Imperial College London

Causal nonlinear quantum optics

Stefan Scheel, Imperial College London

Dirk–Gunnar Welsch, Friedrich–Schiller-Universität Jena

Vilnius, September 2006

Outline:

- Motivation: single-photon sources, diamagnetic materials etc.

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- QED in linear dielectrics — a reminder
- Nonlinear interaction Hamiltonian and nonlinear noise polarization

A brief guide towards nonlinear quantum optics

- Vacuum QED: Maxwell's equations in free space without matter, Lorentz invariant

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- Nonlinear QED: . . .

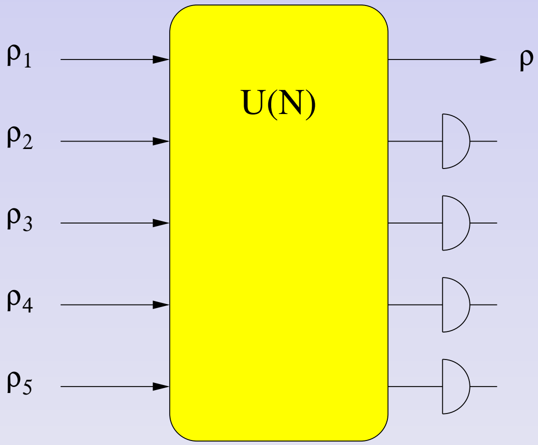
Goal: to provide a consistent theory for quantizing the electromagnetic field in nonlinear dielectrics with dispersion and absorption

Some practical motivation, if needed...

single-photon and entangled-photon sources needed in

- all-optical quantum information processing
- metrology, for building luminosity standards

QIP with linear optics still needs highly nonlinear elements such as single-photon sources and detectors!



Enhancing single-photon efficiency by post-selection does not work!
Neither does enhancing the detection efficiency by post-selection.
$\Rightarrow$ Better sources and detectors are needed!

D.W. Berry, S. Scheel, B.C. Sanders, and P.L. Knight, Phys. Rev. A **69**, 031806(R) (2004); D.W. Berry, S. Scheel, C.R. Myers, B.C. Sanders, P.L. Knight, and R. Laflamme, New J. Phys. **6**, 93 (2004); P. Kok, IEEE: Sel. Top. Quantum Electron. **9**, 1498 (2003).

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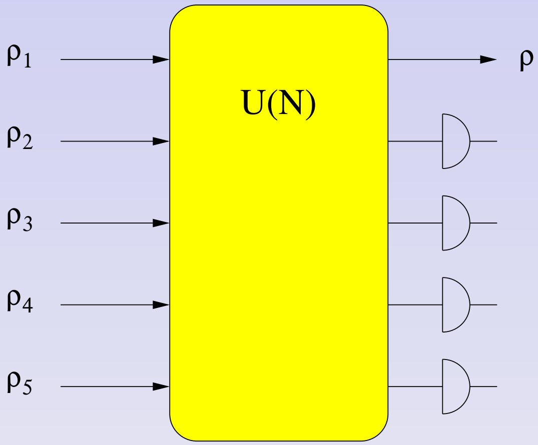

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How well can single-photon sources be made?

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Causality and unavoidable decoherence

Causality in macroscopic electrodynamics: Kramers–Kronig relations

$$\varepsilon_I(\omega) = \frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty} d\omega' \frac{\varepsilon_R(\omega') - 1}{\omega' - \omega} \qquad \equiv \qquad \varepsilon(\omega) - 1 = \frac{1}{i\pi}[\varepsilon(\omega) - 1] * \mathcal{P}\frac{1}{\omega}$$

(Kramers–Kronig relations also exist for nonlinear susceptibilities!)

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$$\boxed{\text{ability to manipulate}} \iff \boxed{\text{decoherence}}$$

S. Scheel, Phys. Rev. A **73**, 013809 (2006).

Diamagnetism and superconductivity

Goal: find phenomenological description of e.m. field interaction with macroscopically large systems

Linear response theories:

- linear dielectric media
- paramagnetic materials (paramagnetism is present even without external magnetic fields)

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Nonlinear response theories:

- nonlinear dielectric media
- diamagnetic materials (diamagnetism is *induced* by external magnetic fields)
- superconductivity (perfect diamagnetism)

Naive extension of vacuum QED

mode expansion of the field operators

$$\hat{\mathbf{E}}(\mathbf{r}) = i\sum_\lambda \omega_\lambda \mathbf{A}_\lambda(\mathbf{r})\hat{a}_\lambda + \text{h.c.}$$

in terms of bosonic modes with $[\hat{a}_\lambda, \hat{a}^\dagger_{\lambda'}] = \delta_{\lambda\lambda'}$

Naive introduction of a (complex) $n(\omega)$ leads to decaying commutation rules!

$$[\hat{a}(\mathbf{r},\omega), \hat{a}^\dagger(\mathbf{r}',\omega')] = e^{-n_I(\omega)\omega/c|\mathbf{r}-\mathbf{r}'|}\delta(\omega-\omega')$$

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—

Solution: introduce Langevin noise!
Example: damped harmonic oscillator $\langle\hat{a}(t)\rangle = \langle\hat{a}(t')\rangle\; e^{-\Gamma(t-t')}$

But: relation for expectation values cannot hold for operators!
$\Rightarrow$ add Langevin noise $\hat{f}$ with $\langle\hat{f}\rangle = 0$

$$\dot{\hat{a}} = -\Gamma\hat{a} + \hat{f}$$

Langevin force takes care of ETCR

Classical macroscopic electrodynamics

Classical electrodynamics with media without external sources:

$$\begin{aligned} \boldsymbol{\nabla}\cdot\mathbf{B}(\mathbf{r},t) &= 0\,, \quad \boldsymbol{\nabla}\times\mathbf{E}(\mathbf{r},t) = -\dot{\mathbf{B}}(\mathbf{r},t)\,, \\ \boldsymbol{\nabla}\cdot\mathbf{D}(\mathbf{r},t) &= 0\,, \quad \boldsymbol{\nabla}\times\mathbf{H}(\mathbf{r},t) = \dot{\mathbf{D}}(\mathbf{r},t) \end{aligned}$$

need to be supplemented by constitutive relations $\mathbf{D}[\mathbf{E}]$ and $\mathbf{H}[\mathbf{B}]$

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$$\mathbf{D}(\mathbf{r},t) = \varepsilon_0\mathbf{E}(\mathbf{r},t) + \mathbf{P}(\mathbf{r},t)$$

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

$\Rightarrow$ causal response to electric field
But: what about fluctuations associated with the dissipation?

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But: what about fluctuations associated with the dissipation?

use results of Leontovich–Rytov theory

$\Rightarrow$ noise polarization $\mathbf{P}_N(\mathbf{r},t)$ is a Langevin force!

Langevin forces as fundamental fields

Helmholtz equation for electric field:

$$\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\mathbf{E}(\mathbf{r},\omega)-\frac{\omega^2}{c^2}\varepsilon(\mathbf{r},\omega)\mathbf{E}(\mathbf{r},\omega)=\mu_0\omega^2\mathbf{P}_N(\mathbf{r},\omega)$$

Solution in terms of classical dyadic Green function:

$$\mathbf{E}(\mathbf{r},\omega)=\mu_0\omega^2\int d^3\mathbf{s}\,\boldsymbol{G}(\mathbf{r},\mathbf{s},\omega)\cdot\mathbf{P}_N(\mathbf{s},\omega)$$

Can express all e.m. fields in terms of
Langevin forces and dyadic Green function!

calculating Green function is a classical scattering problem

(and is better left to electrical engineers who know better how to solve it. . .)

L. Knöll, S. Scheel, and D.-G. Welsch, in *Coherence and Statistics of Photons and Atoms* (Wiley, New York, 2001).

Field quantization

Regard the fundamental field $\mathbf{f}(\mathbf{r},\omega)$ as a bosonic vector field with the ETCR

$$\left[\hat{\mathbf{f}}(\mathbf{r},\omega),\hat{\mathbf{f}}^{\dagger}(\mathbf{r}',\omega')\right] = \delta(\mathbf{r}-\mathbf{r}')\delta(\omega-\omega')\boldsymbol{U}$$

relation between fundamental fields and noise polarization:

$$\hat{\mathbf{P}}_N(\mathbf{r},\omega) = i\sqrt{\frac{\hbar\varepsilon_0}{\pi}\varepsilon_I(\mathbf{r},\omega)}\,\hat{\mathbf{f}}(\mathbf{r},\omega)$$

Schrödinger operator of the electric field:

$$\boxed{\begin{aligned}\hat{\mathbf{E}}(\mathbf{r}) &= \int_0^\infty d\omega\,\hat{\mathbf{E}}(\mathbf{r},\omega) + \text{h.c.}\\ \hat{\mathbf{E}}(\mathbf{r},\omega) &= i\sqrt{\frac{\hbar}{\varepsilon_0\pi}}\frac{\omega^2}{c^2}\int d^3\mathbf{s}\,\sqrt{\varepsilon_I(\mathbf{s},\omega)}\,\boldsymbol{G}(\mathbf{r},\mathbf{s},\omega)\cdot\hat{\mathbf{f}}(\mathbf{s},\omega)\end{aligned}}$$

compare with $\hat{\mathbf{E}}(\mathbf{r}) = i\sum_\lambda \omega_\lambda \mathbf{A}_\lambda(\mathbf{r})\hat{a}_\lambda + \text{h.c.}$: generalized mode decomposition in terms of bosonic variables that describe collective excitation!

Mesocopic justification — Hopfield model

e.m. field		polarization		heat bath
$\mathbf{A}$, $\mathbf{E}$	$\longleftrightarrow$	$\mathbf{X}$, $\mathbf{P}$	$\longleftrightarrow$	$\mathbf{Y}_\omega$, $\mathbf{Q}_\omega$

J.J. Hopfield, Phys. Rev. **112**, 1555 (1958); B. Huttner and S.M. Barnett, Phys. Rev. A **46**, 4306 (1992); L.G. Suttorp and M. Wubs, **70**, 013816 (2004).

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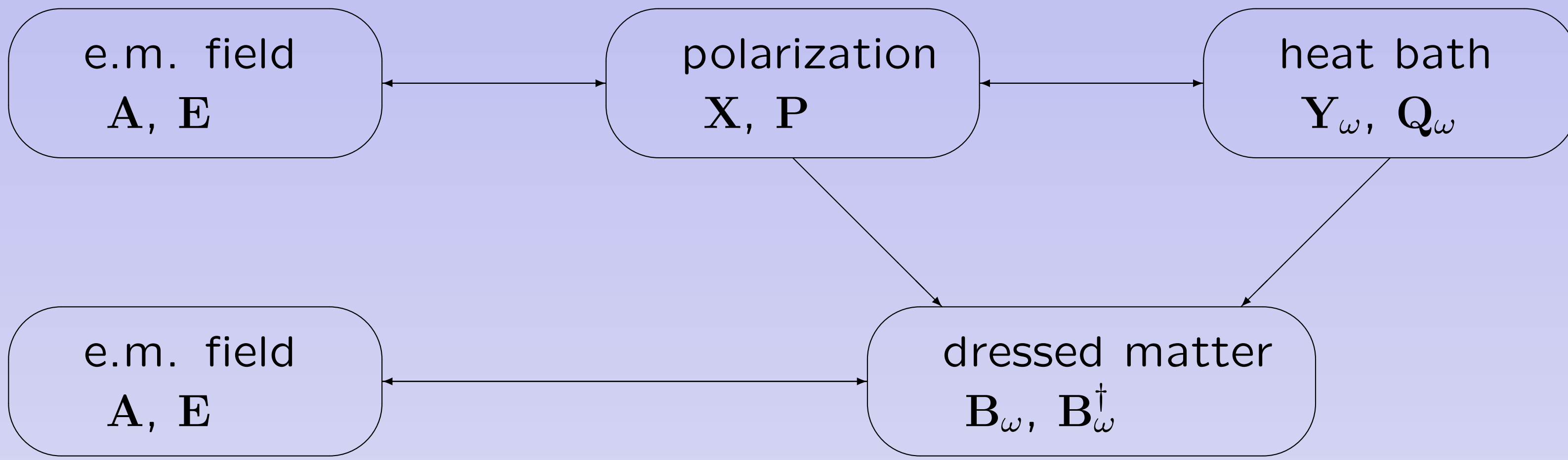

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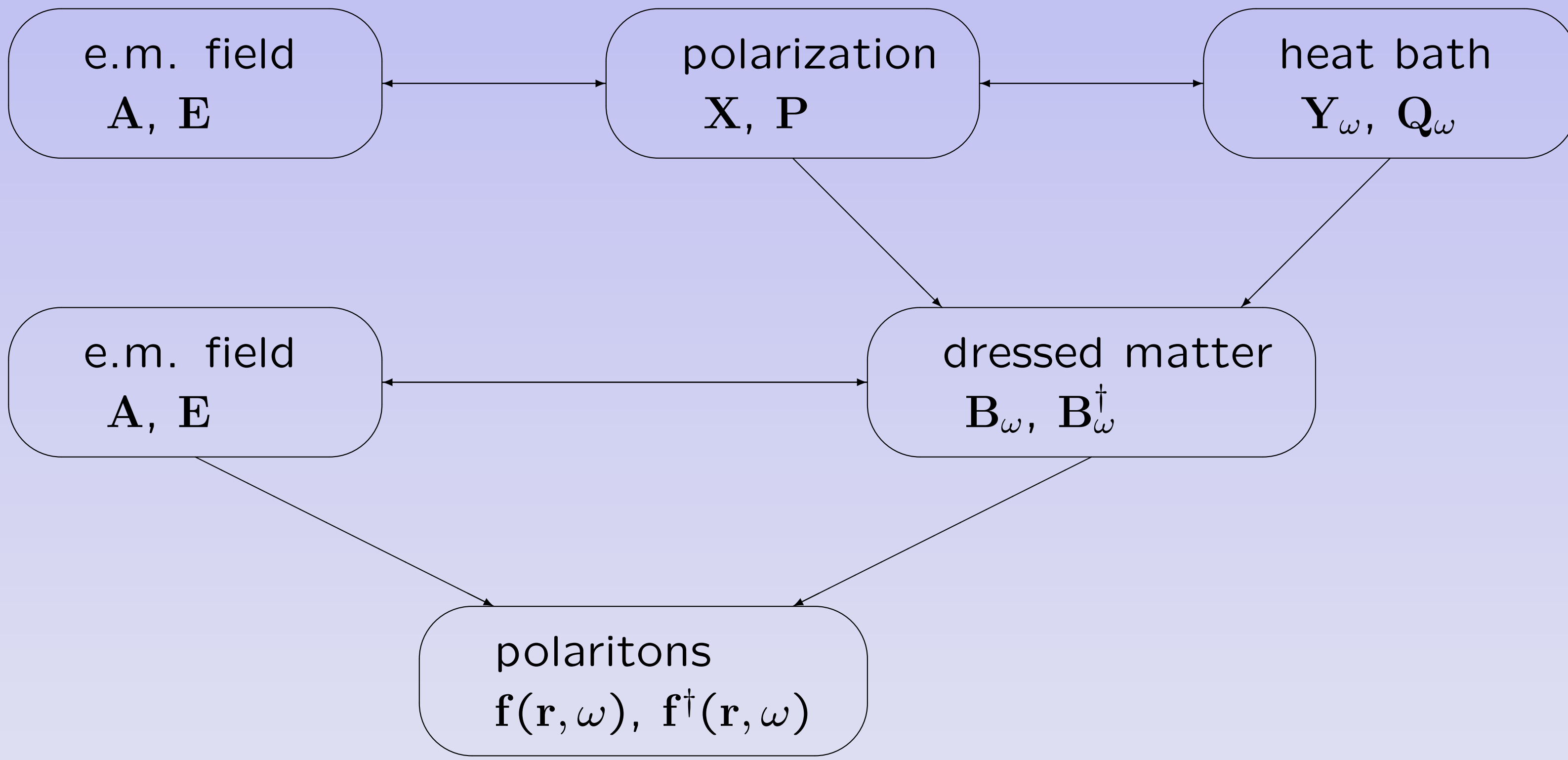

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Consistency checks

- ETCR between electric field and magnetic induction $\surd$

$$\left[\hat{\mathbf{E}}(\mathbf{r}),\hat{\mathbf{B}}(\mathbf{r}')\right] = -\frac{i\hbar}{\varepsilon_0}\boldsymbol{\nabla}\times\delta(\mathbf{r}-\mathbf{r}')\boldsymbol{U}$$

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$$\left[\hat{\mathbf{E}}(\mathbf{r}),\hat{\mathbf{B}}(\mathbf{r}')\right] = -\frac{i\hbar}{\varepsilon_0}\boldsymbol{\nabla}\times\delta(\mathbf{r}-\mathbf{r}')\boldsymbol{U}$$

- consistent with fluctuation-dissipation theorem $\surd$

$$\langle 0|\hat{\mathbf{E}}(\mathbf{r},\omega)\hat{\mathbf{E}}^\dagger(\mathbf{r}',\omega')|0\rangle = \frac{\hbar\omega^2}{\pi\varepsilon_0 c^2}\mathrm{Im}\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\delta(\omega-\omega')$$

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- Maxwell's equations follow from bilinear Hamiltonian $\surd$

$$\hat{H} = \int d^3\mathbf{r}\int_0^\infty d\omega\,\hbar\omega\,\hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)\cdot\hat{\mathbf{f}}(\mathbf{r},\omega)$$

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What is it all good for?

entanglement degradation in quantum communication:

- Entanglement degradation of Gaussian states in optical fibres
  S. Scheel and D.-G. Welsch, PRA **64**, 063811 (2001)
- Kraus operator decomposition of a lossy beam splitter
  S. Scheel, K. Nemoto, W.J. Munro, and P.L. Knight, PRA **68**, 032310 (2003)

atomic decoherence processes:

- Spontaneous emission in LHMs and near carbon nanotubes
  T.D. Ho, S.Y. Buhmann, L. Knöll, D.-G. Welsch, S. Scheel, and J. Kaestel, PRA **68**, 043816 (2003); I.V. Bondarev and P. Lambin, PRB **70**, 035407 (2004)
- Thermal spin flips in atom chips and spatial decoherence
  P.K. Rekdal, S. Scheel, P.L. Knight, and E.A. Hinds, PRA **70**, 013811 (2004); S. Scheel, P.K. Rekdal, P.L. Knight, and E.A. Hinds, PRA **72**, 042901 (2005); R. Fermani, S. Scheel, and P.L. Knight, PRA **73**, 032902 (2006)

Interatomic and intermolecular forces

- Casimir–Polder forces
  S.Y. Buhmann, L. Knöll, D.-G. Welsch, and T.D. Ho, PRA **70**, 052117 (2004)
- Casimir forces
  C. Raabe and D.-G. Welsch, PRA **71**, 013814 (2005)

Nonlinear (cubic) Hamiltonian

Two ways to approach nonlinear interaction:

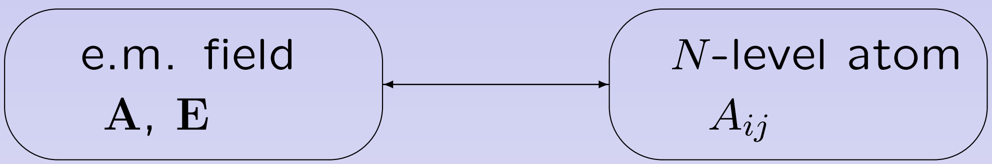
- microscopic theory with anharmonic oscillators
- macroscopic *ansatz* for Hamiltonian

$\Rightarrow$ microscopic justification needed!

Derivation of approximate interaction Hamiltonians:

- collection of N-level atoms non-resonantly coupled to electromagnetic field
- pick out nonlinear process of interest (e.g. three-wave mixing, Kerr effect, etc.)
- effectively remove dependencies on atomic quantities
- approximate interaction Hamiltonian containing only photonic operators

Nonlinear (cubic) Hamiltonian



- free e.m. field coupled to N-level atoms

Nonlinear (cubic) Hamiltonian

approximate H_{int}
$a_1^2 a_2^\dagger,\ a_1^{\dagger 2} a_2$

e.m. field
$\mathbf{A},\ \mathbf{E}$

N-level atom
A_{ij}

- free e.m. field coupled to N-level atoms
- derive approximate interaction Hamiltonian (e.g. three-wave mixing)

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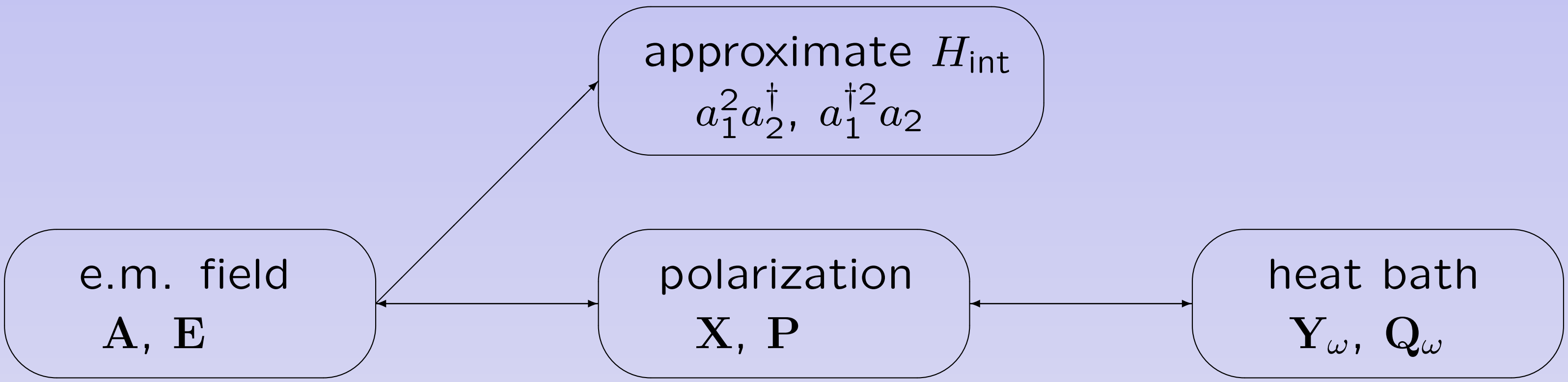

- free e.m. field coupled to N-level atoms
- derive approximate interaction Hamiltonian (e.g. three-wave mixing)
- approximate N-level atoms by harmonic oscillators, coupled to bath

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e.m. field
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dressed matter
$\mathbf{B}_\omega,\ \mathbf{B}_\omega^\dagger$

- free e.m. field coupled to N-level atoms
- derive approximate interaction Hamiltonian (e.g. three-wave mixing)
- approximate N-level atoms by harmonic oscillators, coupled to bath
- diagonalize the matter Hamiltonian

Nonlinear (cubic) Hamiltonian

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$a_1^2 a_2^\dagger,\ a_1^{\dagger 2} a_2$

$\updownarrow$

polaritons
$\mathbf{f}(\mathbf{r},\omega),\ \mathbf{f}^\dagger(\mathbf{r},\omega)$

- free e.m. field coupled to N-level atoms
- derive approximate interaction Hamiltonian (e.g. three-wave mixing)
- approximate N-level atoms by harmonic oscillators, coupled to bath
- diagonalize the matter Hamiltonian
- diagonalize the bilinear part of the total Hamiltonian

Nonlinear (cubic) Hamiltonian

approximate H_{int}
$a_i \mapsto \mathbf{f}(\mathbf{r},\omega_i)$

$\updownarrow$

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$\mathbf{f}(\mathbf{r},\omega)$, $\mathbf{f}^\dagger(\mathbf{r},\omega)$

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- diagonalize the matter Hamiltonian
- diagonalize the bilinear part of the total Hamiltonian
- re-express photonic operators by dynamical variables

Nonlinear (cubic) Hamiltonian

Find interaction Hamiltonian consistent with microscopic considerations:

$$\boxed{\hat{H}_{NL} = \int d\mathbf{1}\, d\mathbf{2}\, d\mathbf{3}\, \alpha_{ijk}(\mathbf{1},\mathbf{2},\mathbf{3})\hat{\mathbf{f}}_i^\dagger(\mathbf{1})\hat{\mathbf{f}}_j(\mathbf{2})\hat{\mathbf{f}}_k(\mathbf{3}) + \text{h.c.}} \qquad \boldsymbol{k} \equiv (\mathbf{r}_k, \omega_k)$$

most general normal-ordered form of the nonlinear interaction energy corresponding to a $\chi^{(2)}$ medium

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most general normal-ordered form of the nonlinear interaction energy corresponding to a $\chi^{(2)}$ medium

Observation #1: Faraday's law

$$\boldsymbol{\nabla} \times \hat{\mathbf{E}}(\mathbf{r}) = -\dot{\hat{\mathbf{B}}}(\mathbf{r}) = -\frac{1}{i\hbar}\left[\hat{\mathbf{B}}(\mathbf{r}), \hat{H}_L + \hat{H}_{NL}\right]$$

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$$\boldsymbol{\nabla} \times \hat{\mathbf{E}}(\mathbf{r}) = -\dot{\hat{\mathbf{B}}}(\mathbf{r}) = -\frac{1}{i\hbar}\left[\hat{\mathbf{B}}(\mathbf{r}), \hat{H}_L + \hat{H}_{NL}\right]$$

But (transverse) electric field and magnetic induction are purely electromagnetic fields without knowledge of the interaction. Therefore, their functional form is as in the linear theory!

Nonlinear (cubic) Hamiltonian

Find interaction Hamiltonian consistent with microscopic considerations:

$$\boxed{\hat{H}_{NL} = \int d\mathbf{1}\, d\mathbf{2}\, d\mathbf{3}\, \alpha_{ijk}(\mathbf{1},\mathbf{2},\mathbf{3})\hat{\mathbf{f}}_i^\dagger(\mathbf{1})\hat{\mathbf{f}}_j(\mathbf{2})\hat{\mathbf{f}}_k(\mathbf{3}) + \text{h.c.}} \qquad \boldsymbol{k} \equiv (\mathbf{r}_k, \omega_k)$$

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Derivation of the nonlinear polarization

Observation #2: Ampere's law, keep only terms linear in α_{ijk},
other terms contribute to higher-order nonlinear processes!

$$\boldsymbol{\nabla} \times \boldsymbol{\nabla} \times \hat{\mathbf{E}}(\mathbf{r}) = \quad -\mu_0 \ddot{\hat{\mathbf{D}}}_L(\mathbf{r}) \quad -\mu_0 \ddot{\hat{\mathbf{P}}}_{NL}(\mathbf{r})$$

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$$
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& \Downarrow & & \\
& +\frac{\mu_0}{\hbar^2}\left[\left[\hat{\mathbf{D}}_L(\mathbf{r}),\hat{H}_L\right],\hat{H}_L\right] & & +\frac{\mu_0}{\hbar^2}\left[\left[\hat{\mathbf{D}}_L(\mathbf{r}),\hat{H}_L\right],\hat{H}_{NL}\right] \\
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Particular solution: $\boxed{\left[\hat{\mathbf{D}}_L(\mathbf{r}),\hat{H}_{NL}\right] = -\left[\hat{\mathbf{P}}_{NL}(\mathbf{r}),\hat{H}_L\right]}$

General solution includes commutants with $\hat{H}_L$ which are functionals of the number density operator $\hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)\hat{\mathbf{f}}(\mathbf{r},\omega)$.

But: commutants must vanish to avoid divergences $\Rightarrow$ Particular solution is general solution!

Derivation of the nonlinear polarization

Neglected terms in one particular order appear as additional contributions in higher orders.

Example: $\chi^{(3)}$

- terms $\left[\hat{\mathbf{D}}_L(\mathbf{r}), \hat{H}_{NL}^{(3)}\right]$ and $\left[\hat{\mathbf{P}}_{NL}^{(3)}(\mathbf{r}), \hat{H}_L\right]$ are trilinear in the dynamical variables

- contributions from $\chi^{(2)}$ such as $\left[\left[\hat{\mathbf{D}}_L(\mathbf{r}), \hat{H}_{NL}^{(2)}\right], \hat{H}_{NL}^{(2)}\right]$, $\left[\left[\hat{\mathbf{P}}_{NL}^{(2)}(\mathbf{r}), \hat{H}_L\right], \hat{H}_{NL}^{(2)}\right]$ and $\left[\left[\hat{\mathbf{P}}_{NL}^{(2)}(\mathbf{r}), \hat{H}_{NL}^{(2)}\right], \hat{H}_L\right]$ are also trilinear

- double commutators are quadratic in $\chi^{(2)}$ and can be neglected only if $|\chi^{(2)}|^2/|\chi^{(3)}| \ll 1$

$\Rightarrow$ hierarchy as known from classical nonlinear optics

Derivation of the nonlinear polarization

Solve formally for nonlinear polarization:

$$\hat{\mathbf{P}}_{NL}(\mathbf{r}) = -\frac{1}{i\hbar}\mathcal{L}_L^{-1}\left[\hat{\mathbf{D}}_L(\mathbf{r}), \hat{H}_{NL}\right]$$

Liouvillian $\mathcal{L}_L$ generated by Hamiltonian $\hat{H}_L$: $\mathcal{L}_L \bullet = 1/(i\hbar)[\bullet, \hat{H}_L]$

By decomposition of the linear displacement into its reactive and noise parts, we can identify the noise contribution to the nonlinear polarization:

$$\boxed{\hat{\mathbf{P}}_{NL}^{(N)}(\mathbf{r}) = -\frac{1}{i\hbar}\mathcal{L}_L^{-1}\left[\hat{\mathbf{P}}_L^{(N)}(\mathbf{r}), \hat{H}_{NL}\right]}$$

$\hat{\mathbf{P}}_{NL}^{(N)}(\mathbf{r})$ vanishes if $\varepsilon_I(\mathbf{r},\omega) \to 0$, i.e. if there is no noise!

Inversion of the Liouvillian:

$$\hat{\mathbf{P}}_{NL}(\mathbf{r}) = \frac{i}{\hbar}\lim_{s\to 0}\int_0^\infty d\tau\, e^{-s\tau} e^{-\frac{i}{\hbar}\hat{H}_L\tau}\left[\hat{\mathbf{D}}_L(\mathbf{r}), \hat{H}_{NL}\right] e^{\frac{i}{\hbar}\hat{H}_L\tau}$$

Classical nonlinear polarization

Definition of the nonlinear polarization within framework of response theory:

$$P_{NL,l}(\mathbf{r},t)=\varepsilon_0\int\limits_{-\infty}^{t}d\tau_1 d\tau_2\,\chi^{(2)}_{lmn}(\mathbf{r},t-\tau_1,t-\tau_2)E_m(\mathbf{r},\tau_1)E_n(\mathbf{r},\tau_2)+P^{(N)}_{NL,l}(\mathbf{r},t)$$

We have to match this expression to what we have derived before!
In that way we find functional relation $\chi^{(2)}_{lmn}\leftrightarrow\alpha_{ijk}$.

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Slowly-varying amplitude approximation: only three relevant field amplitudes with mid-frequencies $\Omega_1 = \Omega_2 + \Omega_3, \Omega_2, \Omega_3$, taken out of the integral at t:

$$\tilde{P}^{(++)}_{NL,l}(\mathbf{r},\Omega_1) = \varepsilon_0 \chi^{(2)}_{lmn}(\mathbf{r},\Omega_2,\Omega_3) \tilde{E}_m(\mathbf{r},\Omega_2) \tilde{E}_n(\mathbf{r},\Omega_3) + \tilde{P}^{(N,++)}_{NL,l}(\mathbf{r},\Omega_1)$$

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Now insert expression for electric field in terms of Green function $\boldsymbol{G}(\mathbf{r},\mathbf{s},\omega)$ and dynamical variables $\hat{\mathbf{f}}(\mathbf{r},\omega)$ and compare...

Comparison with classical polarization

find solution to integral equation

$$\int d^3\mathbf{s}\sqrt{\varepsilon_I(\mathbf{s},\Omega_1)}\alpha_{mjk}(\mathbf{s},\Omega_1,\mathbf{s}_2,\Omega_2,\mathbf{s}_3,\Omega_3)G_{lm}(\mathbf{r},\mathbf{s},\Omega_1) =$$

$$\frac{\hbar^2}{i\pi c^2}\sqrt{\frac{\pi}{\hbar\varepsilon_0}}\frac{\Omega_2^2\Omega_3^2}{\Omega_1\varepsilon(\mathbf{r},\Omega_1)}\sqrt{\varepsilon_I(\mathbf{s}_2,\Omega_2)\varepsilon_I(\mathbf{s}_3,\Omega_3)}\chi^{(2)}_{lmn}(\mathbf{r},\Omega_2,\Omega_3)G_{mj}(\mathbf{r},\mathbf{s}_2,\Omega_2)G_{nk}(\mathbf{r},\mathbf{s}_3,\Omega_3)$$

Fredholm integral equation is solved by inverting the integral kernel

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Fredholm integral equation is solved by inverting the integral kernel

Helmholtz operator: $H_{ij}(\mathbf{r},\omega)=\partial_i^r\partial_j^r-\delta_{ij}\Delta^r-(\omega^2/c^2)\varepsilon(\mathbf{r},\omega)\delta_{ij}$

inverts Green function: $H_{ij}(\mathbf{r},\omega)G_{jk}(\mathbf{r},\mathbf{s},\omega)=\delta_{ik}\delta(\mathbf{r}-\mathbf{s})$

$$\boxed{\begin{aligned}&\alpha_{ijk}(\mathbf{r},\Omega_1,\mathbf{s}_2,\Omega_2,\mathbf{s}_3,\Omega_3)=\frac{\hbar^2}{i\pi c^2}\sqrt{\frac{\pi}{\hbar\varepsilon_0}}\frac{\Omega_2^2\Omega_3^2}{\Omega_1}\sqrt{\frac{\varepsilon_I(\mathbf{s}_2,\Omega_2)\varepsilon_I(\mathbf{s}_3,\Omega_3)}{\varepsilon_I(\mathbf{r},\Omega_1)}}\\&\times H_{li}(\mathbf{r},\Omega_1)\left[\frac{\chi^{(2)}_{lmn}(\mathbf{r},\Omega_2,\Omega_3)}{\varepsilon(\mathbf{r},\Omega_1)}G_{mj}(\mathbf{r},\mathbf{s}_2,\Omega_2)G_{nk}(\mathbf{r},\mathbf{s}_3,\Omega_3)\right]\end{aligned}}$$

$\Rightarrow$ sought functional relation between nonlinear coupling α_{ijk} in the nonlinear Hamiltonian $\hat{H}_{NL}$ and measurable nonlinear susceptibility $\chi^{(2)}_{lmn}$

Nonlinear noise polarization

- insert solution for α_{ijk} into expression for $\hat{\mathbf{P}}^{(N)}_{NL}(\mathbf{r})$
- rewrite expression in terms of (slowly-varying) electric fields

$$\boxed{\hat{P}^{(N)}_{NL,m}(\mathbf{r},\Omega_1)=\frac{\varepsilon_0 c^2}{\Omega_1^2}H_{mn}(\mathbf{r},\Omega_1)\left[\frac{\chi^{(2)}_{imn}(\mathbf{r},\Omega_2,\Omega_3)}{\varepsilon(\mathbf{r},\Omega_1)}\tilde{E}_m(\mathbf{r},\Omega_2)\tilde{E}_n(\mathbf{r},\Omega_3)\right]}$$

(Helmholtz operator should not be taken as second-order partial differential operator for consistency with SVAA)

estimate strength of nonlinear noise polarization:

$$\boxed{\frac{\langle\hat{P}^{(N)}_{NL}\rangle}{\langle\hat{P}^{(N)}_{L}\rangle}\sim\frac{|\chi^{(2)}|}{|\varepsilon|}|\mathcal{E}|}$$

$\Rightarrow$ nonlinear noise grows with amplitude of pump fields

S. Scheel and D.-G. Welsch, Phys. Rev. Lett. **96** 073601 (2006);
S. Scheel and D.-G. Welsch, J. Phys. B: At. Mol. Opt. Phys. **39**, S711 (2006).

Summary and outlook

- macroscopic quantum theory for $\chi^{(2)}$ interactions that includes absorption and dispersion
- gives a cubic Hamiltonian with a nonlinear coupling constant that can be related to the nonlinear susceptibility
- can treat inhomogeneous situations easily because all geometrical information is contained in Green functions
- theory leads to a nonlinear noise polarization that has hitherto been ignored
- extension to higher-order nonlinearities in the same way; lower-order processes contribute to noise polarization

- application to entangled-light generation, limits to fidelity
- limits to uses of Kerr and cross-Kerr effects in QIP
- new way of finding nonlinear fluctuation-dissipation theorems