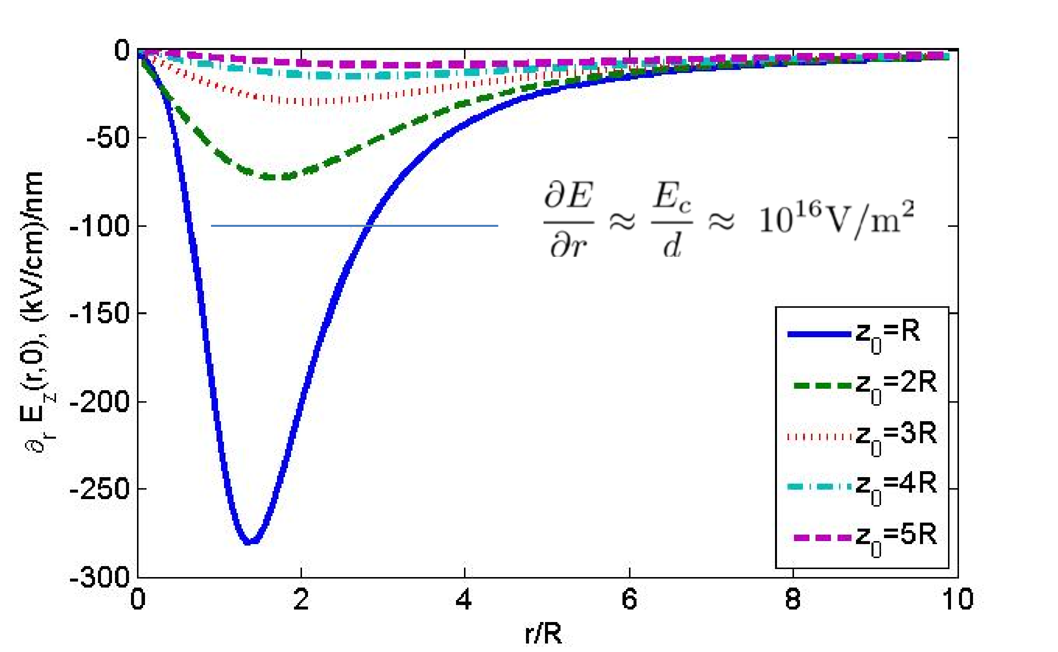
R. C. Smith,[a] D. C. Cox, and S. R. P. Silva
Appl. Phys. Lett. **87**, 103112 (2005)

$$E_c \approx 10^7 \ \mathrm{V/m}$$

Typical Electric field gradient

$$\frac{\partial E}{\partial r} \approx \frac{E_c}{d} \approx \ 10^{16}\mathrm{V/m^2}$$

Detailed numerical calculations confirm these rough estimates (V=10mV, d=1nm)



Coupling constant: shift of the fluorescent resonance per m of CNT displacement

$$\Delta\nu = \alpha\Delta x \qquad \alpha_{\mathrm{Max}} = 10 \ \mathrm{GHz/nm}$$

Strong Coupling

This is 3-4 orders of magnitude larger than the magnetic coupling in NV centers

Hamiltonian

$$H = H_M + H_L + H_{\mathrm{osc}} + H_{\mathrm{osc-M}} + H_{\mathrm{phot}} + H_\xi$$

$|2\rangle$

$|1\rangle$

Molecule: $H_M = \hbar\omega_0\hat{n}$ $\qquad \hat{\pi} = |1\rangle\langle 2|, \quad \hat{n} = |2\rangle\langle 2|$

Laser: $H_L = \hbar\Omega\hat{\pi}e^{i\omega_L t} + H.c.$

Mechanical Oscillator: $H_{\mathrm{osc}} = p_x^2/2m + m\omega_M^2 x^2/2$

Coupling Hamiltonial: $H_{\mathrm{osc-M}} = \hbar\alpha x\hat{n}$

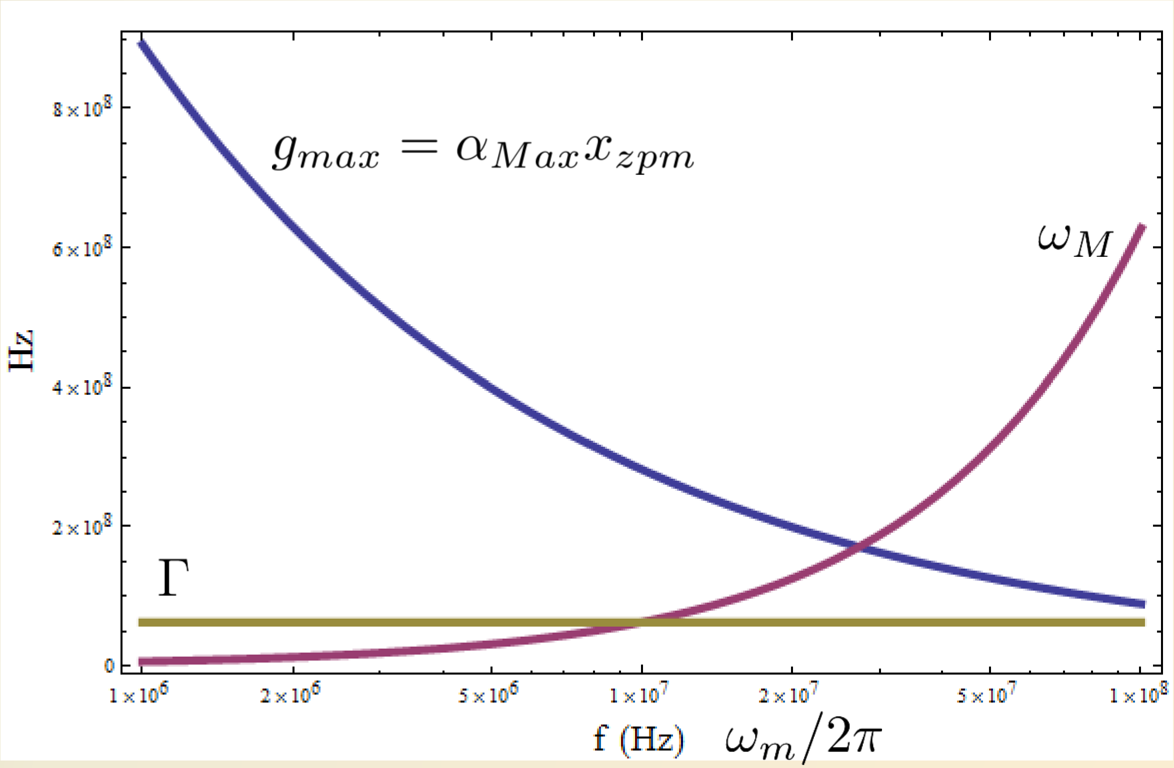
$$\omega(x) = \omega_o + \Delta\vec{\mu}\cdot\vec{E}(x)/\hbar \approx \omega_o + \Delta\vec{\mu}\cdot\vec{E}(0)/\hbar + \alpha x \qquad \Delta\vec{\mu} = \langle 2|\vec{\mu}|2\rangle - \langle 1|\vec{\mu}|1\rangle$$

Coupling of the Molecule and of the Oscillator to the environnement leading to a width Γ and a damping ϒ $\qquad H_\xi + H_{\mathrm{phot}}$

Comparison of frequency scales

For a SW carbon nanotube of 1 μm

$$m = 10^{-21}\ \mathrm{Kg} \qquad x_{zpm} = \sqrt{\frac{\hbar}{2m\omega_M}}$$



Ultra strong coupling since $g \gg \Gamma, \omega_M$

Bloch Equations description

$$\frac{d\sigma_{12}(t)}{dt} = -i\left[\delta + \alpha x(t)\right]\sigma_{12}(t) - \frac{\Gamma}{2}\sigma_{12}(t) + i\Omega\left(2\sigma_{22}(t) - 1\right),$$

$$\frac{d\sigma_{22}(t)}{dt} = -2\Omega\Im\left[\sigma_{12}(t)\right] + \Gamma\left[\sigma_{22}^{(\mathrm{eq})} - \sigma_{22}(t)\right]$$

Ω Laser-Molecule coupling constant $\sigma_{ij} = \langle i|\hat{\rho}|j\rangle$ $\delta = \omega_0 - \omega_L$

The fluorescence signal is given by σ_{22}

$$\frac{d^2x(t)}{dt^2} + \gamma\frac{dx(t)}{dt} + \omega_M^2 x(t) = \frac{\xi(t)}{m} - \frac{\hbar\alpha}{m}\sigma_{22}(t)$$

$\langle\xi(t)\rangle = 0$ $\langle\xi(t)\xi(t')\rangle = 2m\gamma k_B T\delta_D(t-t')$

Classical Thermal fluctuations

$k_B T \gg \hbar\omega_M$

Weak coupling to the laser $\Omega/\Gamma \ll 1$

To order Ω^2

$$\sigma_{22}(t) = \Omega^2 \left| \int_{-\infty}^{t} dt_1 e^{-i\left(\delta - \frac{i}{2}\Gamma\right)(t-t_1) - i\alpha \int_{t_1}^{t} d\tau x(\tau)} \right|^2$$

After averaging over the fluctuations of x for $\gamma \ll \omega_M, \Gamma$

$$\langle \sigma_{22} \rangle = \Omega^2 \frac{\Gamma + \Gamma_\phi}{\Gamma} \sum_{n=-\infty}^{+\infty} \frac{e^{-\frac{\Gamma_\phi}{2\gamma}} I_n\left(\zeta^2\right)}{\left(\delta + n\omega_M\right)^2 + \frac{1}{4}\left(\Gamma + \Gamma_\phi\right)^2}$$

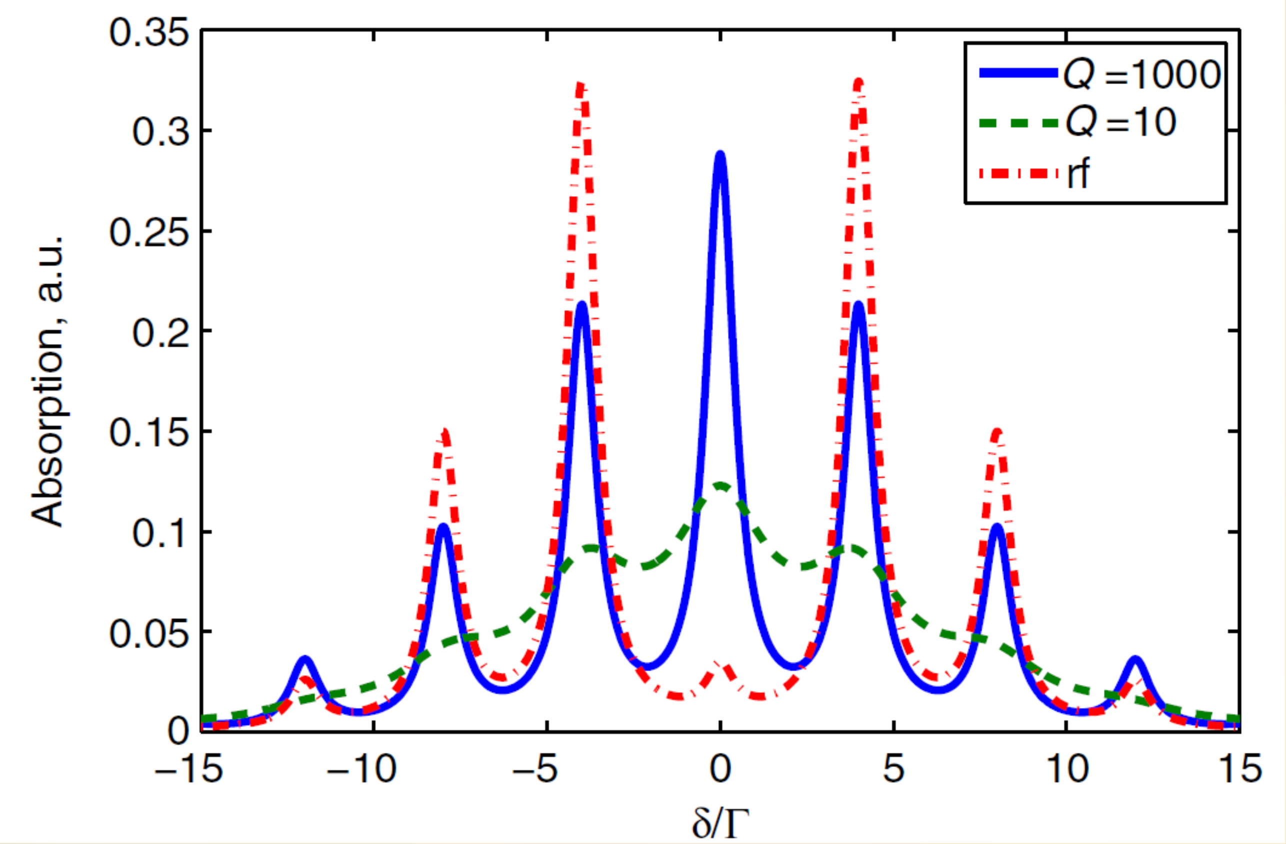
$\zeta^2 = \frac{\alpha^2 \langle x^2 \rangle}{\omega_M^2}$ $\quad I_n(z)$ modified Bessel functions

$\Gamma_\phi = 2\gamma\zeta^2 \quad Q = \omega_M/\gamma \gg 1$

For thermal motion: $\langle x^2 \rangle = k_B T / m\omega_M^2$

Comparison with coherent drive: $\overline{\sigma_{22}} = \Omega^2 \sum_{n=-\infty}^{+\infty} \frac{\left[J_n\left(2\zeta\right)\right]^2}{\left(\delta + n\omega_M\right)^2 + \frac{1}{4}\Gamma^2}$

Side bands



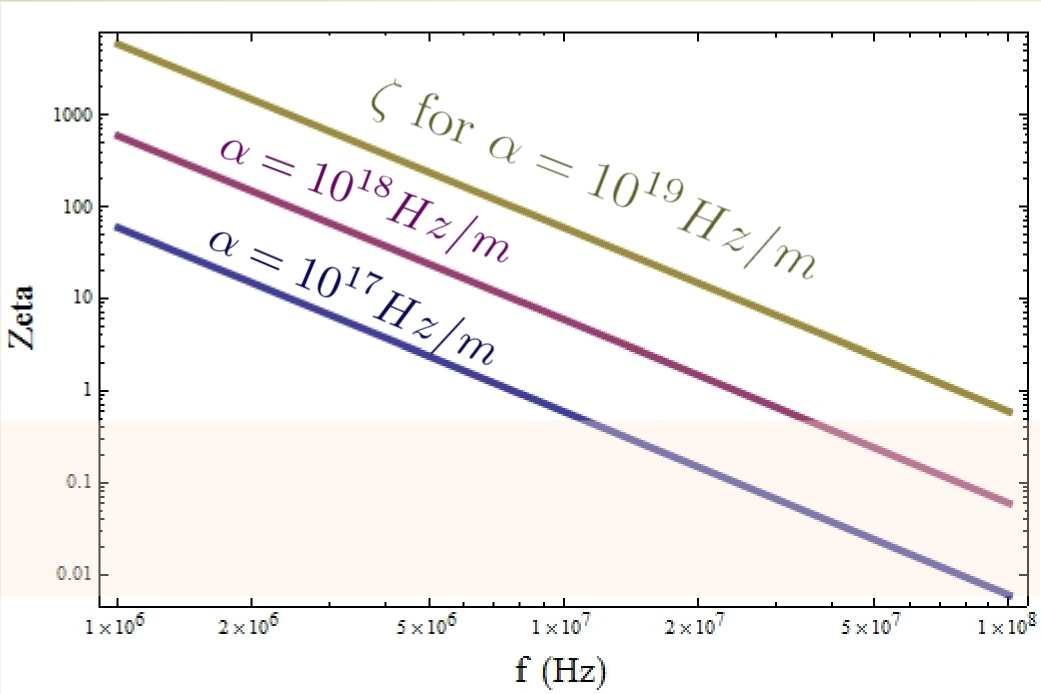
$$\zeta = 1.5,\ \omega_M = 4\Gamma$$

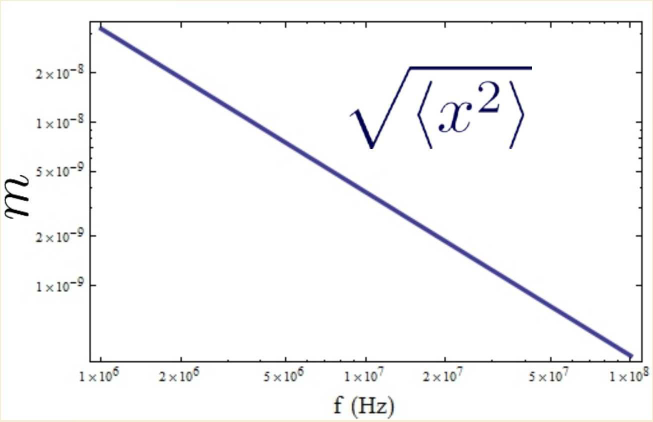
The ratio of the peak intensity gives direct access to the fluctuation intensity

$$I_{n+1}\left(\frac{\Gamma_\phi}{2\gamma}\right)/I_n\left(\frac{\Gamma_\phi}{2\gamma}\right) \approx \alpha^2\langle x^2\rangle/2(n+1)\omega_M^2$$

Practicals

$$T = 4\ \mathrm{K} \qquad \zeta^2 = \frac{\alpha^2 \langle x^2 \rangle}{\omega_M^2} \qquad \langle x^2 \rangle = \frac{k_B T}{m \omega_M^2} \qquad m = 10^{-21}\ \mathrm{Kg}$$





Γ $\qquad$ $\omega_M / 2\pi$

Real Time position measurement

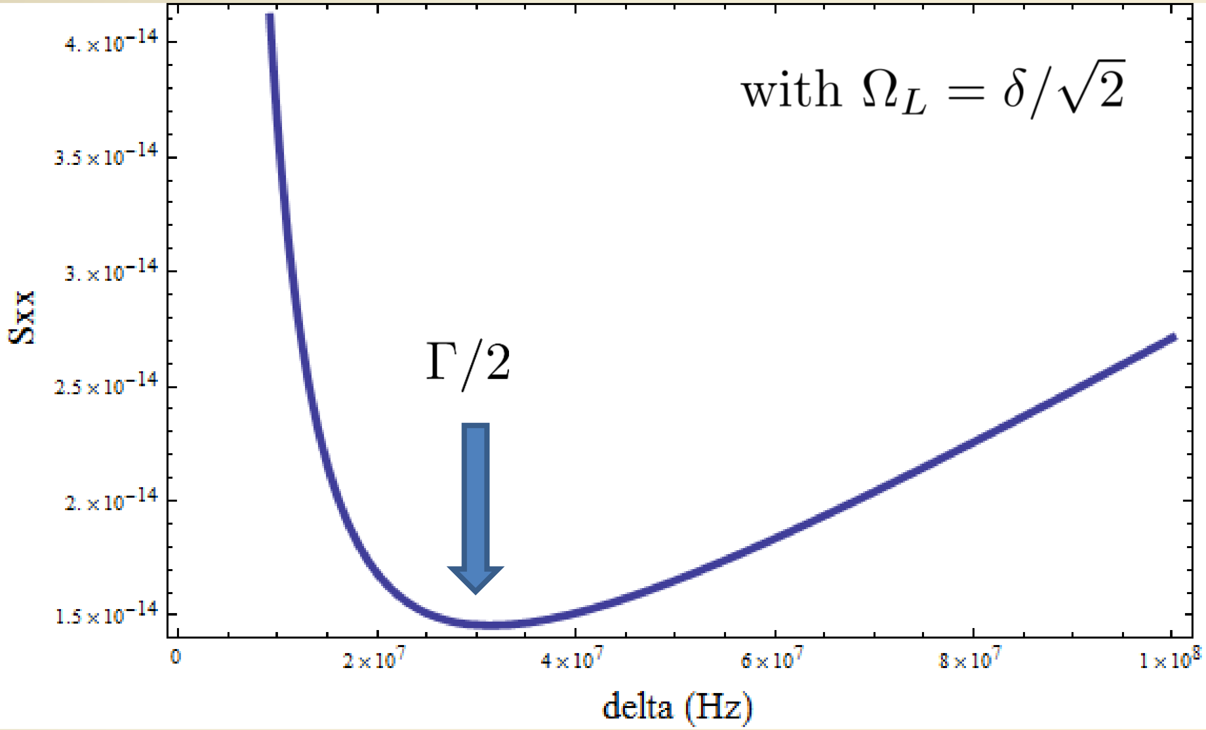
Adiabatic limit $$\sigma_{22}(t) = \frac{\Omega_L^2}{[\delta + \alpha x(t)]^2 + \frac{\Gamma^2}{4} + 2\Omega_L^2} + \ldots$$

Response to displacement: $$G = \left.\frac{\partial \langle I \rangle}{\partial x(t)}\right|_{x(t)=0} = -\frac{2\eta\Gamma\Omega_L^2\delta\alpha}{(\delta^2 + \frac{\Gamma^2}{4} + 2\Omega_L^2)^2}$$

$$\overline{S}_{xx} \equiv \sqrt{\frac{S_{II}(0)}{G^2}} = \frac{1}{2\Omega\alpha\sqrt{\eta\Gamma}} \frac{\left(\delta^2 + \frac{\Gamma^2}{4} + 2\Omega_L^2\right)^{\frac{3}{2}}}{|\delta|}$$

$$\text{minimum for } |\delta| = \Omega_L\sqrt{2} = \Gamma/2$$

Typical Sensitivity

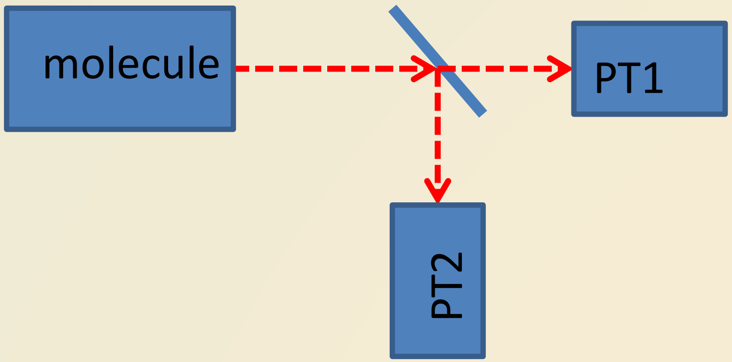


$\Gamma/2\pi = 10^7$ Hz, $\eta = 0.01$, $\alpha = 10^{19}$ Hz/m we obtain $\overline{S}_{xx} \sim 10^{-14}\mathrm{m}/\sqrt{\mathrm{Hz}}$

Best optical detection $10^{-20}\mathrm{m}/\sqrt{\mathrm{Hz}}$ best with NEMS $10^{-16}\mathrm{m}/\sqrt{\mathrm{Hz}}$

Second order correlation function

$$g^{(2)}(t,t') = \frac{\langle \hat{\pi}^\dagger(t')\hat{\pi}^\dagger(t)\hat{\pi}(t)\hat{\pi}(t')\rangle_x}{\langle \hat{\pi}^\dagger(t')\hat{\pi}(t')\rangle_x \langle \hat{\pi}^\dagger(t)\hat{\pi}(t)\rangle_x} \qquad \hat{\pi} = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}$$



Evaluation by Quantum Regression Theorem

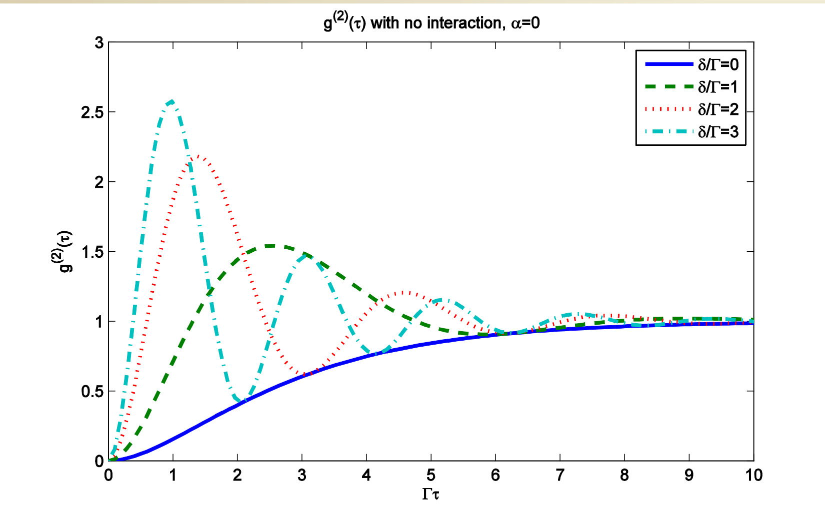
At lowest order in Ω_L

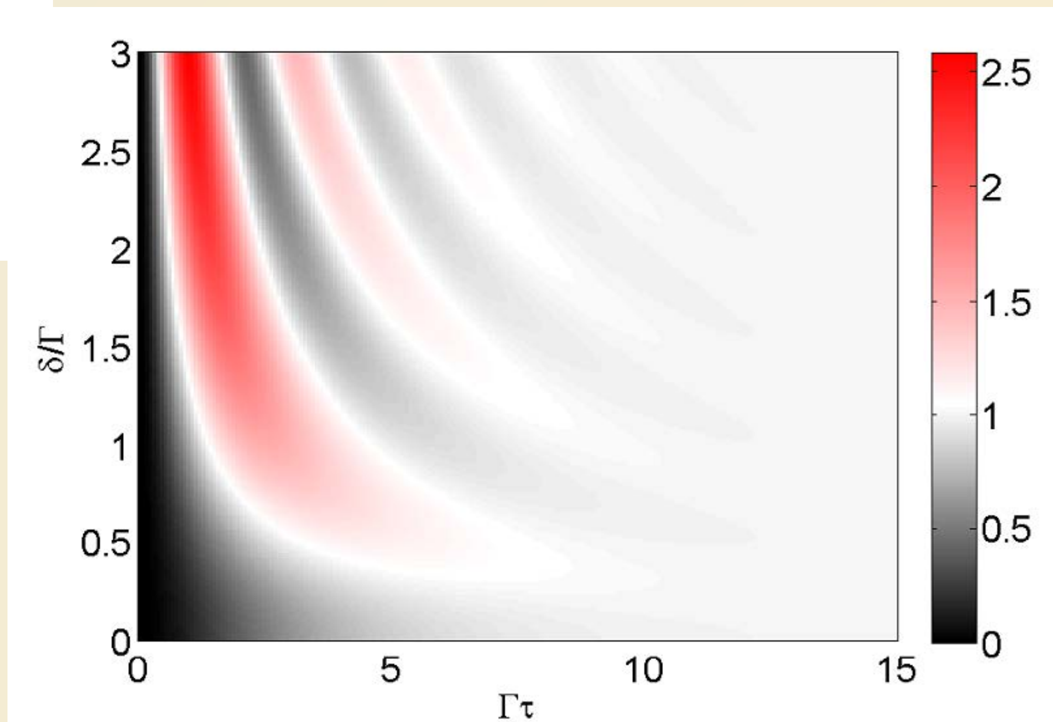
$$g^{(2)}(t,t') = \frac{\left\langle \sigma_{22}(t')\Omega_L^2 \left| \int_{t'}^{t} dt_1 e^{-i\left(\delta - i\frac{\Gamma}{2}\right)(t-t_1) - i\int_{t_1}^{t} d\tau \alpha x(\tau)} \right|^2 \right\rangle_x}{\langle \sigma_{22}(t)\rangle_x \langle \sigma_{22}(t')\rangle_x}$$

Non-interacting case

In the absence of interaction with the oscillator:

$$g^{(2)}(\tau) = 1 + e^{-\Gamma\tau} - 2\cos(\delta\tau)\, e^{-\frac{\Gamma\tau}{2}}$$





Long time behavior

$$g^{(2)}(t,t') = \frac{\left\langle \sigma_{22}(t')\Omega_L^2 \left| \int_{t'}^{t} dt_1 e^{-i\left(\delta - i\frac{\Gamma}{2}\right)(t-t_1) - i\int_{t_1}^{t} d\tau \alpha x(\tau)} \right|^2 \right\rangle_x}{\langle \sigma_{22}(t)\rangle_x \langle \sigma_{22}(t')\rangle_x} \quad \Rightarrow \quad g^{(2)}(t,t') = \frac{\langle \sigma_{22}(t')\sigma_{22}(t)\rangle_x}{\langle \sigma_{22}(t)\rangle_x \langle \sigma_{22}(t')\rangle_x}$$

$$|t-t'| \gg \Gamma^{-1}$$

At lowest order in $\zeta \ll 1$:

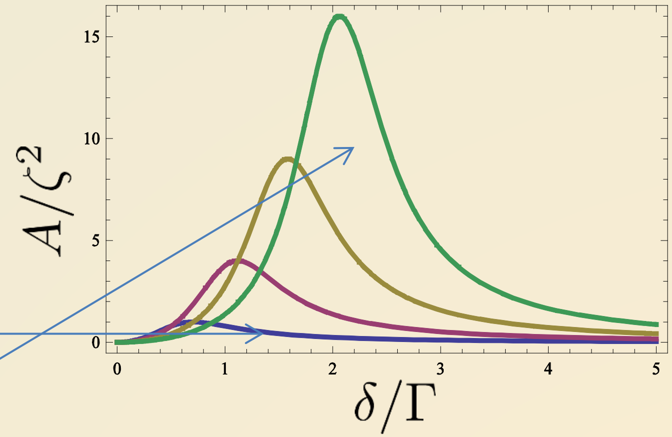
$$g^{(2)}(t,t') = 1 + \alpha^2 \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \frac{S_{xx}(\omega)}{\omega} \frac{2\delta}{(\delta-\omega)^2 + \frac{\Gamma^2}{2}}$$

with $S_{xx}(t) = \langle x(t)x(0)\rangle_x$.

Fast and Slow oscillator limit for large t-t'

Simple form for the underdamped harmonic oscillator: $g^2(\tau) = 1 + A\cos(\omega_M \tau)$

$$A = \zeta^2 \omega_M \delta \left[\frac{1}{(\delta-\omega_M)^2 + \frac{\Gamma^2}{4}} - \frac{1}{(\delta+\omega_M)^2 + \frac{\Gamma^2}{4}} \right]$$



For $\omega_M \ll \Gamma$: $A = \frac{\zeta^2 \delta^2 \omega_M^2}{(\delta^2 + \frac{\Gamma^2}{4})^2}$.

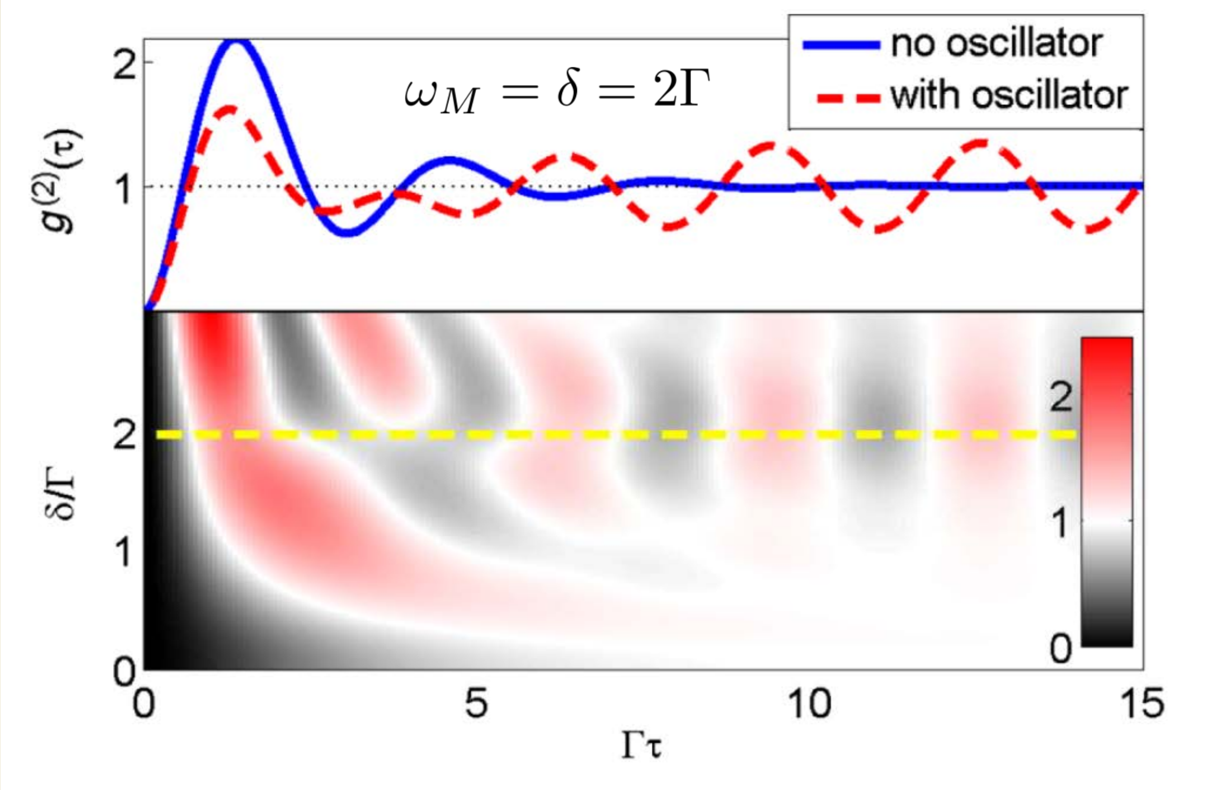
The maximum value is for $\delta = \Gamma/2$: $A_M = \zeta^2 \frac{\omega_M^2}{\Gamma^2} \ll \zeta^2$

In this limit the result is even more general: $g^{(2)} = 1 + (\frac{G}{I})^2 \langle x(\tau) x(0) \rangle$

For $\omega_M \gg \Gamma$ and $\delta \approx \omega_M$: $A = 4\zeta^2 \frac{\omega_M^2}{\Gamma^2} \gg 4\zeta^2$ easier to observe

General result for small ς

We have analytical expression for $\zeta \ll 1$.
From that we can plot the form of $g^{(2)}(\tau)$:

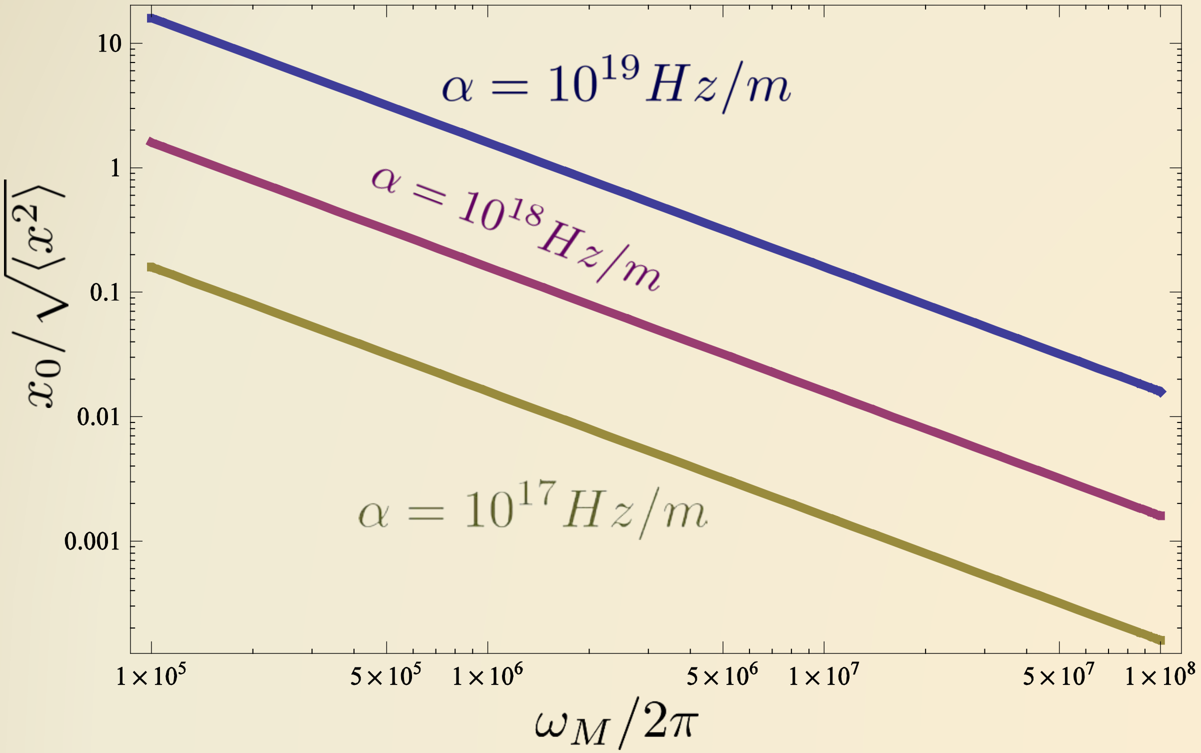


$\omega_M = 2\Gamma$, $\zeta = 0.15$ and $Q = 10^3$.

Back action

Displacement of the equilibrium position: $x_0 = \frac{\hbar\alpha}{(m\omega_M^2)}$

This can be quite large.
For instance for $T = 1\text{K}$ $m = 10^{-21}$ Kg, $\alpha = 10^{19} Hz/m$ $\frac{x_0}{\sqrt{\langle x^2 \rangle}} = 10^7 \text{ Hz}/\omega_M$



In order to estimate back-action we use linear back-action theory (*quantum noise*)

A.A. Clerk, Phys. Rev. B 70, 245306 (2004); A.A. Clerk and S. Bennett, New J. Phys. 7, 238 (2005);

$$T_{osc} = \frac{\gamma T + \gamma_1 T_{TLS}}{\gamma + \gamma_1}$$

$$\gamma_1 = \frac{\hbar \alpha^2}{m\omega_M}\left[S_{nn}(\omega_M) - S_{nn}(-\omega_M)\right]; \qquad T_{TLS} = \frac{\hbar\omega_M}{2k_B}\frac{S_{nn}(\omega_M) + S_{nn}(-\omega_M)}{S_{nn}(\omega_M) - S_{nn}(-\omega_M)};$$

$$S_{nn}(\omega) = \int dt e^{i\omega t}\langle \hat{n}(t)\hat{n}(0)\rangle = \frac{\Omega^2}{\delta^2 + \frac{\Gamma^2}{4}}\frac{\Gamma}{(\delta-\omega)^2 + \frac{\Gamma^2}{4}} \qquad \text{For } \Omega/\Gamma \ll 1$$

validity of linear approach $\gamma_1 \ll \Gamma$

Cooling

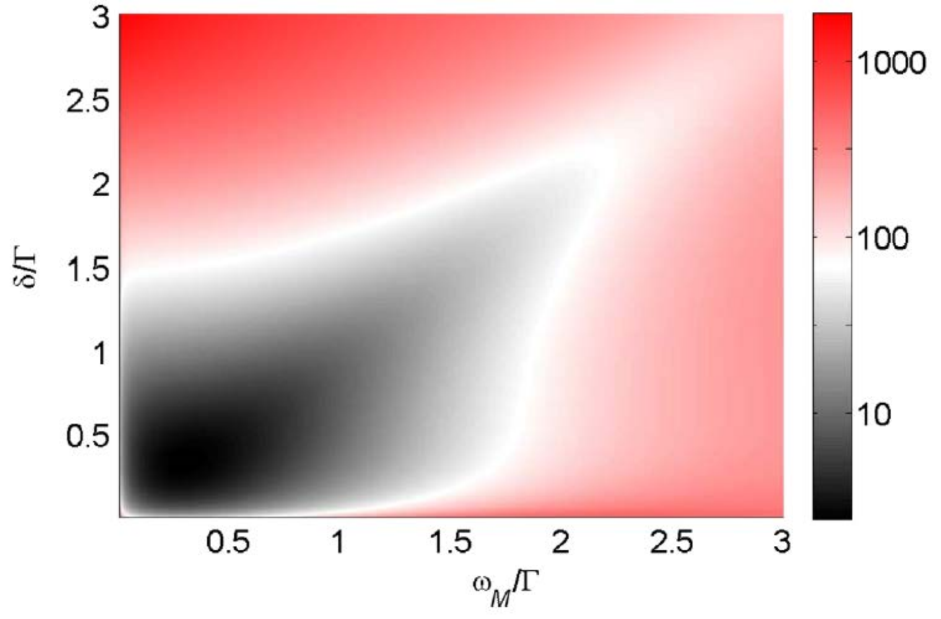
I. Wilson-Rae et al, PRL 2003, I. Martin et al, PRB 2004, F. Marquardt et al., PRL 2007; I. Wilson-Rae et al, PRL 2007, P. Rabl, PRB 2010, …

$$T_{TLS} = \frac{\hbar\omega_M}{k_B \log\left[\frac{(\delta+\omega_M)^2+\Gamma^2/4}{(\delta-\omega_M)^2+\Gamma^2/4}\right]}$$

$$\delta = \sqrt{\omega_M^2 + \Gamma^2/4}$$

for $\omega_M \ll \Gamma$ we have $T_{TLS} \approx \hbar\Gamma/4k_B$

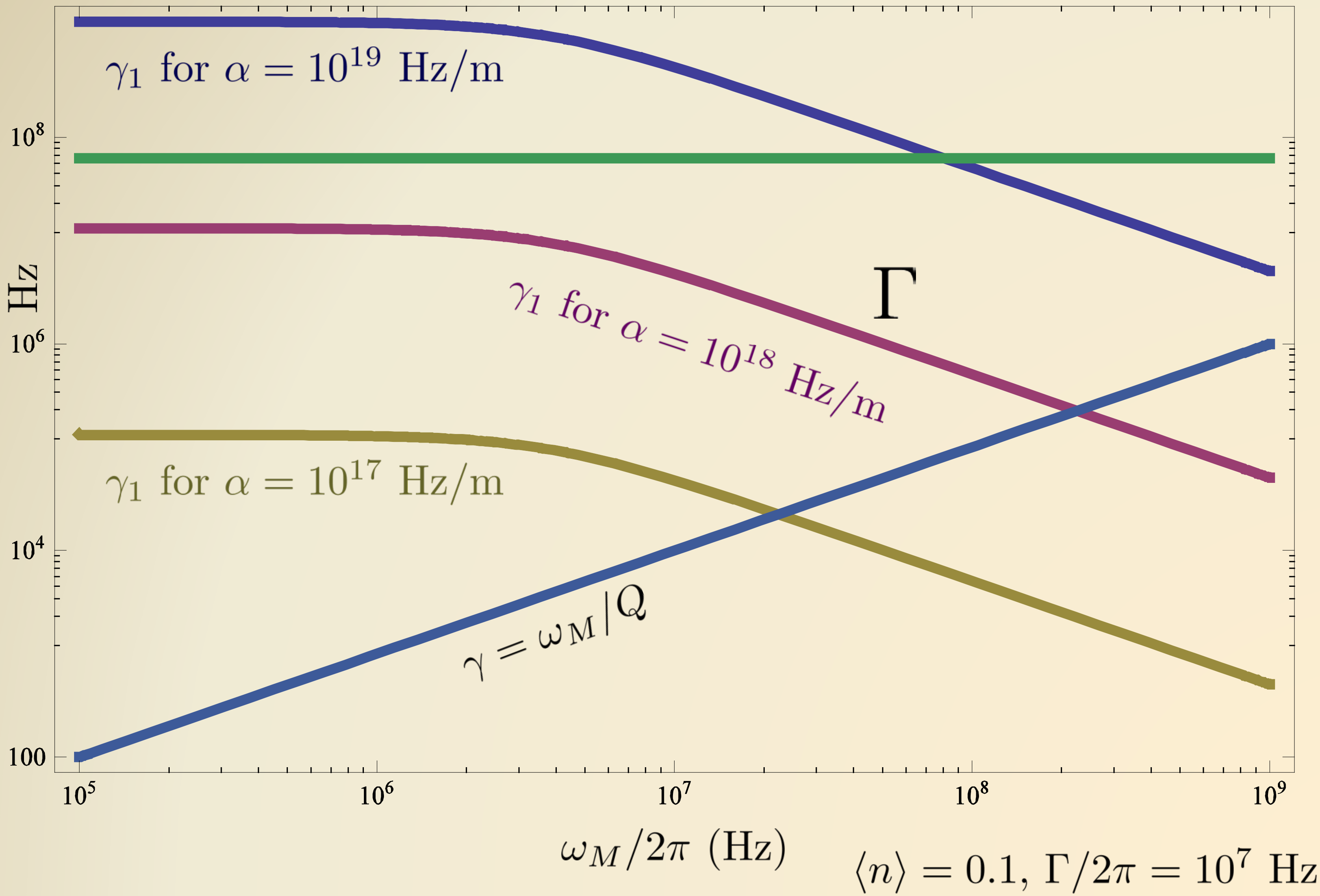
for $\omega_M \gg \Gamma$ we have $T_{TLS} = 2\hbar\omega_M/[k_B \log(4\omega_M/\Gamma)]$



$k_B T/\hbar\omega_M$

$$T = 1\ \mathrm{K},\ m = 10^{-21}\ \mathrm{Kg},\ \alpha = 10^{19}\ \mathrm{Hz/m},\ Q = 10^3$$

Different regimes



Conclusions

- Single Molecule Spectroscopy can be used to detect and manipulate suspended carbon nanotubes
- Strong coupling can be achieved with strong back action

Perspectives

- Dynamics in the strong coupling regime (ongoing work)
- Experiments