

# Quantum optics of macroscopic systems

## QED in dielectrics

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Summer School on Modern Quantum Technologies



# Quantum optics of macroscopic systems

## Menu

### Outline:

- field quantization in free space
- open quantum systems
- fluctuation-dissipation theorem
- field quantization in absorbing dielectrics

# Quantum optics of macroscopic systems

## Maxwell's equations in free space

### Maxwell's equations:

$$\begin{aligned}\nabla \cdot \mathbf{B}(\mathbf{r}) &= 0 & \nabla \times \mathbf{E}(\mathbf{r}) &= -\dot{\mathbf{B}}(\mathbf{r}) \\ \nabla \cdot \mathbf{E}(\mathbf{r}) &= 0 & \nabla \times \mathbf{B}(\mathbf{r}) &= \frac{1}{c^2} \dot{\mathbf{E}}(\mathbf{r})\end{aligned}$$

- introduce vector potential in Coulomb gauge  $\mathbf{B}(\mathbf{r}) = \nabla \times \mathbf{A}(\mathbf{r})$  and  $\mathbf{E}(\mathbf{r}) = -\dot{\mathbf{A}}(\mathbf{r})$  with  $\nabla \cdot \mathbf{A}(\mathbf{r}) = 0$  (transversality condition)

$$\Delta \mathbf{A}(\mathbf{r}, t) = -\frac{1}{c^2} \ddot{\mathbf{A}}(\mathbf{r}, t)$$

- separation of variables (mode decomposition)

$$\mathbf{A}(\mathbf{r}, t) = \sum_{\lambda} \mathbf{A}_{\lambda}(\mathbf{r}) u_{\lambda}(t)$$

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### wave equation for vector potential

$$\Delta \mathbf{A}(\mathbf{r}, t) - \frac{1}{c^2} \ddot{\mathbf{A}}(\mathbf{r}, t) = 0$$

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# Quantum optics of macroscopic systems

## Classical Hamiltonian

- Mode decomposition in cartesian coordinates

$$\mathbf{A}(\mathbf{r}, t) = \sum_{\sigma} \int \frac{d^3 k}{(2\pi)^{3/2}} \mathbf{e}_{\sigma} \left[ u_{\mathbf{k}\sigma} e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)} + u_{\mathbf{k}\sigma}^* e^{-i(\mathbf{k} \cdot \mathbf{r} - \omega t)} \right]$$

- classical Hamiltonian

$$H = \frac{1}{2} \int d^3 r \left[ \epsilon_0 \mathbf{E}^2(\mathbf{r}) + \frac{1}{\mu_0} \mathbf{B}^2(\mathbf{r}) \right] = 2\epsilon_0 \sum_{\sigma} \int d^3 k \omega^2 |u_{\mathbf{k}\sigma}|^2$$

- define  $q_{\mathbf{k}\sigma} = \sqrt{\epsilon_0} (u_{\mathbf{k}\sigma} + u_{\mathbf{k}\sigma}^*)$  and  $p_{\mathbf{k}\sigma} = -i\omega \sqrt{\epsilon_0} (u_{\mathbf{k}\sigma} - u_{\mathbf{k}\sigma}^*)$

- Hamiltonian turns into

$$H = \frac{1}{2} \sum_{\sigma} \int d^3 k (p_{\mathbf{k}\sigma}^2 + \omega^2 q_{\mathbf{k}\sigma}^2) \Rightarrow \text{infinite sum of uncoupled harmonic oscillators with frequencies } \omega = |\mathbf{k}|c$$

# Quantum optics of macroscopic systems

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oscillators with frequencies  $\omega = |\mathbf{k}|c$

# Quantum optics of macroscopic systems

## Quantization

- replace  $c$ -number functions  $q_{k\sigma}, p_{k\sigma}$  by operators  $\hat{q}_{k\sigma}, \hat{p}_{k\sigma}$
- postulate equal-time commutation relations  $[\hat{q}_{k\sigma}, \hat{p}_{k'\sigma'}] = i\hbar\delta(\mathbf{k} - \mathbf{k}')\delta_{\sigma\sigma'}$
- define creation and annihilation operators

$$\hat{a}_\sigma(\mathbf{k}) = \sqrt{\frac{\omega}{2\hbar}} \left( \hat{q}_{k\sigma} + \frac{i\hat{p}_{k\sigma}}{\omega} \right), \quad \hat{a}_\sigma^\dagger(\mathbf{k}) = \sqrt{\frac{\omega}{2\hbar}} \left( \hat{q}_{k\sigma} - \frac{i\hat{p}_{k\sigma}}{\omega} \right)$$

with equal-time commutation relations  $[\hat{a}_\sigma(\mathbf{k}), \hat{a}_{\sigma'}^\dagger(\mathbf{k}')] = \delta(\mathbf{k} - \mathbf{k}')\delta_{\sigma\sigma'}$

- shorthand notation  $\lambda \equiv (\mathbf{k}, \sigma) \Rightarrow \hat{\mathbf{A}}(\mathbf{r}) = \sum_{\lambda} [\mathbf{A}_{\lambda}(\mathbf{r})\hat{a}_{\lambda} + \text{h.c.}]$

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# Quantum optics of macroscopic systems

## Quantized Hamiltonian

$\hat{H} = \frac{1}{2} \sum_{\lambda} \hbar \omega_{\lambda} \left( \hat{a}_{\lambda} \hat{a}_{\lambda}^{\dagger} + \hat{a}_{\lambda}^{\dagger} \hat{a}_{\lambda} \right) = \sum_{\lambda} \hbar \omega_{\lambda} \left( \hat{a}_{\lambda}^{\dagger} \hat{a}_{\lambda} + \frac{1}{2} \right)$  contains constant contribution to total field energy

$$E_0 = \sum_{\lambda} \frac{1}{2} \hbar \omega_{\lambda} = \frac{\hbar c}{2} \sum_{\sigma} \int \frac{d^3 k}{(2\pi)^{3/2}} |\mathbf{k}|$$

- ground-state (vacuum) energy
- exists in the absence of any electromagnetic field excitation
- infinitely large due to summation over infinitely many modes

# Quantum optics of macroscopic systems

## Macroscopic Maxwell's equations

naive approach to field quantization in dielectrics with constant refractive index  $n$ :

- constitutive relations  $\mathbf{D}(\mathbf{r}, t) = \varepsilon_0 \varepsilon \mathbf{E}(\mathbf{r}, t)$ ,  $\mathbf{H}(\mathbf{r}, t) = \frac{1}{\mu_0 \mu} \mathbf{B}(\mathbf{r}, t)$
- Helmholtz equation for spatial mode functions in Coulomb gauge  

$$\Delta \mathbf{A}_\lambda(\mathbf{r}) + \frac{n^2 \omega_\lambda^2}{c^2} \mathbf{A}_\lambda(\mathbf{r}) = 0$$
 with plane-wave solutions  $\exp(i n \mathbf{k}_\lambda \cdot \mathbf{r})$

but: refractive index is always a complex function of frequency!

(absorption only negligible in certain frequency intervals, but not for all frequencies)

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# Quantum optics of macroscopic systems

## Kramers–Kronig relations

causal response of linear susceptibilities  $\Rightarrow$  Kramers–Kronig relations

$$\operatorname{Re} \chi^{(1)}(\omega) = -\frac{1}{\pi} \mathcal{P} \int_{-\infty}^{\infty} d\omega' \frac{\operatorname{Im} \chi^{(1)}(\omega')}{\omega - \omega'}, \quad \operatorname{Im} \chi^{(1)}(\omega) = \frac{1}{\pi} \mathcal{P} \int_{-\infty}^{\infty} d\omega' \frac{\operatorname{Re} \chi^{(1)}(\omega')}{\omega - \omega'}$$

analogously for complex refractive index  $n(\omega) = \eta(\omega) + i\kappa(\omega)$ :

$$\eta(\omega) - 1 = -\frac{2}{\pi} \mathcal{P} \int_{-\infty}^{\infty} d\omega' \frac{\omega' \kappa(\omega')}{\omega^2 - \omega'^2}, \quad \kappa(\omega) = \frac{2\omega}{\pi} \mathcal{P} \int_{-\infty}^{\infty} d\omega' \frac{\eta(\omega') - 1}{\omega^2 - \omega'^2}$$

plane waves  $e^{in(\omega)\mathbf{k}\cdot\mathbf{r}} = e^{i\eta(\omega)\mathbf{k}\cdot\mathbf{r}} e^{-\kappa(\omega)\mathbf{k}\cdot\mathbf{r}}$  are damped, neither orthogonal nor complete!



# Quantum optics of macroscopic systems

## What went wrong?

- damped plane waves cannot be used for mode expansion (no bosonic modes)
- reason: too simple description of light-matter interaction
- refractive index results from (bilinear) coupling of free electromagnetic field to matter
  - ⇒ reduction to degrees of freedom of field alone disturbs conservation of energy
  - ⇒ no Hermitian Hamiltonian

possible solution: describe complete system consisting of field and absorbing matter quantum mechanically and solve full dynamical problem

**but:** impossible due to large number of (unobservable) degrees of freedom

⇒ seek quantum statistical approach to field quantization in absorbing media by using linear response theory

B. Huttner and S.M. Barnett, Phys. Rev. A **46**, 4306 (1992).

# Quantum optics of macroscopic systems

## Damped harmonic oscillator

- consider damped harmonic oscillator mode described by amplitude operators  $\hat{a}$  and  $\hat{a}^\dagger$
- assume time evolution of expectation value  $\langle \hat{a}(t) \rangle = e^{-(i\omega + \Gamma/2)t} \langle \hat{a}(0) \rangle$
- evolution equation for expectation value  $\frac{d}{dt} \langle \hat{a} \rangle = -(i\omega + \Gamma/2) \langle \hat{a} \rangle$
- does **not** hold for operators!

formal solution to  $\dot{\hat{a}} = -(i\omega + \Gamma/2)\hat{a}$  is  $\hat{a}(t) = e^{-(i\omega + \Gamma/2)t} \hat{a}(0)$

**but:** commutation relation  $[\hat{a}(0), \hat{a}^\dagger(0)] = 1 \Rightarrow [\hat{a}(t), \hat{a}^\dagger(t)] = e^{-\Gamma t}$

$\Rightarrow$  amplitude operators lose their bosonic character!

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# Quantum optics of macroscopic systems

## A formal solution (without justification yet)

save commutation relations by adding some operator  $\hat{f}$  with appropriately chosen properties:

$$\dot{\hat{a}} = -(i\omega + \Gamma/2)\hat{a} + \hat{f}$$

choose time-dependent commutation relations

$$\begin{aligned} [\hat{f}(t_1), \hat{f}^\dagger(t_2)] e^{i\omega(t_1-t_2)} &= \Gamma \delta(t_1 - t_2) \\ [\hat{a}(t_1), \hat{f}^\dagger(t_2)] &= 0, (t_2 > t_1) \end{aligned}$$

from formal solution  $\hat{a}(t) = e^{-(i\omega+\Gamma/2)t} \hat{a}(0) + \int_0^\infty dt' e^{-(i\omega+\Gamma/2)(t-t')} \hat{f}(t')$

and  $[\hat{a}(0), \hat{a}^\dagger(0)] = 1 \Rightarrow [\hat{a}(t), \hat{a}^\dagger(t)] = 1$  for all times  $t$

# Quantum optics of macroscopic systems

## Open quantum systems and Langevin equations

phenomenological introduction of operators  $\hat{f}(t)$  can be justified within the framework of open quantum systems

example (Caldeira–Leggett model): coupling of single harmonic oscillator to reservoir of harmonic oscillators

$$\hat{H} = \hbar\omega\hat{a}^\dagger\hat{a} + \sum_i \hbar\omega_i\hat{b}_i^\dagger\hat{b}_i + \hbar \sum_i \left( g_i\hat{a}^\dagger\hat{b}_i + \text{h.c.} \right)$$

e.g. H.-P. Breuer and F. Petruccione, *The theory of open quantum systems* (Oxford, 2002).

# Quantum optics of macroscopic systems

## Open quantum systems and Langevin equations

Heisenberg's equations of motion

$$\dot{\hat{a}} = -i\omega\hat{a} - i\sum_i g_i \hat{b}_i, \quad \dot{\hat{b}}_i = -i\omega_i \hat{b}_i - ig_i^* \hat{a}$$

have formal solution

$$\hat{b}_i(t) = e^{-i\omega_i t} \hat{b}_i(0) - ig_i^* \int_0^t dt' e^{-i\omega_i(t-t')} \hat{a}(t')$$

⇒ obtain effective equation of motion for 'system' oscillator

$$\dot{\hat{a}}(t) = -i\omega\hat{a}(t) - \sum_i |g_i|^2 \int_0^t dt' e^{-i\omega_i(t-t')} \hat{a}(t') - i \sum_i g_i e^{-i\omega_i t} \hat{b}_i(0)$$

$$\stackrel{\text{Markov}}{\simeq} -i\omega\hat{a}(t) - \sum_i |g_i|^2 \hat{a}(t) \left[ \pi\delta(\omega - \omega_i) + i\mathcal{P}\frac{1}{\omega - \omega_i} \right] - i \sum_i g_i e^{-i\omega_i t} \hat{b}_i(0)$$



# Quantum optics of macroscopic systems

## Open quantum systems and Langevin equations

introduce notation

$$\Gamma = 2\pi \sum_i |g_i|^2 \delta(\omega - \omega_i)$$

$$\delta\omega = \sum_i |g_i|^2 \mathcal{P} \frac{1}{\omega - \omega_i}$$

$$\hat{f}(t) = -i \sum_i g_i e^{-i\omega_i t} \hat{b}_i(0)$$

reproduces Langevin equation

$$\dot{\hat{a}}(t) = -(i\omega + i\delta\omega + \Gamma/2)\hat{a}(t) + \hat{f}(t)$$

- Langevin operators  $\hat{f}(t)$  from initial conditions of reservoir operators,  $\langle \hat{f} \rangle = 0$
- damping constant from correlation function  $\Gamma/2 = \int dt e^{i\omega t} [\hat{f}(t), \hat{f}^\dagger(0)]$
- frequency shift  $\delta\omega$  due to interaction with reservoir

# Quantum optics of macroscopic systems

## Linear fluctuation-dissipation theorem

quantum system in thermal equilibrium:  $\hat{G} = \frac{1}{Z} e^{-\beta \hat{H}_0} \left( \beta = \frac{1}{k_B T} \right)$

perturbation out of equilibrium:  $\frac{d}{dt} \hat{\varrho}_I(t) = \frac{i}{\hbar} [\hat{\varrho}_I(t), \hat{H}_I(t)]$

linear response:  $\hat{\varrho}_I(t) = \hat{G} + \frac{i}{\hbar} \int_{-\infty}^t ds [\hat{G}, \hat{H}_I(s)]$

Hamiltonian:  $\hat{H}(t) = \hat{H}_0 - \lambda(t) \hat{V}$  with  $[\hat{G}, \hat{H}_0] = 0$

$\Rightarrow \hat{\varrho}(t) = \hat{G} - \frac{i}{\hbar} \int_{-\infty}^t ds \lambda(s) [\hat{G}, \hat{V}(s-t)]$

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# Quantum optics of macroscopic systems

## Linear fluctuation-dissipation theorem

expectation value of observable  $\hat{O}$ :  $\langle \hat{O} \rangle = \text{Tr}[\hat{G} \hat{O}] + \int_0^\infty d\tau \lambda(t - \tau) \Gamma(\hat{O} \hat{V}; \tau)$

with linear-response function  $\Gamma(\hat{O} \hat{V}; \tau) = \frac{i}{\hbar} \langle [\hat{O}(\tau), \hat{V}] \rangle$

compare  $\Gamma(\hat{A} \hat{B}; \tau) = \langle [\hat{A}(\tau), \hat{B}] \rangle$  with symmetrized correlation function  
 $K(\hat{A} \hat{B}; \tau) = \frac{1}{2} \langle \hat{A}(\tau) \hat{B} + \hat{B} \hat{A}(\tau) \rangle - \langle \hat{A} \rangle \langle \hat{B} \rangle$

Kubo–Martin–Schwinger relation:

$$\hat{A}(\tau) e^{-\beta \hat{H}_0} = e^{-\beta \hat{H}_0} e^{\beta \hat{H}_0} \hat{A}(\tau) e^{-\beta \hat{H}_0} = e^{-\beta \hat{H}_0} \hat{A}(\tau - i\hbar\beta)$$

$$\Rightarrow \langle \hat{B} \hat{A}(\tau) \rangle = \langle \hat{A}(\tau - i\hbar\beta) \hat{B} \rangle$$

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define function  $f(\tau) = \langle \hat{A}(\tau)\hat{B} \rangle - \langle \hat{A} \rangle \langle \hat{B} \rangle$

$$\Gamma(\hat{A}\hat{B}; \tau) = \frac{i}{\hbar} f(\tau) - \frac{i}{\hbar} f(\tau - i\hbar\beta) = \frac{i}{\hbar} \int \frac{d\omega}{2\pi} e^{-i\omega\tau} \tilde{f}(\omega) [1 - e^{-\beta\hbar\omega}]$$

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$$\Rightarrow \tilde{K}(\hat{A}\hat{B}; \omega) = \frac{\hbar}{2i} \coth \frac{\hbar\beta\omega}{2} \tilde{\Gamma}(\hat{A}\hat{B}; \omega) = \hbar \coth \frac{\hbar\beta\omega}{2} \chi_I(\omega)$$

## Fluctuation-dissipation theorem

The strength of the fluctuations of a field is proportional to the dissipation (absorption) strength.

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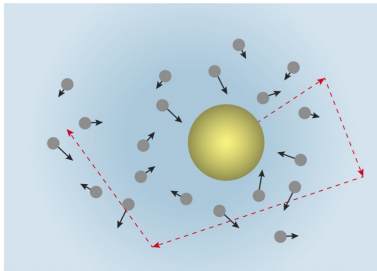
## Fluctuations and dissipation

Example: Brownian motion of a particle

$$\ddot{x}(t) + D\dot{x}(t) + \omega^2 x(t) = \xi(t)$$

dissipation strength  $D$  related to fluctuations of random force  $\xi(t)$ :

$$\langle \xi(t) \rangle = 0, \quad \langle \xi(t)\xi(t') \rangle = 2Dk_B T \delta(t - t')$$



# Quantum optics of macroscopic systems

## Fluctuations in electromagnetism

### Example 1: Linear polarisation, causal response to electric field

$$\mathbf{P}(\mathbf{r}, t) = \varepsilon_0 \int_0^{\infty} d\tau \chi(\mathbf{r}, \tau) \mathbf{E}(\mathbf{r}, t - \tau)$$

$$\mathbf{P}(\mathbf{r}, \omega) = \varepsilon_0 \chi(\mathbf{r}, \omega) \mathbf{E}(\mathbf{r}, \omega)$$

Linear FDT:  $\langle \mathbf{P}(\mathbf{r}, \omega) \mathbf{P}(\mathbf{r}', \omega') \rangle \propto \text{Im} \chi(\mathbf{r}, \omega) \delta(\omega - \omega') \delta(\mathbf{r} - \mathbf{r}')$

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## Fluctuations in electromagnetism

### Example 2: Helmholtz equation

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}, \omega) - \frac{\omega^2}{c^2} \varepsilon(\mathbf{r}, \omega) \mathbf{E}(\mathbf{r}, \omega) = \frac{\omega^2}{c^2 \varepsilon_0} \mathbf{P}_N(\mathbf{r}, \omega)$$

solved by dyadic Green function

$$\mathbf{E}(\mathbf{r}, \omega) = \frac{\omega^2}{c^2 \varepsilon_0} \int d^3 s \mathbf{G}(\mathbf{r}, \mathbf{s}, \omega) \cdot \mathbf{P}_N(\mathbf{s}, \omega)$$

read as linear response relation between electric field and noise polarisation with Green function acting as response function

$$\langle \mathbf{E}(\mathbf{r}, \omega) \otimes \mathbf{E}(\mathbf{r}', \omega') \rangle \propto \text{Im } \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \delta(\omega - \omega')$$

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# Quantum optics of macroscopic systems

## Field quantisation in dielectrics

- need to know field excitations in the presence of dielectrics
- Helmholtz equation with noise polarisation  $\Rightarrow$  no field expansion into modes possible
- promote noise polarisation to operator-valued vector field

$$\hat{\mathbf{P}}_N(\mathbf{r}, \omega) = i\sqrt{\frac{\hbar\epsilon_0}{\pi} \text{Im} \epsilon(\mathbf{r}, \omega)} \hat{\mathbf{f}}(\mathbf{r}, \omega)$$

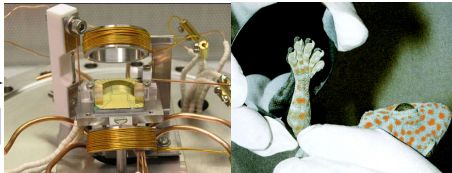
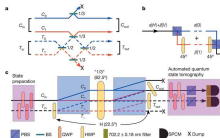
$\hat{\mathbf{f}}(\mathbf{r}, \omega)$ :

- bosonic vector field with equal-time commutation relation  
 $[\hat{\mathbf{f}}(\mathbf{r}, \omega), \hat{\mathbf{f}}^\dagger(\mathbf{r}', \omega')] = \delta(\mathbf{r} - \mathbf{r}')\delta(\omega - \omega')\mathbf{1}$
- collectively excites field and dielectric matter (polariton)
- $|\mathbf{1}(\mathbf{r}, \omega)\rangle = \hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega)|\{0\}\rangle$  is a single-quantum excitation of the **medium-assisted** electromagnetic field

# Quantum optics of macroscopic systems

## What is it good for?

- quantized electromagnetic field in dielectrics: use for quantum description of light and light-matter interactions in macroscopic environments
- only experimentally accessible linear-response functions needed (permittivity, refractive index, transmission/reflection coefficients etc.)
- study entanglement transport through linear-optical devices (optical fibres, beam splitters etc.)
- influence of materials on atomic properties (lifetimes, line widths, dispersion forces etc.)



Review: S. Scheel and S.Y. Buhmann, Macroscopic quantum electrodynamics – concepts and applications, Acta Phys. Slovaca **58**, 675-810 (2008).