Imperial College London

Quantum theory of light and noise polarization in nonlinear optics

Stefan Scheel, Imperial College London

Dirk–Gunnar Welsch, Friedrich–Schiller-Universität Jena

Olomouc, 21.10.2005

Outline:

- Motivation: single-photon and entangled-photon sources

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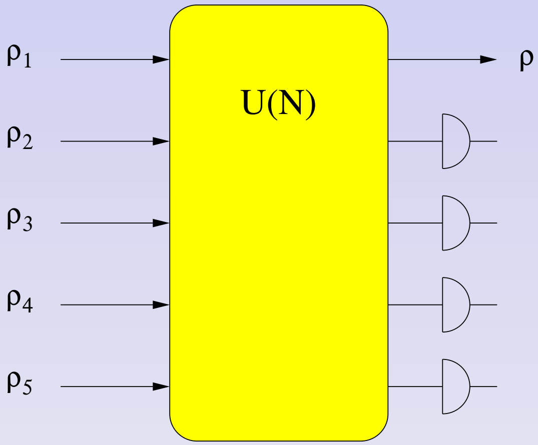
- Motivation: single-photon and entangled-photon sources
- QED in linear dielectrics — a reminder
- Nonlinear interaction Hamiltonian and nonlinear noise polarization

Some 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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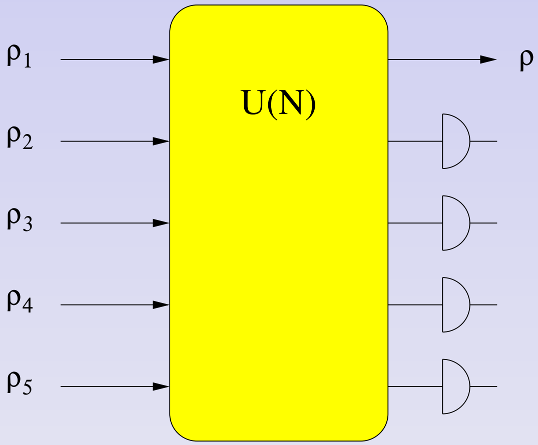

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How well can single-photon sources be made?

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Causality and unavoidable decoherence

Causality in macroscopic electrodynamics: Kramers–Kronig relations

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

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S. Scheel, *quant-ph/0508189*.

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ability to manipulate $\Longleftrightarrow$ decoherence

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QED in linear dielectrics — a reminder

Physical situation: dielectric material whose polarization responds linearly and locally to the electric field

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

noise polarization $\mathbf{P}_L^{(N)}(\mathbf{r},t)$ is there to satisfy fluctuation-dissipation theorem

Helmholtz equation for e.m. field without external sources gets a source term!

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

Unique solution to the Helmholtz equation:

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

$\boldsymbol{G}(\mathbf{r},\mathbf{s},\omega)$: dyadic Green function of the associated classical scattering problem

QED in linear dielectrics — a reminder

Quantization is performed by introducing dynamical variables $\hat{\mathbf{f}}(\mathbf{r},\omega)$ as

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

with bosonic commutation rules $\left[\hat{\mathbf{f}}(\mathbf{r},\omega),\hat{\mathbf{f}}^\dagger(\mathbf{r}',\omega')\right] = \delta(\omega-\omega')\delta(\mathbf{r}-\mathbf{r}')\boldsymbol{I}$.

The Hamiltonian generating Maxwell's equations is bilinear:

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

The electromagnetic field operators then read

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

$$\hat{\mathbf{D}}_L(\mathbf{r},\omega) = \varepsilon_0\varepsilon(\mathbf{r},\omega)\hat{\mathbf{E}}(\mathbf{r},\omega) + \hat{\mathbf{P}}_L^{(N)}(\mathbf{r},\omega), \quad \hat{\mathbf{D}}_L(\mathbf{r}) = \int_0^\infty d\omega\hat{\mathbf{D}}_L(\mathbf{r},\omega) + \text{h.c.}$$

Nonlinear (cubic) Hamiltonian

Two ways to approach nonlinear interaction:

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

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

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

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

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Derivation of the nonlinear polarization

Observation #2: Ampere's law, keep only terms linear in α_{ijk}

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

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$$
\begin{aligned}
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& \quad\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: $\left[\hat{\mathbf{D}}_L(\mathbf{r}),\hat{H}_{NL}\right] = -\left[\hat{\mathbf{P}}_{NL}(\mathbf{r}),\hat{H}_L\right]$

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

- Linear functionals of this type are included in particular solution.
- Higher-order functionals have to be excluded for consistency with the approximation that $\hat{\mathbf{P}}_{NL}(\mathbf{r})$ stays bilinear in the dynamical variables.

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:

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

$\hat{\mathbf{P}}_{NL}^{(N)}(\mathbf{r})$ 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:

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

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

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

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

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

- application to entangled-light generation, limits to fidelity
- extension to Kerr nonlinearities (rather trivial)

S. Scheel and D.-G. Welsch, *quant-ph/0508122*.