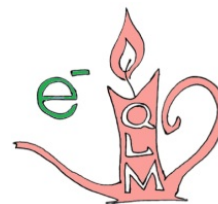


Introduction to the ultra-strong coupling regime

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Southampton



**QUANTUM
LIGHT &
MATTER**

Ultrastrong elevator pitch

From the weak to the ultrastrong

Experimental results

Open quantum systems

Ultrastrong phenomenology

Ultrastrong elevator pitch

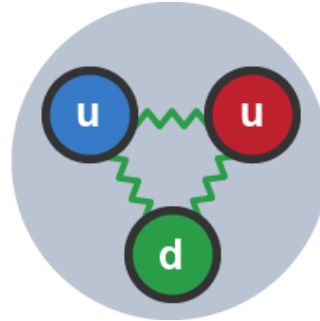
Fundamental interactions

Strong interaction

Mass of up quark: 2.3 MeV

Mass of down quark: 4.8 MeV

Mass of a proton: 938 MeV



99% of proton mass
is due to interaction

(virtual quark-gluon plasma)

Electromagnetic interaction

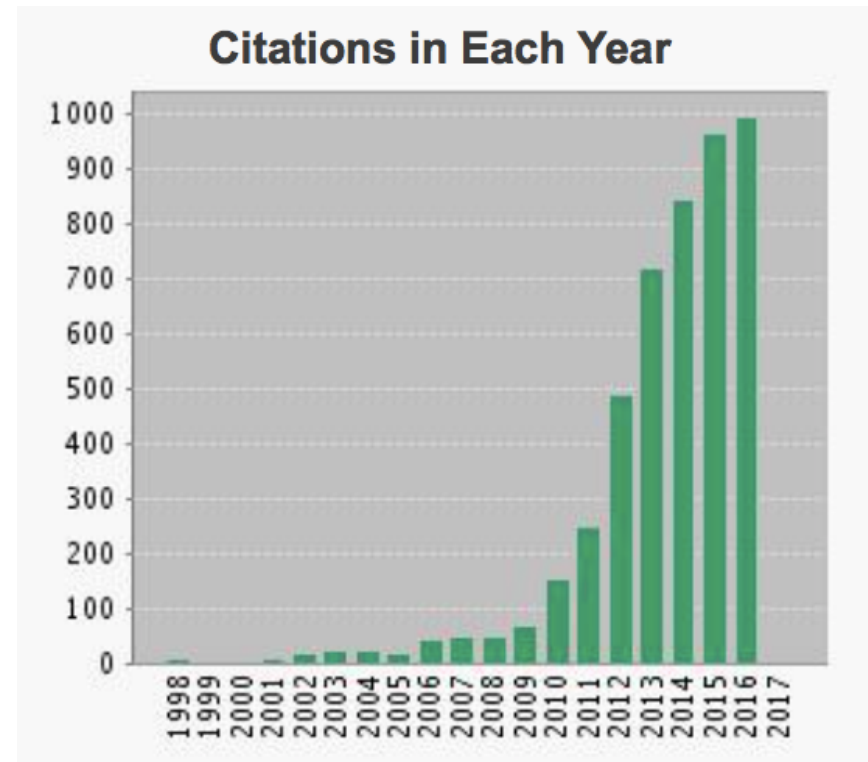
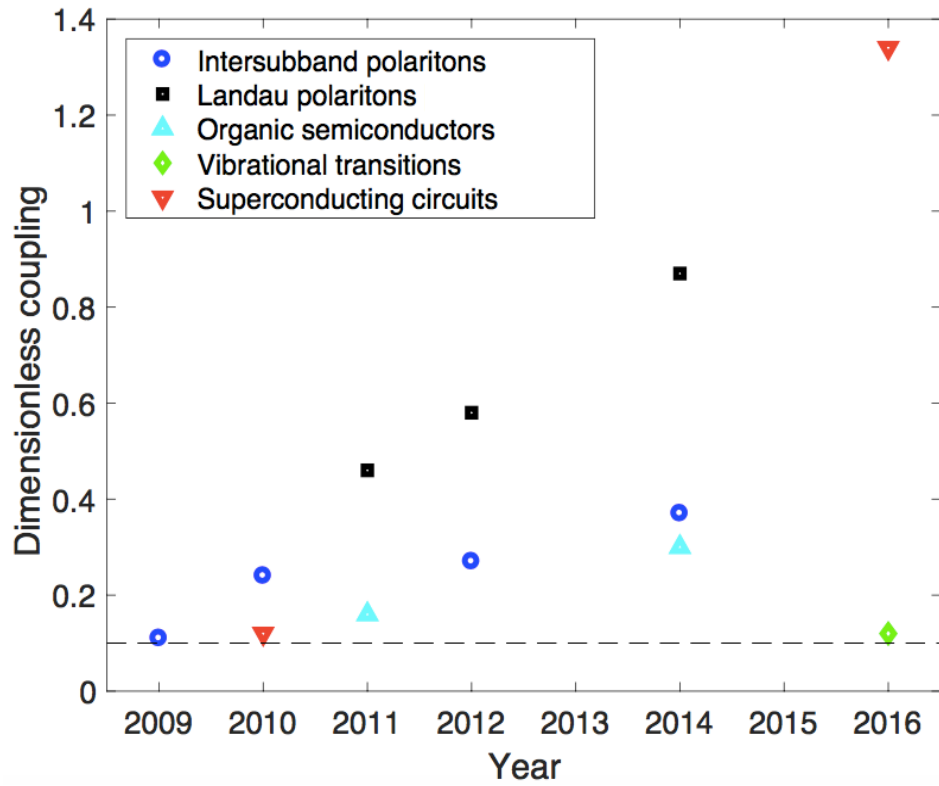
In light-matter interaction the dimensionless coupling constant is $\alpha \simeq \frac{1}{137}$

Low order perturbation theory works well (photon absorption and emission)

The interaction strength Ω_R is much smaller than the bare frequency ω_0

Ultrastrong coupling

Ultrastrong light-matter coupling regime: Ω_R/ω_0 non negligible



Ultrastrong coupling

Ground state contains virtual photons:

- Quantum phase transitions
- Quantum vacuum radiation
- Topologically protected ground states
- Increase in electrical conductivity
- Modified electroluminescent properties
- Change in chemical properties
- Change in structural molecular properties
- Modified lasing
- Vacuum nonlinear processes
- ...

From the weak to the ultrastrong

The single atom Hamiltonian

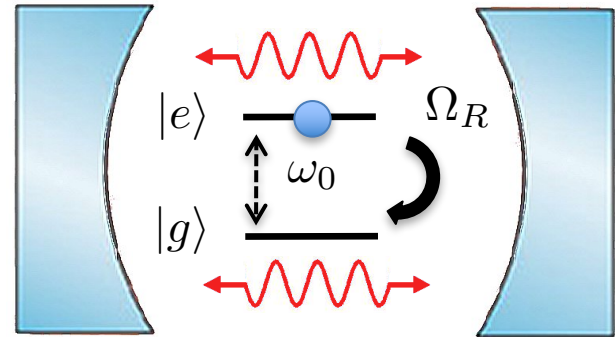
$$H = H_{\text{field}} + \frac{(\mathbf{p} - e\mathbf{A}(\mathbf{r}))^2}{2m} + V(\mathbf{r})$$



$$H = H_{\text{field}} + \frac{\mathbf{p}^2}{2m} + V(\mathbf{r}) - \frac{e\mathbf{p}\mathbf{A}(\mathbf{r})}{m} + \frac{e^2\mathbf{A}(\mathbf{r})^2}{2m}$$



$$H = \underbrace{\omega_c a^\dagger a + \omega_0 |e\rangle\langle e|}_{H_0} + \underbrace{\Omega_R (a^\dagger + a) (|e\rangle\langle g| + |g\rangle\langle e|) + \frac{\Omega_R^2}{\omega_0} (a^\dagger + a)^2}_{H_{\text{int}}}$$



Rotating wave approximation

Resonant terms

Connect states whose energy difference is $\simeq 0$

$$H_{\text{int}} = \hbar\Omega_R \underbrace{(a + a^\dagger)}_{\text{red}} \underbrace{(|e\rangle\langle g| + |g\rangle\langle e|)}_{\text{blue}} + \frac{\hbar\Omega_R^2}{\omega_0} \underbrace{(a + a^\dagger)}_{\text{red}} \underbrace{(a + a^\dagger)}_{\text{blue}}$$

Antiresonant terms

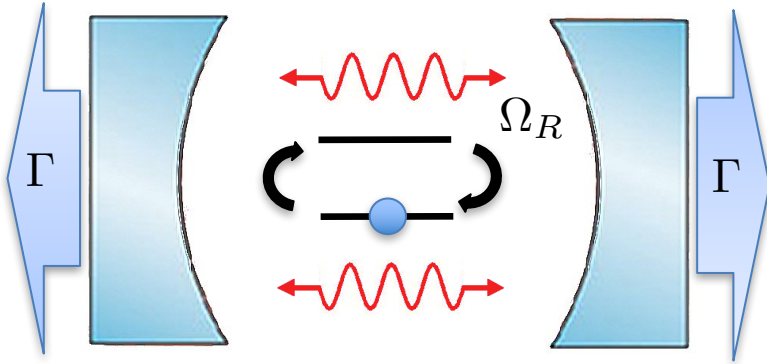
Connect states whose energy difference is $\simeq 2\omega_0$

Fermi golden rule $\Gamma = \frac{2\pi}{\hbar} \sum_f |\langle i|H_{\text{int}}|f\rangle|^2 \delta(\hbar\omega_i - \hbar\omega_f)$

The simpler RWA Hamiltonian gives the same results within first order perturbation

$$H_{\text{int}}^{\text{RWA}} = \hbar\Omega_R \underbrace{(a|e\rangle\langle g| + a^\dagger|g\rangle\langle e|)}_{\substack{\text{Absorption} \\ \downarrow}} + \frac{2\hbar\Omega_R^2}{\omega_0} \underbrace{a^\dagger a}_{\substack{\text{Photon renormalisation (second order)} \\ \downarrow}} \\ \uparrow \\ \text{Emission}$$

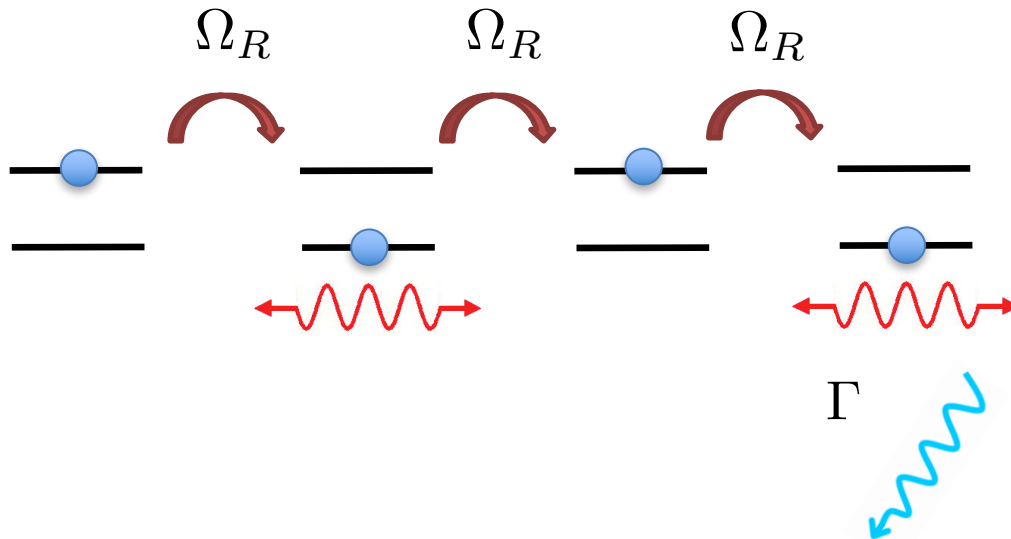
Strong coupling (time domain)



Fermi golden rule: first order perturbation.

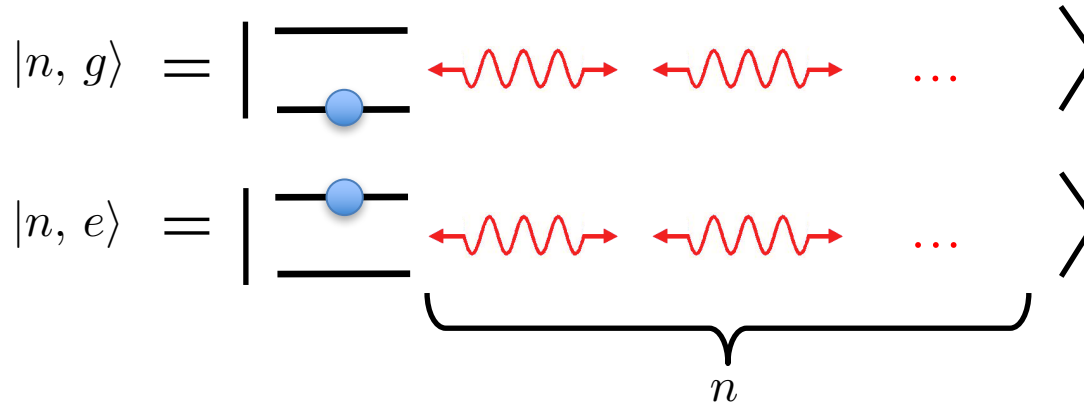
It cannot account for higher order processes, *i.e.* reabsorption. Valid if $\Omega_R < \Gamma$

If $\Omega_R > \Gamma$ the emitted photons is trapped long enough to be reabsorbed



Jaynes-Cummings model

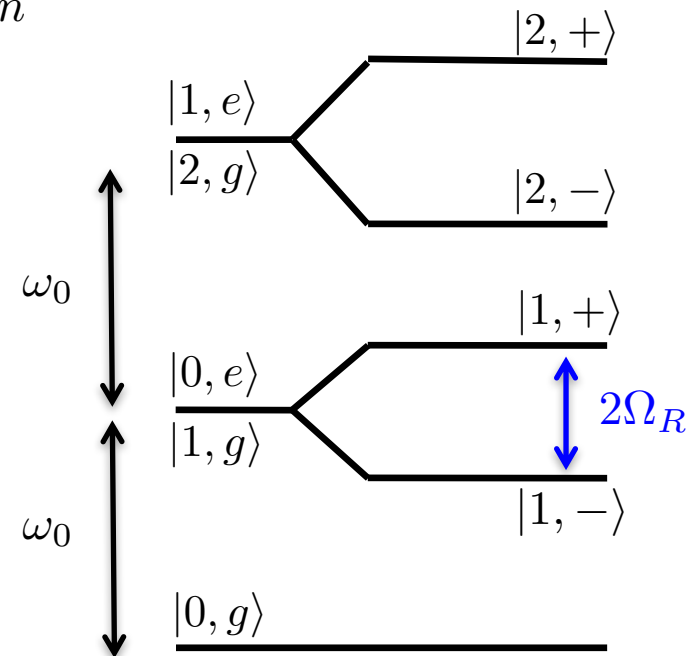
$$H_{\text{JC}} = \hbar\omega_0 a^\dagger a + \hbar\omega_0 |e\rangle\langle e| + \hbar\Omega_R (a|e\rangle\langle g| + a^\dagger|g\rangle\langle e|)$$



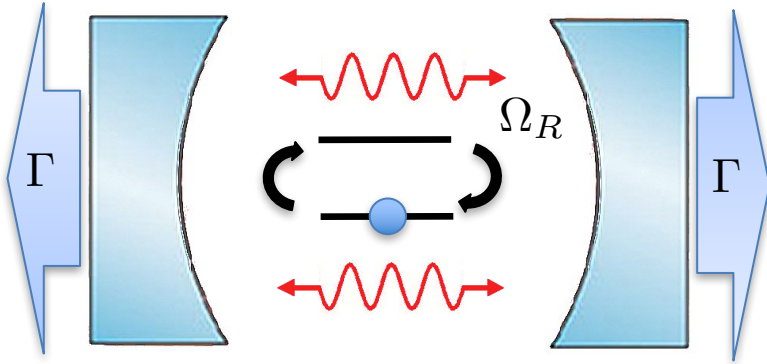
$|n, g\rangle$ and $|n - 1, e\rangle$ form a closed subspace:

$$H_{\text{JC}}^n = \hbar \begin{bmatrix} \omega_0 & \sqrt{n}\Omega_R \\ \sqrt{n}\Omega_R & \omega_0 \end{bmatrix}$$

whose eigenvalues are $|n, -\rangle$ and $|n, +\rangle$,
 split at resonance of $2\sqrt{n}\hbar\Omega_R$
 First order perturbation is exact!



Strong coupling (frequency domain)



The coupling splits the degenerate levels, creating the Jaynes-Cummings ladder.

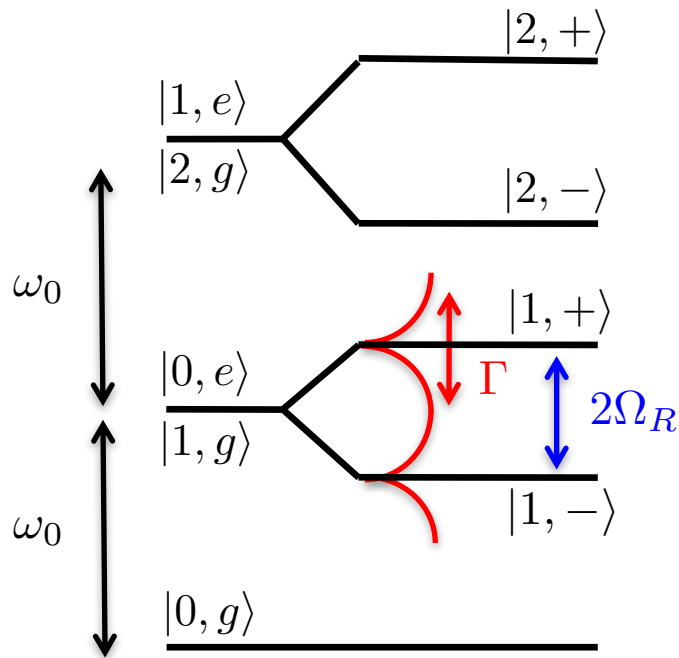
The losses give the resonances a finite width

Strong coupling: $\Omega_R > \Gamma$

Condition to spectroscopically resolve the resonant splitting.

In the strong coupling regime we cannot consider transitions between uncoupled modes, *e.g.*, $|0, e\rangle \longrightarrow |1, g\rangle$.

We are obliged to consider the dressed states, $|1, -\rangle$, $|1, +\rangle$, etc...



Perturbation theory

Let us do perturbation using the full Hamiltonian

$$H_{\text{QRM}} = \underbrace{\hbar\omega_0 a^\dagger a + \hbar\omega_0 |e\rangle\langle e|}_{H_0} + \underbrace{\hbar\Omega_R (a + a^\dagger)(|e\rangle\langle g| + |g\rangle\langle e|)}_{H_{\text{int}}}$$

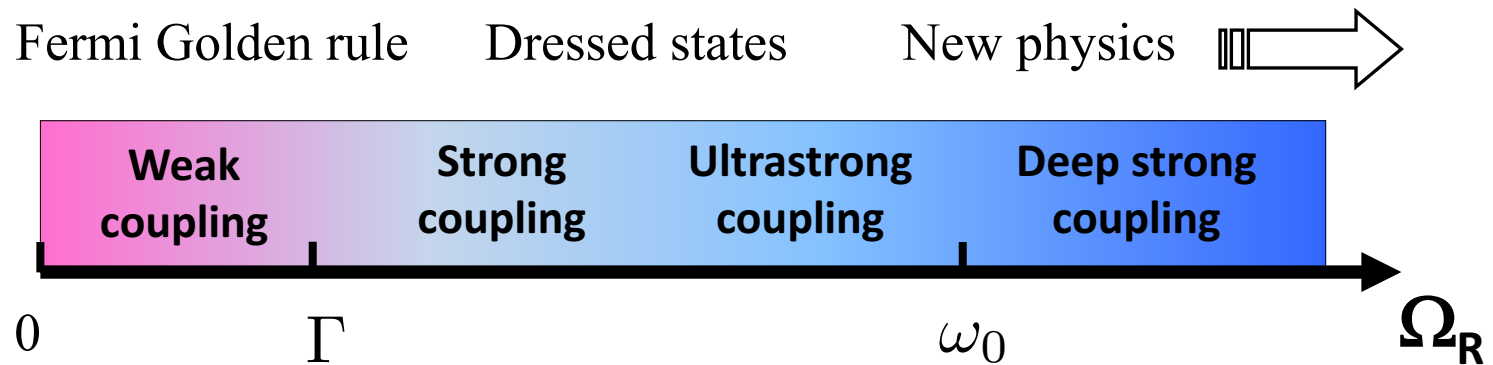
First order perturbation: $\Delta E_\phi^{(1)} \propto \Omega_R$

Second order perturbation: $\Delta E_\phi^{(2)} = \sum_{|\psi\rangle \neq |\phi\rangle} \frac{|\langle\phi|H_{\text{int}}|\psi\rangle|^2}{E_\phi - E_\psi} \propto \frac{\Omega_R^2}{\omega_0} = \Omega_R \times \frac{\Omega_R}{\omega_0}$

1. The second order contribution is due to antiresonant terms
2. It becomes non negligible when $\frac{\Omega_R}{\omega_0}$ is non negligible

Ultrastrong coupling regime

Coupling regimes



Is ultrastrong coupling possible?

Hydrogen atom

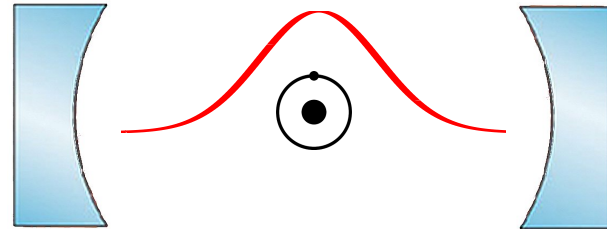
$$E_n = -\frac{Ry}{n^2}$$

Wavelength

$$\lambda = \frac{2\pi c}{\omega_0}$$

Dimensionless volume

$$\tilde{V} = \frac{V}{(\lambda/2)^3}$$



We end up with

$$\frac{\Omega_R}{\omega_0} = \frac{\alpha^{3/2}}{n\pi\sqrt{\tilde{V}}}$$

← Coupling

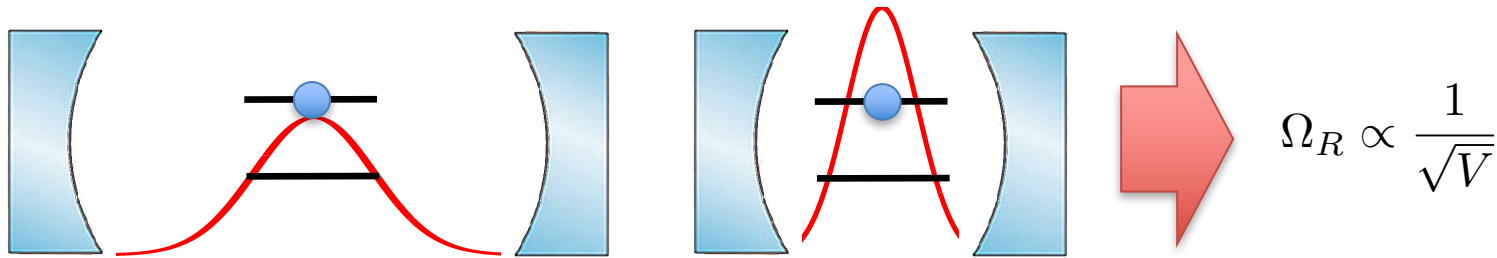
← Overlap

Three ways to ultrastrong coupling

- Reducing \tilde{V}
- Increasing the number of dipoles
- Coupling to currents ($\alpha^{-1/2}$)

Reducing the mode volume

Mode confinement: smaller cavity = larger coupling



The field is an harmonic oscillator

$$H_{\text{field}} = \frac{\epsilon E^2}{2} + \frac{\mu H^2}{2} = u_E + u_M$$

Consider an electromagnetic mode

$$\sin\left(\frac{2\pi}{l}x - \omega_0 t\right) = \sin\left(\frac{2\pi}{l}x - \frac{2\pi}{\lambda}ct\right)$$

Maxwell equation $\nabla \times \mathbf{H} = \epsilon \frac{\partial}{\partial t} \mathbf{E}$ leads to $u_M = \frac{l^2}{\lambda^2} u_E$

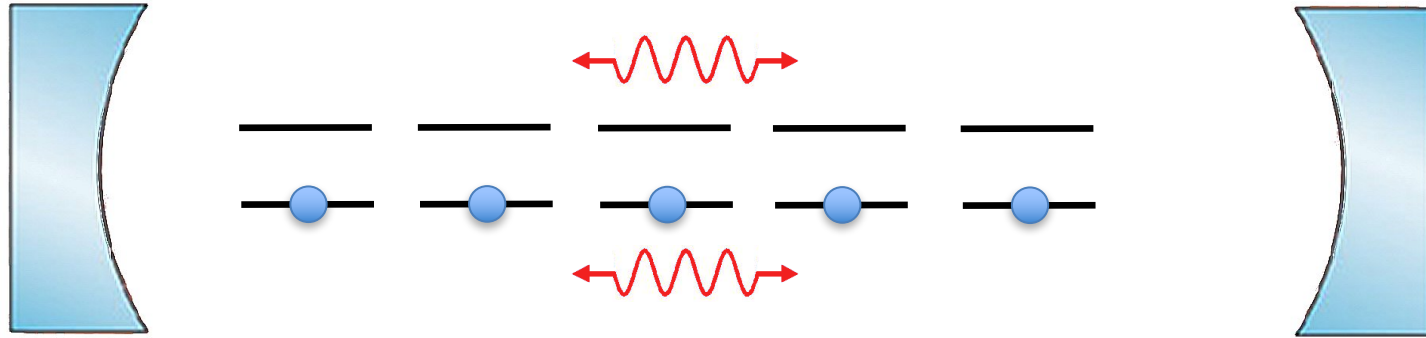
Solution: store energy as kinetic energy $u_E = u_M + u_K$ (plasmons, phonon polaritons)

Sub-wavelength confinement is lossy!

J. Khurgin, Nat. Nanotech. 10, 2 (2015)

Increasing the number of dipoles

$N \gg 1$ two level systems in a cavity



$$H_{\text{Dicke}} = \hbar\omega_0 a^\dagger a + \sum_{j=1}^N \hbar\omega_0 |e_j\rangle\langle e_j| + \sum_{j=1}^N \hbar\Omega_R (a + a^\dagger) (|e_j\rangle\langle g_j| + |g_j\rangle\langle e_j|)$$

Coherent operators: $b = \frac{1}{\sqrt{N}} \sum_{j=1}^N |g_j\rangle\langle e_j|$ State with n systems in the excited state

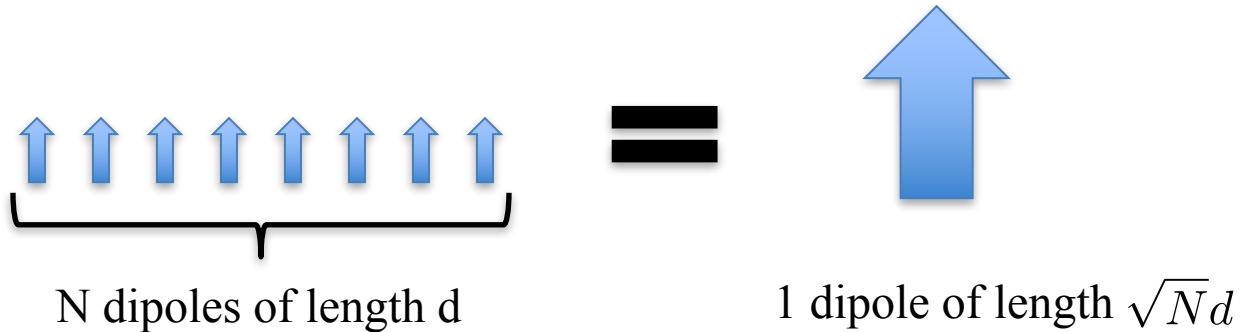
Bosons in the limit $N \gg n$: $\langle n|[b, b^\dagger]|n\rangle = 1 - \frac{2n}{N}$

In the one excitation subspace $|g_j\rangle = |g\rangle$

Enhanced coupling

$$H_{\text{Dicke}} = \hbar\omega_0 a^\dagger a + \hbar\omega_0 b^\dagger b + \hbar\Omega_R \sqrt{N} (a + a^\dagger) (b + b^\dagger)$$

Collective coupling



Superradiance: more dipoles = larger coupling

Formal procedure: Holstein-Primakoff transformation *Phys. Rev.* **58**, 1098, (1940)

Partition function: $Z = (1 + e^{-\beta\omega_0})^N = \sum_{m=0}^N \binom{N}{m} e^{-m\beta\omega_0}$

If they are *indistinguishable* instead:

Partition function
of a bosonic field

$$Z = \sum_{m=0}^N \binom{N}{m} e^{-m\beta\omega_0} = \sum_{m=0}^N e^{-m\beta\omega_0} \rightarrow \frac{1}{1 - e^{-\beta\omega_0}}$$

↙

The RWA Polariton

We want to solve the full Dicke model but let us start by the RWA version

$$H_{\text{Dicke}}^{\text{RWA}} = \hbar\tilde{\omega}_c a^\dagger a + \hbar\omega_0 b^\dagger b + \hbar\tilde{\Omega}_R(ab^\dagger + a^\dagger b) \quad \tilde{\Omega}_R \propto \sqrt{N}$$

Introducing the polaritonic operators: $p_j = x_j a + y_j b$, $j \in [\text{LP}, \text{UP}]$

We can diagonalise the Hamiltonian as: $H_{\text{Dicke}}^{\text{RWA}} = \sum_{j \in [\text{LP}, \text{UP}]} \hbar\omega_j p_j^\dagger p_j$

With $|x_j|^2 + |y_j|^2 = 1$, in order to have $[p_j, p_i^\dagger] = \delta_{i,j}$

A linear system of N interacting bosonic fields is (almost) always equivalent to a system of N non-interacting bosonic fields

J. J. Hopfield, Phys. Rev. 112, 1555 (1958)

The Ultrastrong Polariton

Now the same without RWA. We want to put the Hamiltonian

$$H_{\text{Dicke}} = \hbar\omega_c a^\dagger a + \hbar\omega_0 b^\dagger b + \hbar\tilde{\Omega}_R (a + a^\dagger)(b + b^\dagger)$$

In the diagonal form

$$H_{\text{Dicke}} = \sum_{j \in [\text{LP}, \text{UP}]} \hbar\omega_j p_j^\dagger p_j$$

The previous transformation: $p_j = x_j a + y_j b$ is not enough, as we cannot generate the antiresonant terms multiplying p_j^\dagger and p_j

We need instead a transformation that mixes creation and annihilation operators

$$p_j = x_j a + y_j b + z_j a^\dagger + w_j b^\dagger \quad (\text{non conservation of the bare excitation number})$$

In order to have $[p_j, p_i^\dagger] = \delta_{i,j}$, the coefficients have to respect the condition

$$|x_j|^2 + |y_j|^2 - |z_j|^2 - |w_j|^2 = 1$$

The minuses imply that the coefficients **are not bounded!**

Virtual photons

The ground state is the state annihilated by the annihilation operators

We call $|0\rangle$ the ground state of the uncoupled light-matter system

$$a|0\rangle = b|0\rangle = 0$$

From $p_j = x_j a + y_j b + z_j a^\dagger + w_j b^\dagger$ we have $p_j|0\rangle \neq 0$

The coupling modifies the ground state

We introduce the ground state of the coupled system $|G\rangle$

$$p_j|G\rangle = 0$$

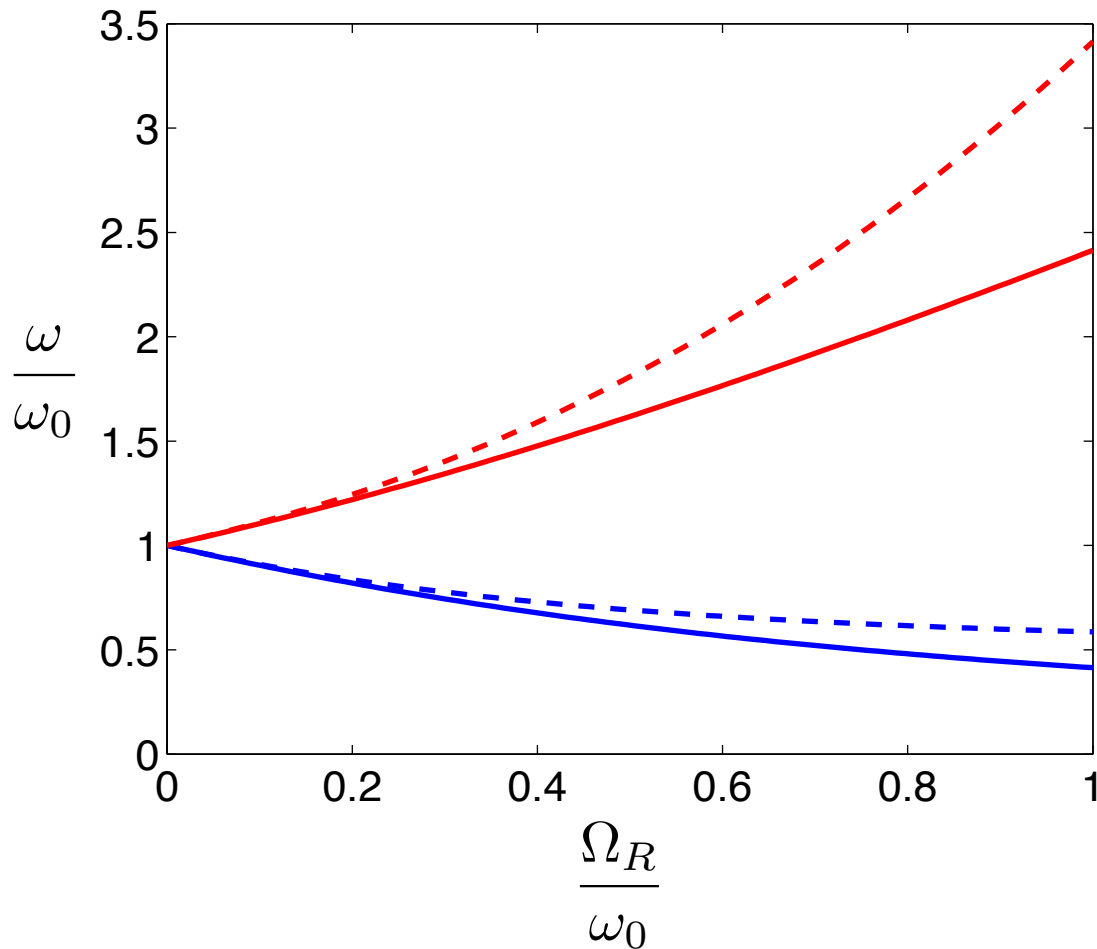
We have then $\langle G|a^\dagger a|G\rangle = |z_{\text{LP}}|^2 + |z_{\text{UP}}|^2 \neq 0 \propto \frac{\Omega_R^2}{\omega_0^2} + O\left(\frac{\Omega_R^3}{\omega_0^3}\right)$

The ground state contains a population of bound photons

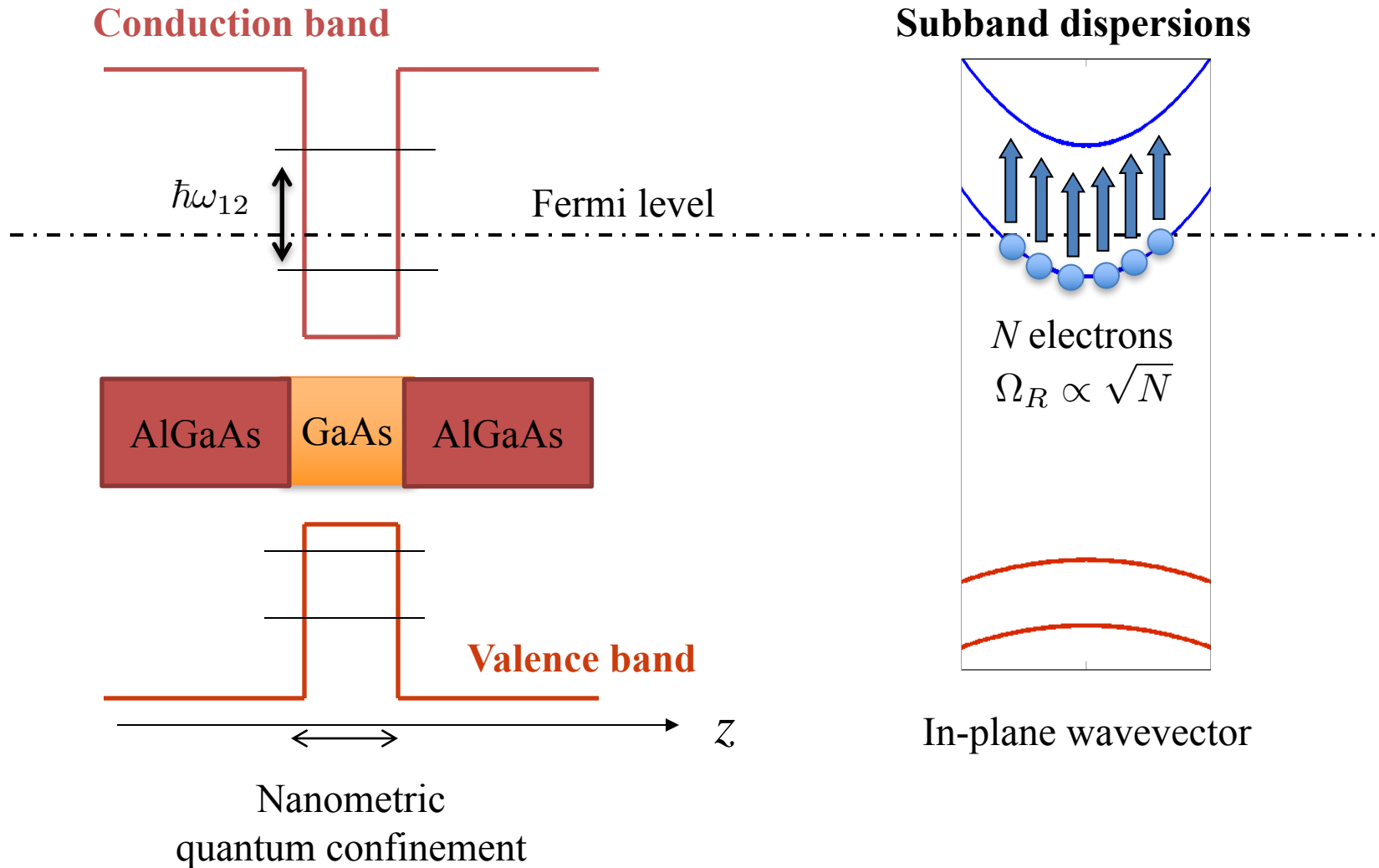
Experimental results

Spectroscopic evidence

- - - Lower polariton RWA
- - - Upper polariton RWA
- Lower polariton
- Upper polariton



Doped quantum well

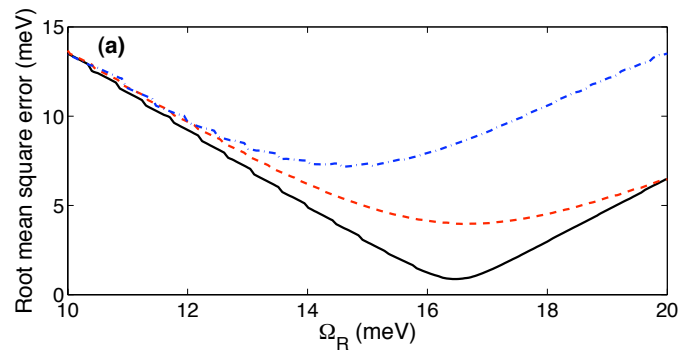


First observation

PHYSICAL REVIEW B **79**, 201303(R) (2009)

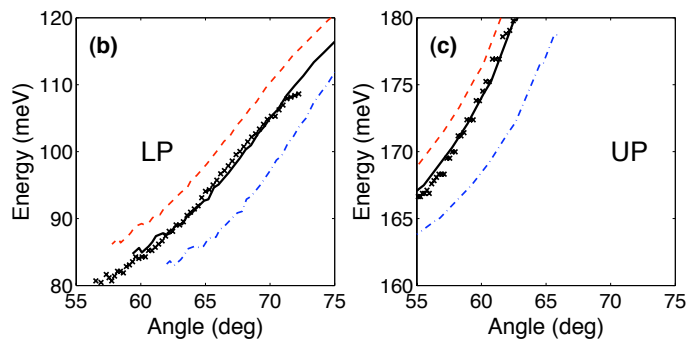
Signatures of the ultrastrong light-matter coupling regime

Aji A. Anappara,¹ Simone De Liberato,^{2,3} Alessandro Tredicucci,^{1,*} Cristiano Ciuti,² Giorgio Biasiol,⁴
Lucia Sorba,¹ and Fabio Beltram¹



$$H_{\text{int}} = \Omega_R(a^\dagger + a)(b^\dagger + b) + \frac{\Omega_R^2}{\omega_0}(a^\dagger + a)^2$$

$$H_{\text{int}}^{\text{RWA}} = \Omega_R(a^\dagger b + b^\dagger a) + \frac{2\Omega_R^2}{\omega_0}a^\dagger a$$

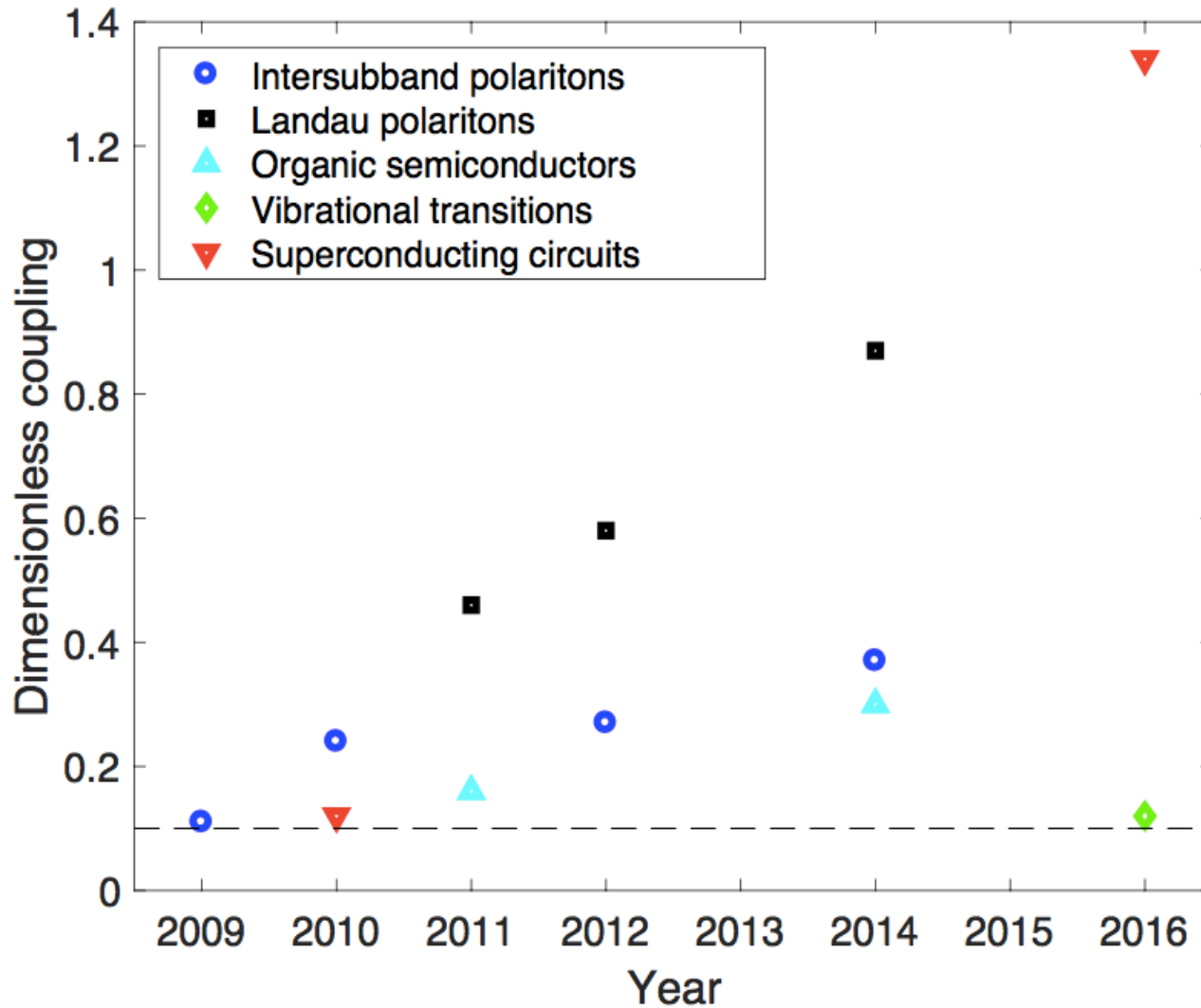


$$H_{\text{int}}^{\text{RWA}'} = \Omega_R(a^\dagger b + b^\dagger a)$$

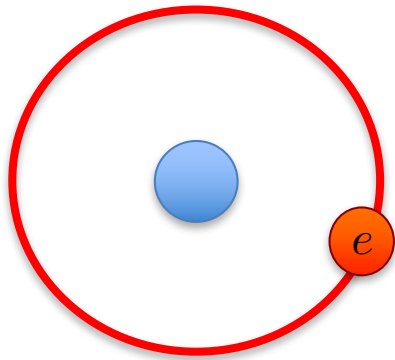
The best fit gives: $\frac{\Omega_R}{\omega_0} = 0.11$

Conventionally taken as threshold (but it is crossover)

First observation

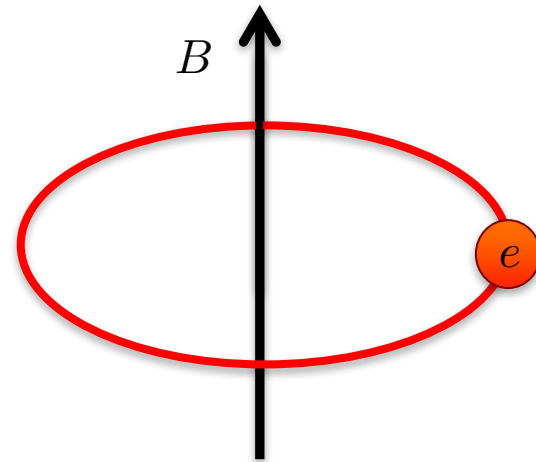


Landau polaritons: a naïf idea



Hydrogenoid atom

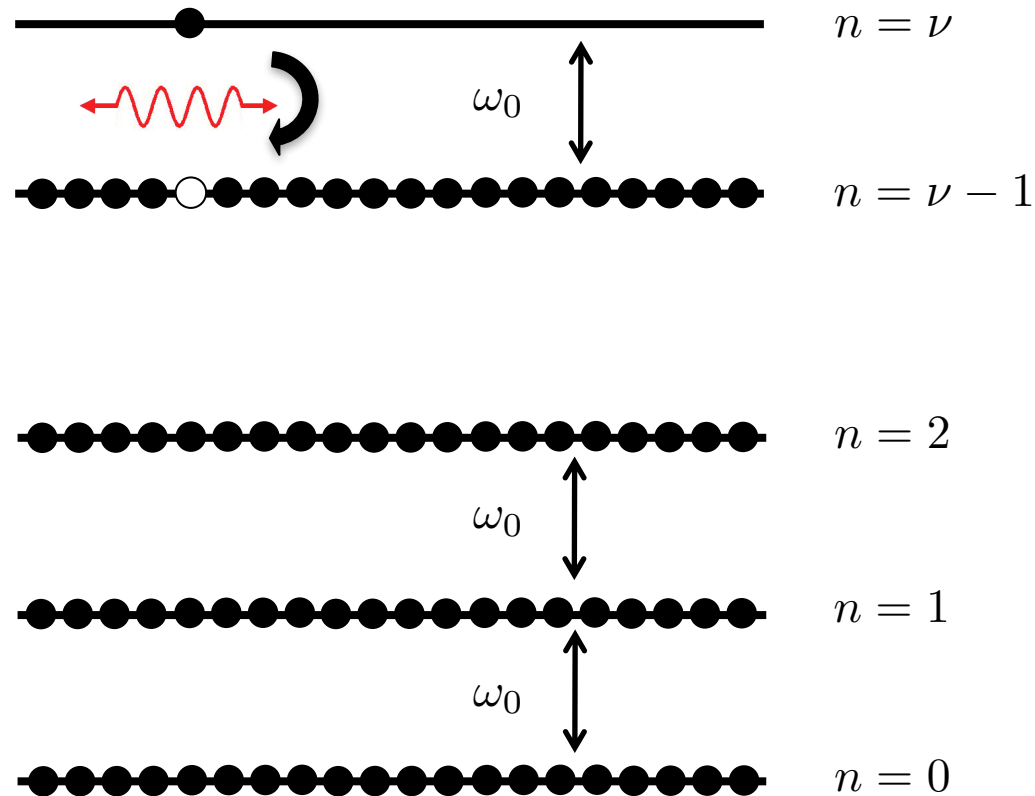
$$r = \frac{n^2 \hbar^2}{e^2 m}$$



Cyclotron orbit

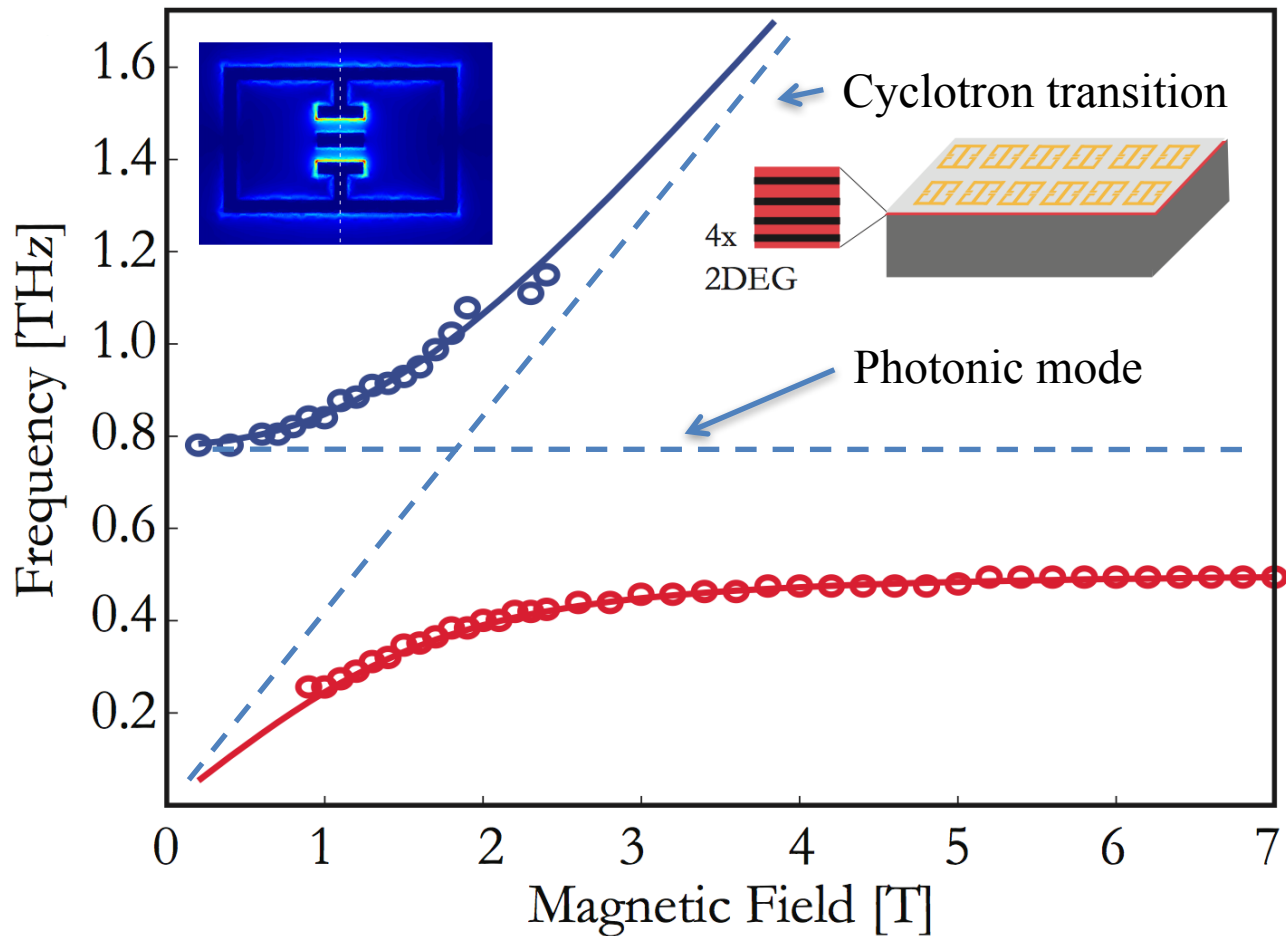
$$r = \frac{mv}{eB}$$

A more realistic description



$$\frac{\Omega_R}{\omega_0} \simeq \sqrt{\alpha \nu n_{\text{QW}}} \propto \frac{1}{\sqrt{B}}$$

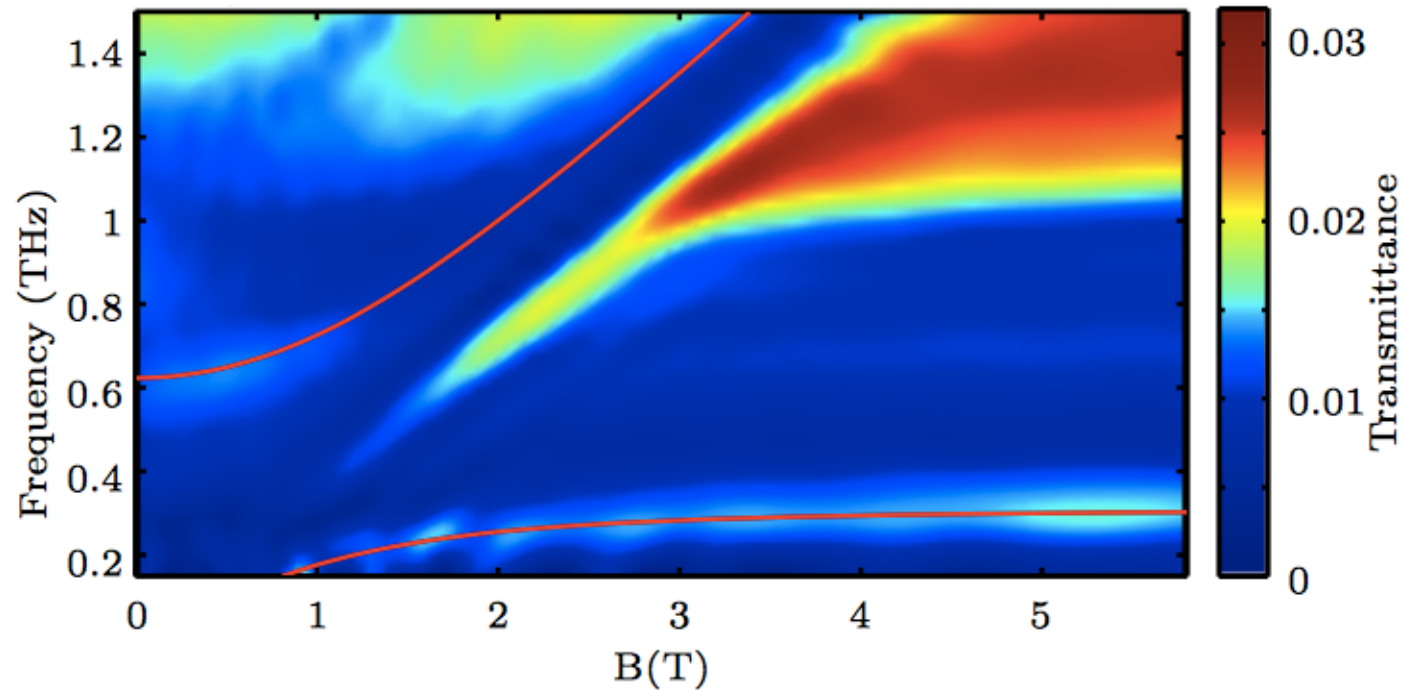
Experimental observation



G. Scalari et al., Science 335, 1323 (2012)

$$\frac{\Omega_R}{\omega_0} = 0.58$$

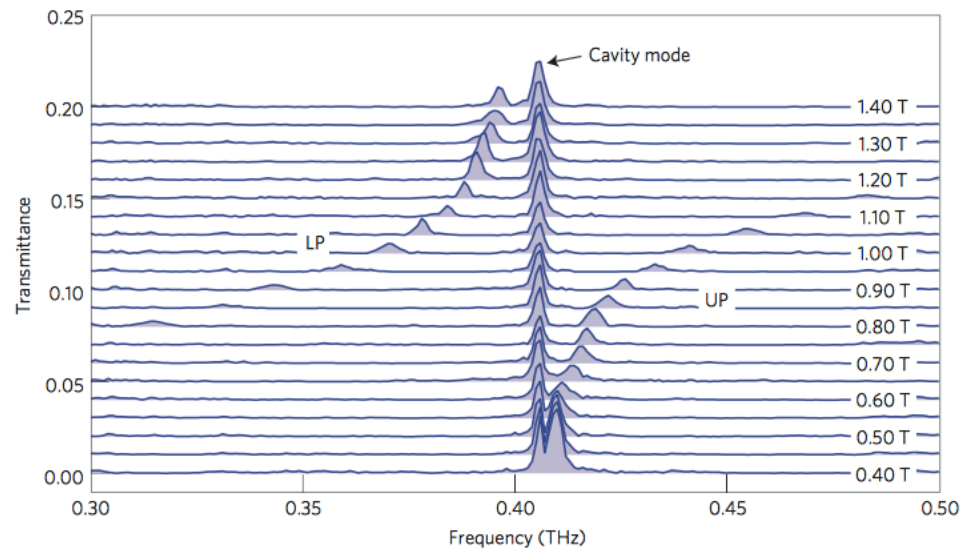
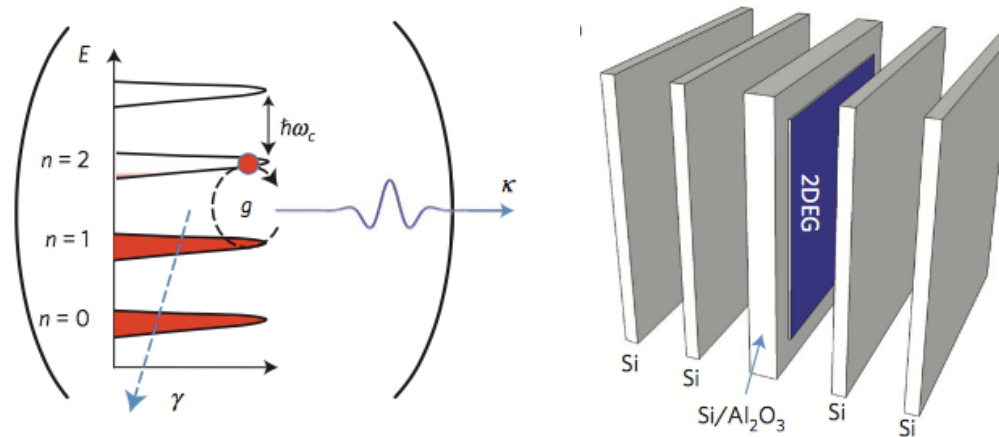
Experimental observation



C. Maissen et al., Phys. Rev. B **90**, 205309 (2014)

$$\frac{\Omega_R}{\omega_0} = 0.87$$

Experimental observation



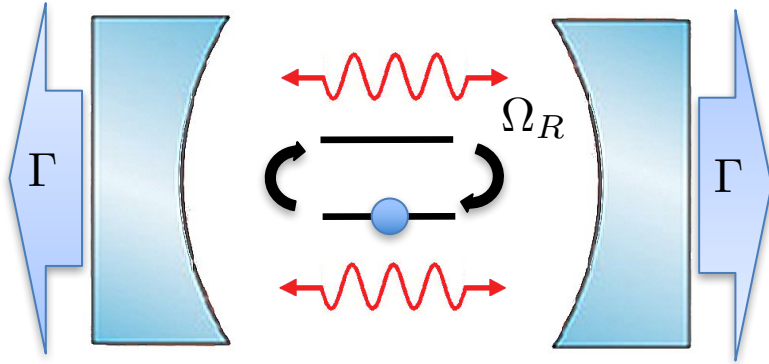
Q. Zhang et al., Nat. Phys. 12, 1005 (2016)

$$\frac{\Omega_R}{\omega_0} = 0.12$$

Open quantum systems



Open quantum systems



Number of emitted photons

$$n_{\text{out}} = \Gamma \langle a^\dagger a \rangle$$

Number of photons inside the cavity

Escape rate

Except that: $\langle G | a^\dagger a | G \rangle = |z_{\text{LP}}|^2 + |z_{\text{UP}}|^2 \neq 0 \propto \frac{\Omega_R^2}{\omega_0^2} + O\left(\frac{\Omega_R^3}{\omega_0^3}\right)$

Emission of photons out of the ground state. **Wrong!**

Open quantum systems

Master equation $\frac{\partial \rho}{\partial t} = -i[H, \rho] + \mathcal{L}(\rho)$

Lindblad operator $\mathcal{L}(\rho) = \frac{\Gamma}{2}(2a\rho a^\dagger - a^\dagger a\rho - \rho a^\dagger a)$

Standard ground state $\mathcal{L}(|0\rangle\langle 0|) = \frac{\Gamma}{2}(2a|0\rangle\langle 0|a^\dagger - a^\dagger a|0\rangle\langle 0| - |0\rangle\langle 0|a^\dagger a) = 0$

Ultrastrong ground state $\mathcal{L}(|G\rangle\langle G|) = \frac{\Gamma}{2}(2a|G\rangle\langle G|a^\dagger - a^\dagger a|G\rangle\langle G| - |G\rangle\langle G|a^\dagger a) \neq 0$

The ground state is not stable! **Wrong!**

Real Lindblad operator $\mathcal{L}(\rho) = U\rho a^\dagger + a\rho U^\dagger - a^\dagger U\rho - \rho U^\dagger a$

Integral operator $U = \int_0^\infty dt g(t) e^{-iHt} a e^{iHt}$

Normally one assumes $e^{-iHt} a e^{iHt} \simeq a e^{i\omega_0 t}$ $\tilde{g}(\omega)$ bath's density of states

Leading to $U = \frac{\Gamma}{2} a$

Open quantum systems

We write the jump operator a on the eigenbasis of H $a = \sum_{\alpha,\beta} |\alpha\rangle a_{\alpha,\beta} \langle\beta|$

$$U = \int_0^\infty dt g(t) e^{-iHt} a e^{iHt} = \sum_{\alpha,\beta} |\alpha\rangle \langle\beta| \int_0^\infty a_{\alpha,\beta} e^{i(\omega_\alpha - \omega_\beta)t} g(t) dt$$

$$U = \sum_{\alpha,\beta} a_{\alpha,\beta} |\alpha\rangle \langle\beta| \int_{-\infty}^\infty d\omega \frac{\tilde{g}(\omega)}{2\pi} \int_0^\infty dt e^{i(\omega_\beta - \omega_\alpha - \omega)t}$$

$$U = \sum_{\alpha,\beta} a_{\alpha,\beta} |\alpha\rangle \langle\beta| \int_{-\infty}^\infty d\omega \frac{\tilde{g}(\omega)}{2\pi} \left[\pi \delta(\omega_\alpha - \omega_\beta + \omega) - \frac{i}{\omega_\alpha - \omega_\beta + \omega} \right]$$

$$U = \sum_{\alpha,\beta} \frac{\tilde{g}(\omega_\beta - \omega_\alpha)}{2} a_{\alpha,\beta} |\alpha\rangle \langle\beta|$$

Resonance shift
 $[H, \rho] \rightarrow [\tilde{H}, \rho]$

There is no density of states at negative frequencies! $\tilde{g}(\omega < 0) = 0$

$$U|G\rangle = \sum_{\alpha} \frac{\tilde{g}(\omega_G - \omega_\alpha)}{2} a_{\alpha,G} |\alpha\rangle = 0 \quad \Rightarrow \quad \mathcal{L}(|G\rangle\langle G|) = 0$$

Open quantum systems

Take home message:

**On the shelf tools and approximations fail in the ultrastrong coupling regime
Always rederive everything from scratch! (From the Lagrangian)**

Bibliography:

*S. De Liberato, D. Gerace, I. Carusotto, and C. Ciuti, Phys. Rev. A **80**, 053810 (2009)*

*F. Beaudoin, J. M. Gambetta, and A. Blais, Phys. Rev. A **84**, 043832 (2011)*

*M. Bamba and T. Ogawa, Phys. Rev. A **88**, 013814 (2013)*

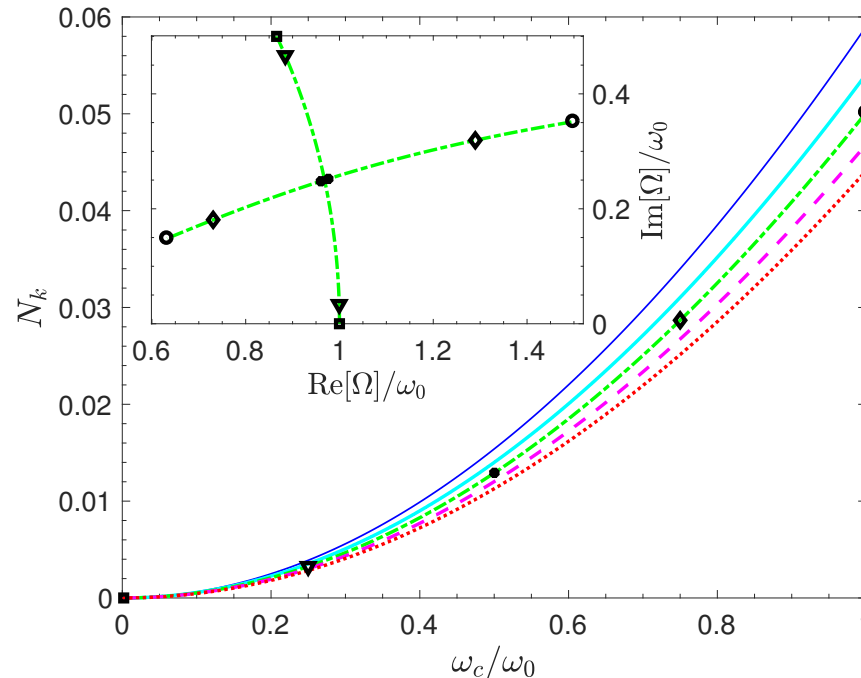
*S. De Liberato, Phys. Rev. A **89**, 017801 (2014)*

*M. Bamba and T. Ogawa, Physical Review A **89**, 023817 (2014)*

Open quantum systems

What about the virtual photons?

They remain also if the system is not in the strong coupling regime



Ultrastrong coupling physics is largely independent from Γ

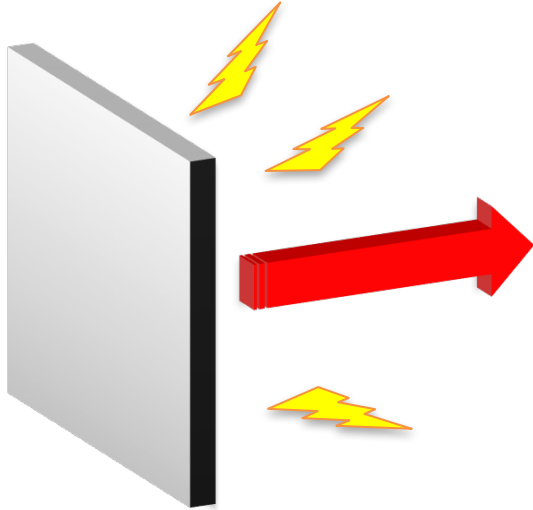
Virtual photons in the ground state of a dissipative system

S. De Liberato

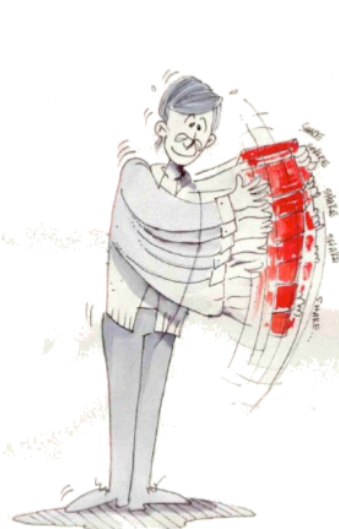
To appear in Nature Communication

Ultrastrong phenomenology

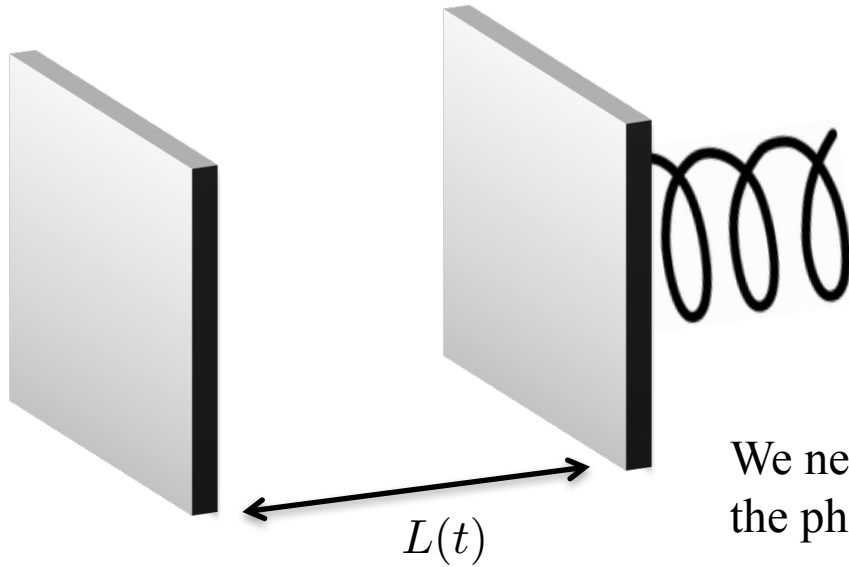
Dynamical Casimir effect



A mirror accelerated in vacuum emits photons
(due to friction with vacuum fluctuations)



Dynamical Casimir effect



A mirror, accelerated in the vacuum, emits photons

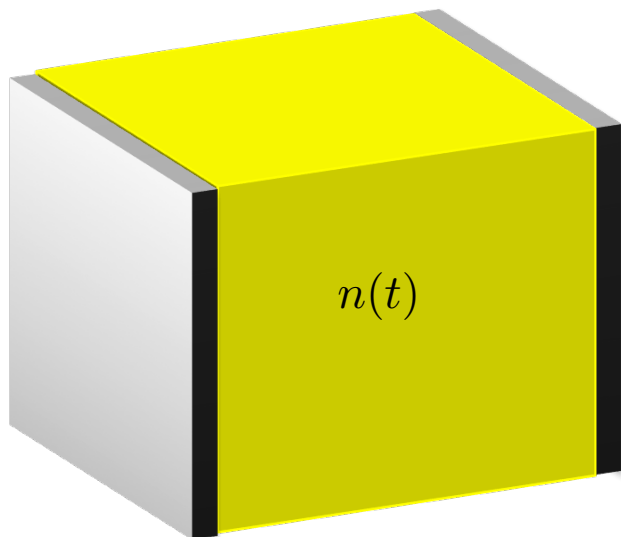
We need the oscillation frequency comparable with the photon one

Impossible for a mechanic spring!

Or, we can keep the length fixed, and change the dielectric constant

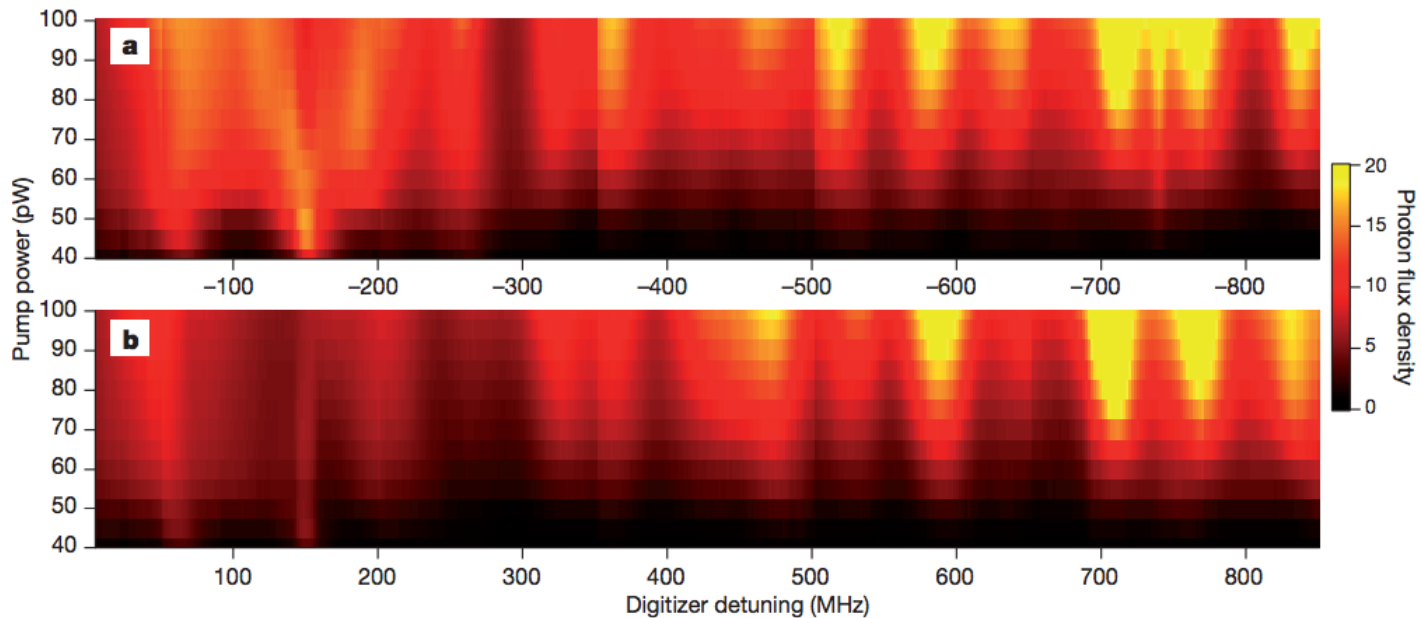
$$L_{\text{opt}}(t) = n(t)L$$

No moving parts!



First observation

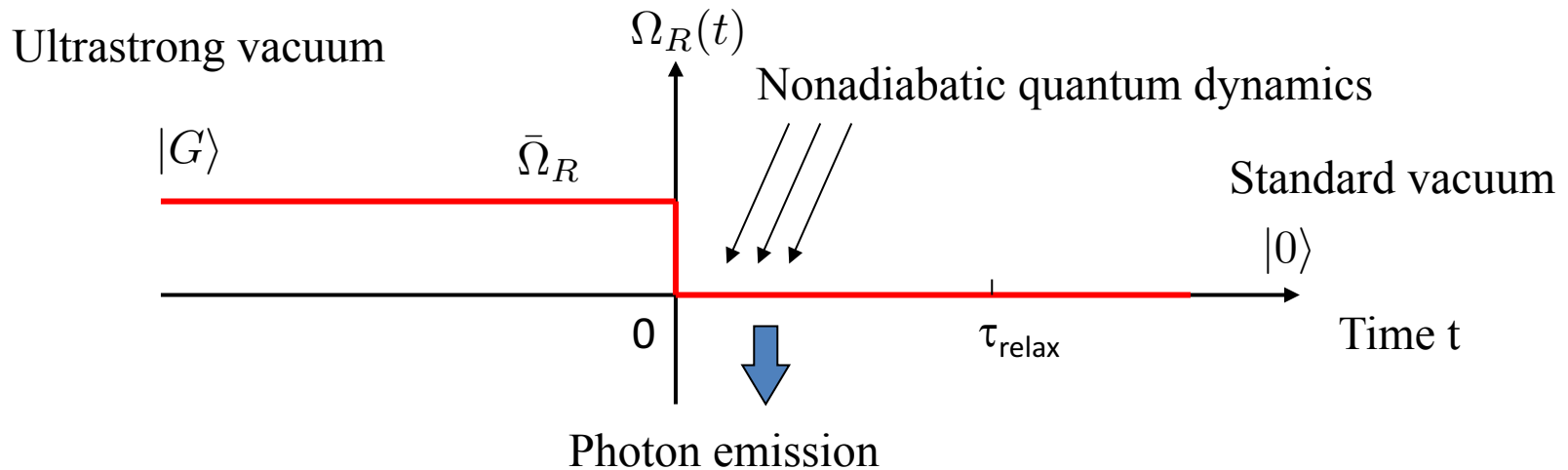
Observed in 2011 using superconducting circuits



C. M. Wilson et al., Nature 479, 376 (2011)

Quantum vacuum emission

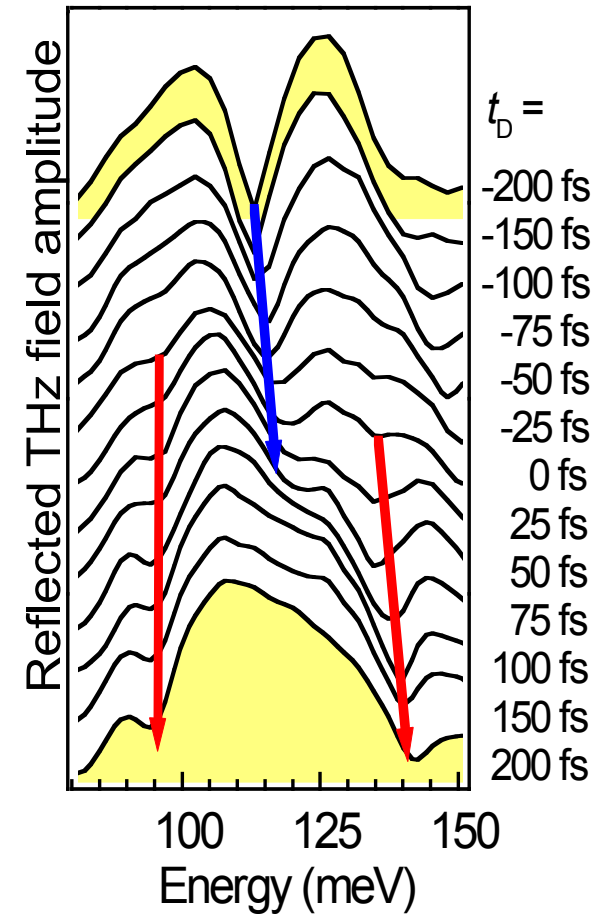
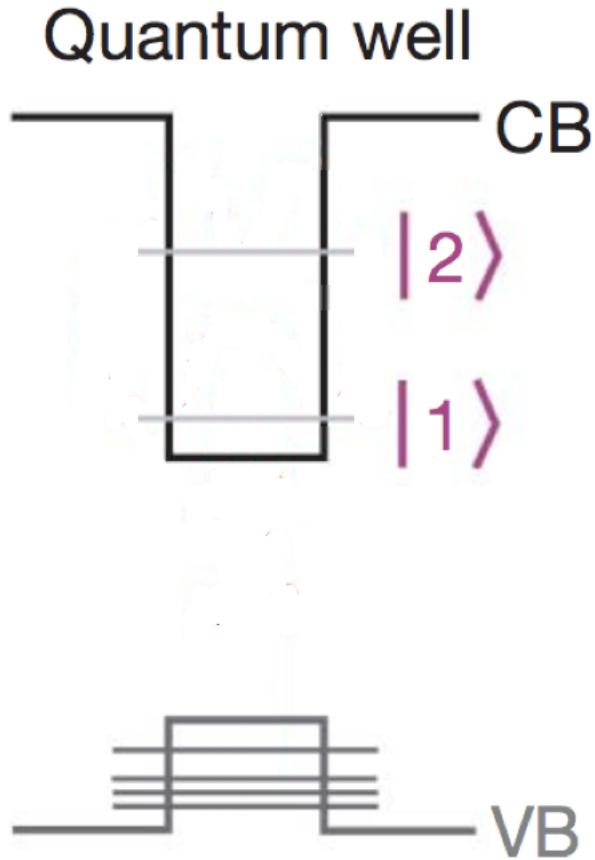
A manifestation of ground state virtual photons



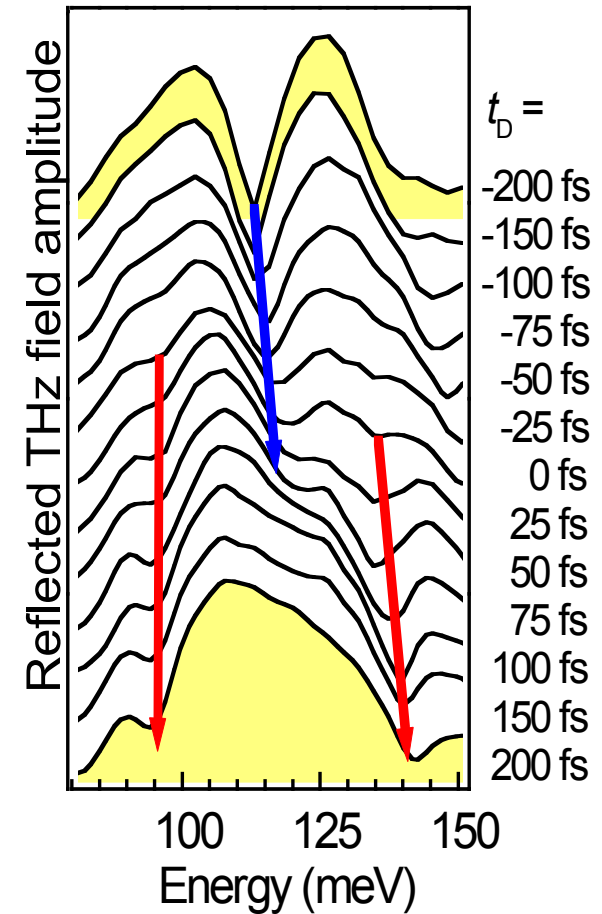
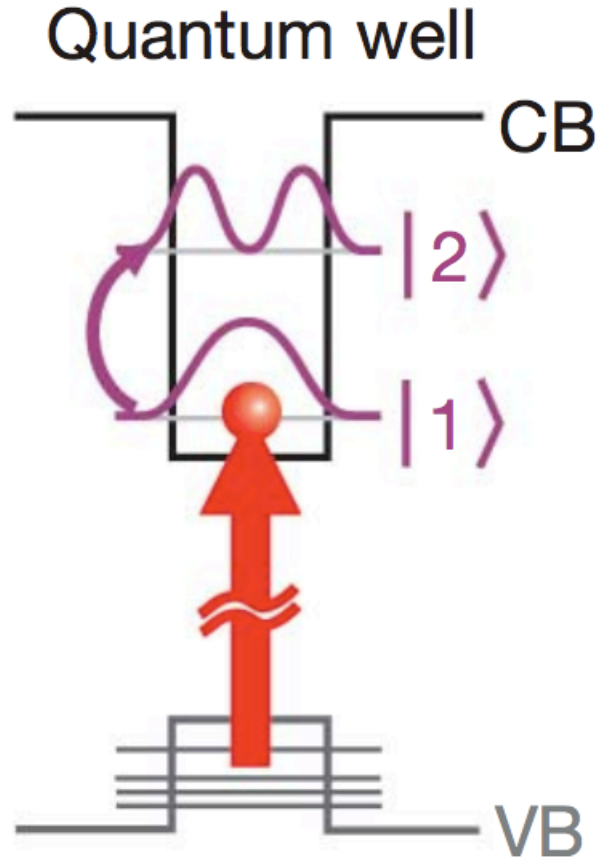
C. Ciuti, G. Bastard, and I. Carusotto, Phys. Rev. B 72, 115303 (2005)

S. De Liberato, C. Ciuti, and I. Carusotto, Phys. Rev. Lett. 98, 103602 (2007)

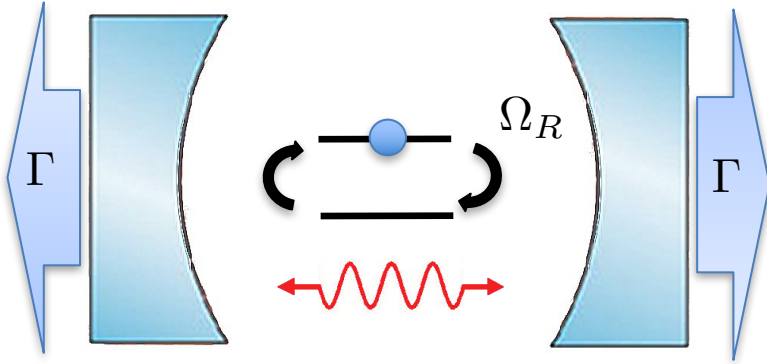
Nonadiabatic modulation



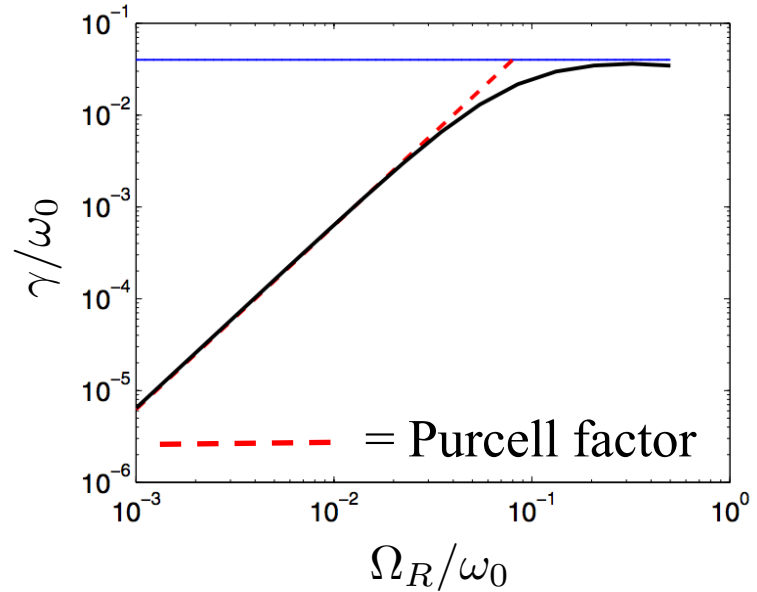
Nonadiabatic modulation



Purcell effect



How the emission rate γ depends on Ω_R ?



*C. Ciuti and I. Carusotto,
Phys. Rev. A 74, 033811 (2006)*

Weak and Strong coupling regimes: quadratic dependency upon Ω_R

Ultrastrong coupling regime: saturation

Purcell effect

$$H = \omega_c a^\dagger a + \omega_0 b^\dagger b + \Omega(a^\dagger + a)(b^\dagger + b) + \frac{\Omega^2}{\omega_0}(a^\dagger + a)^2$$

$$H = H_{\text{field}} + \frac{\mathbf{p}^2}{2m} + V(\mathbf{r}) - \frac{e\mathbf{p}\mathbf{A}(\mathbf{r})}{m} + \frac{e^2\mathbf{A}(\mathbf{r})^2}{2m}$$

Intensity of the field at the location of the dipoles

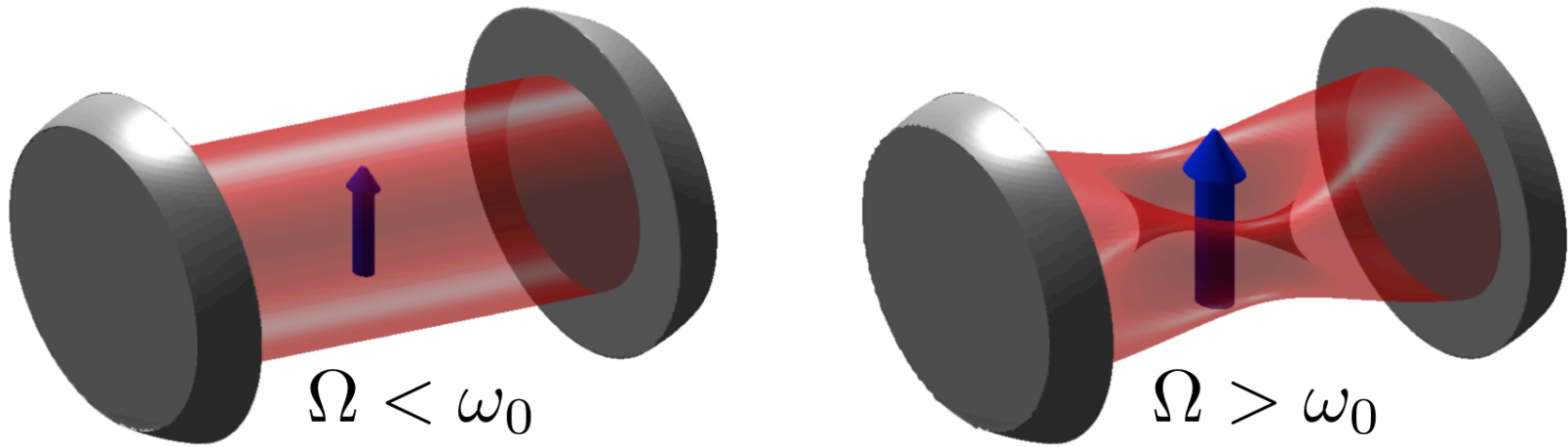
If $\frac{\Omega_R}{\omega_0} > 1$ the last term, **always positive**, becomes dominant

The low energy modes need to minimize the field location over the dipoles

Polariton modes will be $\left\{ \begin{array}{l} \text{pure photon modes that avoid the dipoles} \\ \text{pure matter mode} \end{array} \right.$

Light and matter decouple in the deep strong coupling regime

Purcell effect



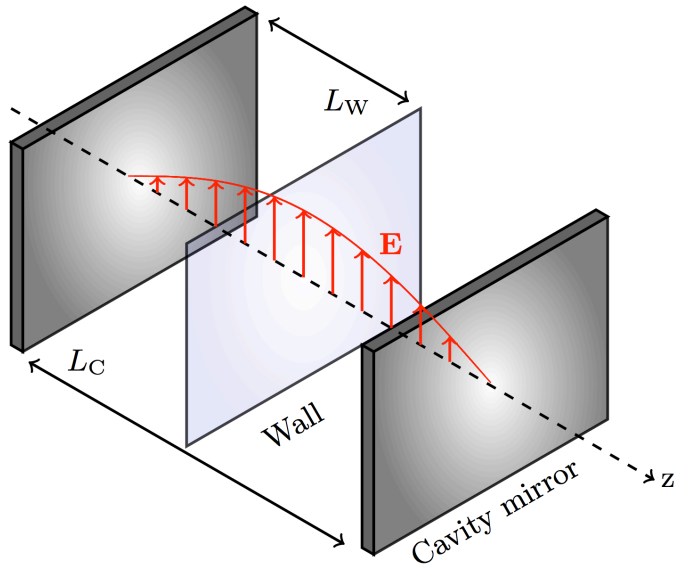
The photonic field avoids the dipoles

Light-matter interaction is due to **local** interactions



Light and matter do not exchange energy

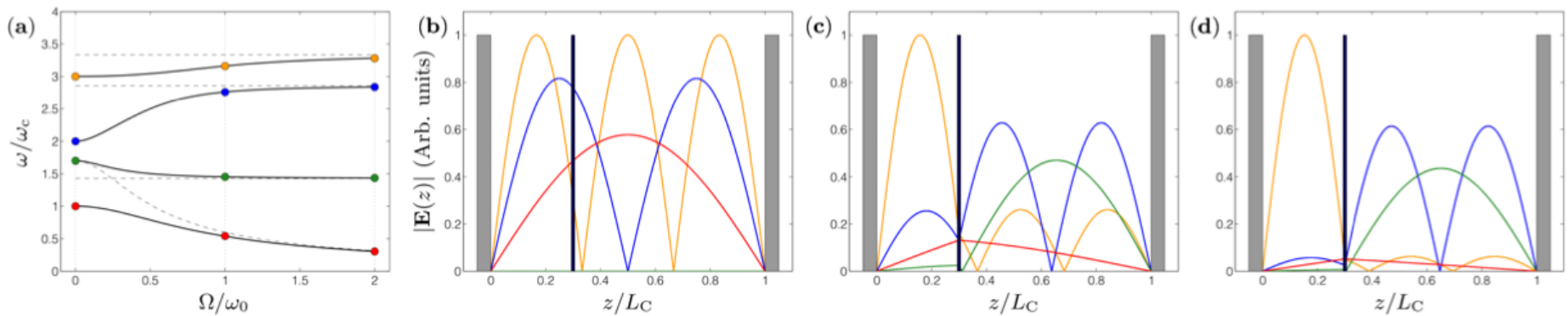
Purcell effect



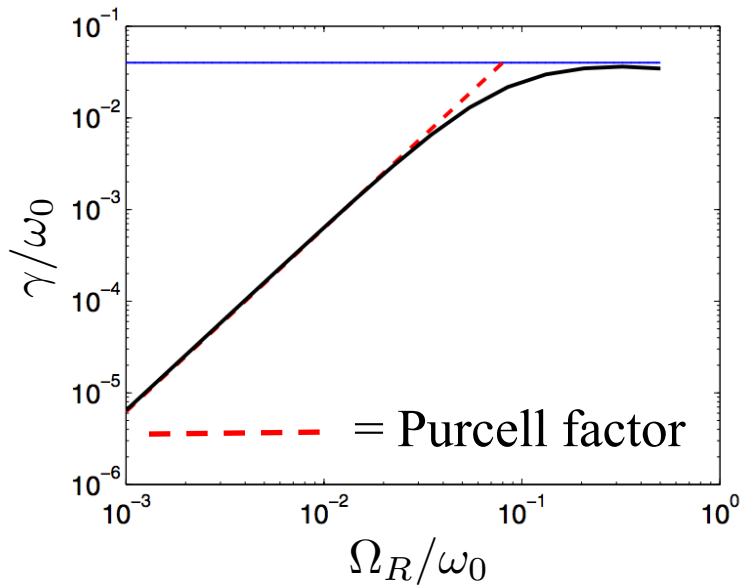
Example: a two-dimensional metallic cavity enclosing a wall of in-plane dipoles

S. De Liberato, Phys. Rev. Lett. **112**, 016401 (2014)

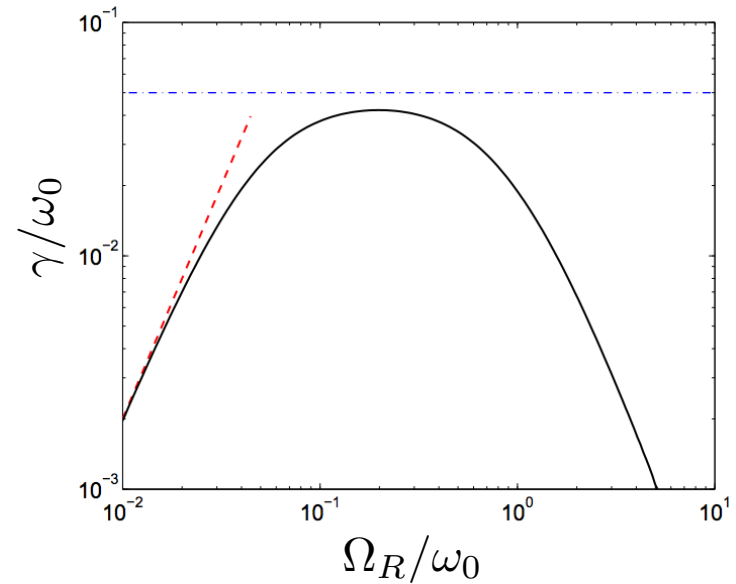
The wall becomes a metallic mirror



Purcell effect



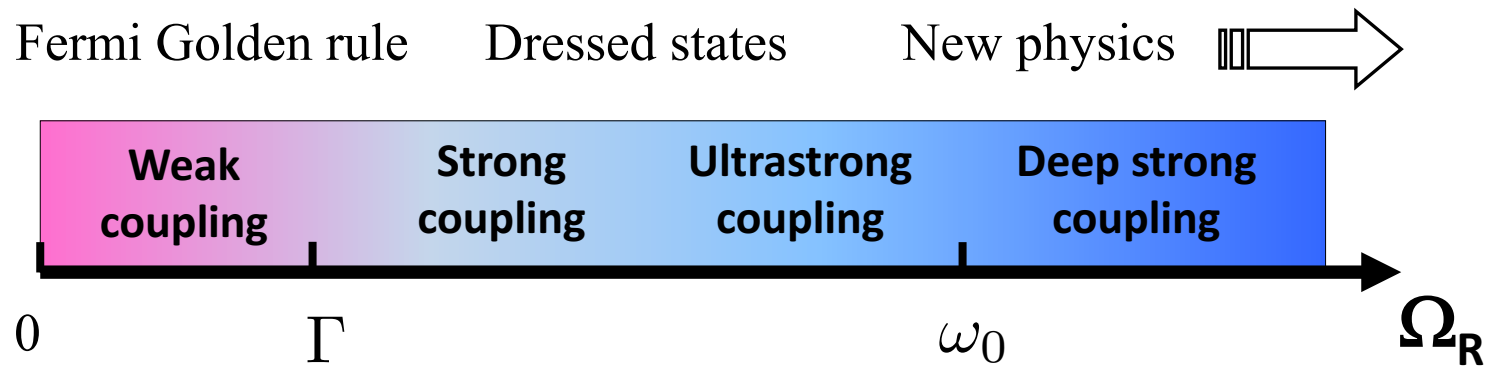
C. Ciuti and I. Carusotto,
Phys. Rev. A **74**, 033811 (2006)



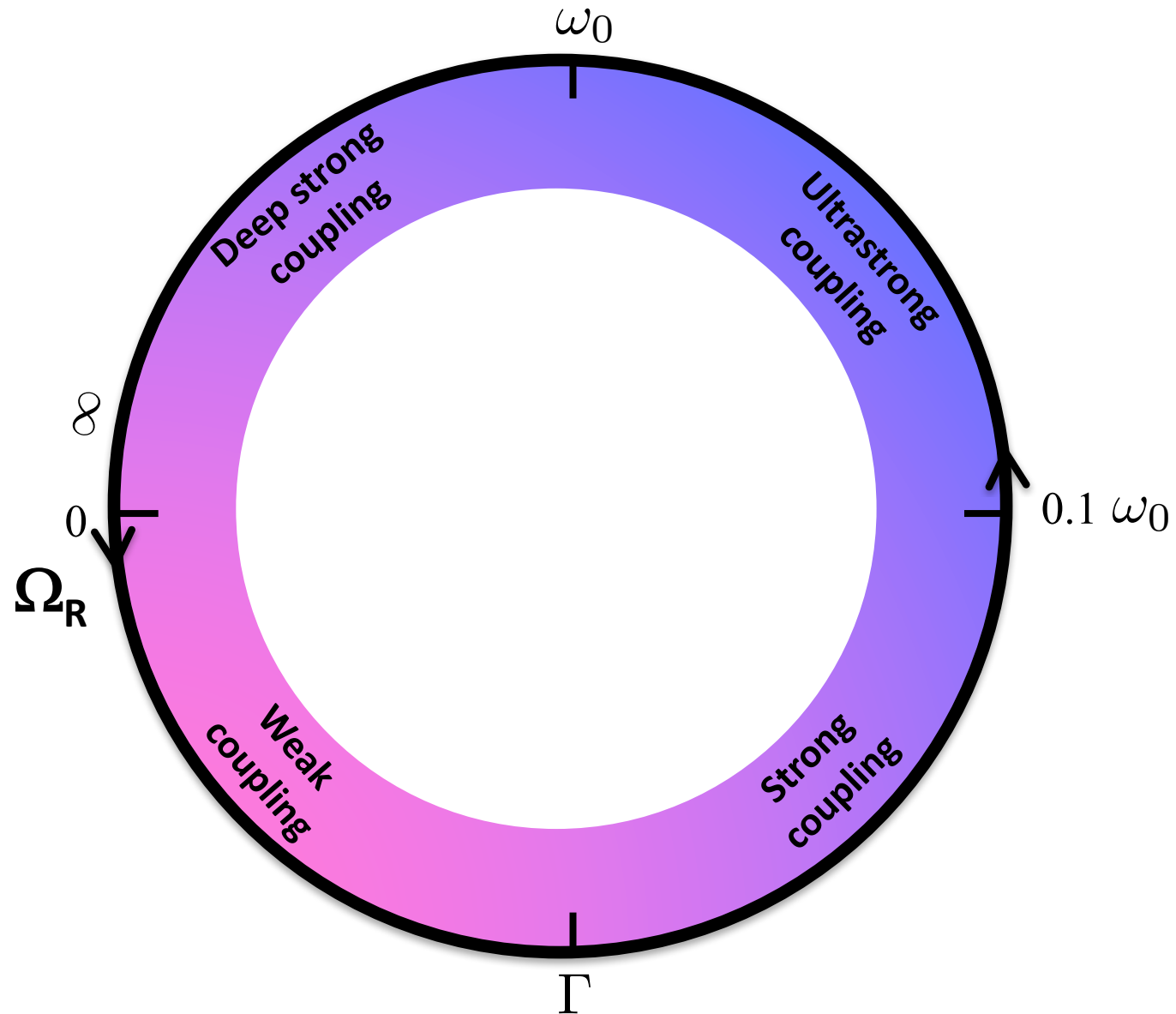
S. De Liberato,
Phys. Rev. Lett. **112**, 016401 (2014)

Breakdown of the Purcell effect!

Light-matter coupling



Light-matter decoupling



Thank you for your attention