Claudiu Genes Max Planck Institute for the Science of Light (Erlangen, Germany)

Quantum Physics of Light-Matter Interactions

Summer Semester 2019, FAU, Erlangen

26 April 2019

Class structure

- Lectures: 12 each of two hours (classes cancelled on bridge days: May 31 and June 21)

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- Exam: 90 minutes written/10 minutes oral examination

Fundamentals of light-matter interactions

- The two-level system approximation
- Light-matter interaction – dipole and rotating wave approximations
- The master equation for spontaneous emission
- Solving the master equation – optical Bloch equations
- Fundamentals of cavity quantum electrodynamics – the Jaynes Cummings Hamiltonian and quantum Langevin equations

The laser

- Laser threshold
- Laser photon statistics

Effects of light onto motion

- The full quantum optical force
- Doppler cooling, gradient cooling
- Ion traps, ion cooling, ion logic
- Cavity optomechanics

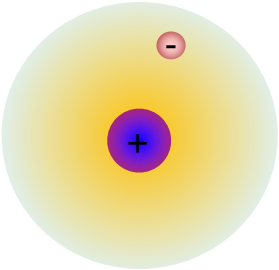
Superradiance/subradiance

- Collective decay of coupled quantum emitters
- subradiantly enhanced energy transfer

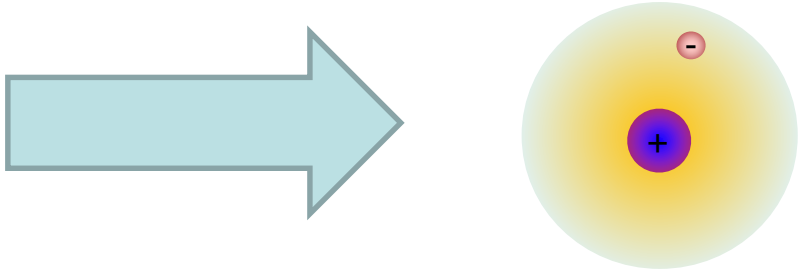
Molecules: vibronic coupling

- The Holstein Hamiltonian
- Emission and absorption spectra, energy transfer

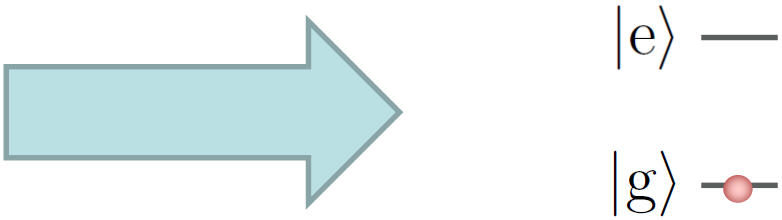
Consider a quantum emitter: atom, molecule, quantum dot etc



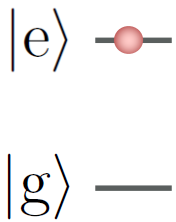
How does it interact with electromagnetic waves?



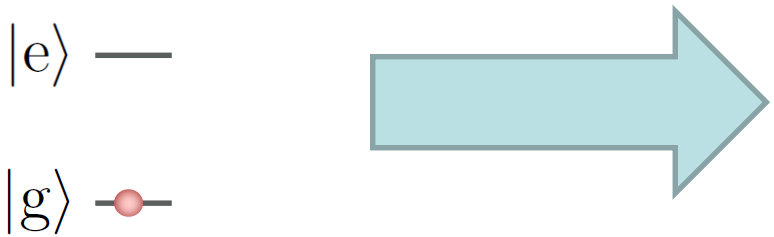
Simplified picture – two level system



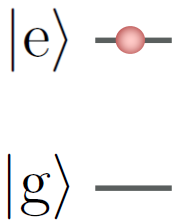
Light absorption



Light emission

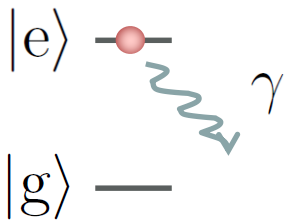


There is also a quantum vacuum of electromagnetic modes (quantization in a big box)



Story in short

Effect – spontaneous emission – gives rise to finite lifetime (non-zero linewidth) for electronic transitions



- Master equation description of the dynamics

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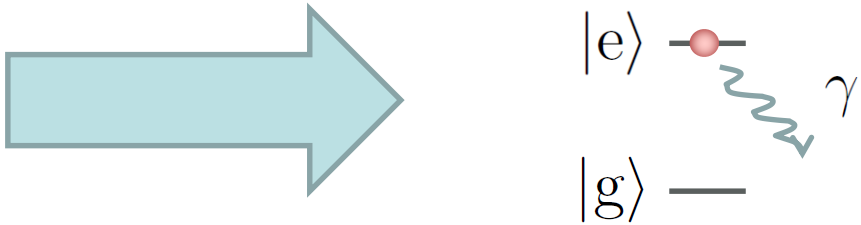
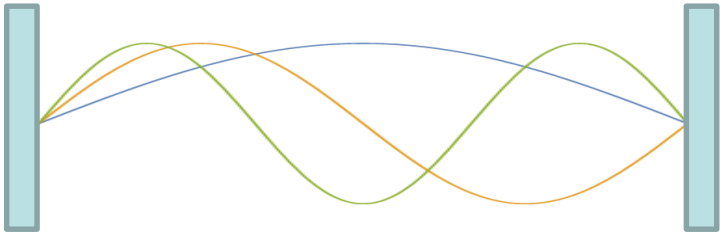


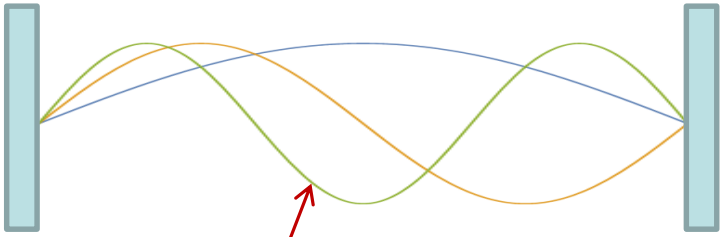
- Master equation description of the dynamics
- Optical Bloch equations

Optical cavity

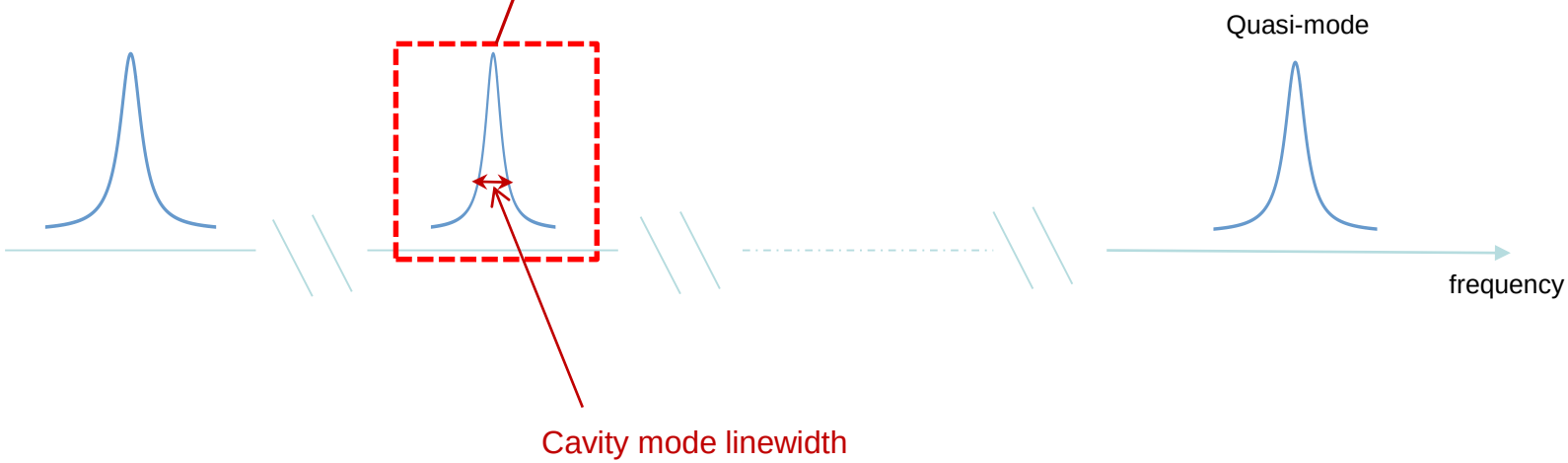


- Multiple resonances

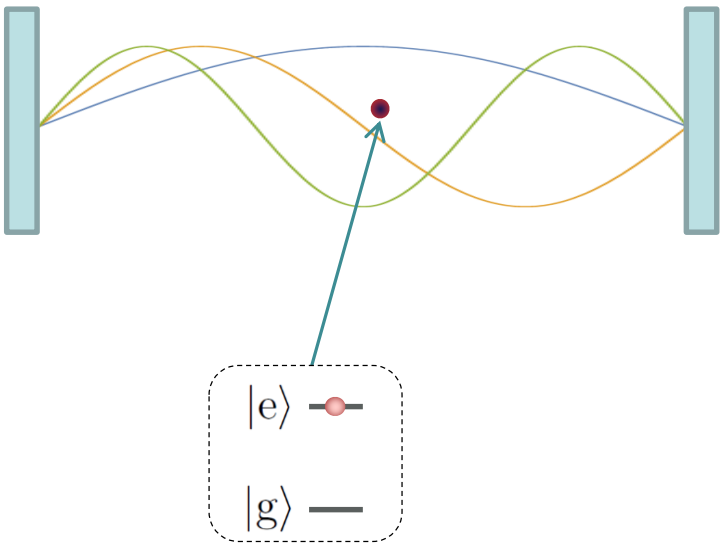
Optical cavity



- Multiple resonances
- Lorentzian profile – enhanced density of optical modes around resonances



Atom in cavity- Jaynes Cummings dynamics

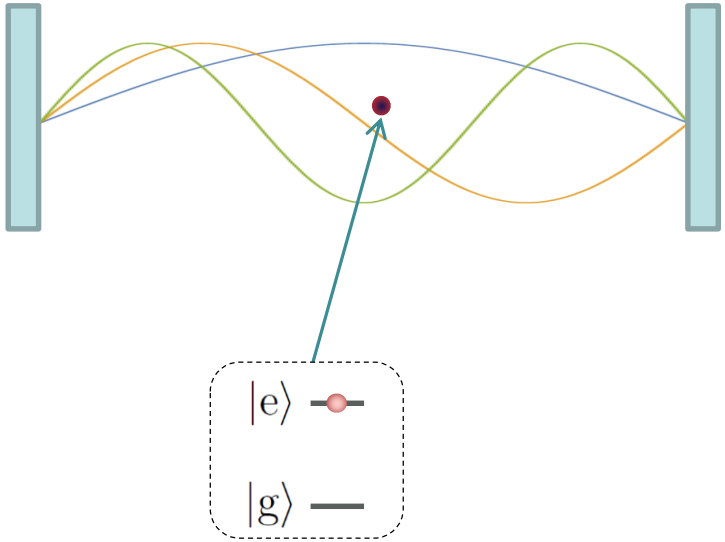


The Jaynes-Cummings Hamiltonian

$$H_{JC} = \hbar g \left[a^\dagger \sigma + a\sigma^\dagger\right]$$

$$g = \frac{1}{\hbar}\mathcal{E}_0 d_{eg} = \left[\frac{\omega}{\hbar\epsilon_0 \ell \mathcal{S}}\right]^{1/2} d_{eg}$$

Enhancement of photon-emitter scattering – cavity quantum electrodynamics



- Jaynes-Cummings Hamiltonian
- Quantum Langevin equation for the cavity field
- Input-output relations

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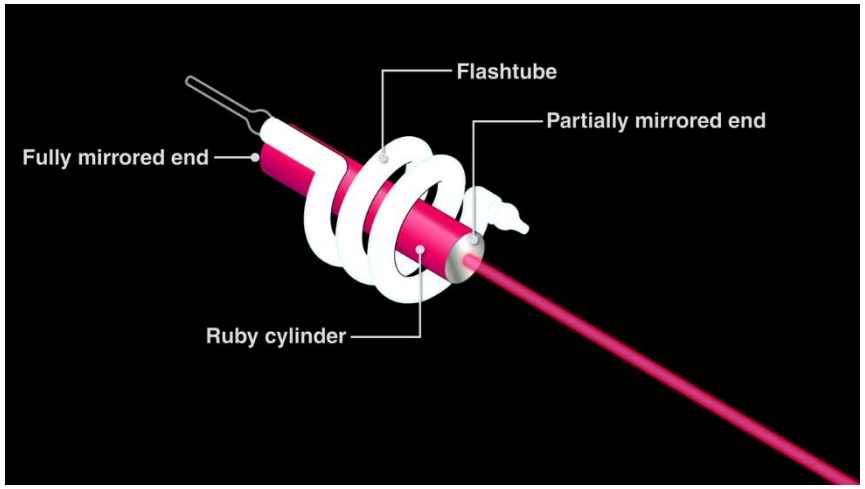
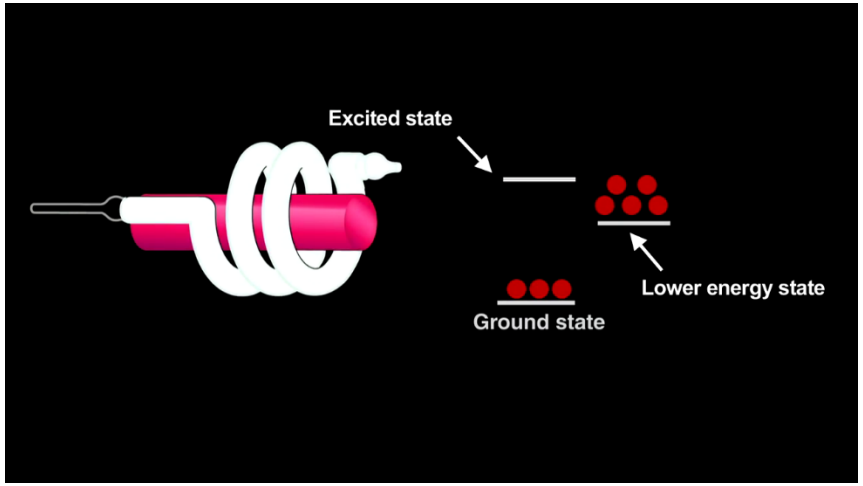
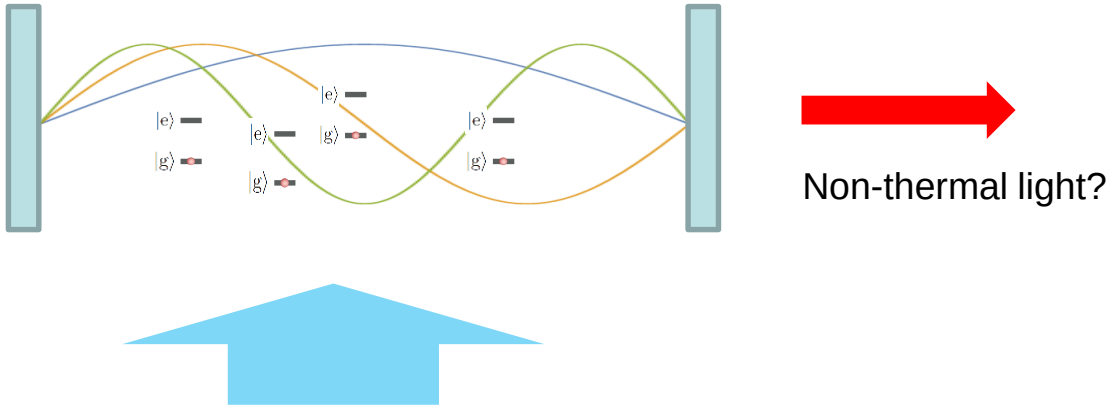




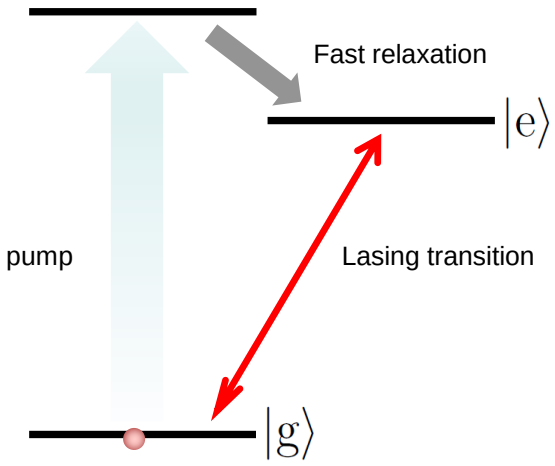
Source: youtube: How a laser works

Can you build a (theoretical) laser?

Enhancement of photon-emitter scattering – optical cavities

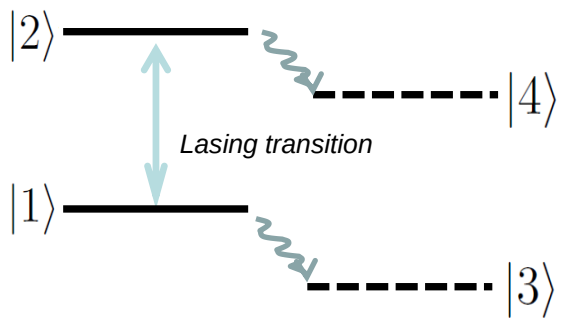


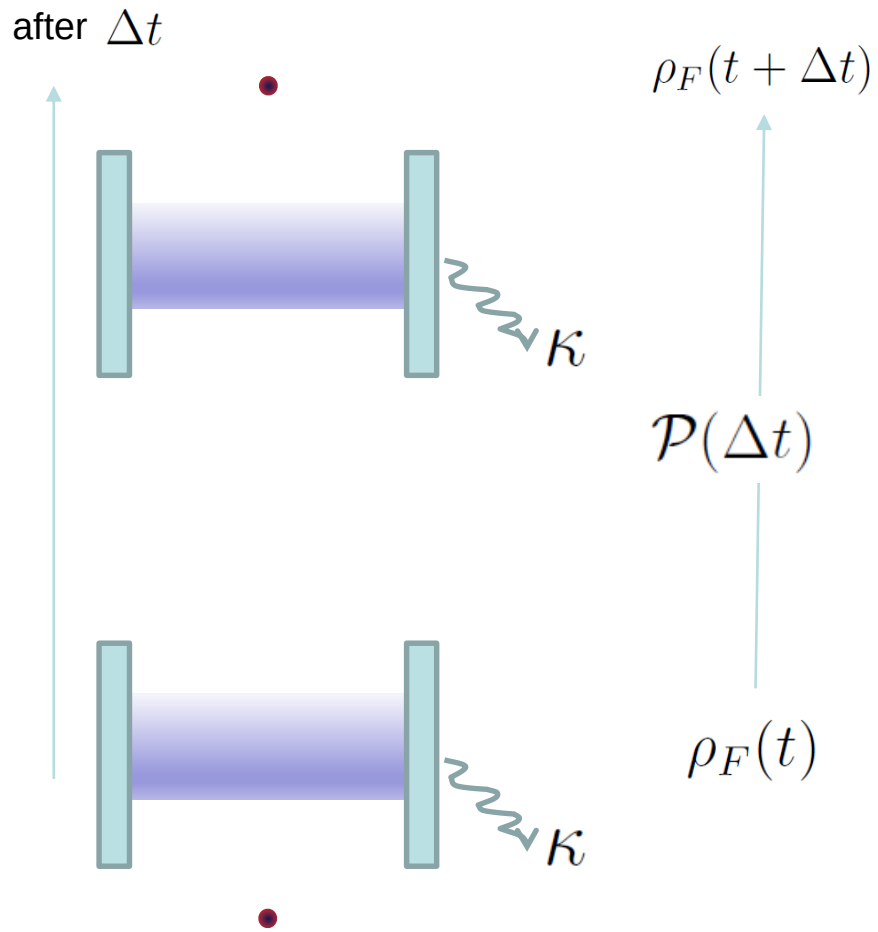
Model 1



Laser threshold

Model 2





Laser photon statistics

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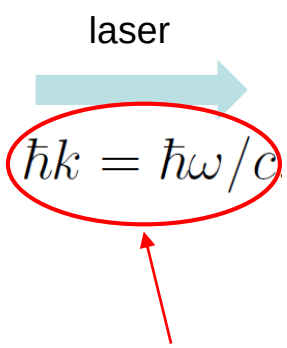
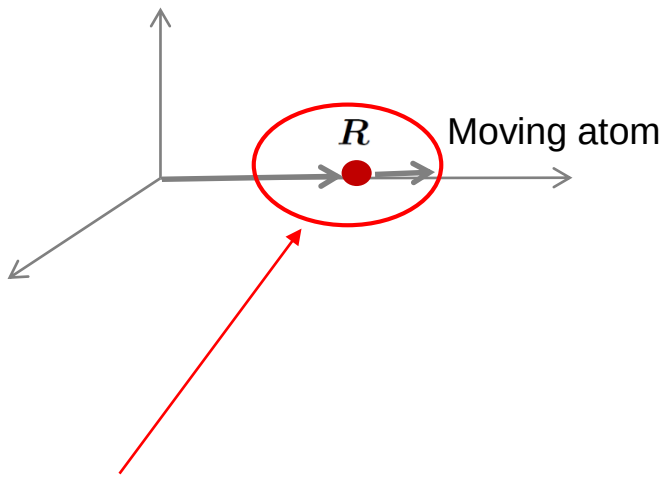
Light can affect motion !

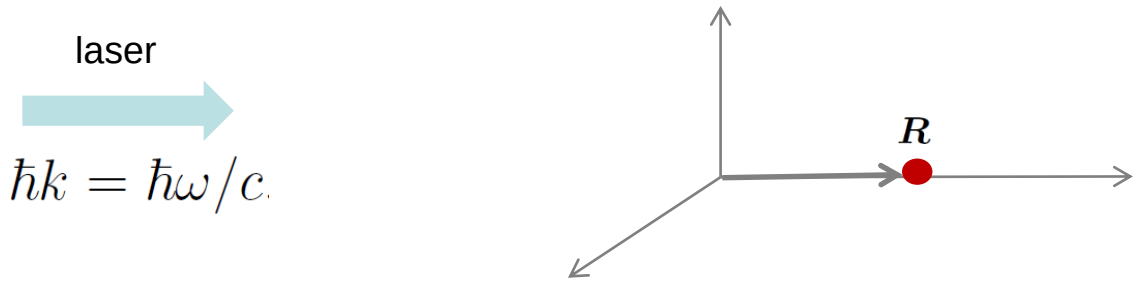


Light carries momentum



Recoil momentum – recoil energy

$$\hbar\omega_{rec} = \frac{(\Delta p)^2}{2m} = \frac{(\hbar k)^2}{2m}$$

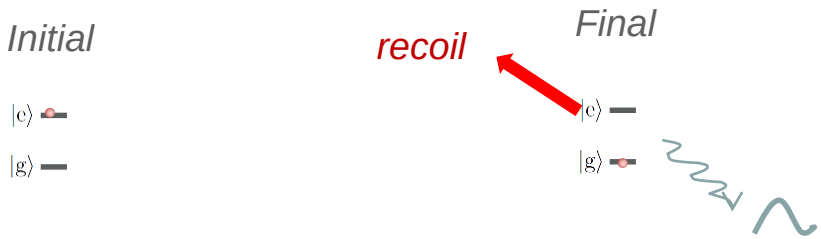


Possible processes:

- Stimulated absorption

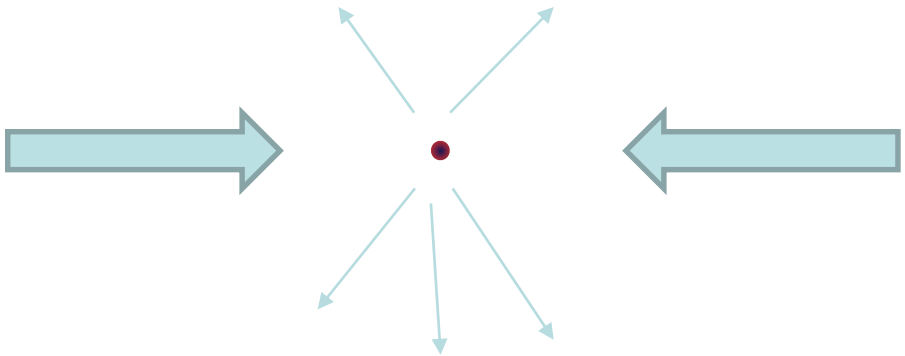
- Stimulated absorption

- Spontaneous emission



Light can be used to control motion ! (for example cooling)

Counterpropagating fields



Spontaneous emission to other directions

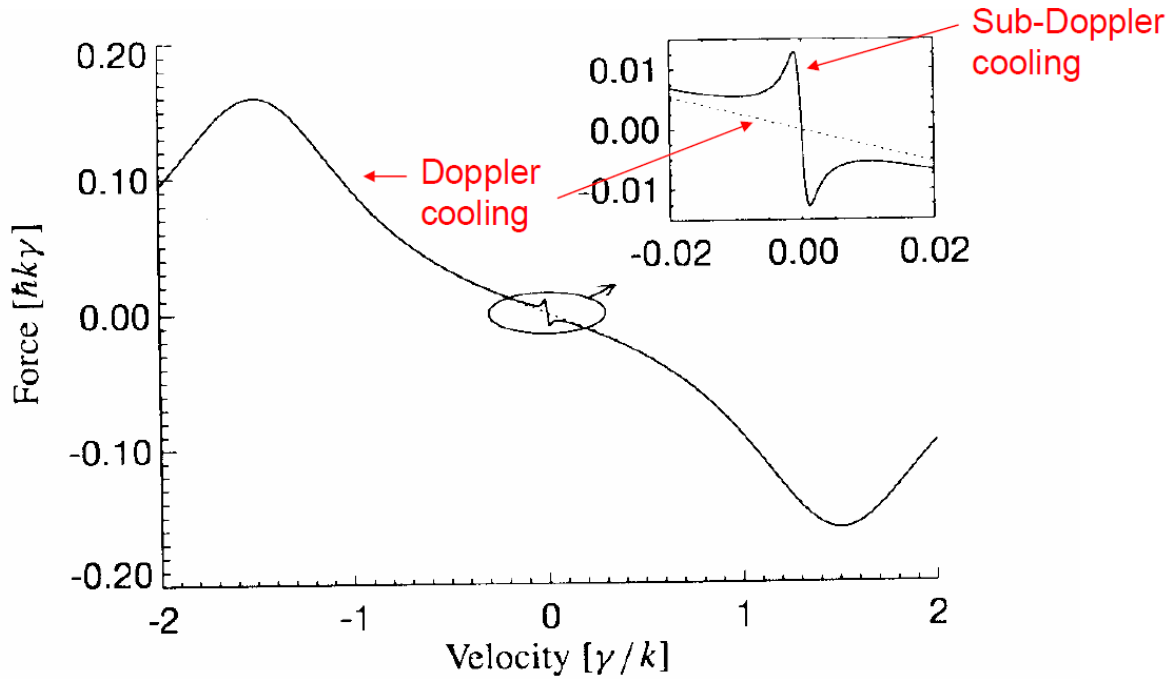
$$\boldsymbol{F}_{standing}(z) = (F_0 + \beta v)\,\hat{z} + (-F_0 + \beta v)\,\hat{z} = 2\beta v \hat{z}$$

$$\beta = \hbar k_L^2 \frac{2\gamma\Omega^2\Delta}{[\gamma^2 + \Delta^2]^2}$$

Effective Langevin equation

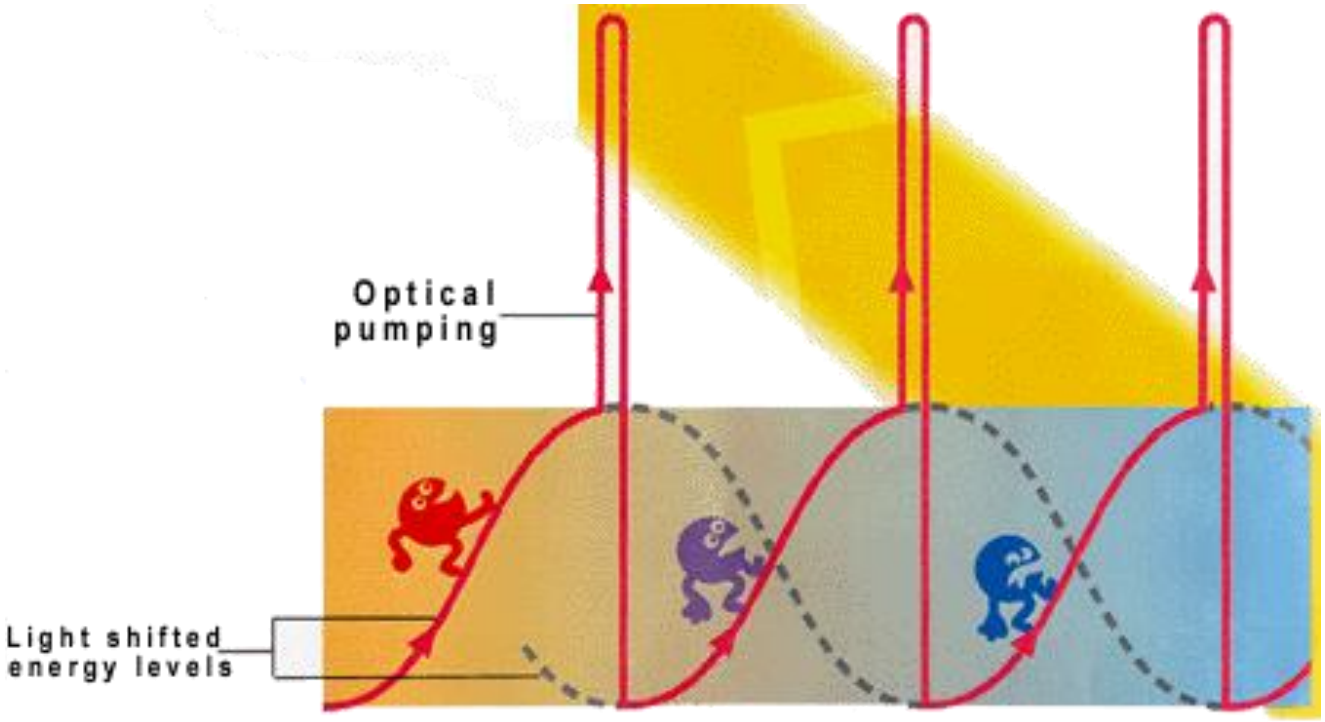
$$\frac{d\hat{p}}{dt} = -f(\hat{p}) + \hat{p}_{in}$$

Polarization gradient cooling force



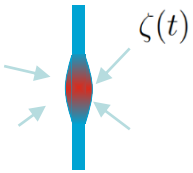
H.J. Metcalf, P. van der Straten, Laser Cooling and Trapping, Springer, 1999

More complicated cooling schemes (Sysiphus cooling)



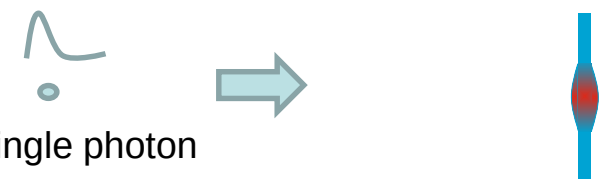
Light can control motion even at the macroscale !

Macroscopic mechanical resonator



Can it be brought close to the quantum ground state of vibrations?

Free space



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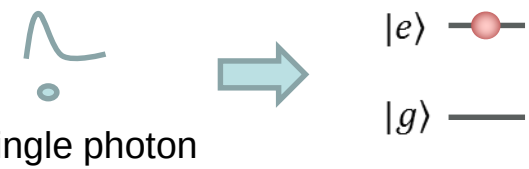
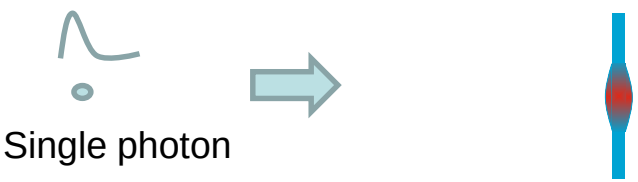
- ✓ Single pass
- ✓ Weak interaction



- *Atoms*
- *Fluorescent molecules*
- *Ions*
- *Quantum dots*

Free space



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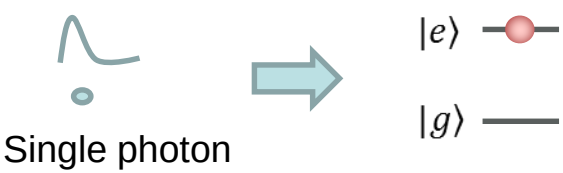

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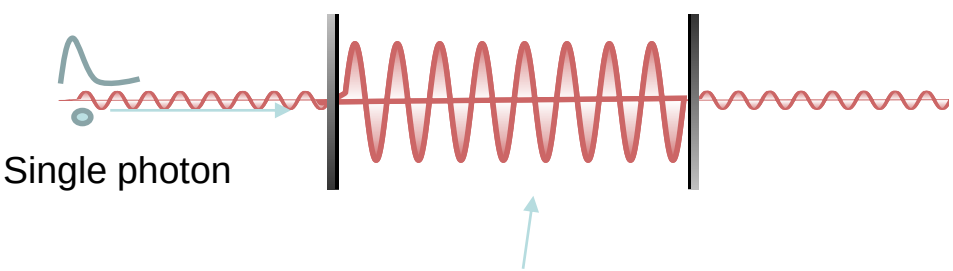
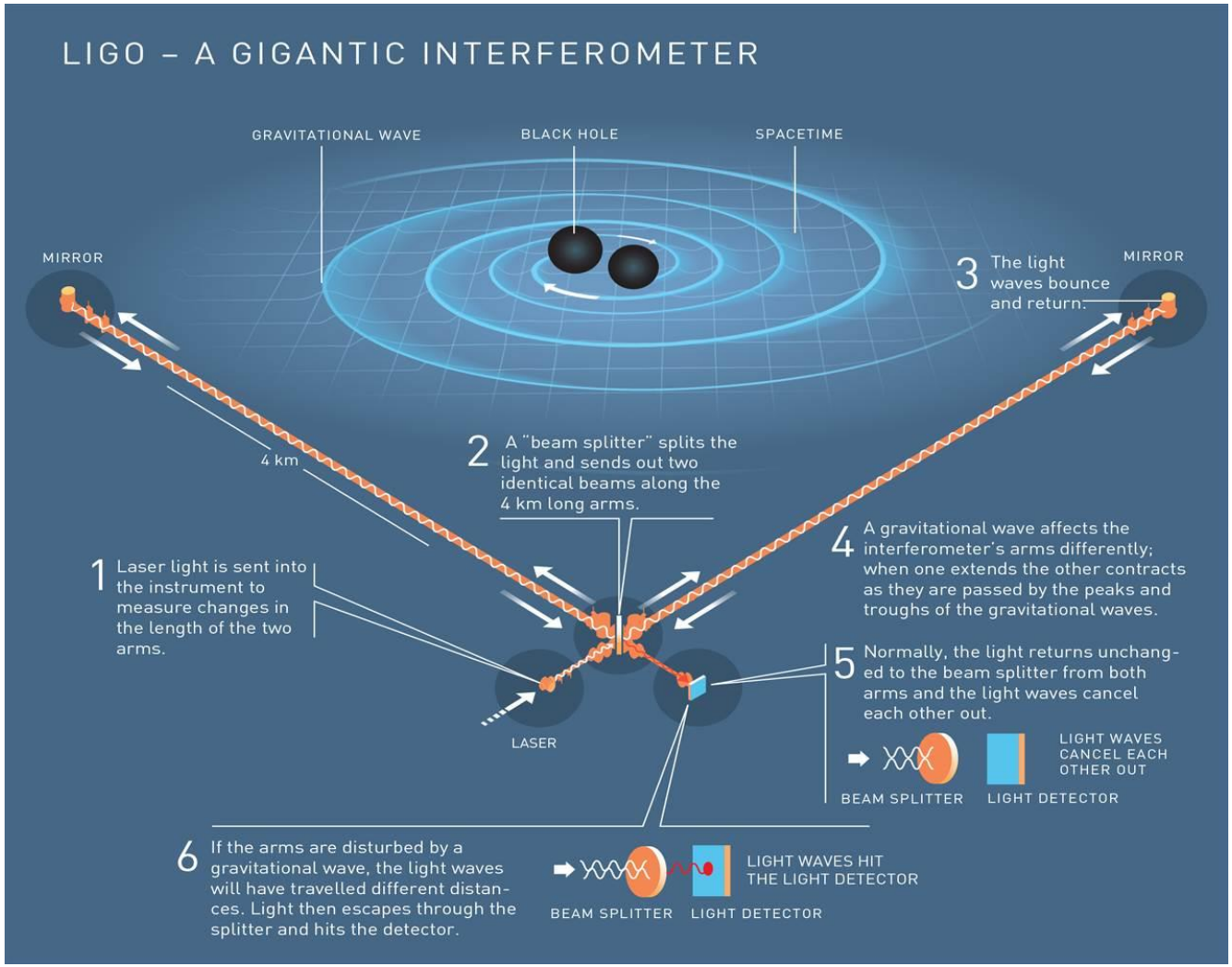
Optical cavity



Standing wave modes (like on a guitar string)

- ✓ Multiple bounces (100000 or more)
- ✓ Stronger interaction

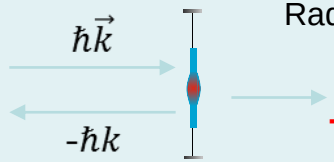
Original motivation: improving precision in the detection of gravitation waves



Nobel Prize 2017: Kip Thorne, Barry Barish, Rainer Weiss

Goal: use light to control motion of massive mechanical resonators at the quantum level.

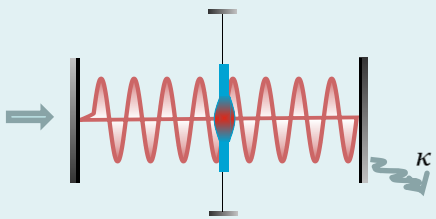
Free space



Radiation pressure force

Tiny effect

Cavity enhancement

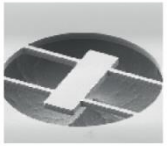


- Dynamical system – back action of motion onto the cavity field
- Time-delayed equations
- Both amplification and cooling of mechanical motion achievable
- Nonlinear dynamics – chaos, self-oscillations

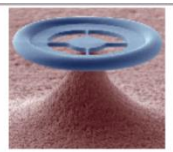
kg

- LIGO, VIRGO
- Detection of gravitational waves

Towards nano-optomechanics

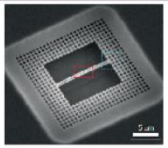


Suspended micro-mirrors



Microtoroid

Suspended membrane



Photonic crystal nano beam (1 D)

Basic science

- Testing the classical-quantum boundary at the large mass scale

Technology

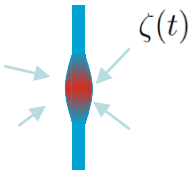
- Ultra-sensitive displacement detection
- inertial sensors

Cavity Optomechanics, M. Aspelmeyer, T. Kippenberg, and F. Marquardt, Phys. Rev. Mod. (2014)

A macroscopic resonator in a thermal environment

$$\dot{q} = \omega_m p,$$

$$\dot{p} = -\gamma_m p - \omega_m q + \zeta(t)$$



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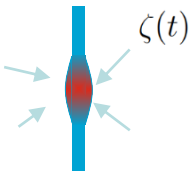
Stochastic noise term

Induced damping



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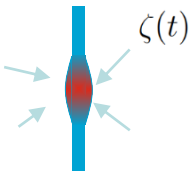

Noise correlations

Thermal noise spectrum

$$\langle \zeta(t)\zeta(t')\rangle = \frac{\gamma_m}{\omega_m}\int_{-\infty}^{\infty} d\omega e^{-i\omega(t-t')}\omega\left[\coth\frac{\hbar\omega}{2k_BT}+1\right] = \frac{1}{2\pi}\int_{-\infty}^{\infty} d\omega e^{-i\omega(t-t')}S_{th}(\omega)$$

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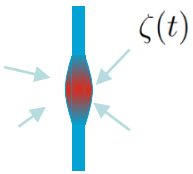

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$$\begin{cases} S_{th}(\omega_m) = 2\gamma_m(\bar{n}+1) \\ S_{th}(-\omega_m) = 2\gamma_m\bar{n} \\ \bar{n} \simeq \dfrac{k_BT}{\hbar\omega_m} \end{cases}$$

$$\gamma_m = \frac{S_{th}(+\omega_m) - S_{th}(-\omega_m)}{2}$$

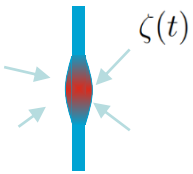
$$\bar{n} = \frac{S_{th}(-\omega_m)}{2\gamma_m}$$

A macroscopic resonator in a thermal environment

$$\dot{q} = \omega_m p,$$
$$\dot{p} = -\gamma_m p - \omega_m q + \zeta(t)$$

Stochastic noise term

Induced damping



Equipartition!

$$\langle q^2(t)\rangle = \bar{n} + \frac{1}{2}$$
$$\langle p^2(t)\rangle = \bar{n} + \frac{1}{2}$$

Noise correlations

Thermal noise spectrum

$$\langle \zeta(t)\zeta(t')\rangle = \frac{\gamma_m}{\omega_m}\int_{-\infty}^{\infty} d\omega e^{-i\omega(t-t')}\omega\left[\coth\frac{\hbar\omega}{2k_BT}+1\right] = \frac{1}{2\pi}\int_{-\infty}^{\infty} d\omega e^{-i\omega(t-t')}S_{th}(\omega)$$

$$\begin{cases} S_{th}(\omega_m) = 2\gamma_m(\bar{n}+1) \\ S_{th}(-\omega_m) = 2\gamma_m\bar{n} \\ \bar{n} \simeq \dfrac{k_BT}{\hbar\omega_m} \end{cases}$$

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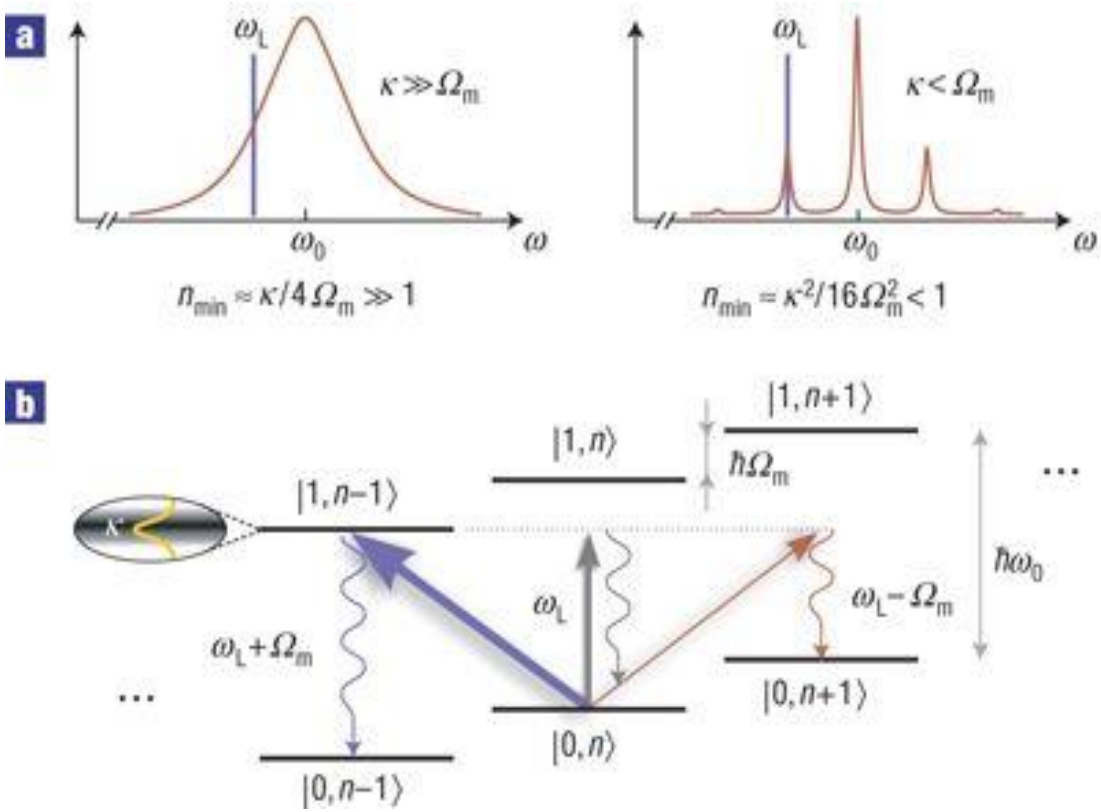
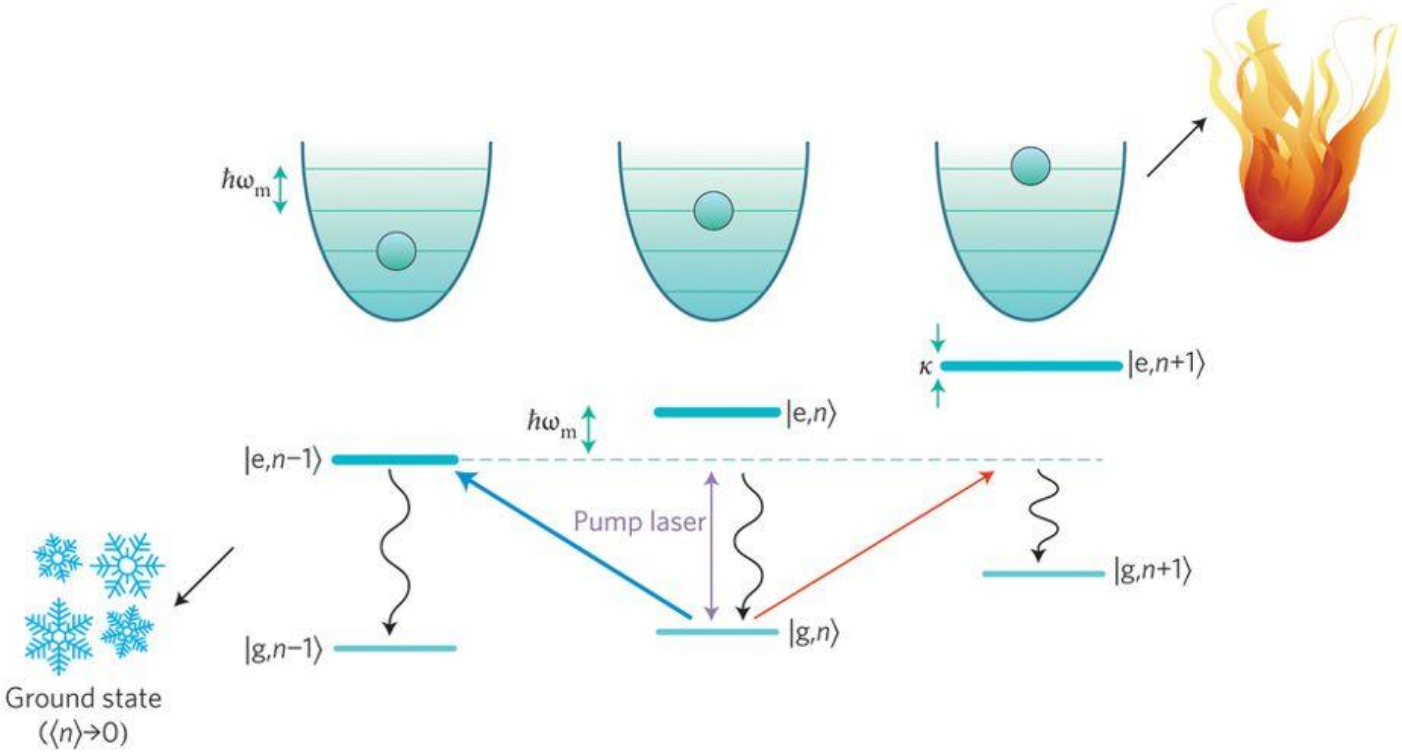
$$\bar{n} = \frac{S_{th}(-\omega_m)}{2\gamma_m}$$



Resolved-sideband cooling of a micromechanical oscillator, A. Schliesser, R. Rivière, G. Anetsberger, O. Arcizet & T. J. Kippenberg, *Nature Physics*, **4**, 415 (2008)

Cooling scheme analogous to sideband cooling of trapped ions!



Frozen Motion, Ania Bleszynski Jayich, News and Views, Nature Physics 11, 710, 2015

Fundamentals of light-matter interactions

- The two-level system approximation
- Light-matter interaction – dipole and rotating wave approximations
- The master equation for spontaneous emission
- Solving the master equation – optical Bloch equations
- Fundamentals of cavity quantum electrodynamics – the Jaynes Cummings Hamiltonian and quantum Langevin equations

The laser

- Laser threshold
- Laser photon statistics

Effects of light onto motion

- The full quantum optical force
- Doppler cooling, gradient cooling
- Ion traps, ion cooling, ion logic
- Cavity optomechanics

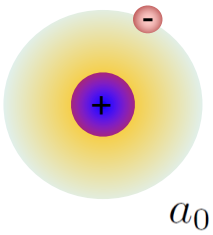
Superradiance/subradiance

- Collective decay of coupled quantum emitters
- Subradiantly enhanced energy transfer

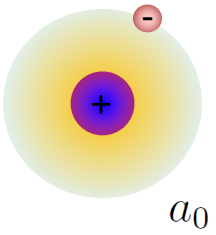
Molecules: vibronic coupling

- The Holstein Hamiltonian
- Emission and absorption spectra, energy transfer

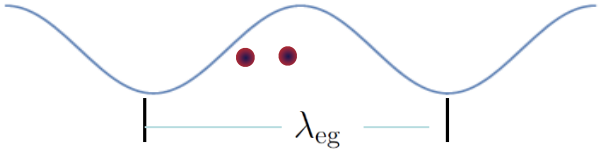
Atomic, molecular system 1



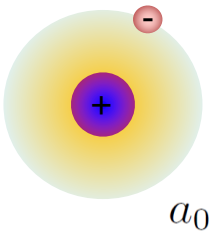
Atomic, molecular system 2



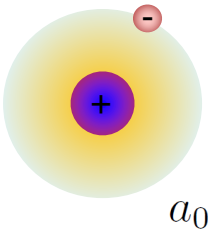
How do atoms decay when they ***see*** each other?



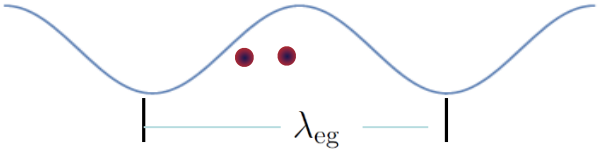
Atomic, molecular system 1



Atomic, molecular system 2



Answer: faster (**superradiance**) or slower (**subradiance**) than independent atoms



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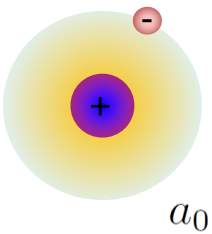
- Subradiantly enhanced energy transfer

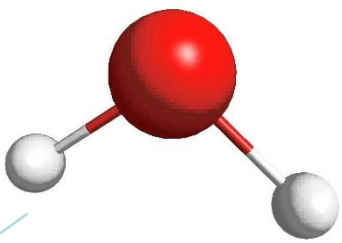
Molecules: vibronic coupling

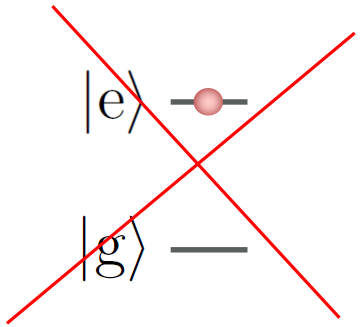
- The Holstein Hamiltonian
- Emission and absorption spectra, energy transfer

Atoms: complicated fine and hyperfine structure

Molecules: fundamentally more complex

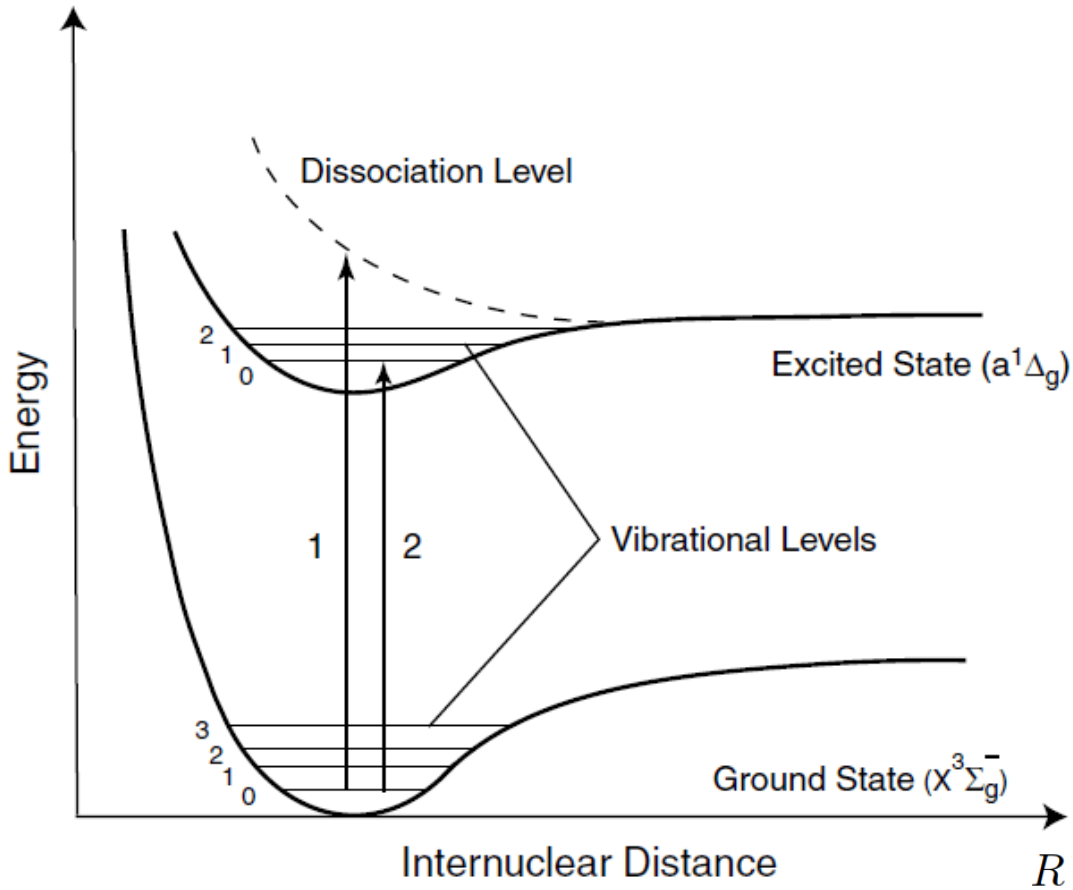




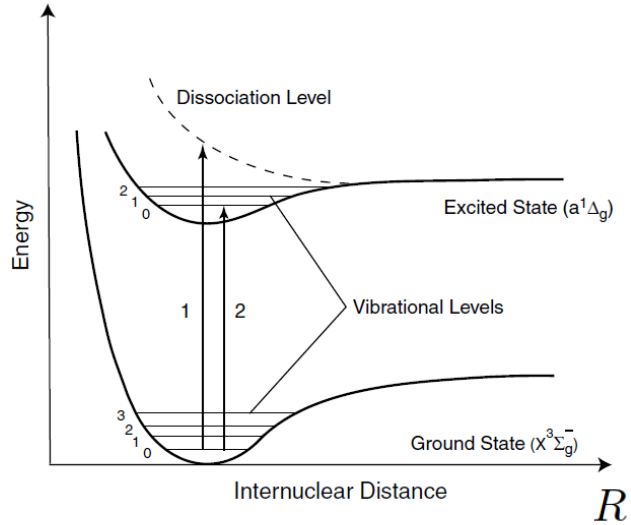


Not always a good enough model

A simple model - diatomic molecules

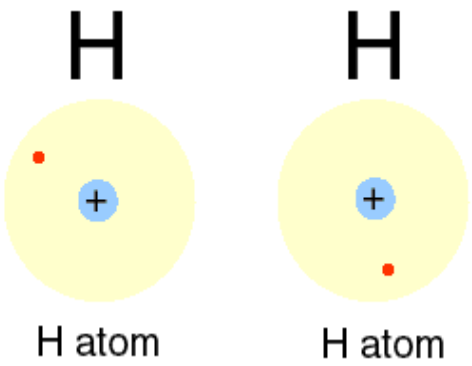


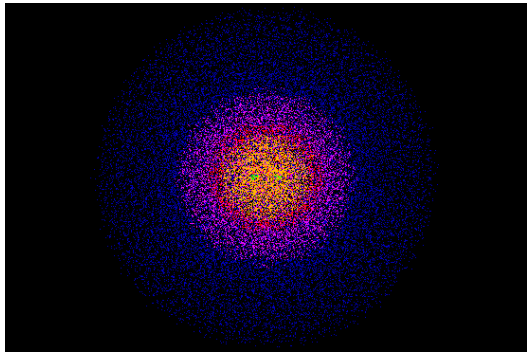
A simple model for electronic to vibrations coupling in diatomic molecules

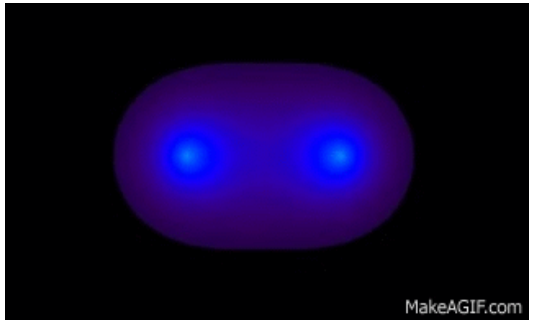


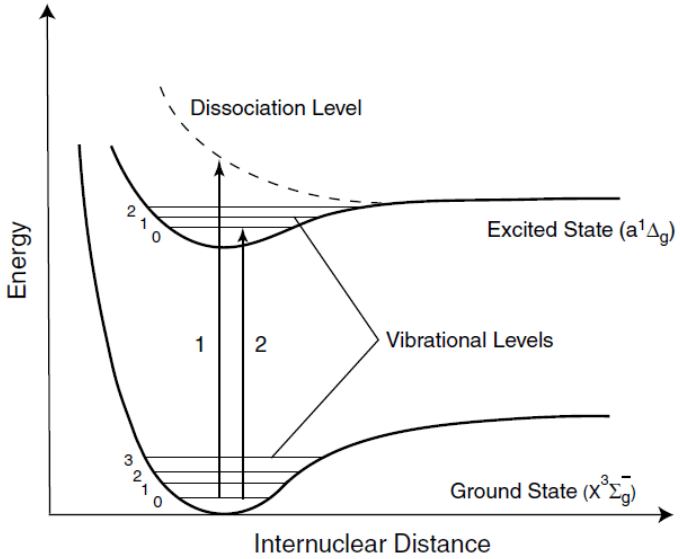
$$H = \left(\omega_e + \frac{p^2}{2\mu} + \frac{1}{2}\mu\nu^2\left(R - R_e\right)^2\right)\sigma^\dagger\sigma + \left(\frac{p^2}{2\mu} + \frac{1}{2}\mu\nu^2\left(R - R_g\right)^2\right)\sigma\sigma^\dagger$$

Molecular systems – coupling to vibrations

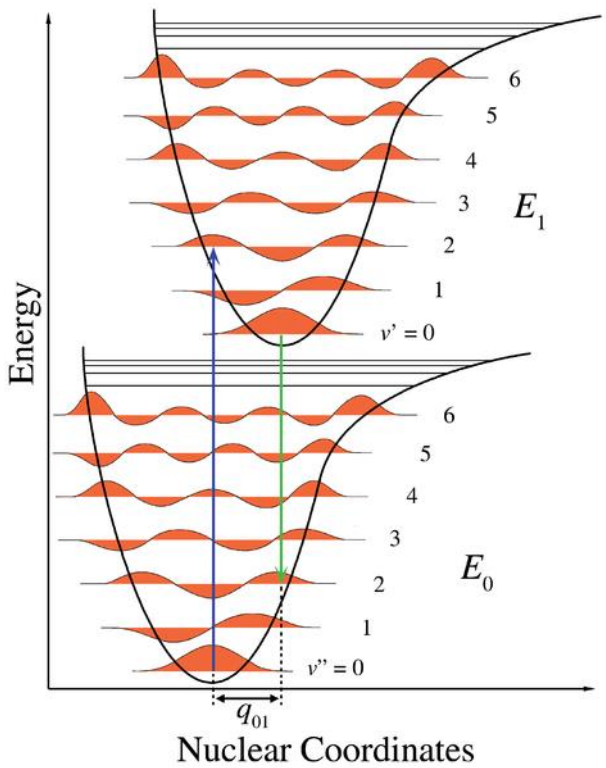


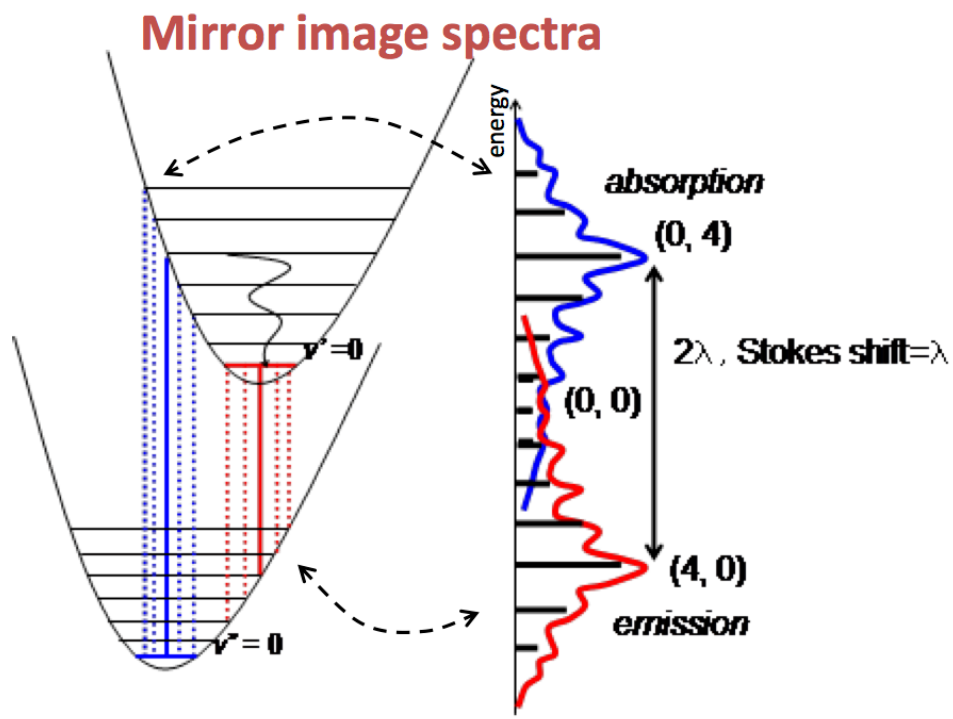






Molecular systems – coupling to vibrations





$$H = \nu b^\dagger b + (\omega_e + \lambda^2 \nu)\sigma^\dagger \sigma - \lambda\nu(b^\dagger + b)\sigma^\dagger \sigma$$

Holstein Hamiltonian

Förster resonance energy transfer

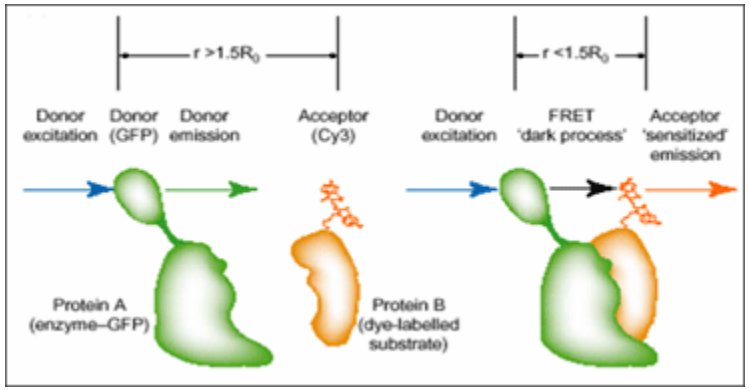
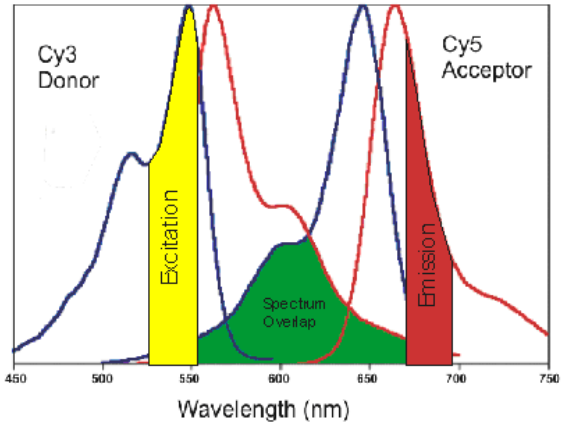
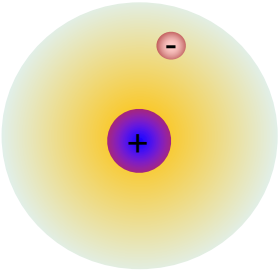


Fig.1 The acceptor and donor fluorophores must be closer than ~ 10 nm for the energy transfer to occur
(adopted from P. I.H. Bastiaens and R. Pepperkok, TIBS 25 (2000) 631).



An Introduction to Fluorescence Resonance Energy Transfer (FRET) Technology and its Application in Bioscience, *Paul Held, Ph.D, Applications Department, BioTek Instruments (2005)*

The two level system
Interaction with a classical light field



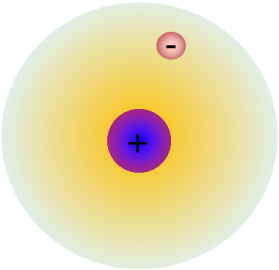
Simplifying assumptions:

- nucleus is heavy and therefore fixed in the origin
- non-relativistic description (Schrödinger equation suffices)
- spinless electron and proton

$$i\hbar\frac{\partial}{\partial t}|\psi\rangle = H|\psi\rangle$$

Radially symmetric potential (Coulomb) – exactly solvable model

$$H = \frac{\hat{\boldsymbol{p}}^2}{2\mu} + V(|\hat{\boldsymbol{r}}|)$$



In Dirac (ket) notation Hamiltonian is diagonalized

$$H = \sum E_{nlm} |nlm\rangle\langle nlm|$$

$$\begin{cases} E_n = -\left[\dfrac{\mu e^4}{32\pi^2\epsilon_0^2\hbar^2}\right]\dfrac{1}{n^2} \\ \\ (1 \leq n < \infty, 0 \leq \ell \leq n-1, -\ell \leq m \leq \ell) \end{cases}$$

$$|100\rangle \qquad |200\rangle \qquad \{|21-1\rangle \pm |211\rangle\} \qquad |210\rangle$$

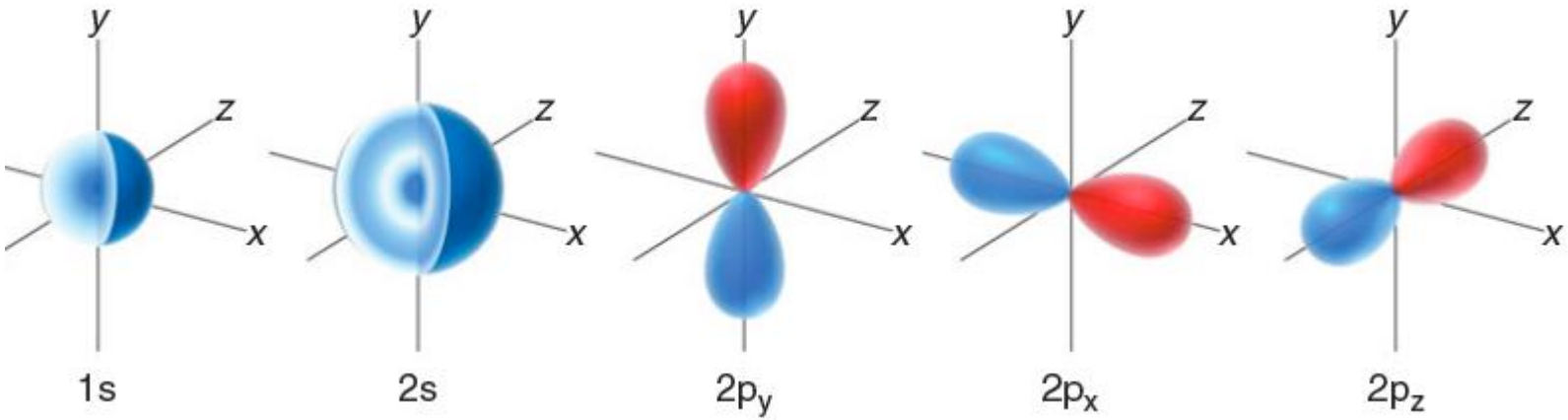
In the position representation

$$i\hbar\frac{\partial}{\partial t}\psi(\boldsymbol{r},t) = \left[-\frac{\hbar^2}{2\mu}\nabla^2 + V(r)\right]\psi(\boldsymbol{r},t)$$

Solutions

$$\psi_{nlm}(r,\theta,\phi) = \langle \boldsymbol{r}|nlm\rangle = R_{nl}(r)Y_{l,m}(\theta,\phi).$$

$|100\rangle$ $|200\rangle$ $\{|21-1\rangle \pm |211\rangle\}$ $|210\rangle$



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Solutions

$$\psi_{nlm}(r,\theta,\phi) = \langle \boldsymbol{r}|nlm\rangle = R_{nl}(r)Y_{l,m}(\theta,\phi).$$

Propagating field

$$\boldsymbol{E}(x,y,z,t) = E_L f(x,y)\cos(k_L z - \omega_L t)\hat{\boldsymbol{\epsilon}}_z = \frac{iE_L}{2}(e^{ik_L z}e^{-i\omega_L t} - e^{-ik_L z}e^{i\omega_L t})\hat{\boldsymbol{\epsilon}}_z$$

Propagating field

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Interaction Hamiltonian (in the dipole approximation)

Classical minimal coupling Hamiltonian

$$H(\boldsymbol{r},t) = \frac{1}{2m}(\boldsymbol{p} + e\boldsymbol{A}(\boldsymbol{r},t))^2 - e\Phi(\boldsymbol{r},t) + V(r)$$

Vector potential

Scalar potential

Propagating field

$$\boldsymbol{E}(x,y,z,t) = E_L f(x,y)\cos(k_L z - \omega_L t)\hat{\boldsymbol{\epsilon}}_z = \frac{iE_L}{2}(e^{ik_L z}e^{-i\omega_L t} - e^{-ik_L z}e^{i\omega_L t})\hat{\boldsymbol{\epsilon}}_z$$

Interaction Hamiltonian (in the dipole approximation)

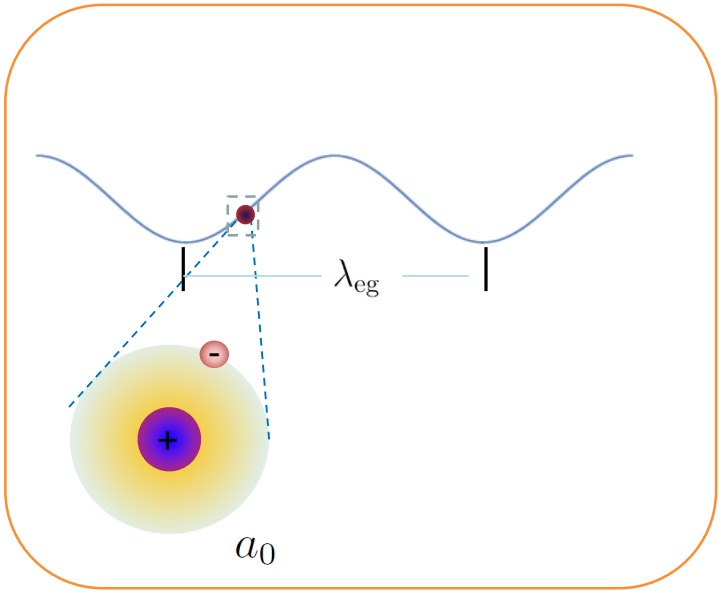
Classical minimal coupling Hamiltonian

$$H(\boldsymbol{r},t) = \frac{1}{2m}(\boldsymbol{p} + e\boldsymbol{A}(\boldsymbol{r},t))^2 - e\Phi(\boldsymbol{r},t) + V(r)$$

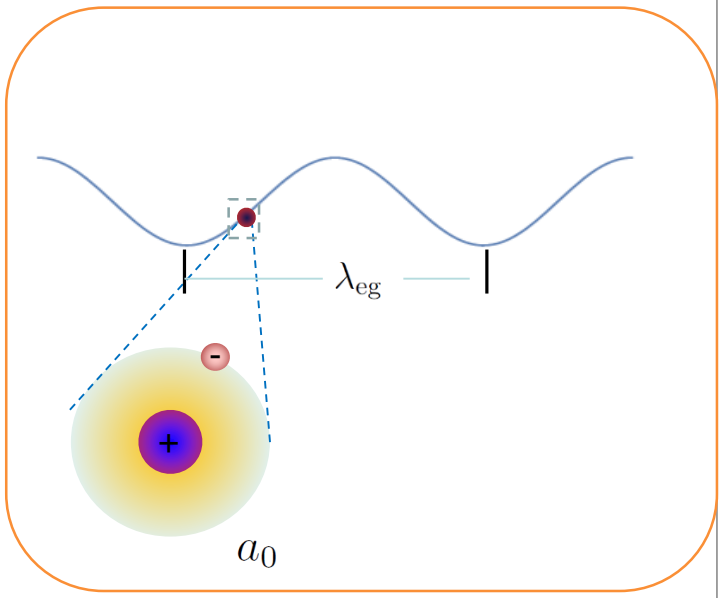
Length gauge transformation + quantization

Dipole Hamiltonian

$$H = \frac{\hat{\boldsymbol{p}}^2}{2\mu} + V(|\hat{\boldsymbol{r}}|) - \hat{\boldsymbol{d}}\cdot\boldsymbol{E}(t)$$



A bit more on the Hamiltonian in the dipole approximation



$$H(\boldsymbol{r},t) = \frac{1}{2m}(\boldsymbol{p} + e\boldsymbol{A}(\boldsymbol{r},t))^2 - e\Phi(\boldsymbol{r},t) + V(r)$$

$$\mathbf{A}(\mathbf{r},t) = \mathbf{A}(t)$$

$$H(\mathbf{r},t) = \frac{\mathbf{p}^2}{2m} + \frac{e}{m}\mathbf{A}(t)\cdot\mathbf{p} + \frac{e^2}{2m}\mathbf{A}^2(t) + V(r)$$

Gauge transformation with $\chi(\mathbf{r},t) = -\mathbf{A}(t)\cdot\mathbf{r}$

$$\begin{cases} \Phi'(\mathbf{r},t) = -\dfrac{\partial\chi(\mathbf{r},t)}{\partial t} = -\dfrac{\partial\mathbf{A}(t)}{\partial t}\mathbf{r} = \mathbf{r}\cdot\mathbf{E}(t) \\ \mathbf{A}'(\mathbf{r},t) = \mathbf{A}(t) + \nabla\chi(\mathbf{r},t) = \mathbf{A}(t) - \nabla(\mathbf{A}(t)\cdot\mathbf{r}) = 0 \end{cases}$$

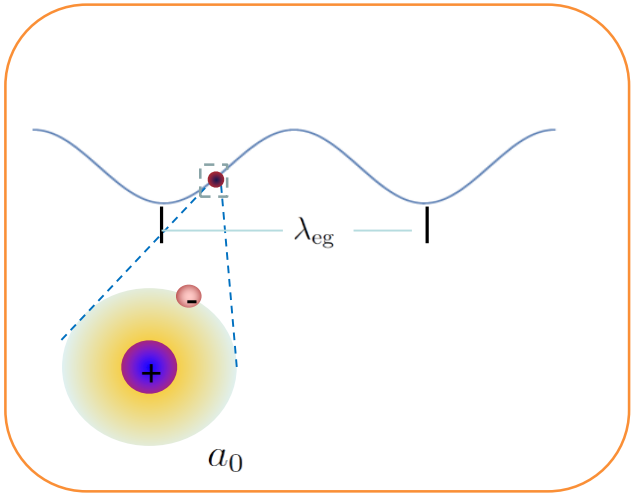
$$H' = \frac{\mathbf{p}^2}{2m} + V(r) - \mathbf{d}\cdot\mathbf{E}(t)$$

In more detail

Dipole Hamiltonian

$$H = \frac{\hat{\boldsymbol{p}}^2}{2\mu} + V(|\hat{\boldsymbol{r}}|) - \hat{\boldsymbol{d}} \cdot \boldsymbol{E}(t)$$

$$-\hat{\boldsymbol{d}} \cdot \boldsymbol{E}(t) = -\frac{iE_L}{2}(e^{-i\omega_L t} - e^{i\omega_L t})\,[-e\hat{\boldsymbol{r}}] \cdot \hat{\boldsymbol{\epsilon}}_z$$



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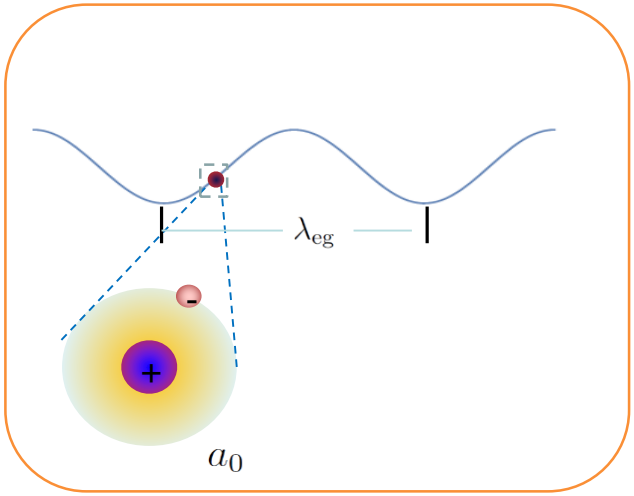


Expanding the dipole operator

$$[e\hat{\boldsymbol{r}}] \cdot \hat{\boldsymbol{\epsilon}}_z = I_\infty\,[e\hat{z}]\,I_\infty = \sum |nlm\rangle\langle nlm|\,[e\hat{z}]\,|n'l'm'\rangle\langle n'l'm'|$$

Dipole matrix elements

Computing dipole matrix elements

$$[e\hat{\boldsymbol{r}}] \cdot \hat{\boldsymbol{\epsilon}}_z = I_\infty \, [e\hat{z}] \, I_\infty = \sum |nlm\rangle\langle nlm| \, [e\hat{z}] \, |n'l'm'\rangle\langle n'l'm'|$$

$$\langle nlm|\hat{z}|n'l'm'\rangle = \langle nlm| \underbrace{\left[\int d\boldsymbol{r}|\boldsymbol{r}\rangle\langle\boldsymbol{r}|\right]}_{\text{unity}} \hat{z} \underbrace{\left[\int d\boldsymbol{r'}|\boldsymbol{r'}\rangle\langle\boldsymbol{r'}|\right]}_{\text{unity}} |n'l'm'\rangle = \int d\boldsymbol{r}\psi^*_{nlm}(\boldsymbol{r}) z \psi_{n'l'm'}(\boldsymbol{r})$$

Computing dipole matrix elements

$$[e\hat{\boldsymbol{r}}]\cdot\hat{\boldsymbol{\epsilon}}_z = I_\infty\,[e\hat{z}]\,I_\infty = \sum |nlm\rangle\langle nlm|\,[e\hat{z}]\,|n'l'm'\rangle\langle n'l'm'|$$

$$\langle nlm|\hat{z}|n'l'm'\rangle = \langle nlm|\underbrace{\left[\int d\boldsymbol{r}|\boldsymbol{r}\rangle\langle\boldsymbol{r}|\right]}_{\text{unity}}\hat{z}\underbrace{\left[\int d\boldsymbol{r}'|\boldsymbol{r}'\rangle\langle\boldsymbol{r}'|\right]}_{\text{unity}}|n'l'm'\rangle = \int d\boldsymbol{r}\psi^*_{nlm}(\boldsymbol{r})z\psi_{n'l'm'}(\boldsymbol{r})$$

Example: 1s to 2p dipole transitions

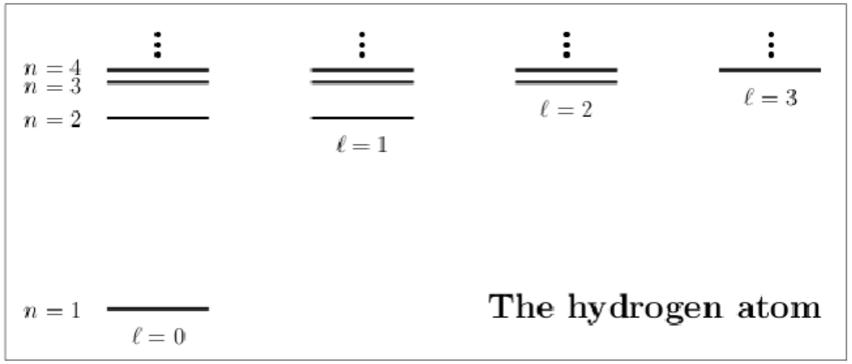
$$\langle 100|\hat{z}|21m\rangle = \int\int\int drd\theta d\phi(r^2\sin\theta)\left[2\frac{1}{a^{3/2}}e^{-r/2a}\frac{1}{\sqrt{4\pi}}\right]z\left[\frac{1}{8\sqrt{3}}\frac{1}{a^{3/2}}\frac{2r}{2a}e^{-r/2a}Y_{1,m}(\theta,\phi)\right] = 2a\delta_{m0}$$

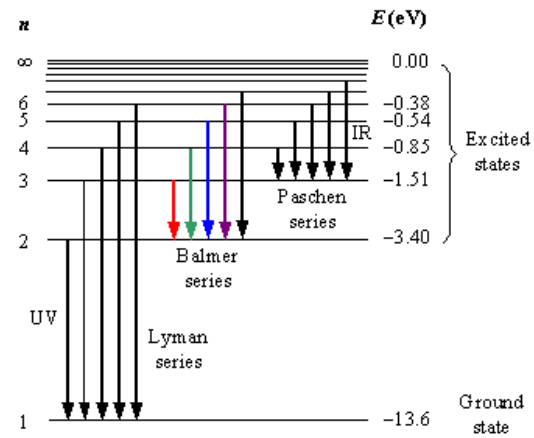
Only 1s to 2p$_z$ transitions allowed

Fulfilling the resonance condition

$$H_{\text{int}}(t) = -\hat{\boldsymbol{d}}(t)\cdot\boldsymbol{E}(t) \propto \langle nlm|\,[\hat{z}]\,|n'l'm'\rangle e^{-i(E_{nlm}-E_{n'l'm'})t} e^{-i\omega_L t} + \ldots$$

Field frequency should fit energetically the energy level difference – resonance condition

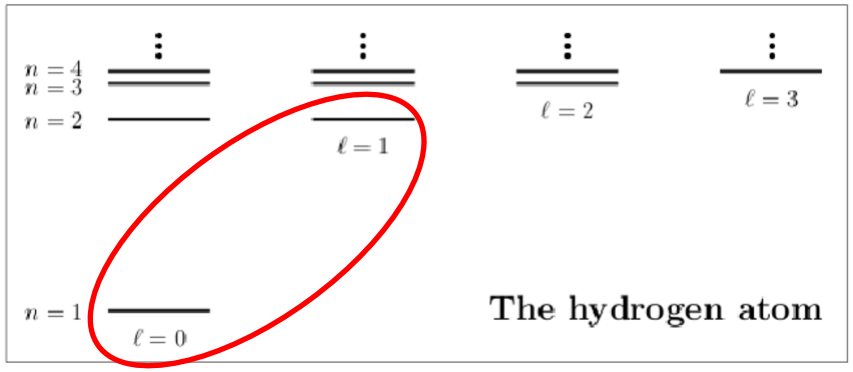




Energy levels of the hydrogen atom with some of the transitions between them that give rise to the spectral lines indicated.

Putting it all together - Roadmap

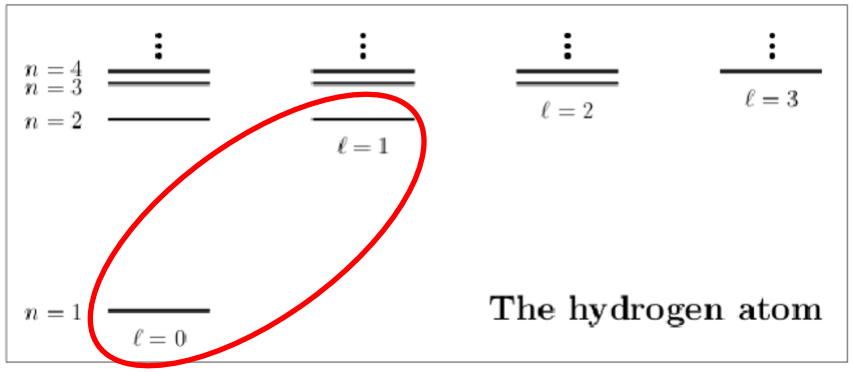
1. Shine a laser field of a given frequency (resonance condition)



Putting it all together - Roadmap

1. Shine a laser field of a given frequency (resonance condition)

2. Choose a given laser polarization (for example linearly polarized in the z direction) (selection rules for dipole-allowed transitions)

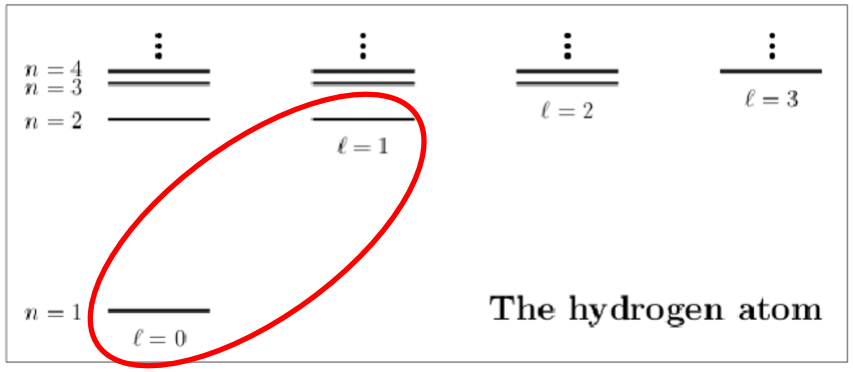


only m=0 allowed

Putting it all together - Roadmap

1. Shine a laser field of a given frequency (resonance condition)

2. Choose a given laser polarization (for example linearly polarized in the z direction) (selection rules for dipole-allowed transitions)



only m=0 allowed

Conclusion

$$|100\rangle\langle 100| + \cancel{|200\rangle\langle 200|} + |21-1\rangle\cancel{\langle 21-1|} + |210\rangle\langle 210| + \cancel{|211\rangle\langle 211|} + \cancel{|300\rangle\langle 300|} + \ldots = \text{Unity}$$

Reduced dynamics in a two-state Hilbert space suffices!

$$\{|100\rangle, |210\rangle\} \rightarrow \{|g\rangle, |e\rangle\}$$

The two level system

Ladder operators

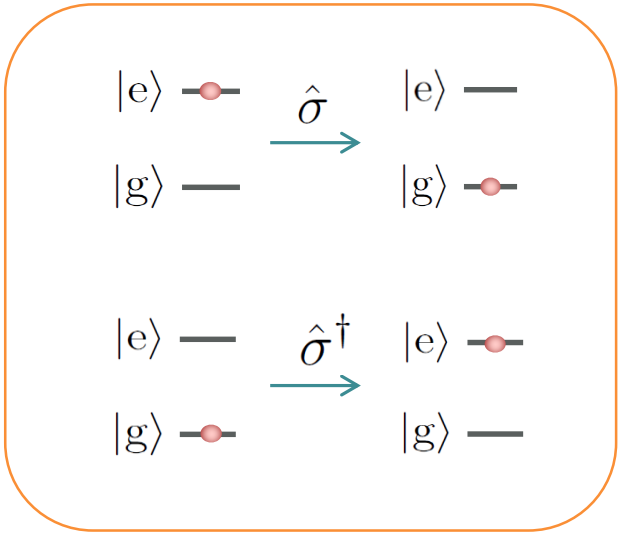
$$\sigma = |g\rangle\langle e| \qquad \sigma^\dagger = |e\rangle\langle g|$$

Projectors

$$\sigma^\dagger\sigma = |e\rangle\langle e| \qquad \sigma\sigma^\dagger = |g\rangle\langle g|$$

Complete system – projectors add to unity

$$\sigma^\dagger\sigma + \sigma\sigma^\dagger = 1$$



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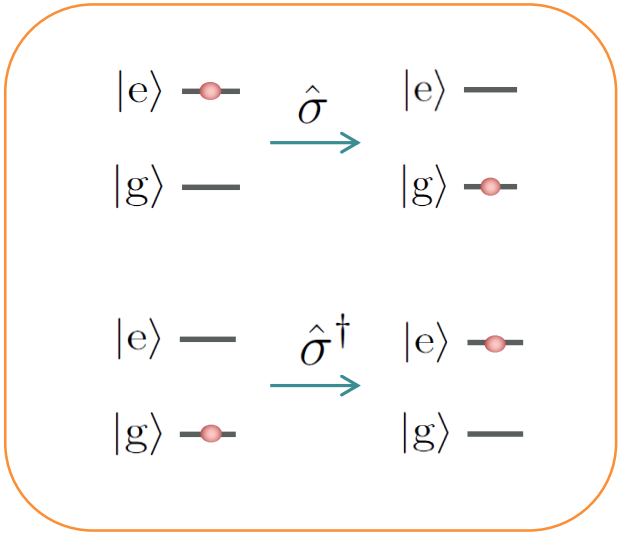

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Free Hamiltonian

$$H = \omega_e|e\rangle\langle e| + \omega_g|g\rangle\langle g| = \omega_e\sigma^\dagger\sigma + \omega_g(1 - \sigma^\dagger\sigma) = \omega_{eg}\sigma^\dagger\sigma + \omega_g 1$$

Constant energy shift - ignore

Dipole operator

$$\hat{\boldsymbol{d}} = [|e\rangle\langle e| + |g\rangle\langle g|]\,\hat{\boldsymbol{d}}\,[|e\rangle\langle e| + |g\rangle\langle g|] = \boldsymbol{d}_{eg}\left[\sigma + \sigma^{\dagger}\right]$$

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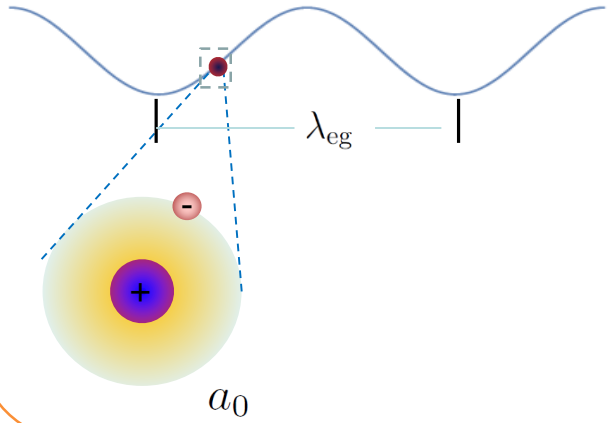
Interaction in the dipole approximation

$$H_{int} = -\boldsymbol{d}_{eg} \cdot \boldsymbol{E}(t)\left[\sigma + \sigma^\dagger\right]$$

$$E(t) = E_0 \sin(\omega_L t)\hat{\epsilon} = \frac{iE_0}{2}(e^{-i\omega_L t} - e^{i\omega_L t})\hat{\epsilon}.$$

$a_0 \ll \lambda_{\mathrm{eg}} \longrightarrow \mathrm{H}_{\mathrm{int}} = -\hat{\mathbf{d}} \cdot \hat{\mathbf{E}}$



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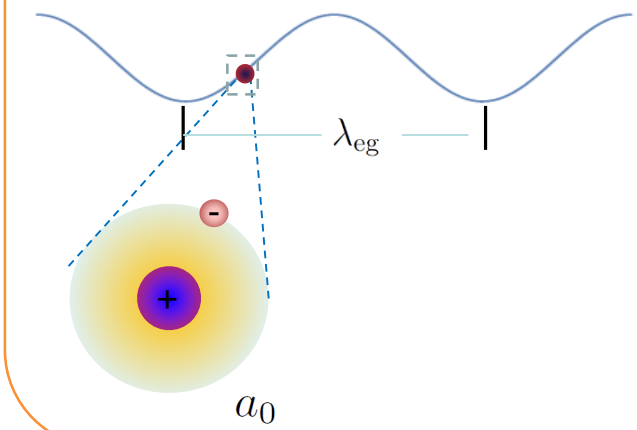

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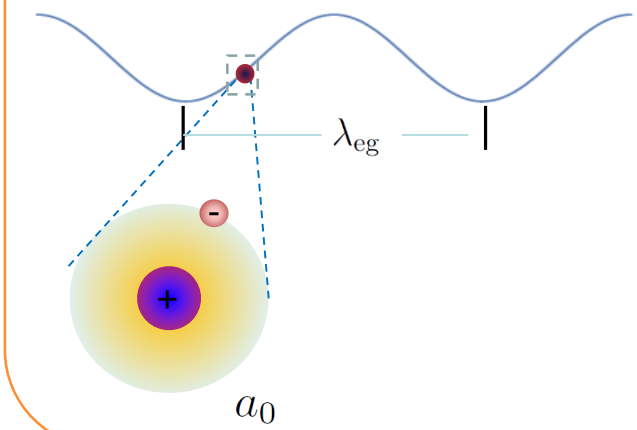

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Free operator evolution

$$\sigma(t) = e^{iH_0 t}\sigma e^{-iH_0 t} = \sigma e^{-i\omega_{eg} t}$$

Dipole operator

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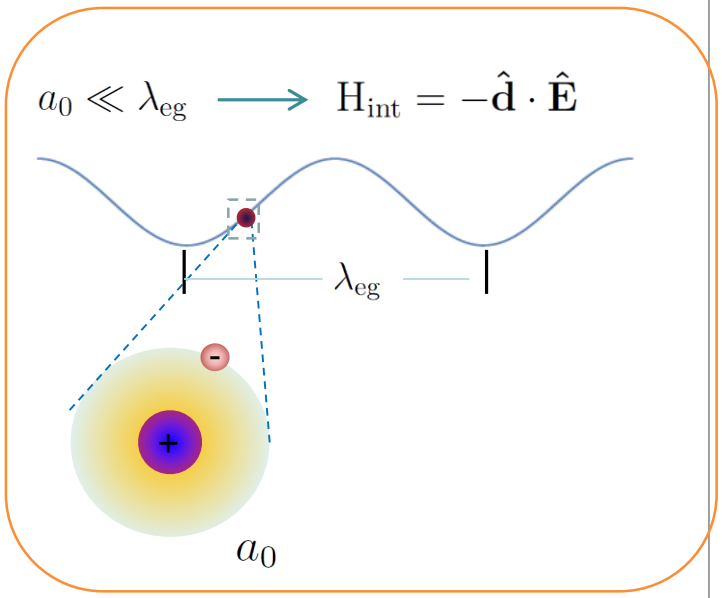

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Free operator evolution

$$\sigma(t) = e^{iH_0 t}\sigma e^{-iH_0 t} = \sigma e^{-i\omega_{eg} t}$$

Fast rotating terms. Discard within the Rotating wave approximation (RWA)

Result

$$H = \hbar\omega_{eg}\sigma^\dagger\sigma + i\Omega\left[\sigma e^{-i\omega_L t} - \sigma^\dagger e^{i\omega_L t}\right]$$

Rabi frequency

$$\Omega = \frac{iE_0\boldsymbol{d}_{eg}\cdot\hat{\epsilon}}{2\hbar}$$

Approximations made on the way

- Imposing resonance and selection rules – reduction to a two-level system
- The dipole approximation (long wavelength approximation)
- Rotating wave approximation (discarding energy non-conserving terms)

HAVE A NICE WEEKEND!

(…after exercise class of course)

Maxell's equations (no sources)

$$\nabla \cdot \mathbf{E} = 0 \qquad \nabla \times \mathbf{E} = -\partial_t \mathbf{B},$$

$$\nabla \cdot \mathbf{B} = 0 \qquad \nabla \times \mathbf{B} = \frac{1}{c}\partial_t \mathbf{E},$$

Vector potential

$$\mathbf{E} = -\partial_t \mathbf{A} \qquad \mathbf{B} = \nabla \times \mathbf{A},$$

Coulomb gauge

$$\nabla \cdot \mathbf{A} = 0$$

Wave equation

$$\nabla^2 \mathbf{A}(\mathbf{r},t) = \frac{1}{c^2}\frac{\partial^2 \mathbf{A}(\mathbf{r},t)}{\partial t^2}$$

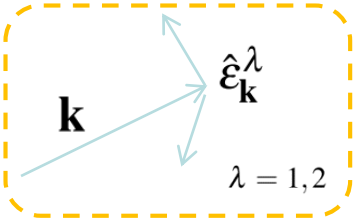
with $\nabla^2 \mathbf{A} = \nabla(\nabla \cdot \mathbf{A}) - \nabla \times \nabla \times \mathbf{A}$

Wave equation

$$\nabla^2 \mathbf{A}(\mathbf{r},t) = \frac{1}{c^2}\frac{\partial^2 \mathbf{A}(\mathbf{r},t)}{\partial t^2}$$

Solutions

$$\mathbf{A}^{(+)}(\mathbf{r},t) = i\sum_{\mathbf{k}} c_{\mathbf{k}}\mathbf{u}_{\mathbf{k}}(\mathbf{r})e^{-i\omega_k t}$$



$$\left[\nabla^2 + \frac{\omega_k^2}{c^2}\right]\mathbf{u}_{\mathbf{k}}(\mathbf{r}) = 0$$