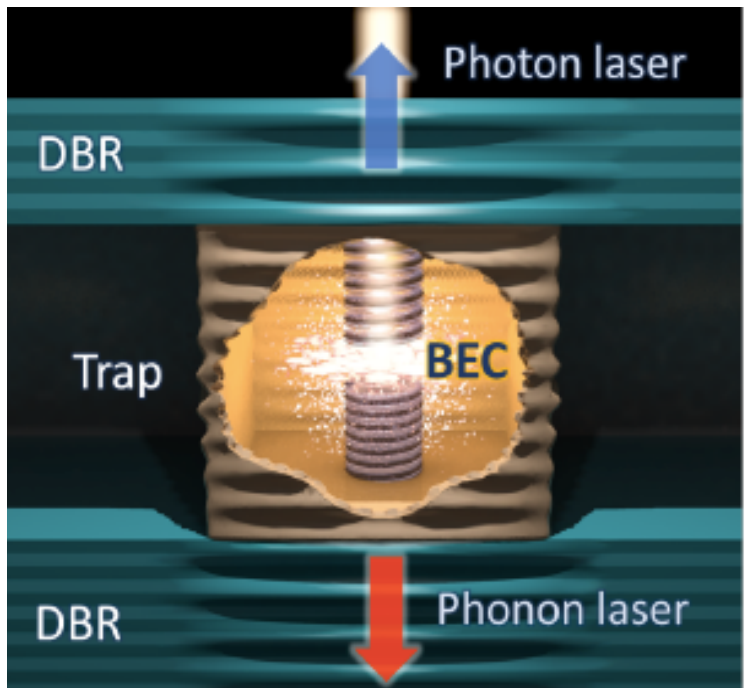
Raman-Brillouin: Una introducción a las interacciones optomecánicas

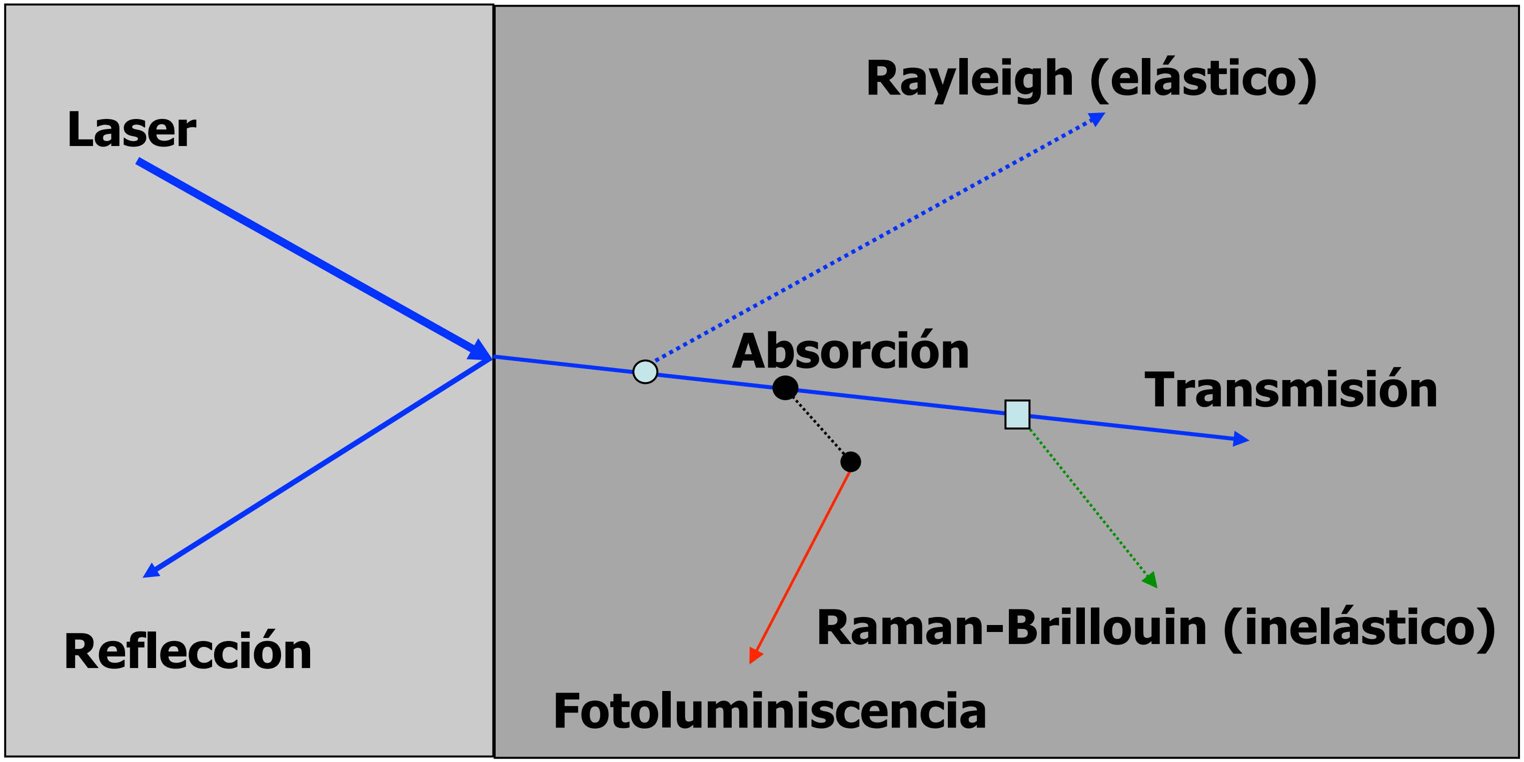
Alex Fainstein

Instituto Balseiro, Bariloche, Argentina

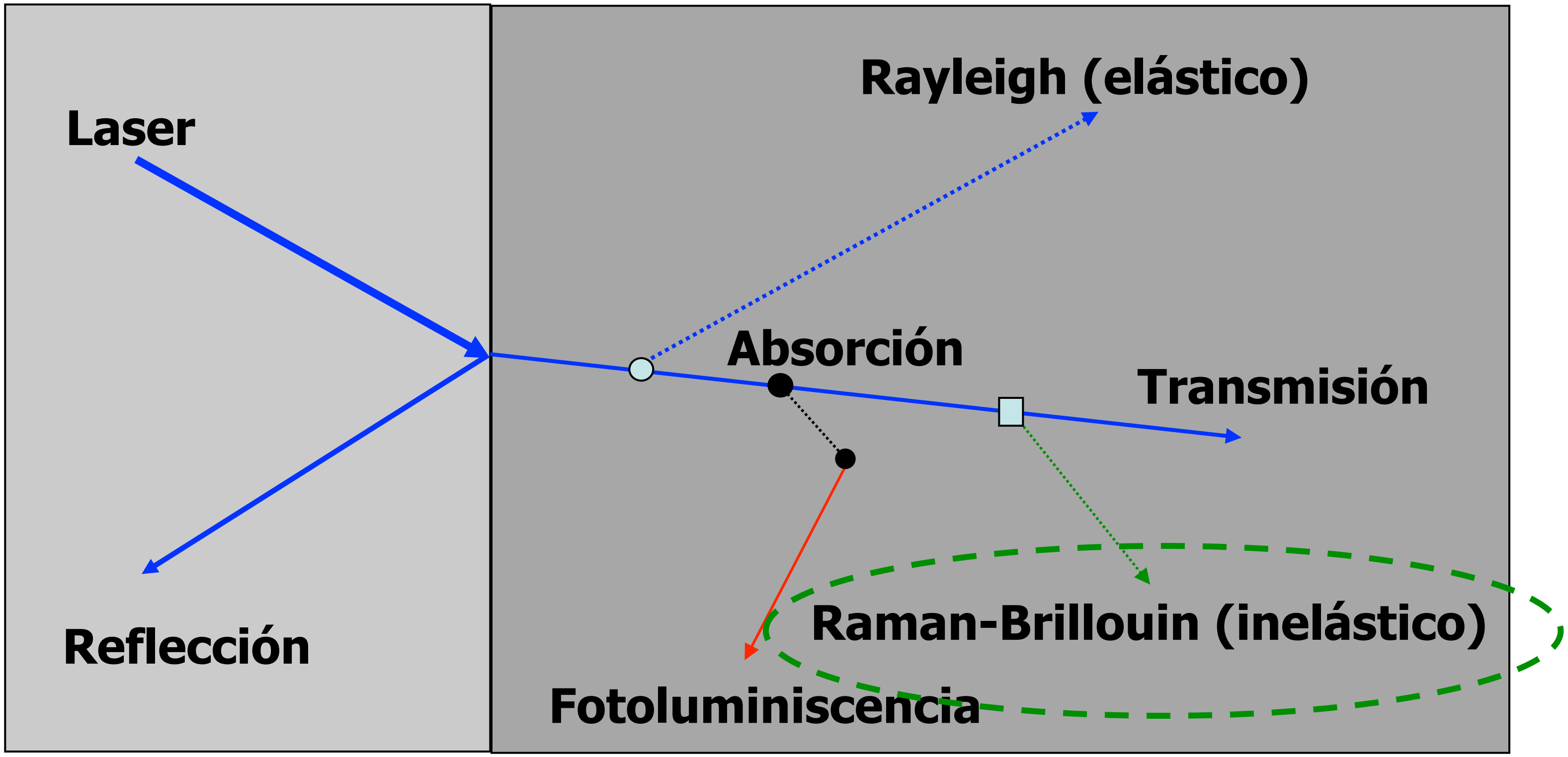
alex.fainstein@gmail.com



Óptica lineal: cómo interactúa la luz con la materia



Óptica lineal: cómo interactúa la luz con la materia



Qué dicen los libros de óptica no-lineal sobre la dispersión Raman?

La dispersión inelástica de luz en la óptica lineal y no-lineal

The *Raman scattering* processes can be described by the equation

$$P_i = \chi_{ij}^{(1)} E_j + \chi_{ijk}^{(2)} E_j E_k + \chi_{ijkl}^{(3)} E_j E_k E_l.$$

La dispersión inelástica de luz en la óptica lineal y no-lineal

The *Raman scattering* processes can be described by the equation

$$P_i = \chi_{ij}^{(1)} E_j + \chi_{ijk}^{(2)} E_j E_k + \chi_{ijkl}^{(3)} E_j E_k E_l.$$

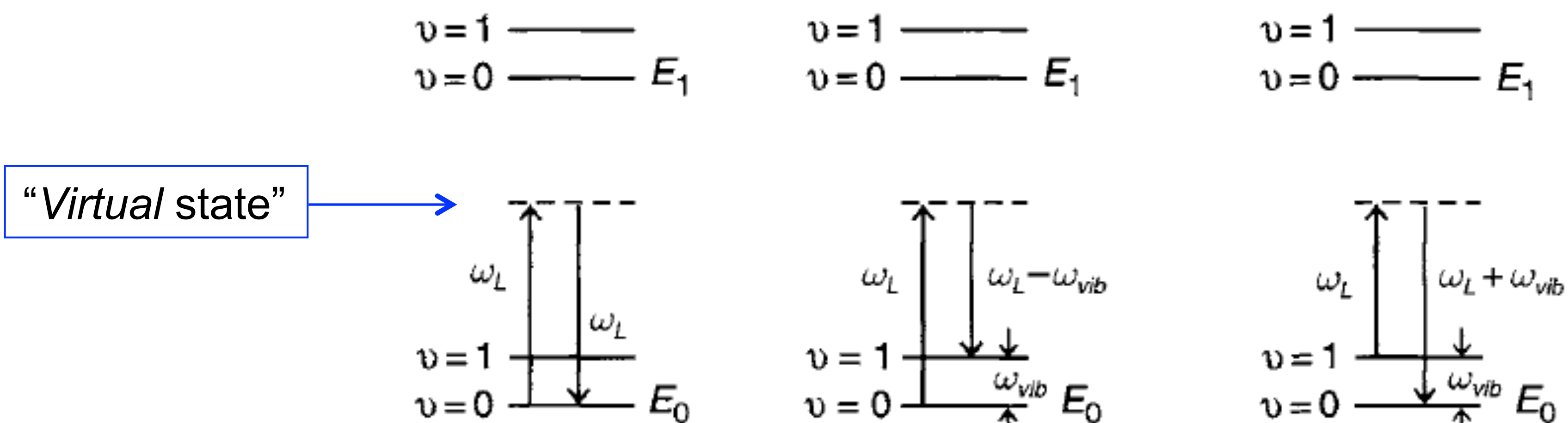
The first term is connected with linear polarization and describes spontaneous, linear Raman scattering, which is studied by the conventional methods of Raman spectroscopy. The second- and third terms describe spontaneous, nonlinear Raman scattering (called *hyper-Raman scattering*). The third is also responsible for the, *stimulated Raman scattering*.

La dispersión inelástica de luz en la óptica lineal y no-lineal

The *Raman scattering* processes can be described by the equation

$$P_i = \boxed{\chi_{ij}^{(1)} E_j} + \chi_{ijk}^{(2)} E_j E_k + \chi_{ijkl}^{(3)} E_j E_k E_l .$$

The first term is connected with linear polarization and describes spontaneous, linear Raman scattering, which is studied by the conventional methods of Raman spectroscopy. The second- and third terms describe spontaneous, nonlinear Raman scattering (called *hyper-Raman scattering*). The third is also responsible for the, *stimulated Raman scattering*.



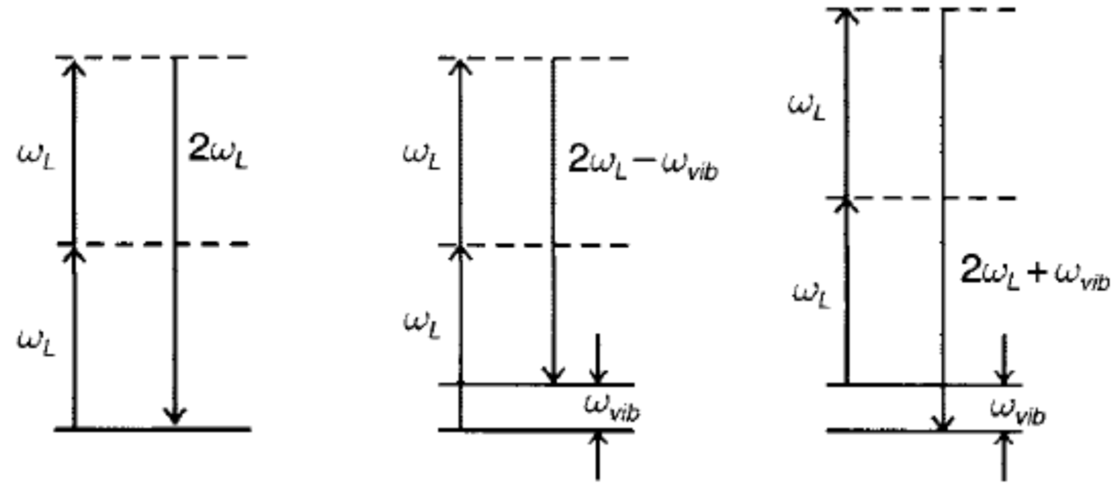
Rayleigh scattering | Raman scattering Stokes | Raman scattering anti-Stokes

LINEAL

La dispersión inelástica de luz en la óptica lineal y no-lineal

“*Virtual* state” →

$$P_i = \boxed{\chi^{(2)}_{ijk} E_j E_k} + \chi^{(3)}_{ijkl} E_j E_k E_l$$



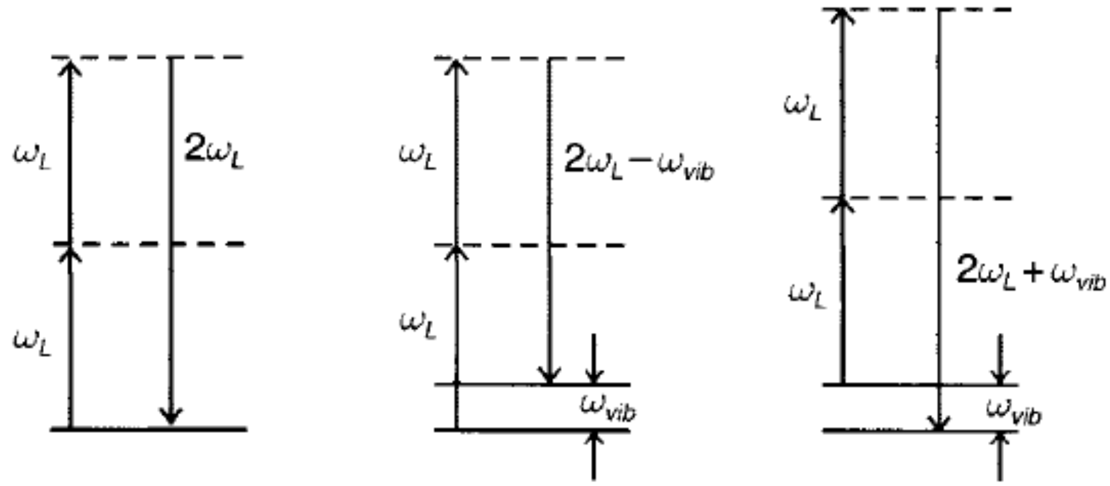
HIPER Rayleigh scattering

Raman scattering Stokes

Raman scattering anti-Stokes

La dispersión inelástica de luz en la óptica lineal y no-lineal

$$P_i = \boxed{\chi^{(2)}_{ijk} E_j E_k} + \chi^{(3)}_{ijkl} E_j E_k E_l$$



HIPER

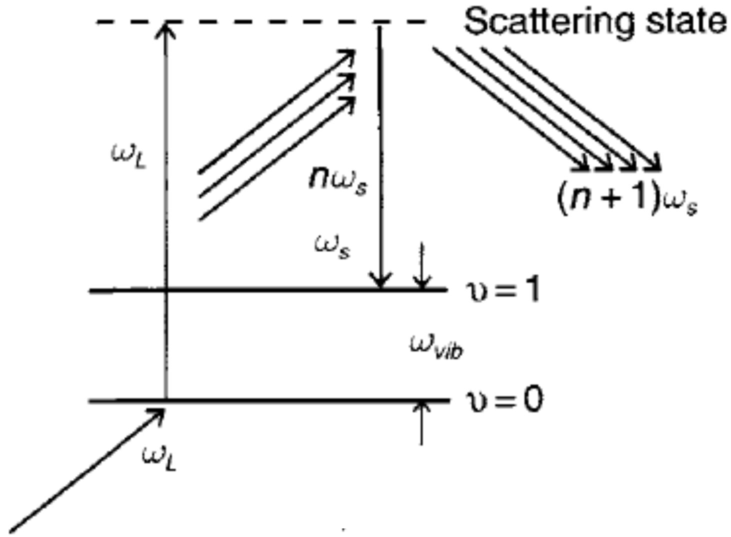
Rayleigh scattering

Raman scattering Stokes

Raman scattering anti-Stokes

"*Virtual* state" →

$$P_i = \chi^{(2)}_{ijk} E_j E_k + \boxed{\chi^{(3)}_{ijkl} E_j E_k E_l}$$



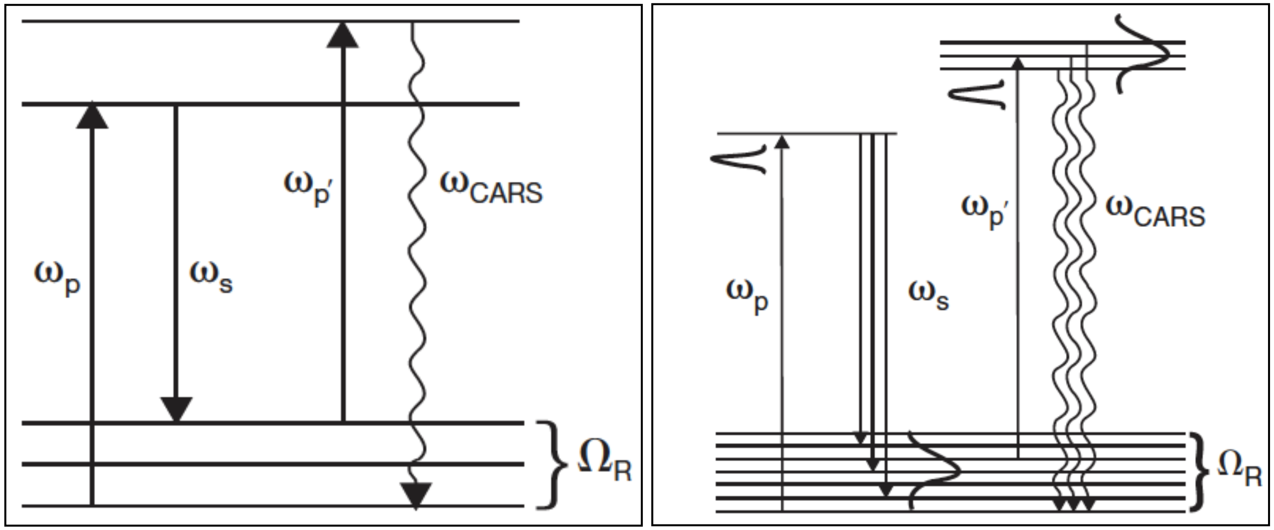
Raman scattering Stokes

ESTIMULADO

Un proceso estimulado $\chi^{(3)}$: CARS

Coherent anti-Stokes Raman scattering

CARS

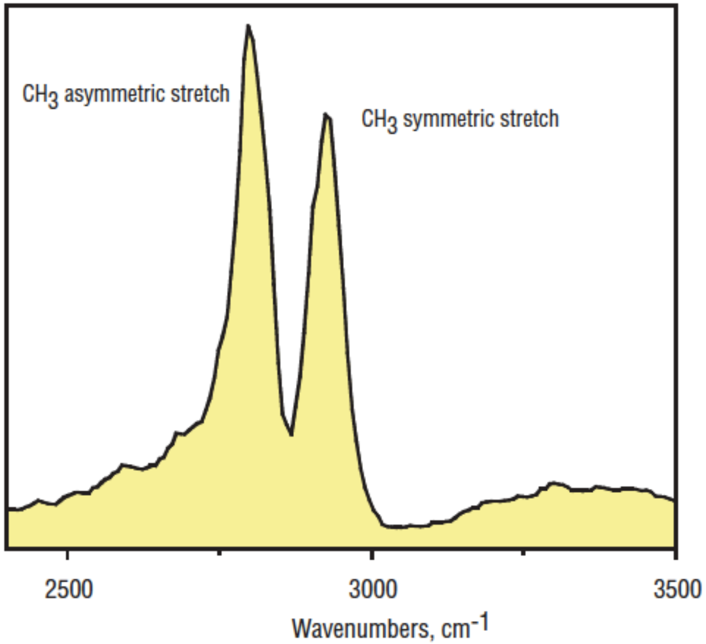


$$\omega_{AS} = \omega_L + \omega_{vib} = \omega_L + (\omega_L - \omega_s)$$

$$\omega_4 = \omega_{AS};\ \omega_1 = \omega_L;\ \omega_2 = \omega_L;\ \omega_3 = \omega_S$$

$$\Delta \boldsymbol{k} = \boldsymbol{k}_L + \boldsymbol{k}_L - \boldsymbol{k}_S - \boldsymbol{k}_{AS} = 2\boldsymbol{k}_L - \boldsymbol{k}_S - \boldsymbol{k}_{AS}$$

$$I_{AS}^{SRS} = const \cdot ((\chi^{(3)}))^2 I_L^2 I_S l^2 \left(\frac{\sin \Delta k l/2}{\Delta k l/2} \right)^2$$



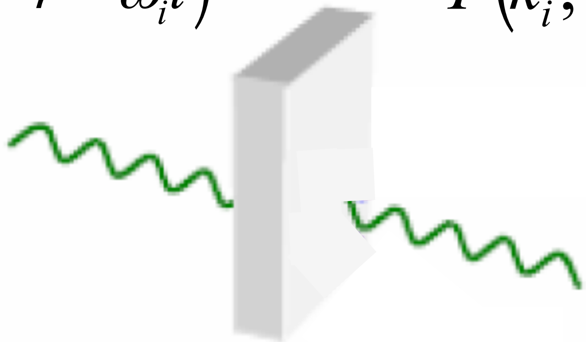
Pero, porqué aparecen bandas laterales?

Y qué es eso de los "estados virtuales"?

Modelo macroscópico de la dispersión Raman

$$\vec{E}_i(\vec{r},t) = \vec{E}_i(\vec{k}_i,\omega_i)\cos(\vec{k}_i \cdot \vec{r} - \omega_i t)$$

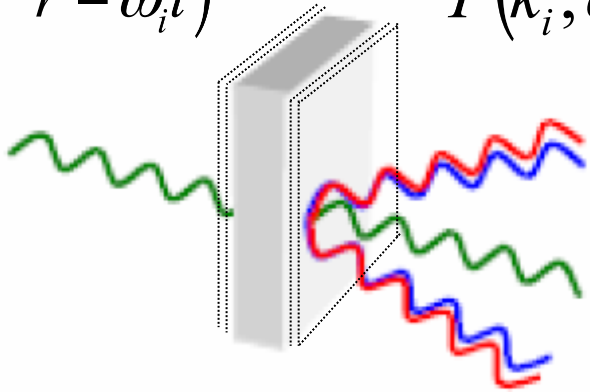
$$\vec{P}(\vec{k}_i,\omega_i) = \tilde{\chi}(\vec{k}_i,\omega_i)\vec{E}_i(\vec{k}_i,\omega_i)$$



Modelo macroscópico de la dispersión Raman

$$\vec{E}_i(\vec{r},t) = \vec{E}_i(\vec{k}_i,\omega_i)\cos(\vec{k}_i\cdot\vec{r}-\omega_i t)$$

$$\vec{P}(\vec{k}_i,\omega_i) = \tilde{\chi}(\vec{k}_i,\omega_i)\vec{E}_i(\vec{k}_i,\omega_i)$$

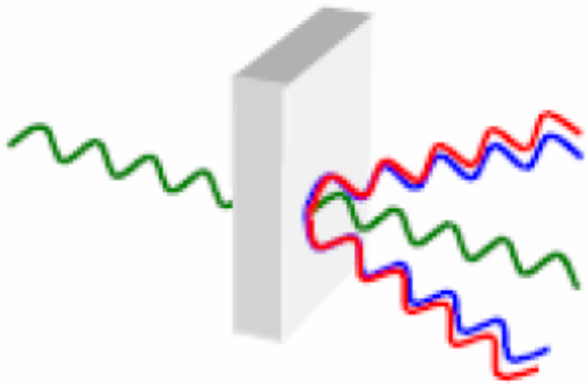


$$\vec{Q}(\vec{r},t) = \vec{Q}(\vec{q},\omega_0)\cos(\vec{q}\cdot\vec{r}-\omega_0 t)$$

$$\tilde{\chi}(\vec{k}_i,\omega_i,\vec{Q}) = \tilde{\chi}_0(\vec{k}_i,\omega_i) + (\partial\tilde{\chi}/\partial\vec{Q})_0\vec{Q}(\vec{r},t) + \ldots$$

$$\boxed{\vec{P}(\vec{r},t,\vec{Q}) = \vec{P}_0(\vec{r},t) + \vec{P}_{ind}(\vec{r},t,\vec{Q})}$$

Modelo macroscópico de la dispersión Raman



$$\vec{P}_0(\vec{r},t) = \tilde{\chi}_0(\vec{k}_i,\omega_i)\vec{E}_i(\vec{k}_i,\omega_i)\cos(\vec{k}_i\cdot\vec{r}-\omega_i t)$$

Dispersión Rayleigh

$$\vec{P}_{ind}(\vec{r},t,\vec{Q}) = (\partial\tilde{\chi}/\partial\vec{Q})_0\vec{Q}(\vec{r},t)\vec{E}_i(\vec{k}_i,\omega_i)\cos(\vec{k}_i\cdot\vec{r}-\omega_i t) =$$

$$\tfrac{1}{2}(\partial\tilde{\chi}/\partial\vec{Q})_0\vec{Q}(\vec{q},\omega_0)\vec{E}_i(\vec{k}_i,\omega_i)\times\left\{\cos\left[(\vec{k}_i+\vec{q})\cdot\vec{r}-(\omega_i+\omega_0)t\right]+\cos\left[(\vec{k}_i-\vec{q})\cdot\vec{r}-(\omega_i-\omega_0)t\right]\right\}$$

Dispersión Raman

$$I_R(\omega_s) \propto \left|\vec{E}_s(\vec{k}_s,\omega_s)(\partial\tilde{\chi}/\partial\vec{Q})_0\vec{Q}(\vec{q},\omega_0)\vec{E}_i(\vec{k}_i,\omega_i)\right|^2$$

Sección eficaz Raman

Susceptibilidad fluctuante

$$\frac{d\eta(\omega_\xi)}{d\Omega} \propto \left| \int E_s^*(z)\, \delta\chi_\xi(z)\, E_i(z)\, dz \right|^2$$

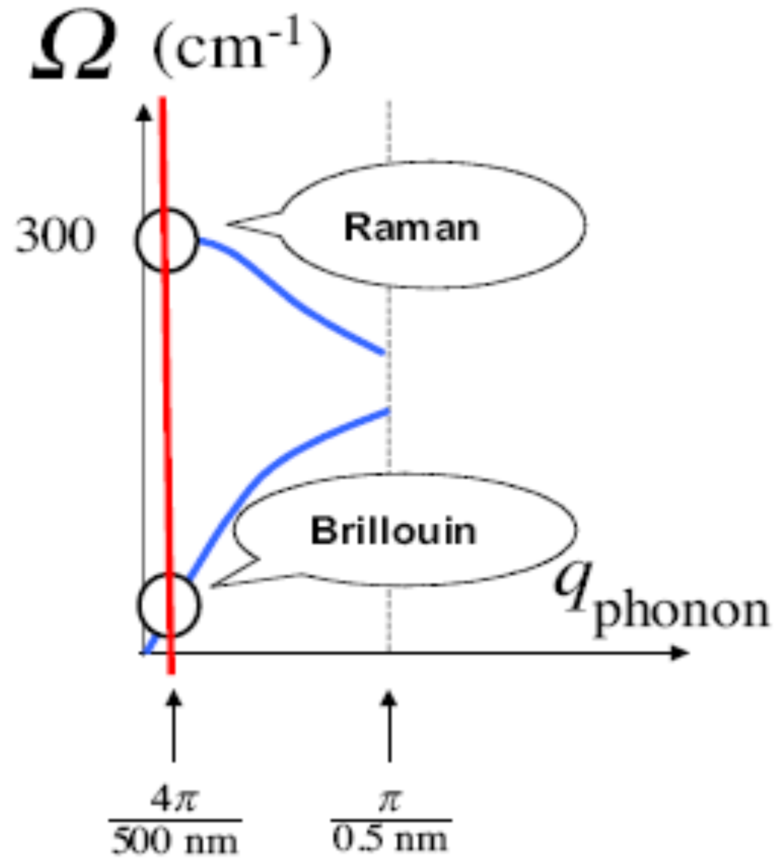
Polarization inducida @ $\omega_L \pm \omega_{ph}$

"Raman": Fonones ópticos $\delta\chi = \frac{\partial \chi}{\partial u} \cdot \Delta u$

"Brillouin": Fonones acústicos $\delta\chi = \frac{\partial \chi}{\partial s} \cdot \Delta s, s = \frac{\partial u}{\partial z}$

Dispersión Raman en cristales: reglas de conservación

$$\begin{cases} \hbar\omega_L = \hbar\omega_S + \hbar\Omega_{\text{phonon}} \\ \hbar\boldsymbol{K}_L = \hbar\boldsymbol{K}_S + \hbar\boldsymbol{q}_{\text{phonon}} \end{cases}$$



Dispersión Raman por fonones acústicos: Mecanismo fotoelástico

$$\frac{d\eta(\omega_\xi)}{d\Omega} \propto \left| \int E_s^*(z)\, \delta\chi_\xi(z)\, E_i(z)\, dz \right|^2 \qquad \delta\chi_s(\mathbf{r},t) = \left.\frac{\partial \chi}{\partial s}\right|_0 \delta s(\mathbf{r},t)$$

$$\boxed{\frac{d\eta(\omega)}{d\Omega} \propto \tfrac{(n_\omega+1)}{\omega} \left| \int E_s^*(z)\, p(z)\, \frac{\partial u(z)}{\partial z}\, E_i(z)\, dz \right|^2}$$

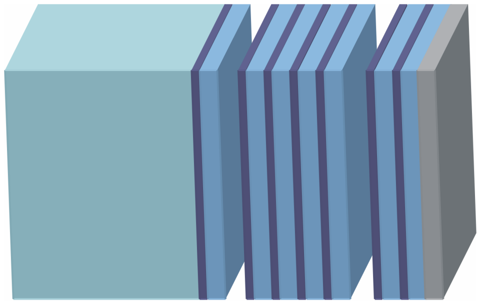
Dispersión Raman por fonones acústicos: Mecanismo fotoelástico

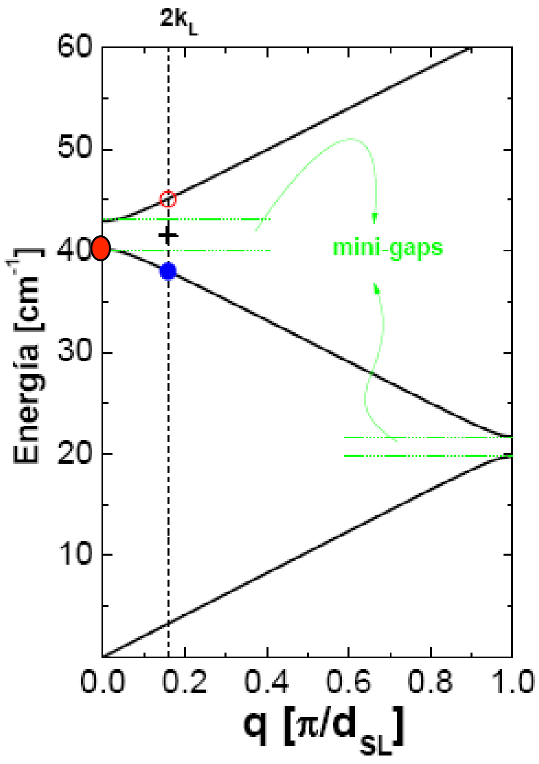
$$\frac{d\eta(\omega_\xi)}{d\Omega} \propto \left| \int E_s^*(z)\, \delta\chi_\xi(z)\, E_i(z)\, dz \right|^2 \qquad \delta\chi_s(\mathbf{r},t) = \left.\frac{\partial\chi}{\partial s}\right|_0 \delta s(\mathbf{r},t)$$

$k_s \;\; \pm nG \;\; +k_{ph} \;\; -k_L$

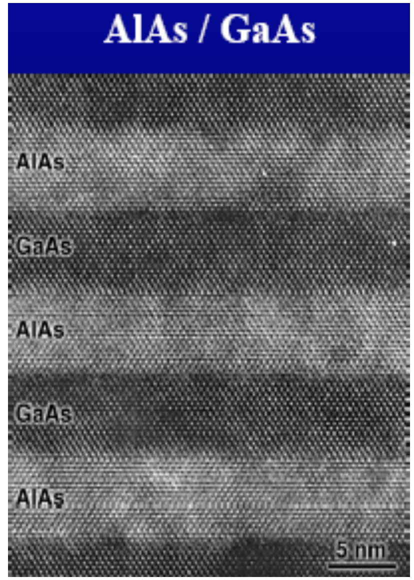
$$\frac{d\eta(\omega)}{d\Omega} \propto \frac{(n_\omega+1)}{\omega} \left| \int E_s^*(z)\, p(z)\, \frac{\partial u(z)}{\partial z}\, E_i(z)\, dz \right|^2$$

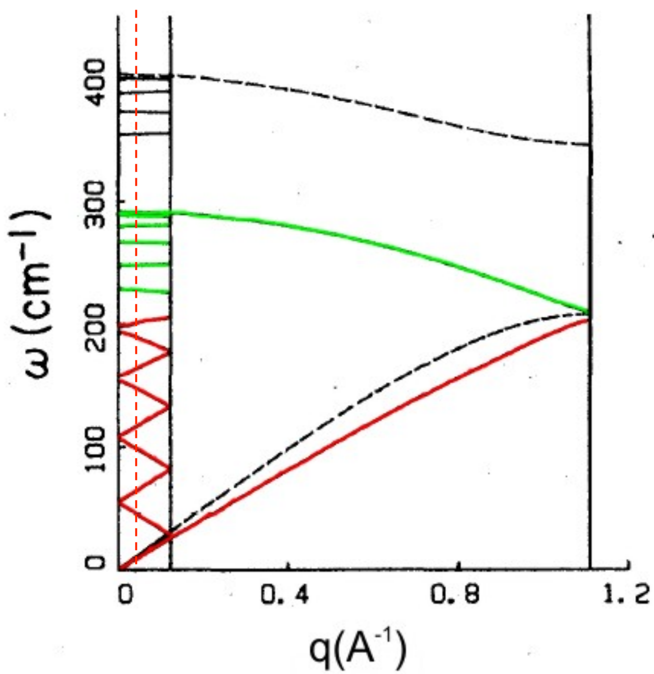
$$p(z) = \sum_n P_n\, e^{inGz} \qquad G = \frac{2\pi}{d_{SL}}$$

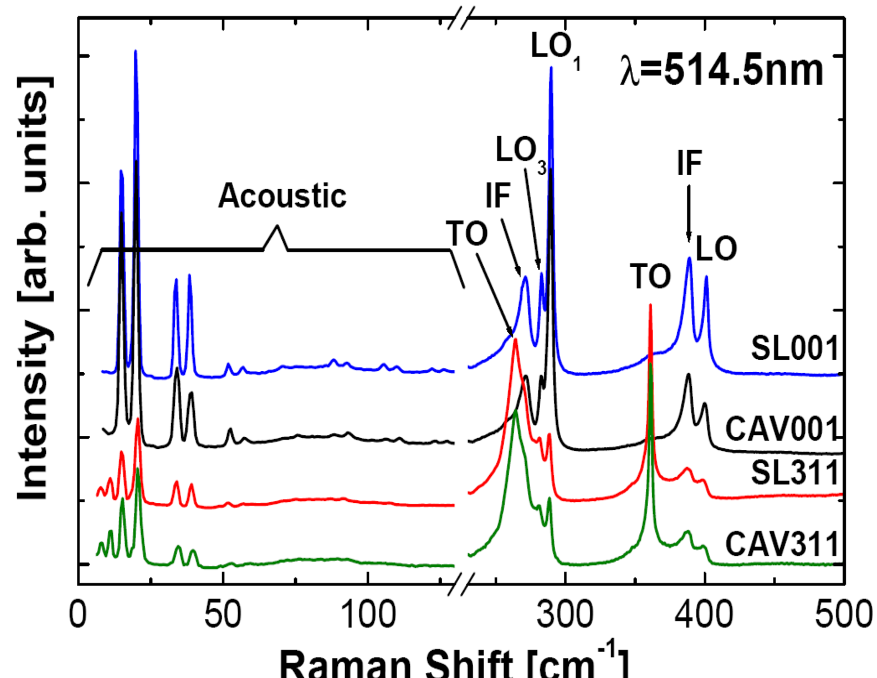




Dispersión Raman en superredes







Dispersión elástica (Rayleigh) e inelástica (Raman)

Fluctuaciones *estáticas* en la función dieléctrica dan lugar a la dispersión Rayleigh (elástica)

Fluctuaciones *dinámicas* en la función dieléctrica dan lugar a la dispersión Raman (inelástica)

Pero, porqué aparecen bandas laterales?

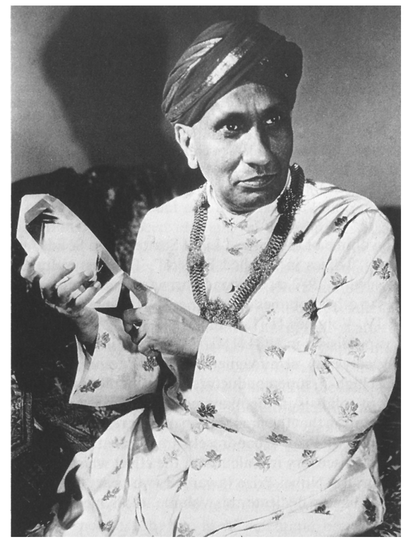
Y qué es eso de los "estados virtuales"?

La *debilidad* de la técnica

Sir C. V. Raman

The Nobel Prize in Physics 1930

"for his work on the scattering of light and for the discovery of the effect named after him"

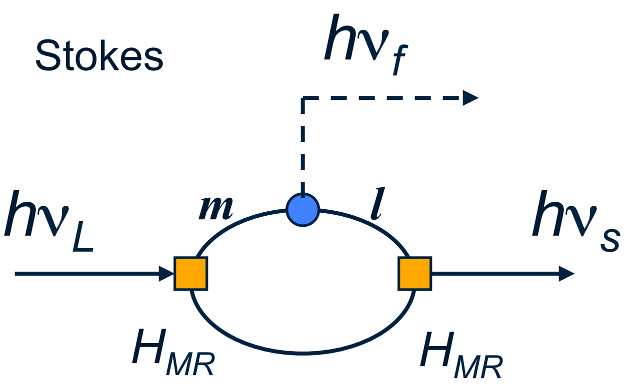


Sir Chandrasekhara Venkata Raman

" The chief difficulty which had hitherto oppressed us in the study of the new phenomenon was its extreme feebleness in general. This was overcome by using a 7-inch refracting telescope in combination with a short-focus lens to condense sunlight into a pencil of very great intensity."

Porqué esta debilidad?, y cómo superar esta debilidad?

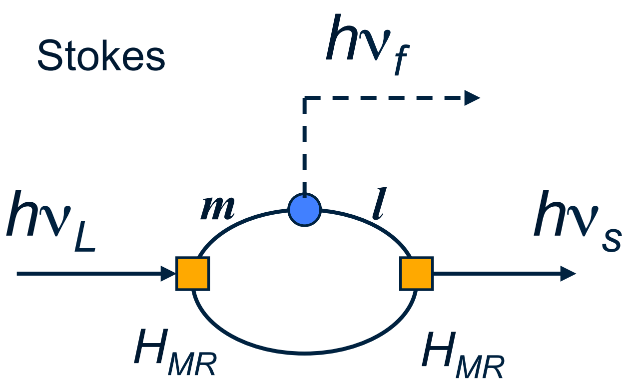
1) Resonancia: Teoría cuántica de la dispersión Raman



3er orden perturbaciones:
2do orden luz-materia
1er orden electrón-fonón

$$\sigma(\omega_i) \propto \left| \sum_{l,m} \frac{\langle f | H_{MR} | l \rangle \langle l | H_{e-v} | m \rangle \langle m | H_{MR} | i \rangle}{(\omega_S - \omega_l - i\gamma_l)(\omega_L - \omega_m - i\gamma_m)} \right|^2$$

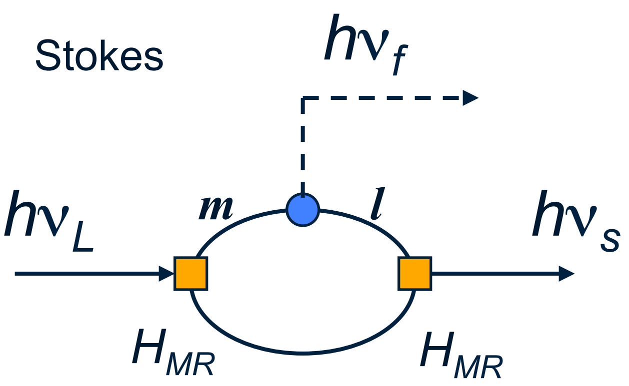
1) Resonancia: Teoría cuántica de la dispersión Raman



3^er orden perturbaciones:
2^do orden luz-materia
1^er orden electrón-fonón

$$\sigma(\omega_i) \propto \left| \sum_{l,m} \frac{\langle f \,|\, H_{MR} \,|\, l \rangle \langle l \,|\, H_{e-v} \,|\, m \rangle \langle m \,|\, H_{MR} \,|\, i \rangle}{(\omega_S - \omega_l - i\gamma_l)(\omega_L - \omega_m - i\gamma_m)} \right|^2 \delta(E_f - E_i)$$

1) Resonancia: Teoría cuántica de la dispersión Raman



3er orden perturbaciones:
2do orden luz-materia
1er orden electrón-fonón

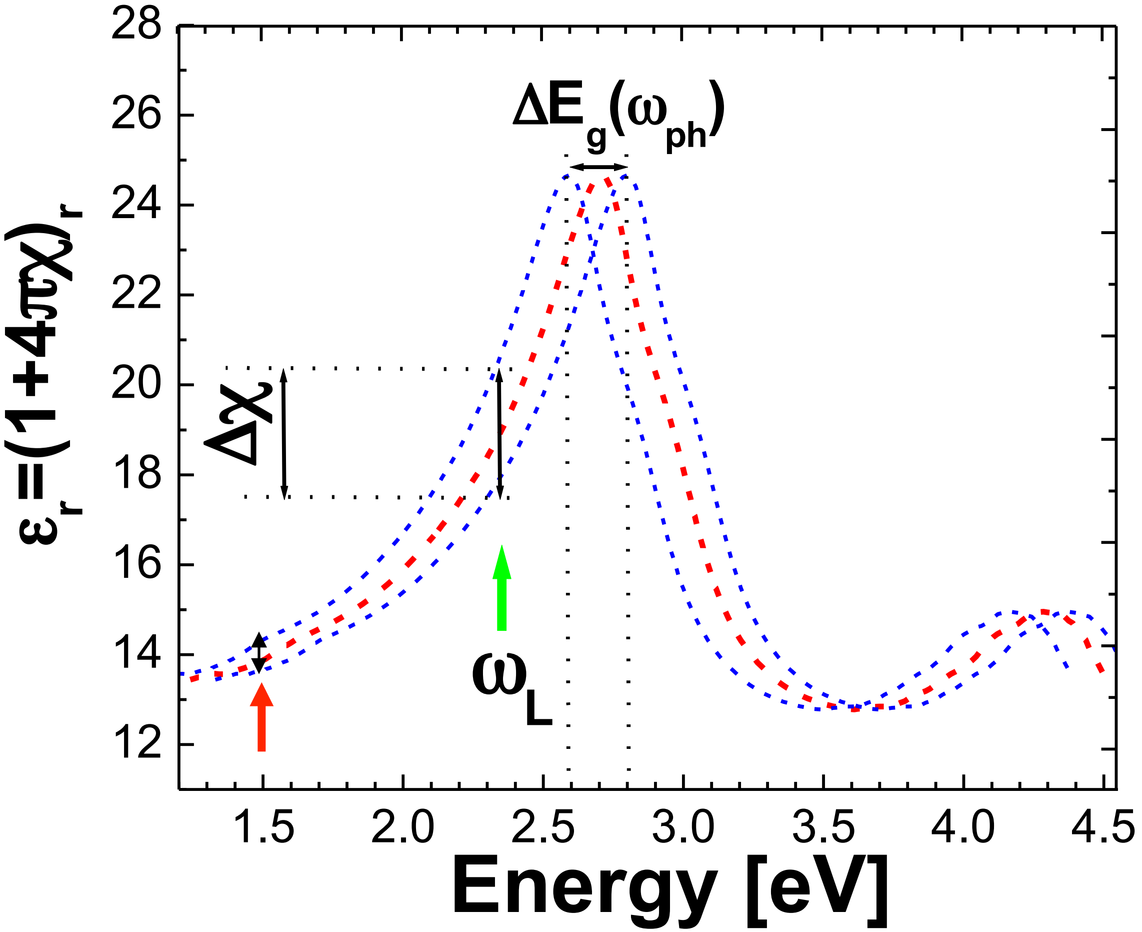
$$\sigma(\omega_i) \propto \left| \sum_{l,m} \frac{\langle f | H_{MR} | l \rangle \langle l | H_{e-v} | m \rangle \langle m | H_{MR} | i \rangle}{(\omega_S - \omega_l - i\gamma_l)(\omega_L - \omega_m - i\gamma_m)} \right|^2 \delta(E_f - E_i)$$

Resonancia saliente

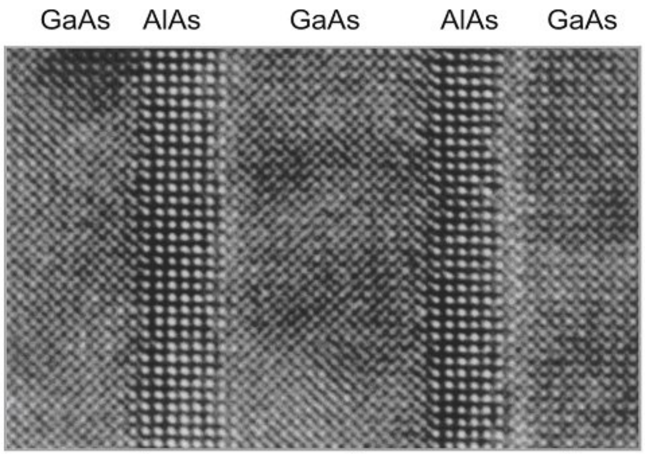
Resonancia entrante

1) Origen de la resonancia en la teoría macroscópica del Raman

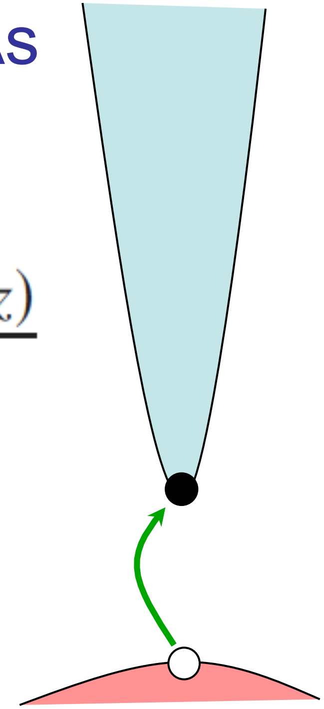
$$\vec{P}_{ind}\left(\vec{r},t,\vec{Q}\right) \propto \left(\partial\tilde{\chi}/\partial\vec{Q}\right) = \left(\partial\tilde{\chi}/\partial\omega_g\right)\left(\partial\omega_g/\partial\vec{Q}\right)$$

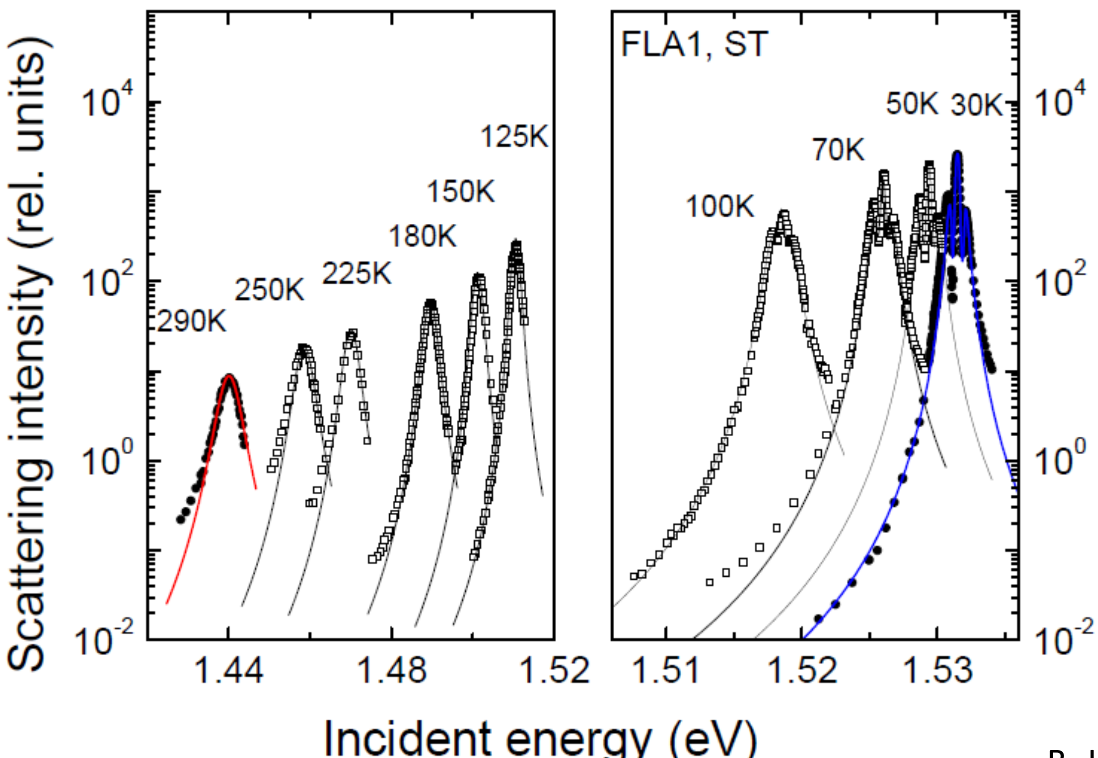


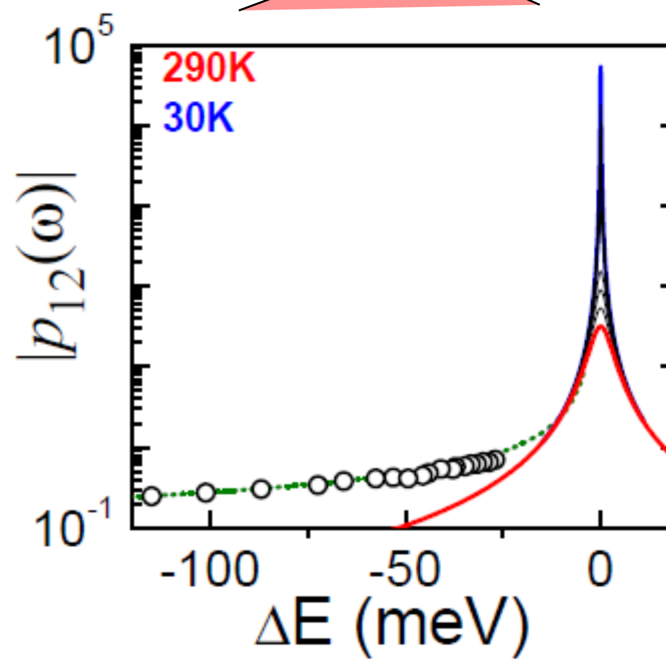
1) Acoplamiento resonante en MQWs de GaAs



$$p(z) = \frac{\partial \chi(z)}{\partial s}$$

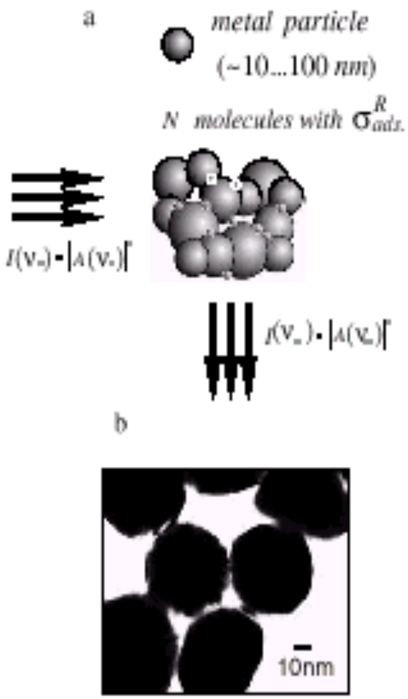




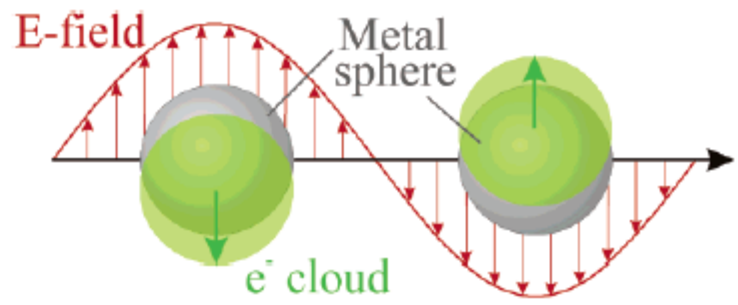


B. Jusserand *et al*, Phys. Rev. Lett. **115**, 267402 (2015)

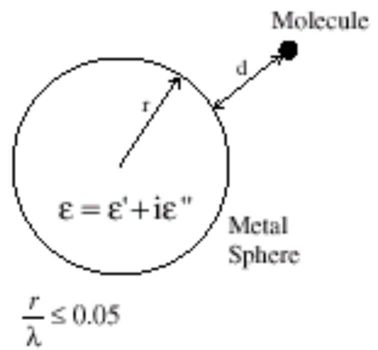
2) SERS: “Surface Enhanced Raman Spectroscopy”



K. Kneipp, J. Phys.: Cond. Matt. **14**, R597 (2002)



L. Kelly, J. Chem. Phys. B **107**, 668 (2003)



$$E_M = E_0 + E_{sp}$$

E : field of a point dipole in the center of the sphere

$$E_{sp} = r^3 \frac{\varepsilon - \varepsilon_0}{\varepsilon + 2\varepsilon_0} E_0 \frac{1}{(r+d)^3}$$

$$\sigma(\omega) \propto \left| \sum_{i,j} \frac{\langle f | H_{MR} | i \rangle \langle i | H_{e-ph} | j \rangle \langle j | H_{MR} | 0 \rangle}{(\omega - \omega_i - i\gamma_i)(\omega - \omega_j - i\gamma_j)} \right|^2$$

Efecto electromagnético --> ~10^5-10^{10}

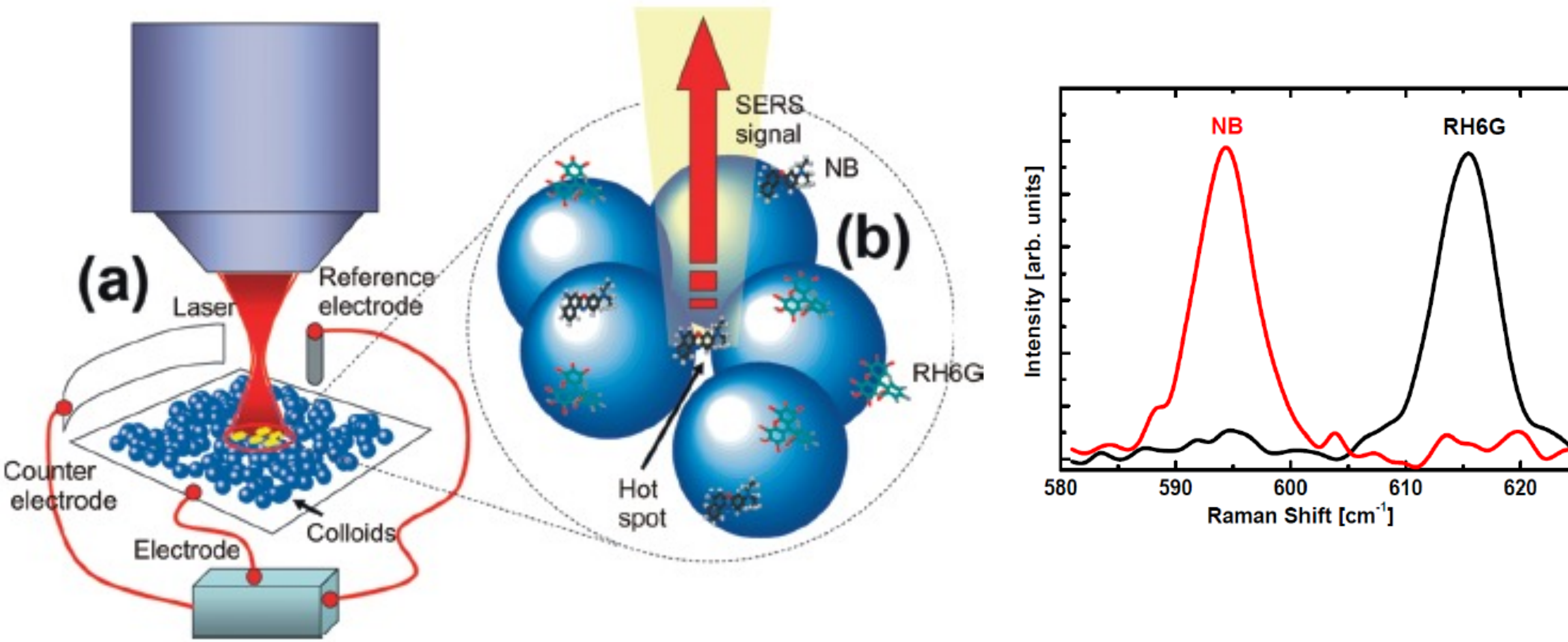
2) SERS de moléculas individuales

J|A|C|S COMMUNICATIONS

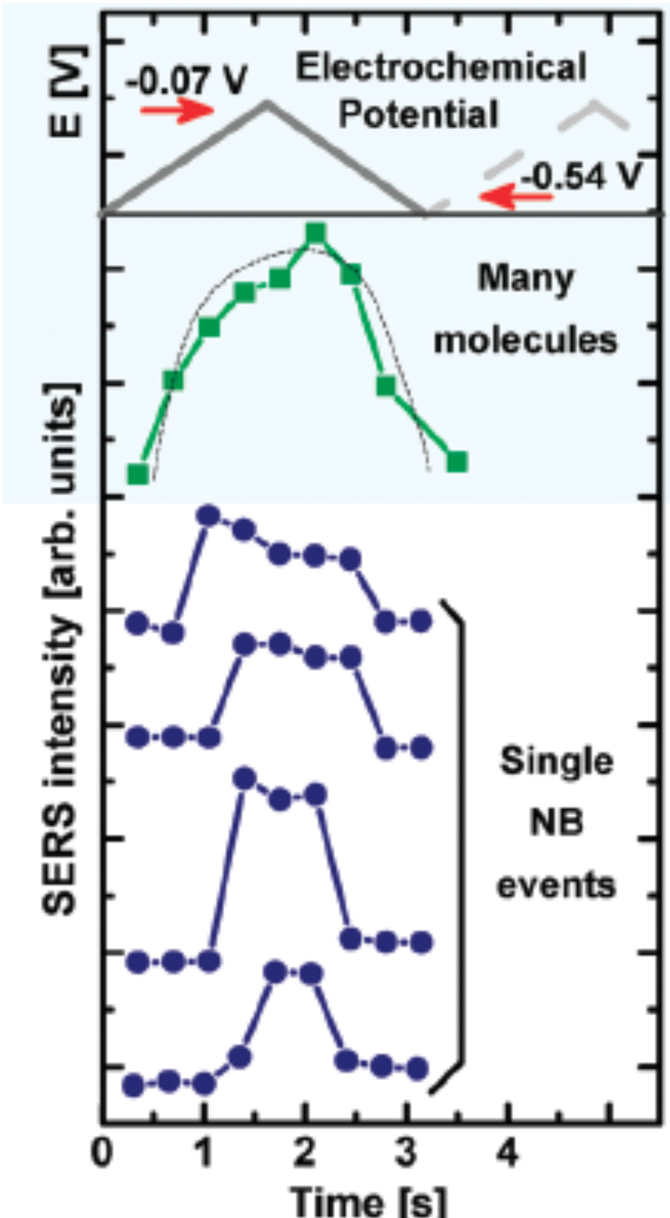
Published on Web 12/07/2010

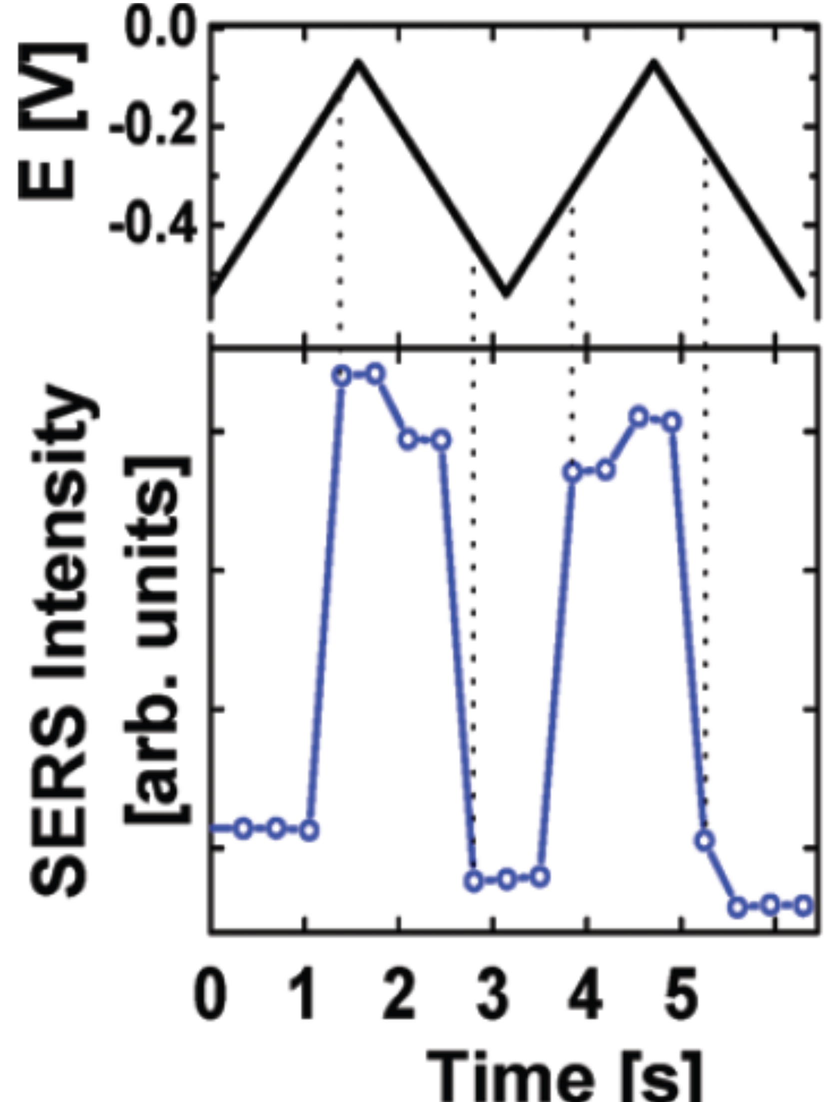
Monitoring the Electrochemistry of Single Molecules by Surface-Enhanced Raman Spectroscopy

Emiliano Cortés,[†] Pablo G. Etchegoin,[*,‡] Eric C. Le Ru,[‡] Alejandro Fainstein,[§] María E. Vela,[†] and Roberto C. Salvarezza[†]

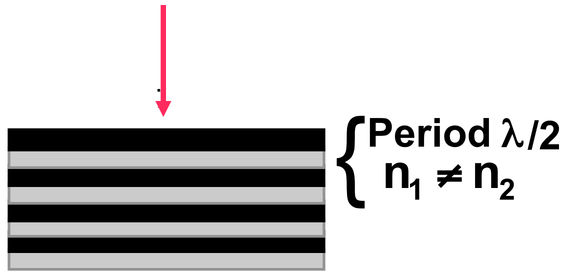


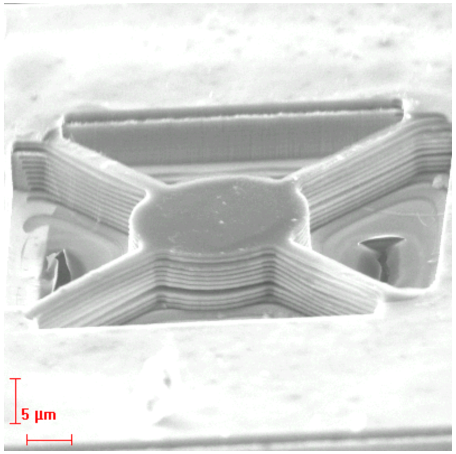
2) Mirando con Raman la carga y descarga de una molécula

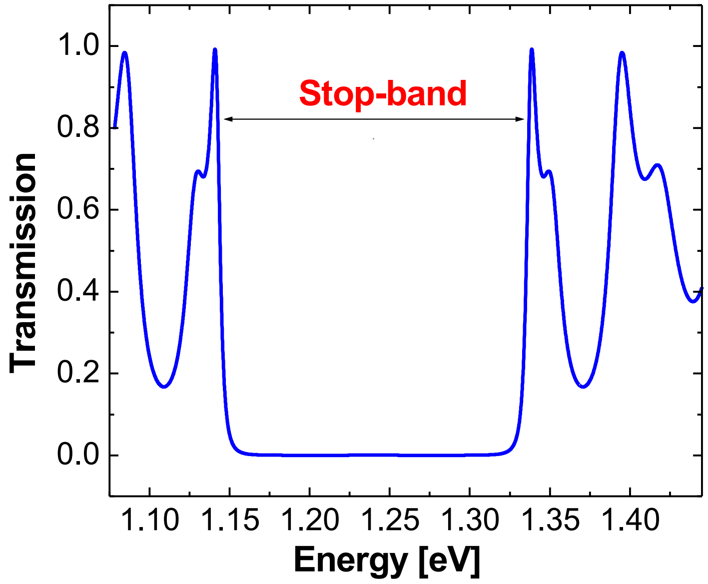




3) Resonadores ópticos: espejos de Bragg (DBRs)



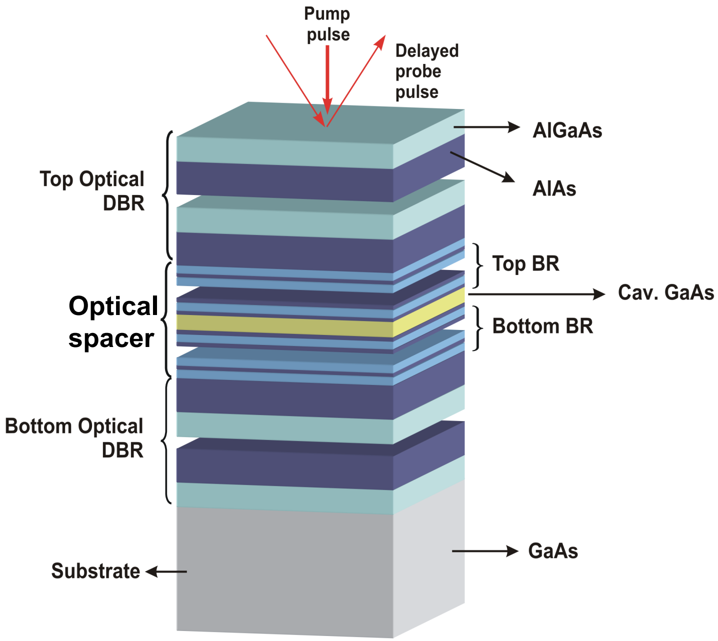


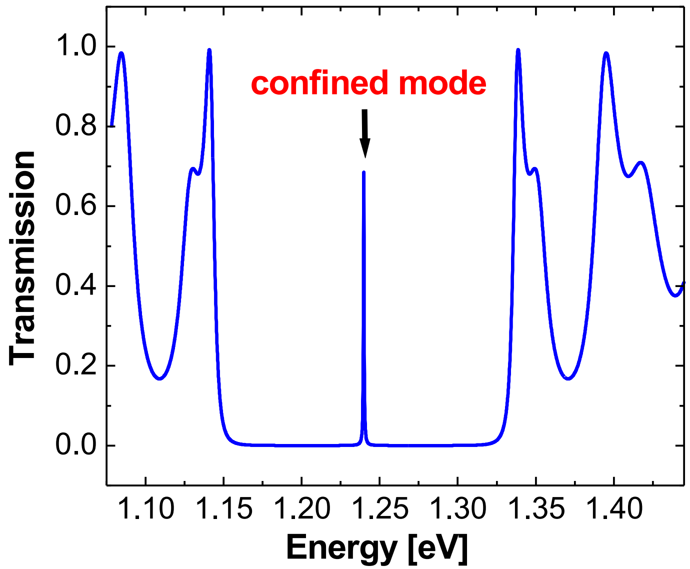


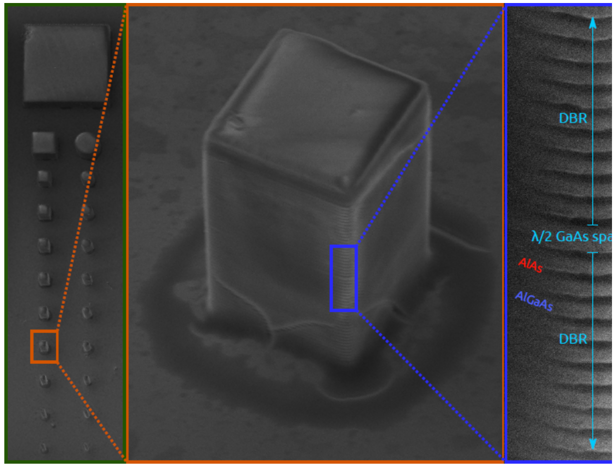
- 1D "Photonic band gap"
- $R \approx 1 - 4(n_1/n_2)^{2N}, n_1 < n_2$
- Stop-band $f(n_1/n_2)$

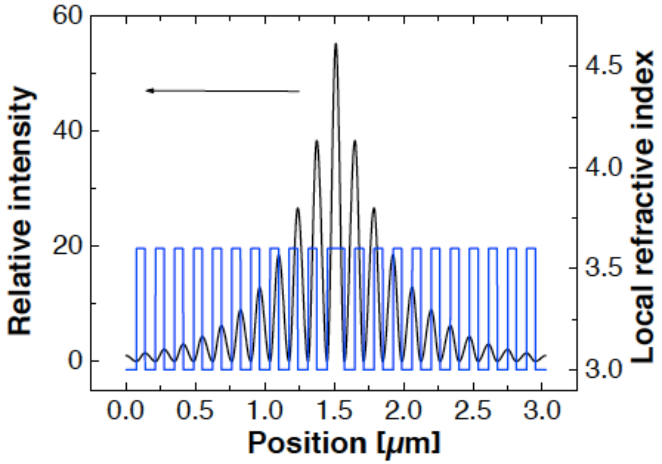
$$R \sim 0.995$$

3) Resonadores ópticos: Microcavidad







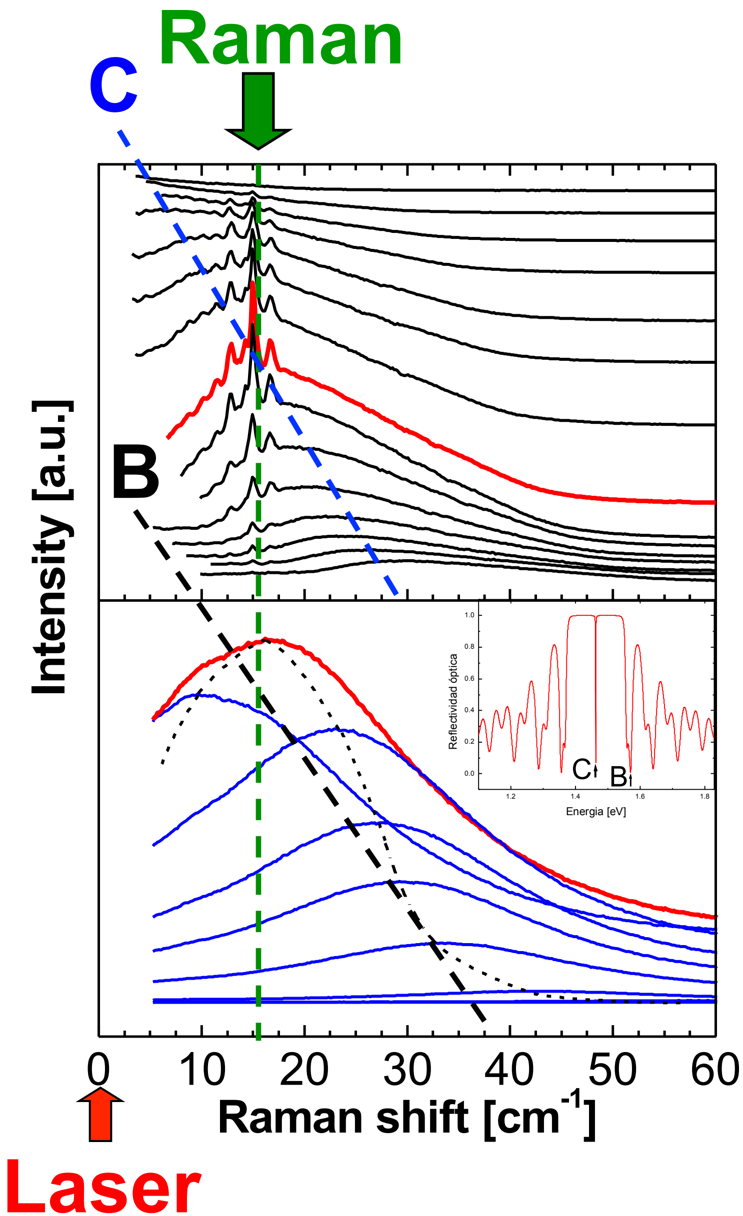


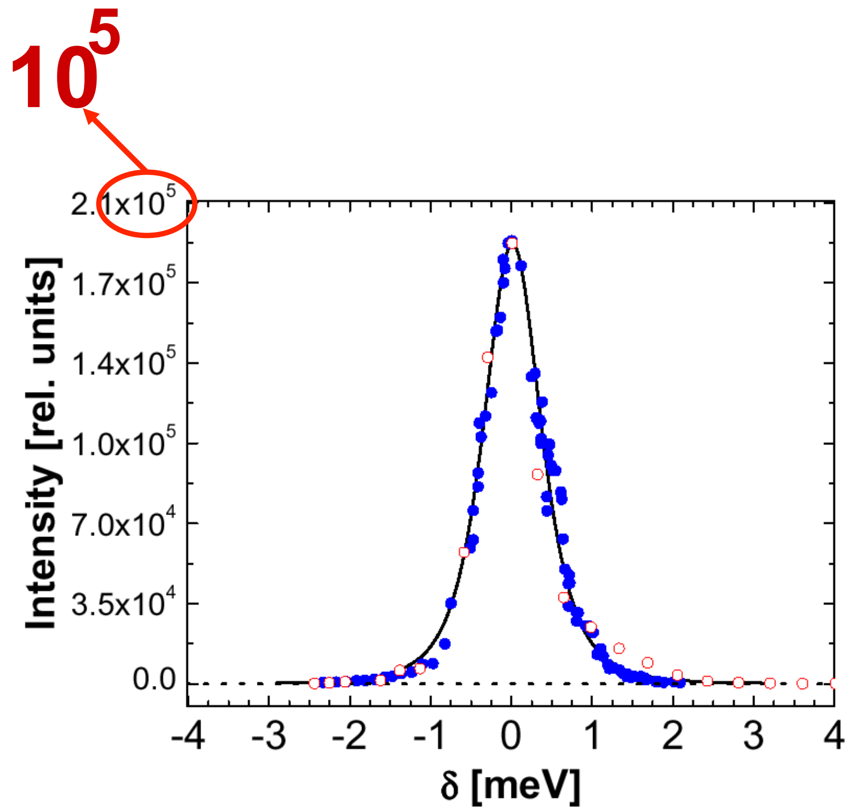
$$F = \pi \frac{L_{eff}}{d} \frac{\sqrt{R}}{(1-R)} \approx 3000$$

$$\tau_c = \frac{L_{eff}}{\mathrm{v}\ln(R)} \approx \frac{L_{eff}}{\mathrm{v}} \times 500$$

A. Fainstein *et al.*,PRL **75**, 3764 (95)
A. Fainstein *et al.*,PRL **78**, 1576 (97)
A. Fainstein *et al.*,PRL **86**, 3411 (01)

3) Resonadores ópticos: doble resonancia Raman

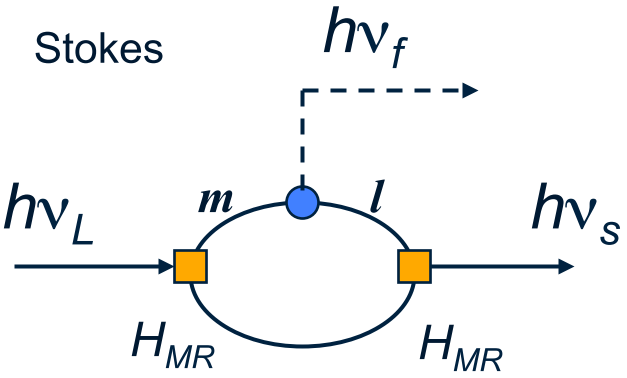




$$\sigma(\omega) \propto \left| \int dz E_s^* E_i \frac{\partial u_\omega(z)}{\partial z} p(z) \right|^2$$

Pero volvamos a la teoría cuántica de la dispersión Raman

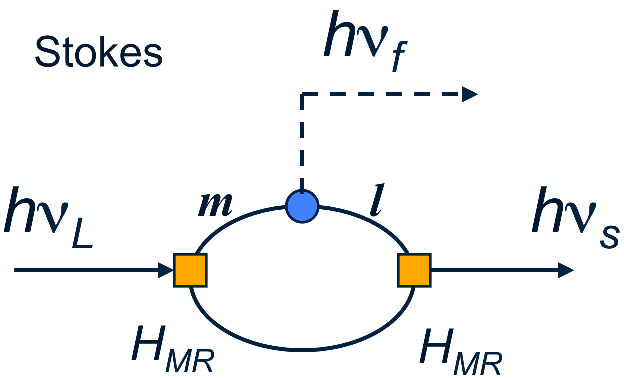
Teoría cuántica de la dispersión Raman resonante



3^{er} orden perturbaciones:
2^{ndo} orden luz-materia
1^{er} orden electrón-fonón

$$\sigma(\omega_i) \propto \left| \sum_{l,m} \frac{\langle f | H_{MR} | l \rangle \langle l | H_{e-v} | m \rangle \langle m | H_{MR} | i \rangle}{(\omega_S - \omega_l - i\gamma_l)(\omega_L - \omega_m - i\gamma_m)} \right|^2$$

Teoría cuántica de la dispersión Raman resonante



3^{er} orden perturbaciones:
2^{do} orden luz-materia
1^{er} orden electrón-fonón

$$\sigma(\omega_i) \propto \left| \sum_{l,m} \frac{\langle f | H_{MR} | l \rangle \langle l | H_{e-v} | m \rangle \langle m | H_{MR} | i \rangle}{(\omega_S - \omega_l - i\gamma_l)(\omega_L - \omega_m - i\gamma_m)} \right|^2$$

$$P_R \propto (N_{Stokes} + 1)(N_{fonon} + 1) N_{laser}$$

Qué es la optomecánica en cavidades?

The gravitational wave detector LIGO

as a huge cavity optomechanical system?

The gravitational wave detector LIGO

as a huge cavity optomechanical system?

Cavity optomechanics as a limiting factor:

Quantum theory of measurement & gravitational wave detection

PHYSICAL REVIEW LETTERS

VOLUME 45 | 14 JULY 1980 | NUMBER 2

Quantum-Mechanical Radiation-Pressure Fluctuations in an Interferometer

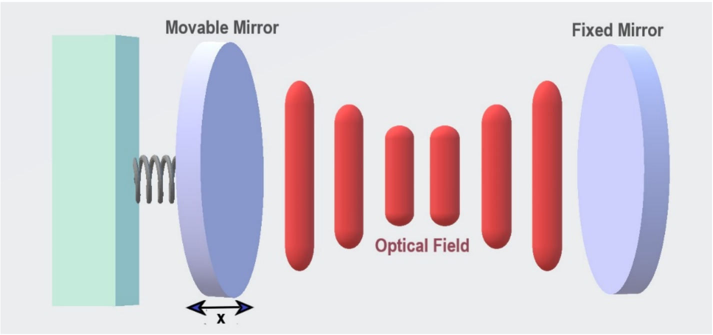
Carlton M. Caves

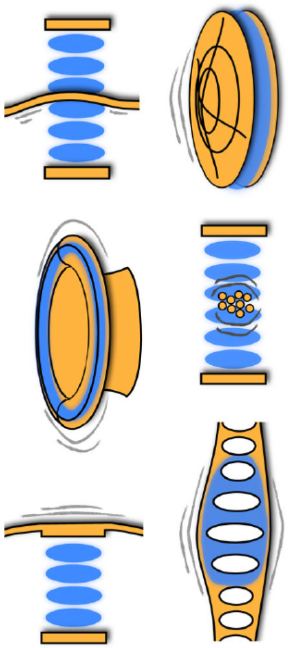
W. K. Kellogg Radiation Laboratory, California Institute of Technology, Pasadena, California 91125

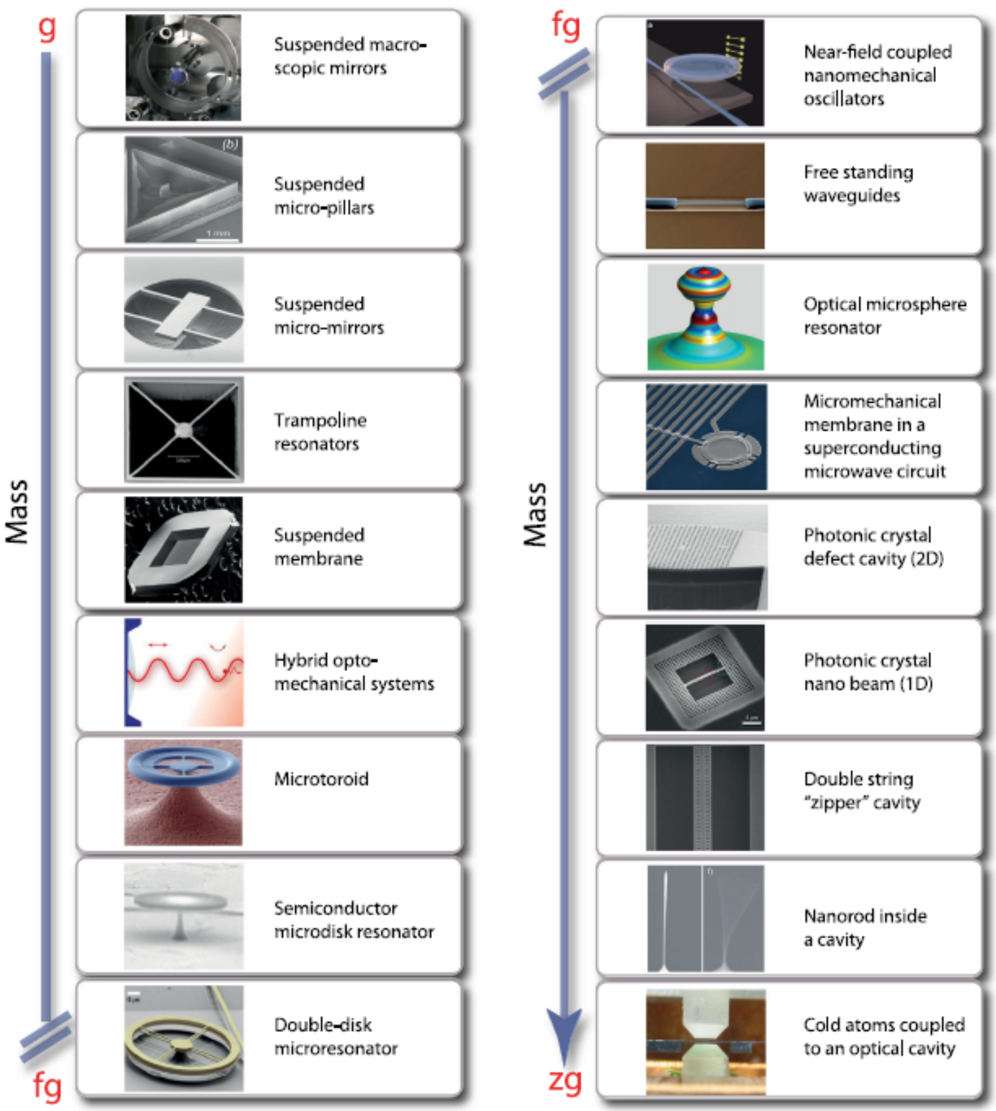
(Received 29 January 1980)

The interferometers now being developed to detect gravitational vaves work by measuring small changes in the positions of free masses. There has been a controversy whether quantum-mechanical radiation-pressure fluctuations disturb this measurement. This Letter resolves the controversy: They do.

Qué es la optomecánica de cavidades?







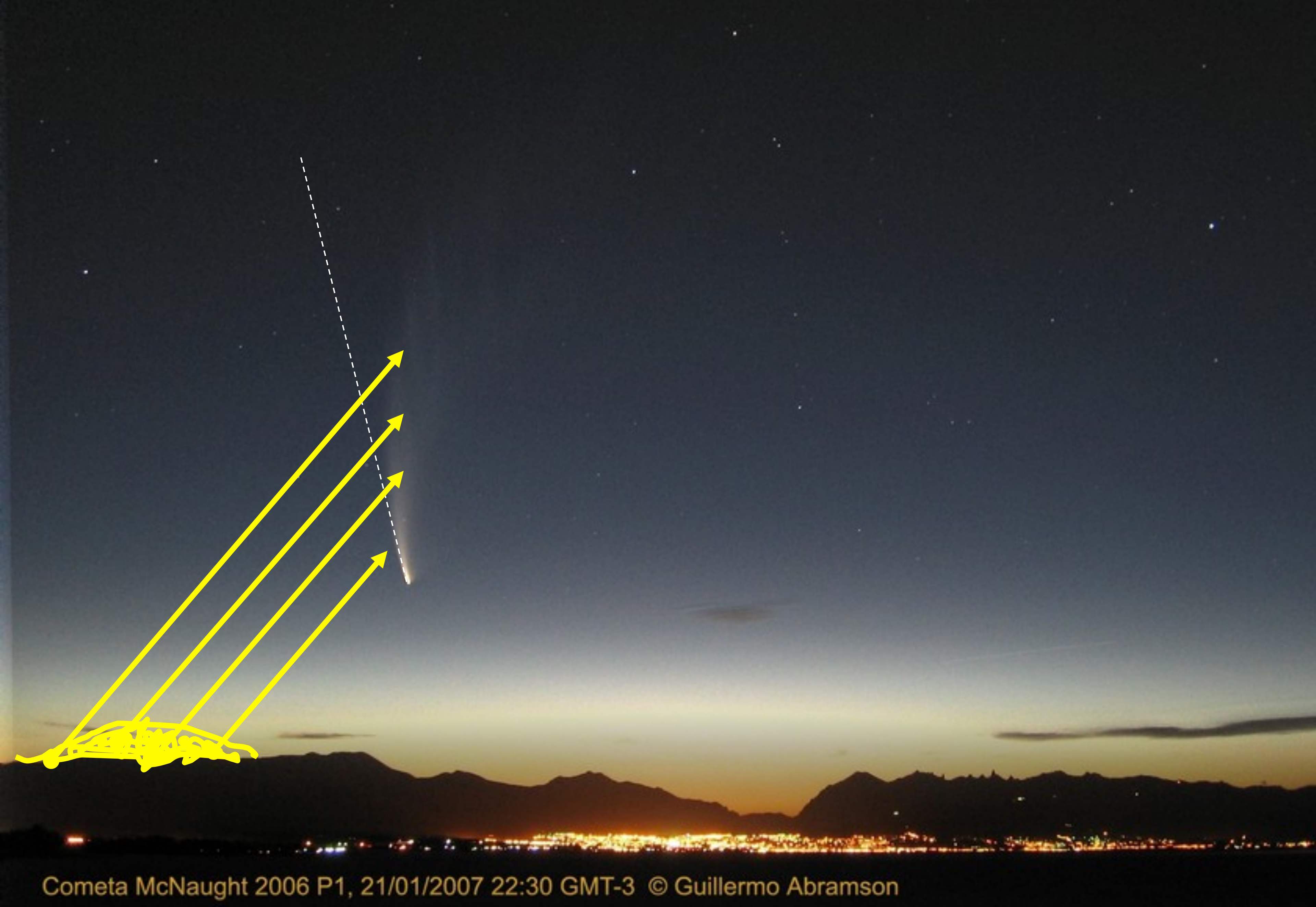
MAspelmeyer, TJK, FM, Rev. Mod. Phys. **86**, 1391 (2014)

Pero, cómo ejerce fuerza la luz sobre la materia?


Cometa McNaught 2006 P1, 21/01/2007 22:30 GMT-3 © Guillermo Abramson

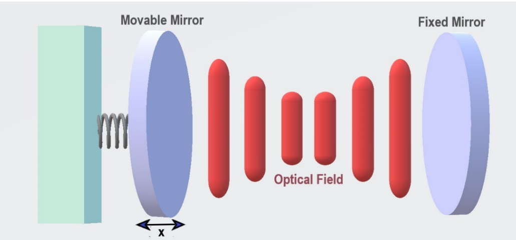

Cometa McNaught 2006 P1, 21/01/2007 22:30 GMT-3 © Guillermo Abramson

Presión de radiación



Y con esto, qué?

La auto-oscilación mecánica



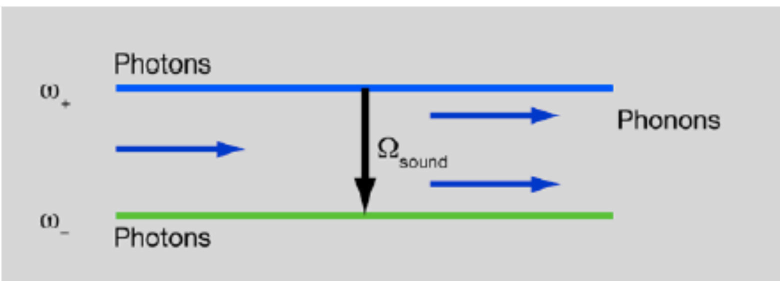
MA, TJK, FM, Rev. Mod. Phys. **86**, 1391 (2014)

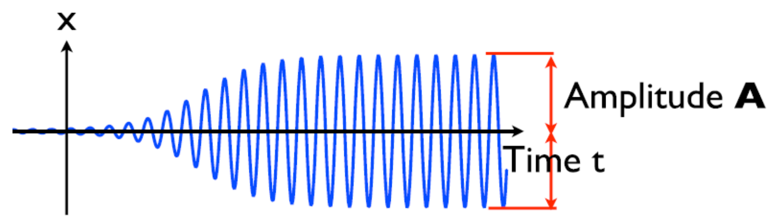
$$\hat{H}_0 = \hbar\omega_{\text{cav}}\hat{a}^\dagger\hat{a} + \hbar\Omega_m\hat{b}^\dagger\hat{b} \quad \hat{H}_{\text{int}} = -\hbar g_0\hat{a}^\dagger\hat{a}(\hat{b}+\hat{b}^\dagger)$$

$$\dot{\alpha} = -\frac{\kappa}{2}\alpha + i(\Delta + Gx)\alpha + \sqrt{\kappa_{ex}}\alpha_{\text{in}}$$

$$m_{\text{eff}}\ddot{x} = -m_{\text{eff}}\Omega_m^2 x - m_{\text{eff}}\Gamma_m\dot{x} + \hbar G\left|\alpha\right|^2$$

$$g_0 = Gx_{\text{ZPF}}$$





$$\Gamma_{\text{eff}} = \Gamma_m(1 - C)$$

Umbral para auto-oscilación:
"Cooperatividad optomecánica"
C=1

$$C = 4N\left|g_0^m\right|^2/(\kappa\,\Gamma_m)$$

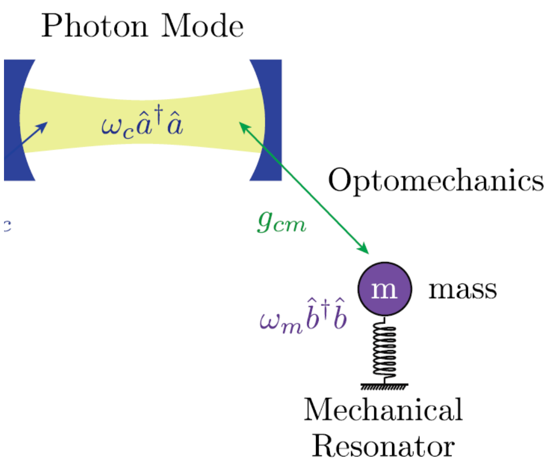
Número de fotones

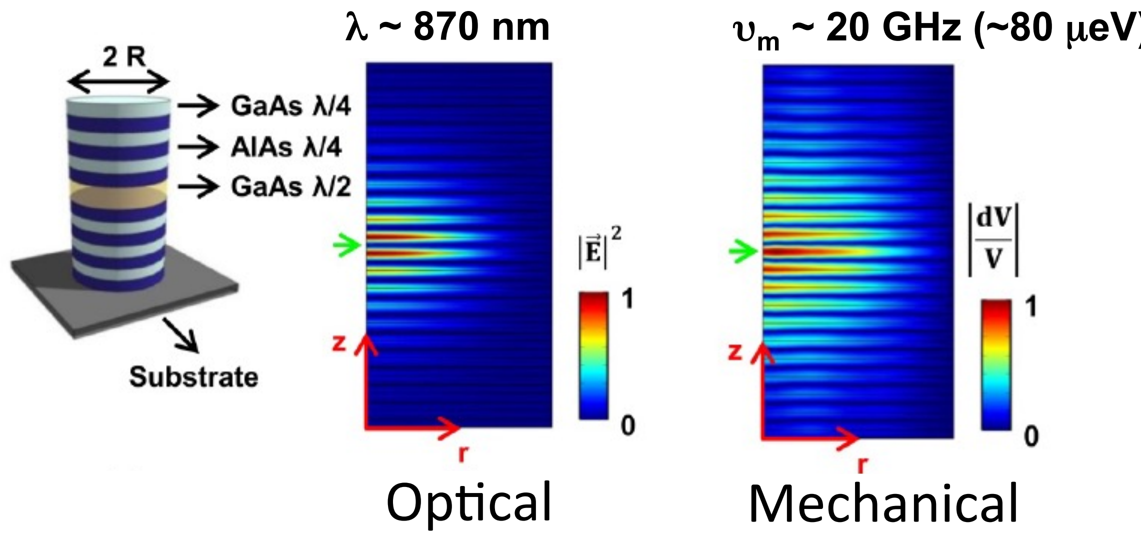
Acoplamiento

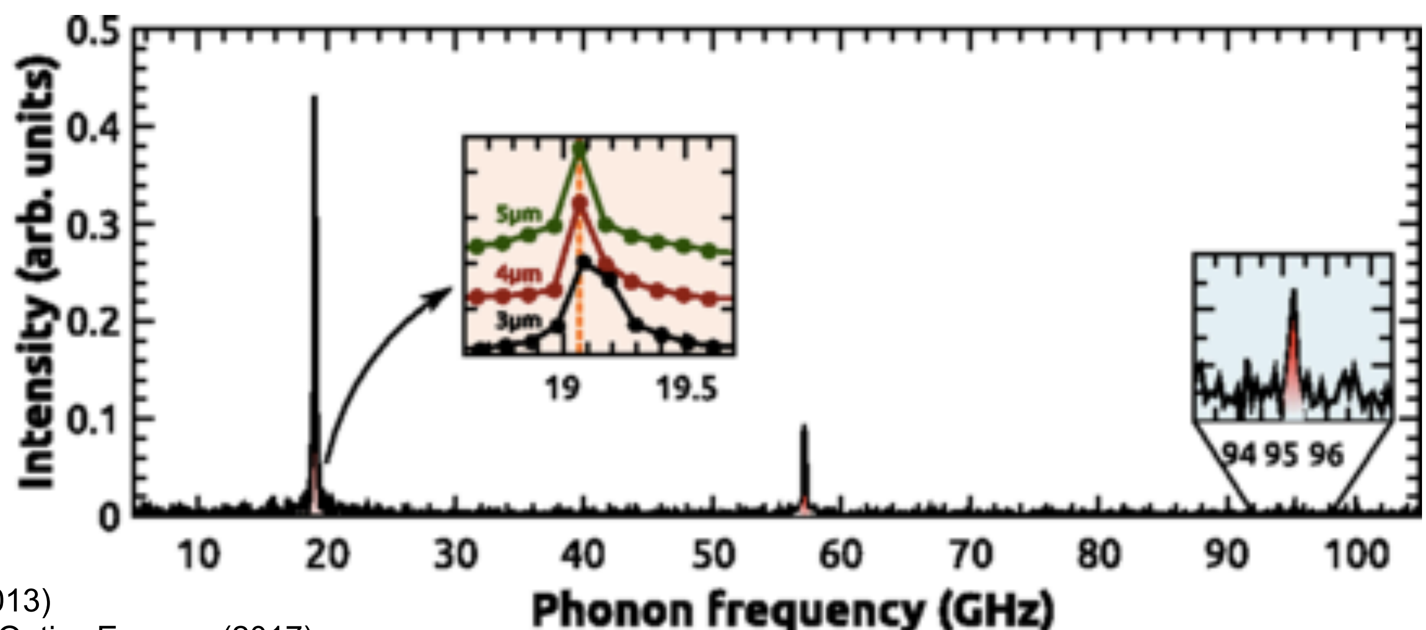
Decay rate del fotón

Decay rate del fonón

Cavidades semiconductoras optimizadas







AF *et al*, PRL (2013)
F. Lamberti *et al*, Optics Express (2017)
S. Anguiano *et al.*, PRL (2017)

Podemos hacer algo mejor?

$$C = 4N\,|g_0^m|^2/(\kappa\,\Gamma_m)$$

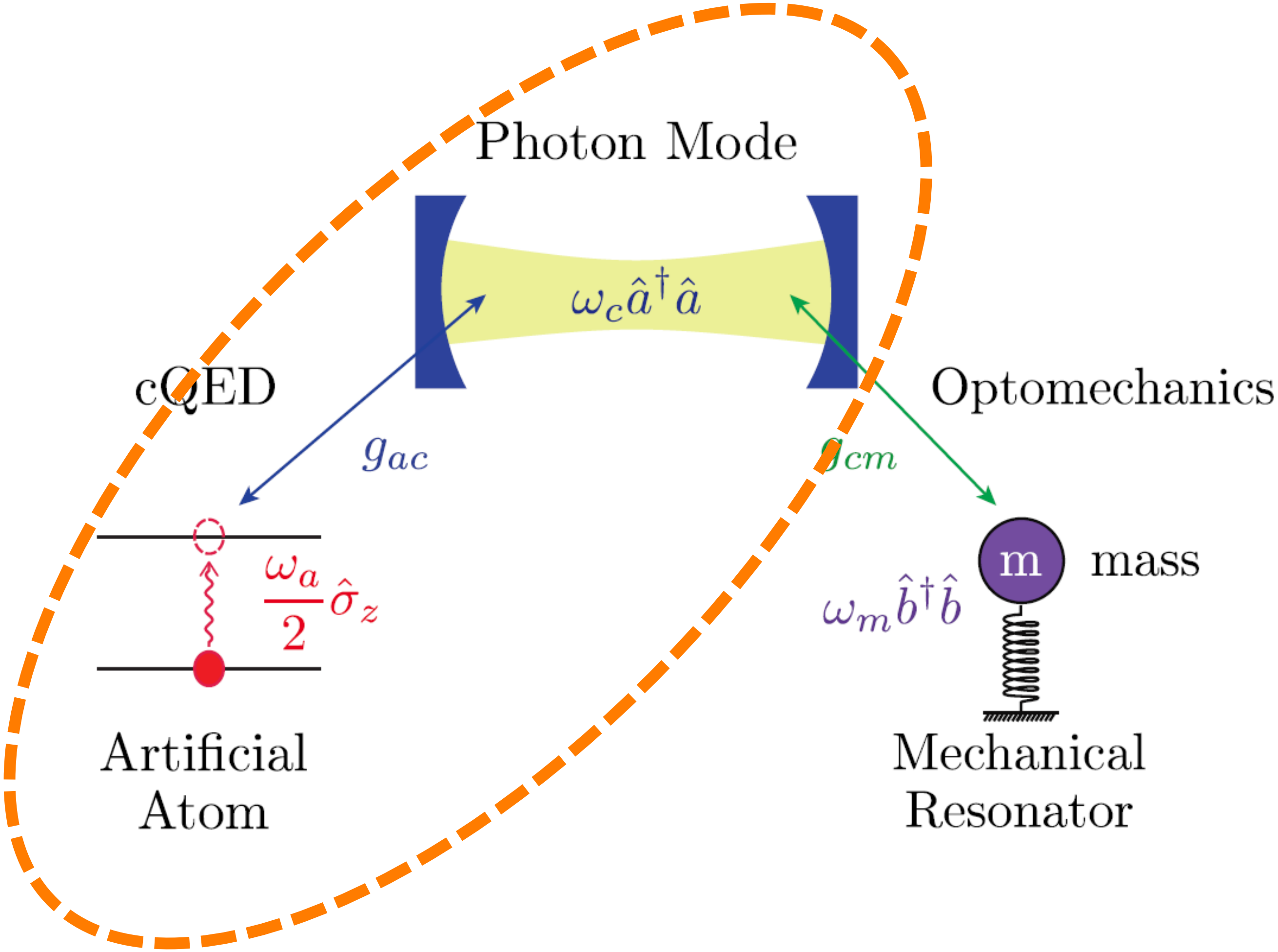
Número de fotones

Acoplamiento

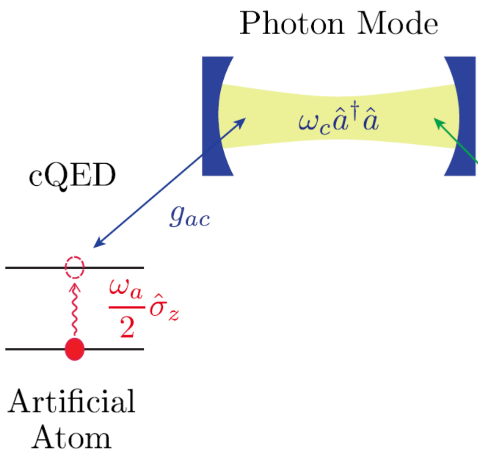
Decay rate del fonón

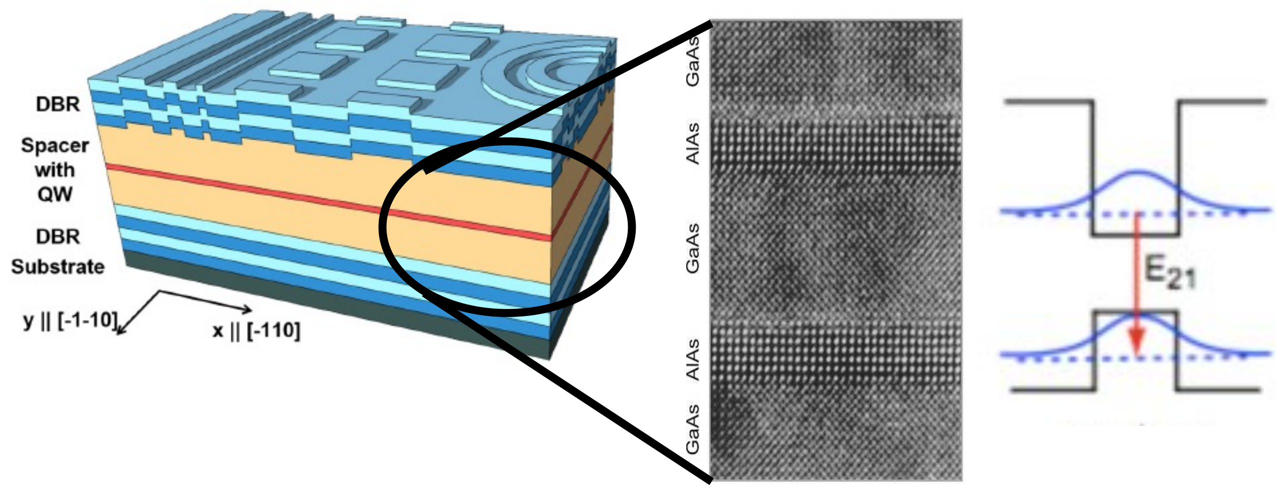
Decay rate del fotón

Conjugando **cQED** + cQOM



Resonadores híbridos: + sistema 2-niveles

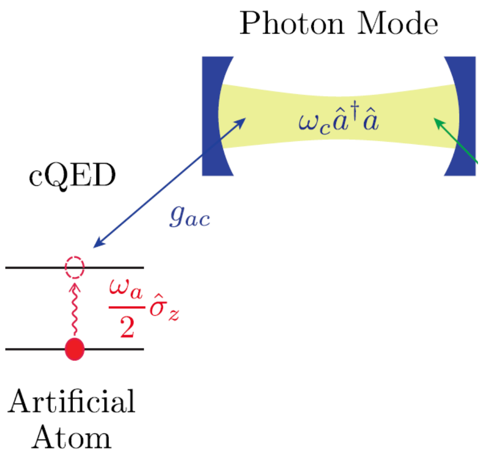


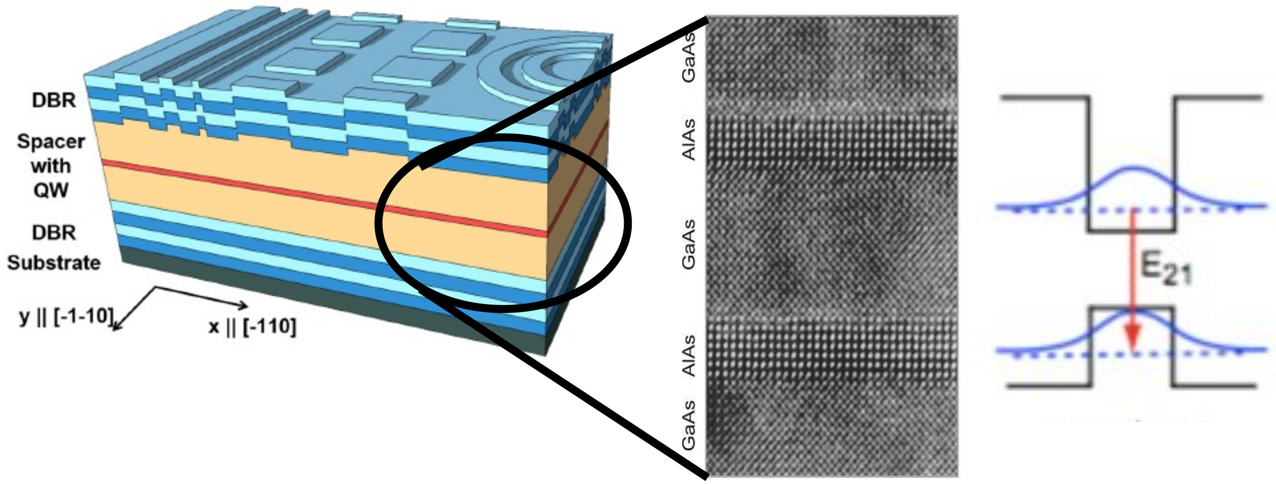


QWs

$$H_{AB} \simeq \hbar g_{\text{eff}}(a^+ b + b^+ a)$$

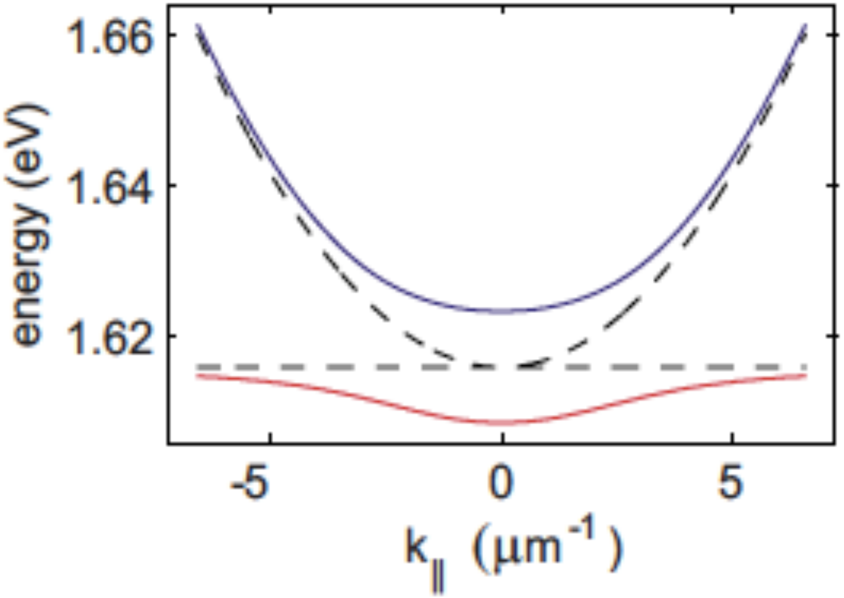
Resonadores híbridos: + sistema 2-niveles



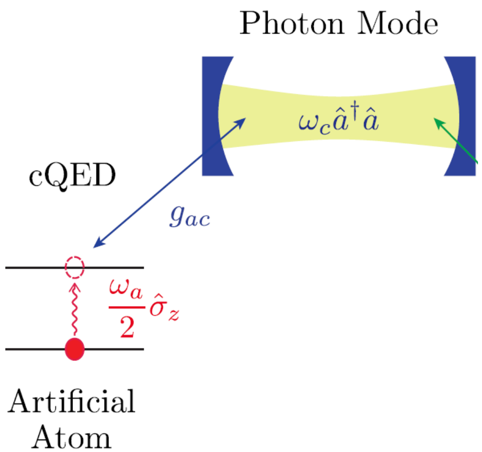


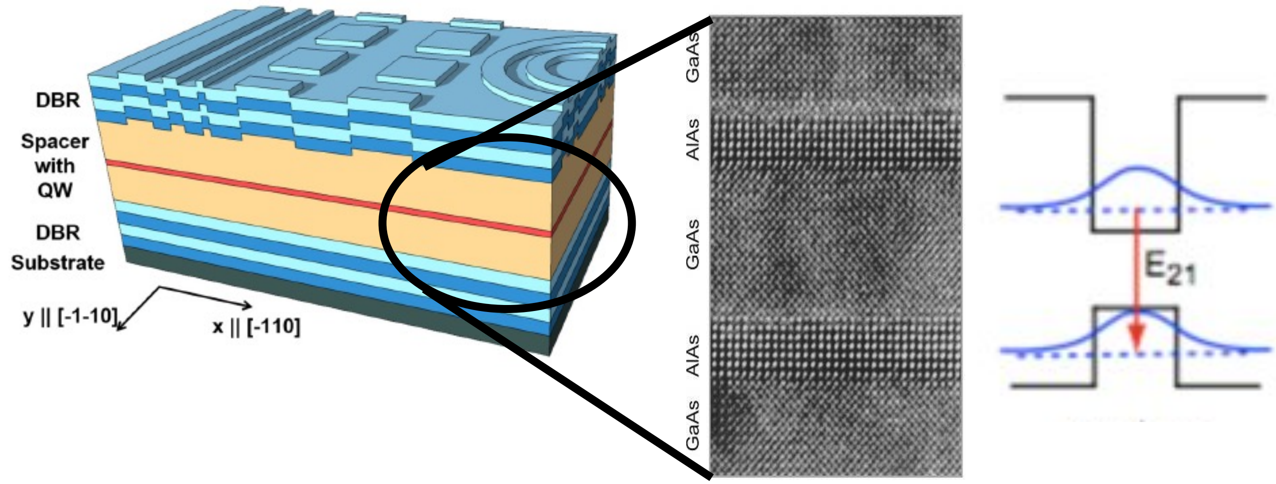
“Strong coupling”

QWs



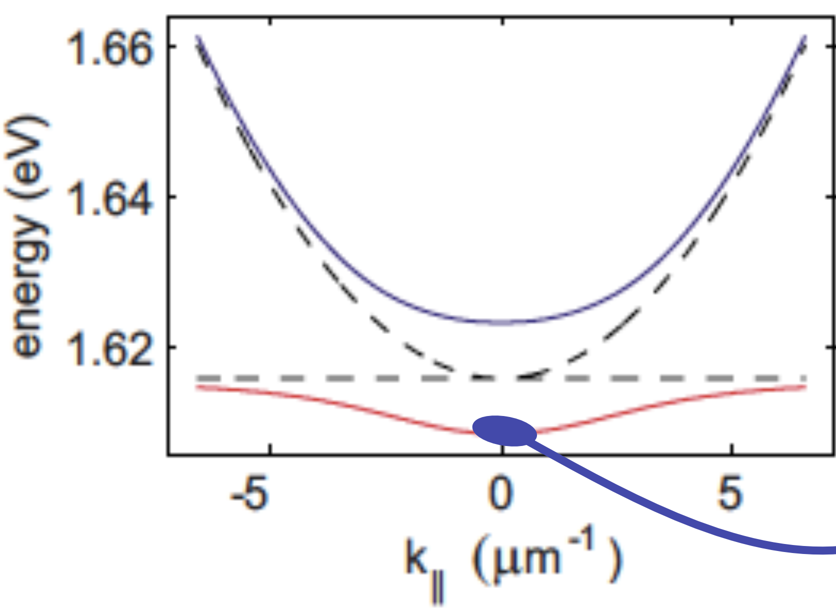
Resonadores híbridos: + sistema 2-niveles

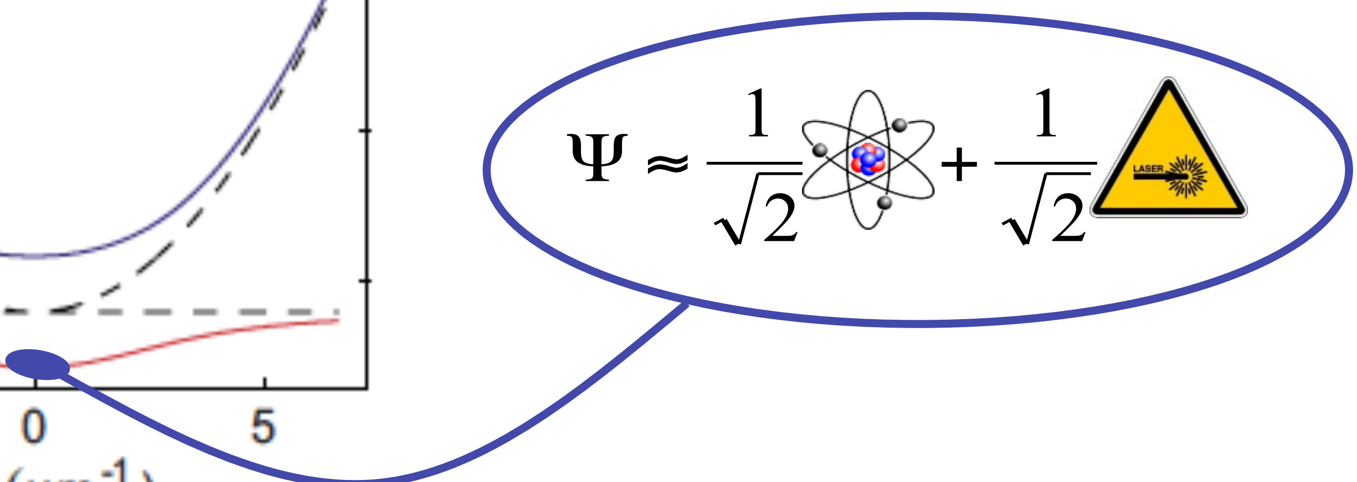




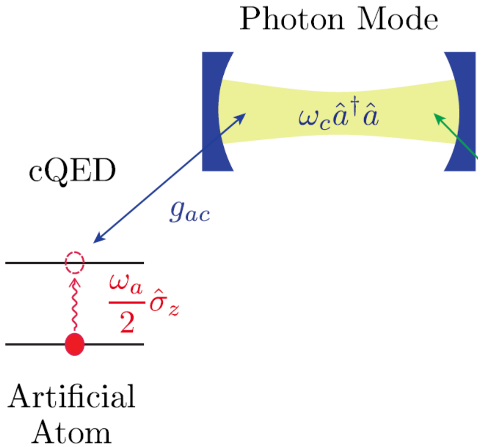
“Strong coupling”

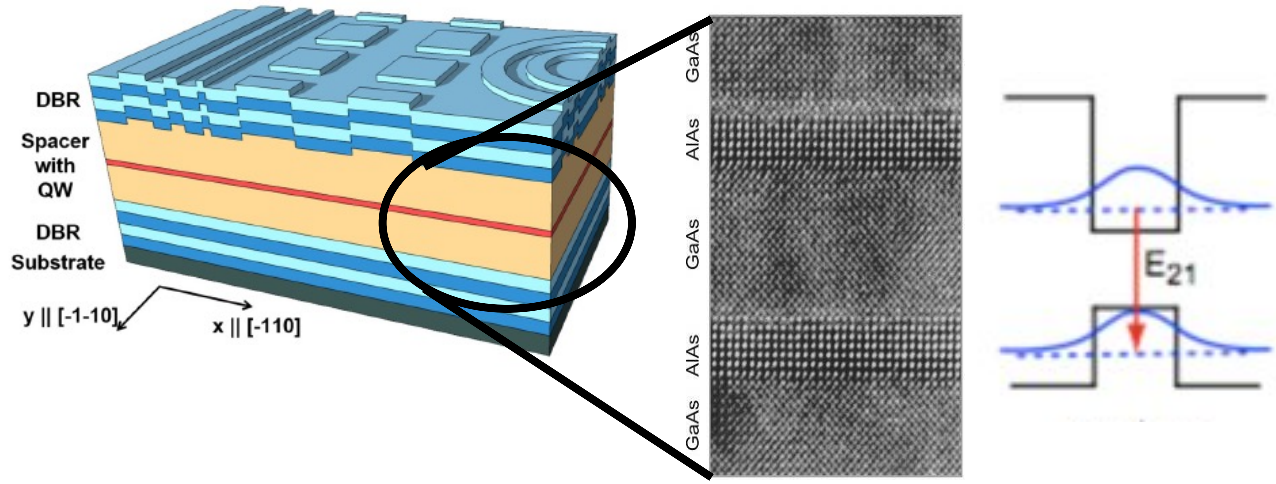
QWs



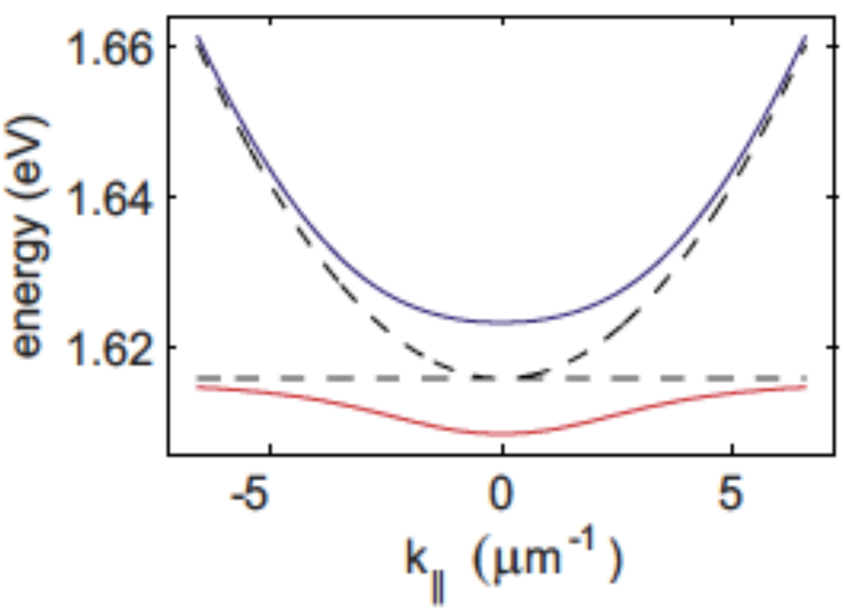


Resonadores híbridos: + sistema 2-niveles

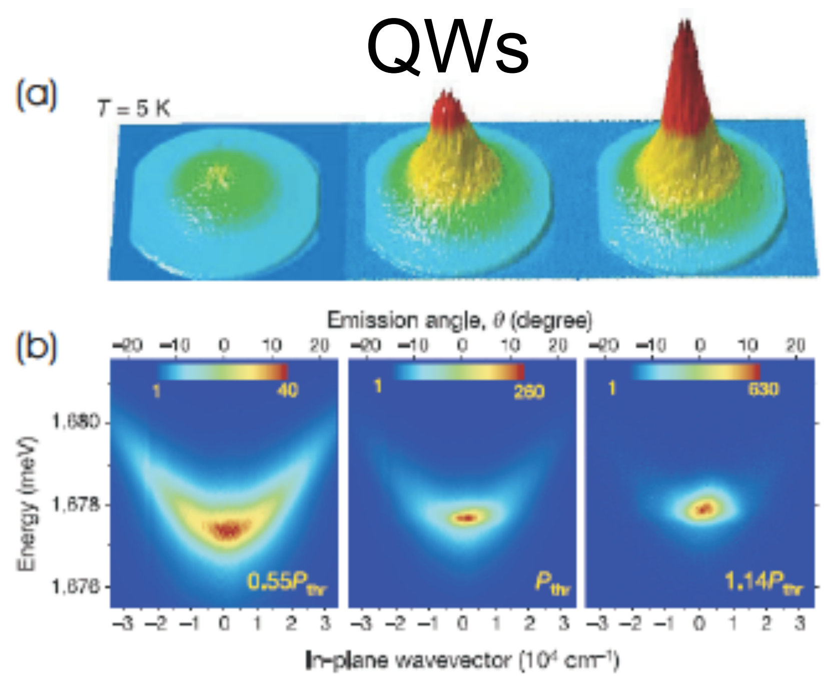




“Strong coupling”



QWs



J. Kasprzak et al., Nature (2006)

Y porqué un BEC de polaritones puede ser interesante?

Y porqué un BEC de polaritones puede ser interesante?

$$\Gamma_{\text{eff}} = \Gamma_m(1 - C)$$

Condición para auto-oscilación C=1

$$C = 4N|g_0^m|^2/(\kappa\,\Gamma_m)$$

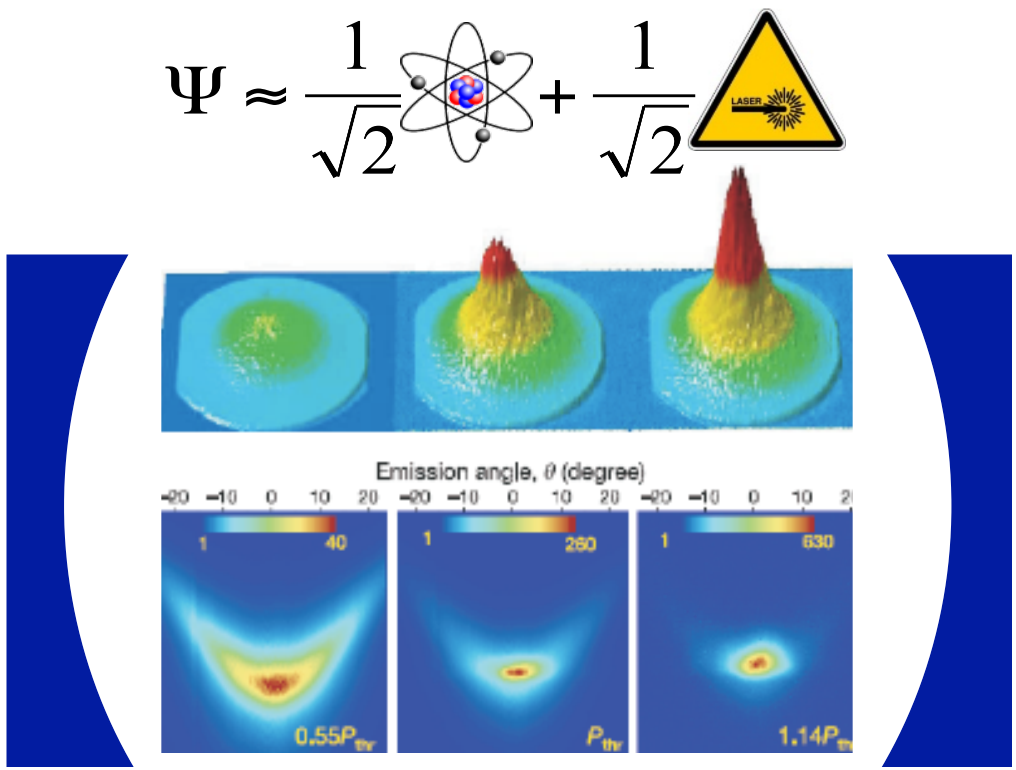
Número de **polaritones** confinados

Acoplamiento

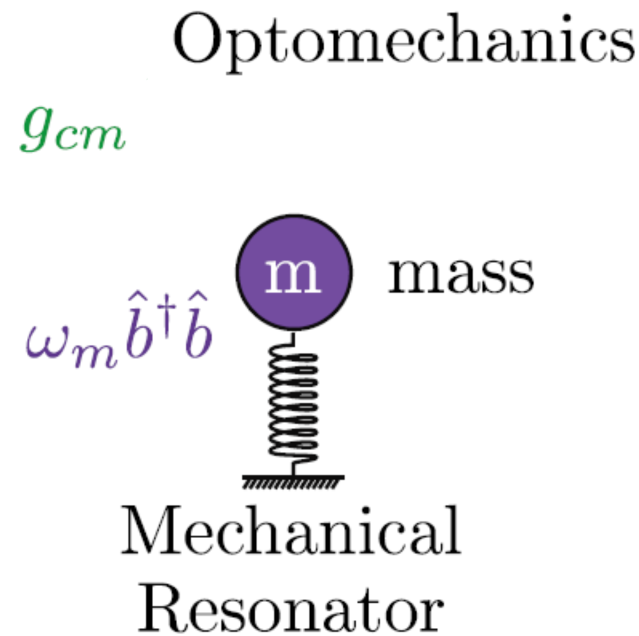
Decay rate de los **polaritones**

Decay rate del fonón

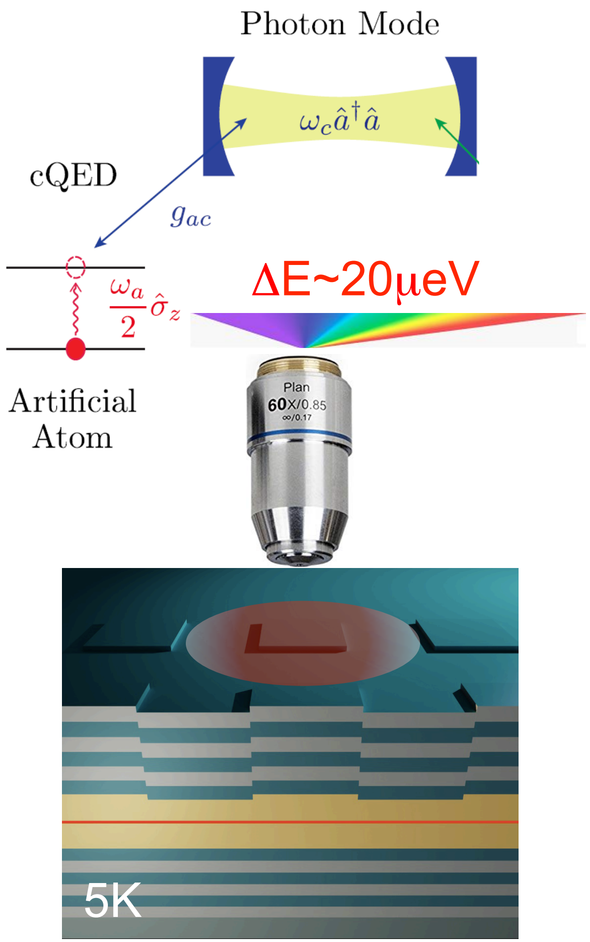
Polariton optomechanics

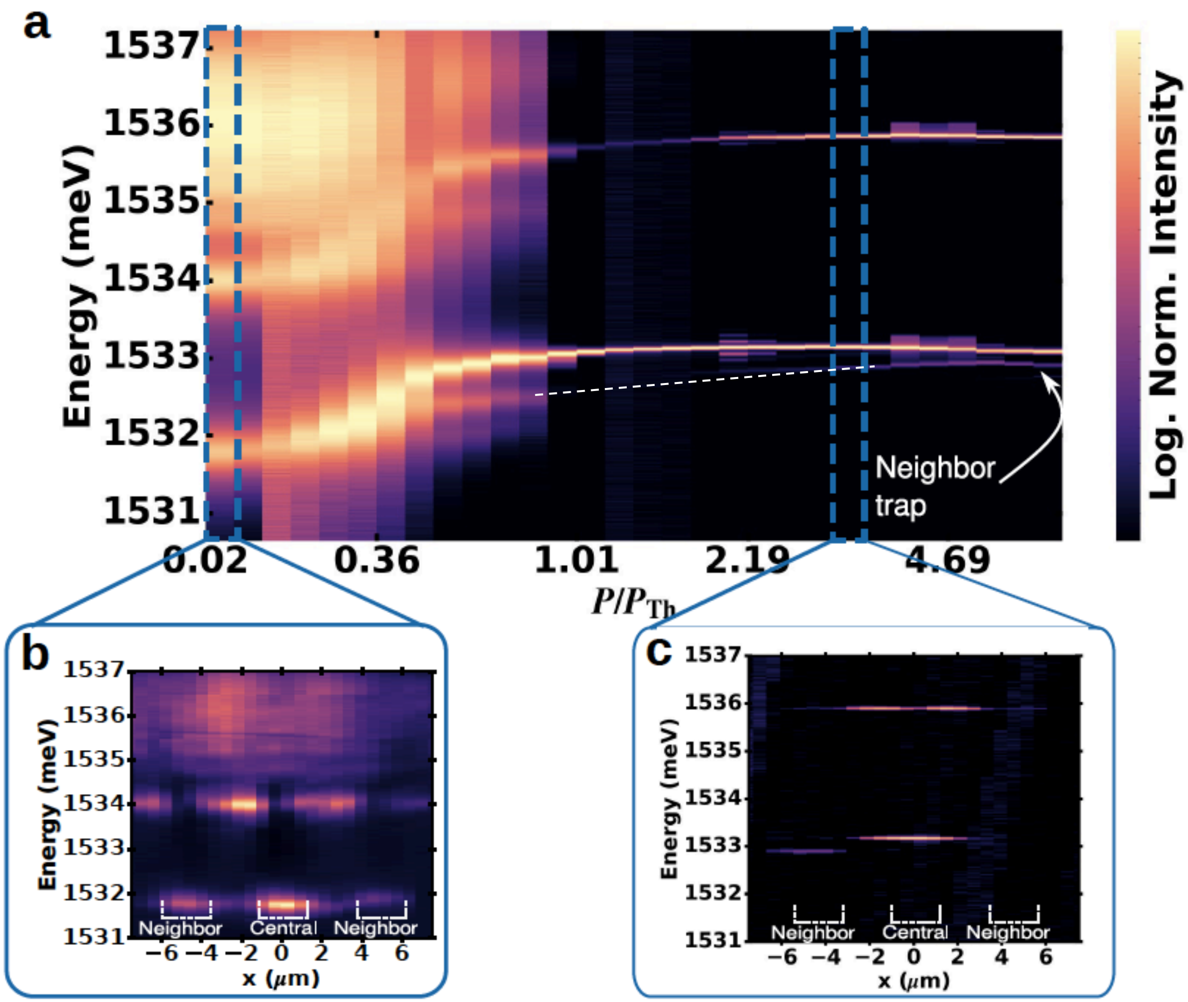


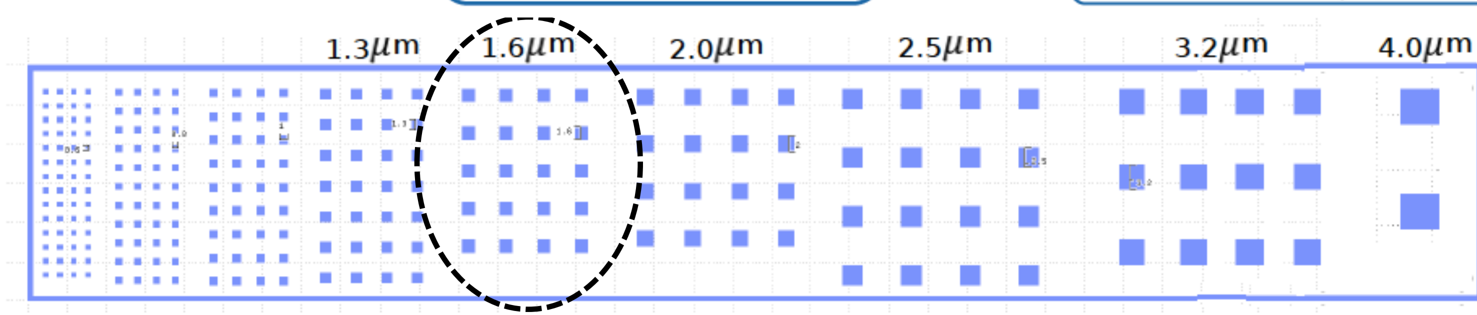
+



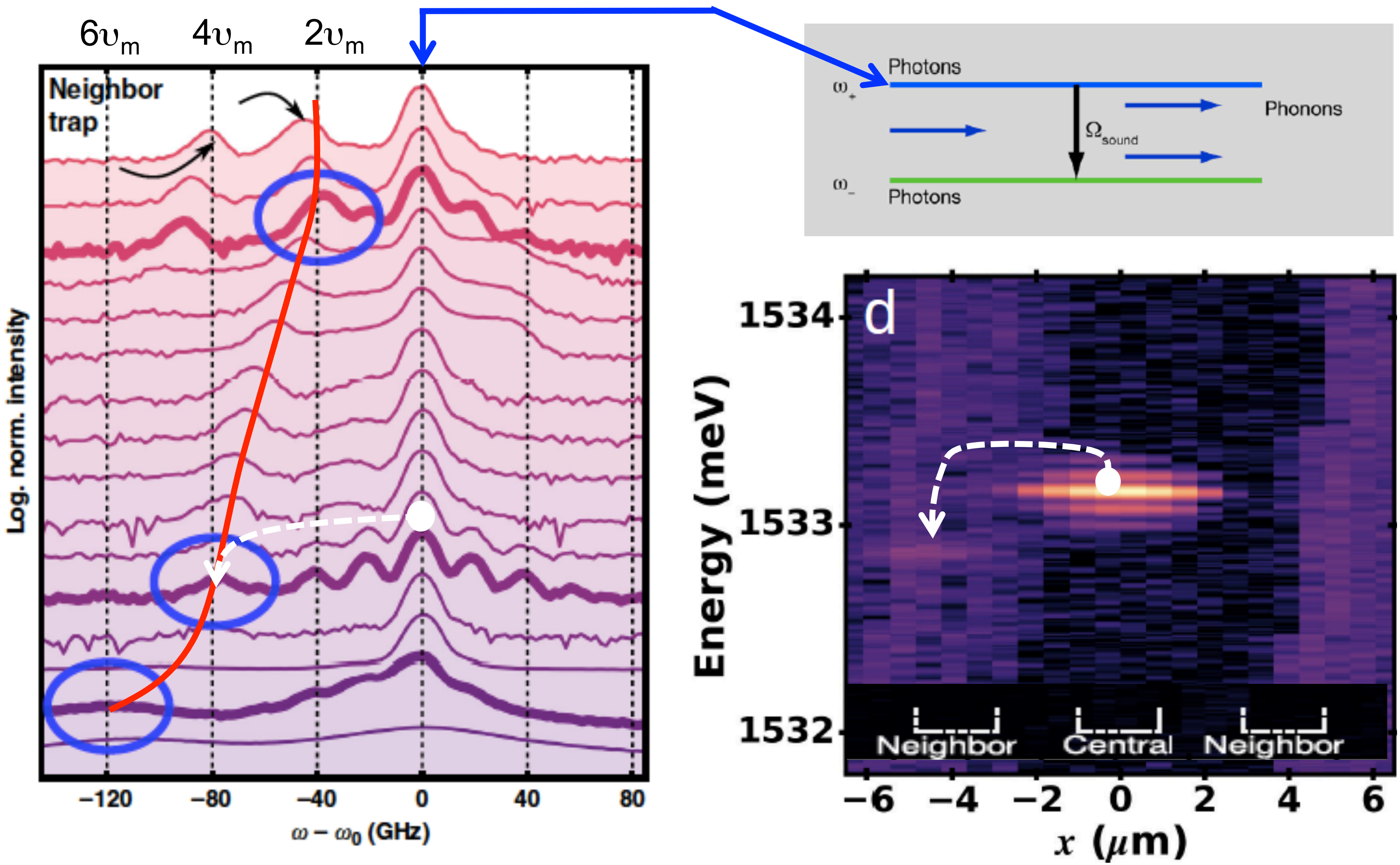
BECs en trampas de polaritones





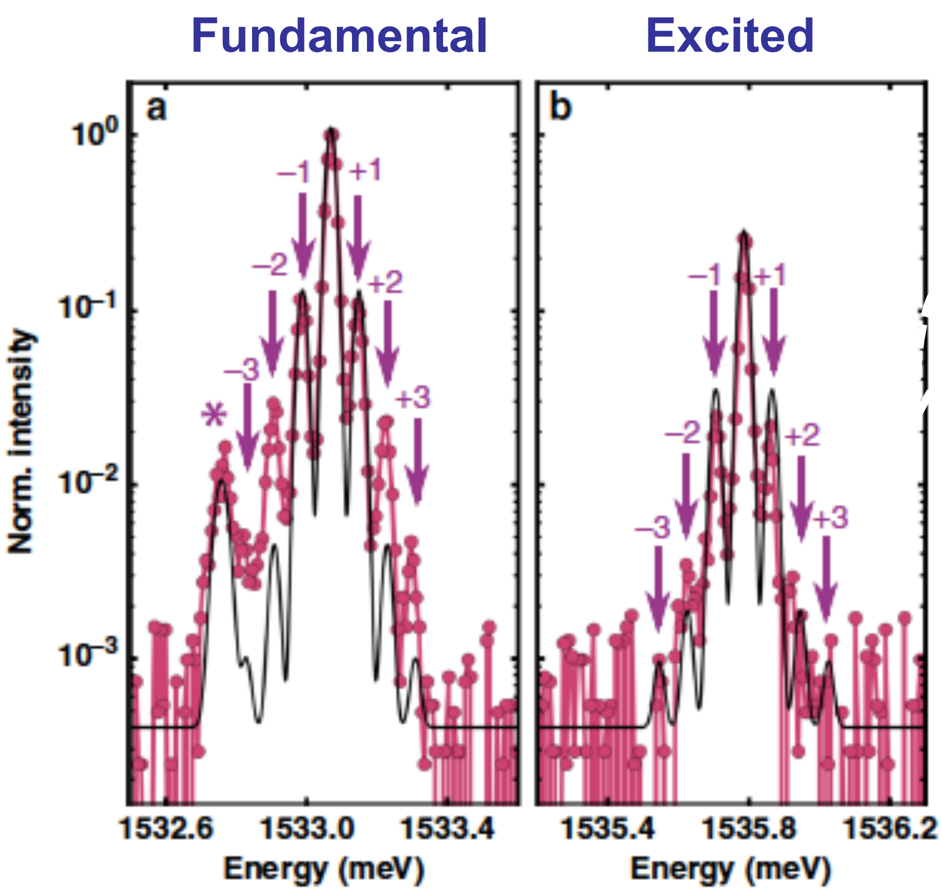


Acoplamiento entre trampas: SASER



Retroacción dinámica: BEC modulado mecánicamente

$$P[\omega] = \sum_{n=-\infty}^{\infty} \frac{J_n^2(\chi)}{\gamma^2 + [\omega - (\omega_0 - n\omega_d)]^2} \qquad \chi = \Delta\omega_{BEC}/\omega_d$$

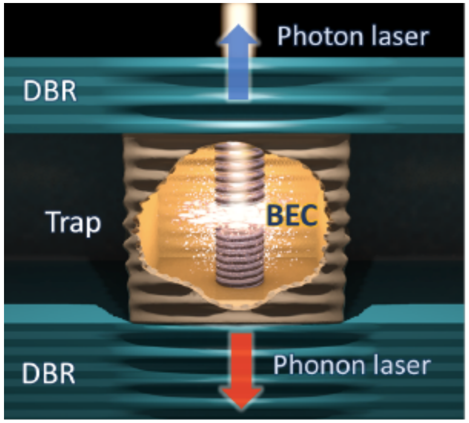


Bandas laterales

$\chi = 0.65$

$\langle N \rangle \sim 2 \times 10^5$

$\langle N \rangle_{Thermal} \sim 5$



That's All Folks!