9, Quantum theory of Nonlinear Optics

1. Degenerate Parametric Amplification
2. Optical Parametric Oscillator
3. Third-Harmonic Generation
4. Four-Wave Mixing
5. Stimulated Raman effect

Ref:

Ch. 16 in *"Quantum Optics,"* by M. Scully and M. Zubairy.

Ch. 8 in *"Quantum Optics,"* by D. Wall and G. Milburn.

Ch. 9 in *"The Quantum Theory of Light,"* by R. Loudon.

Squeezed State

- define the squeezed state as

$$|\Psi_s\rangle = \hat{S}(\xi)|\Psi\rangle,$$

- where the unitary squeeze operator

$$\hat{S}(\xi) = \text{exp}[\frac{1}{2}\xi^* \hat{a}^2 - \frac{1}{2}\xi \hat{a}^{\dagger 2}]$$

where $\xi = r\text{exp}(i\theta)$ is an arbitrary complex number.

- squeeze operator is unitary, $\hat{S}^\dagger(\xi) = \hat{S}^{-1}(\xi) = \hat{S}(-\xi)$,and the unitary transformation of the squeeze operator,

$$\hat{S}^\dagger(\xi)\hat{a}\hat{S}(\xi) = \hat{a}\cosh r - \hat{a}^\dagger e^{i\theta}\sinh r,$$
$$\hat{S}^\dagger(\xi)\hat{a}^\dagger\hat{S}(\xi) = \hat{a}^\dagger\cosh r - \hat{a}e^{-i\theta}\sinh r,$$

- for $|\Psi\rangle$ is the vacuum state $|0\rangle$, the $|\Psi_s\rangle$ state is the *squeezed vacuum*,

$$|\xi\rangle = \hat{S}(\xi)|0\rangle,$$

Squeezed Vacuum State

- for $|\Psi\rangle$ is the vacuum state $|0\rangle$, the $|\Psi_s\rangle$ state is the *squeezed vacuum*,

$$|\xi\rangle = \hat{S}(\xi)|0\rangle,$$

- the variances for squeezed vacuum are

$$\begin{aligned} \Delta\hat{a}_1^2 &= \frac{1}{4}[\cosh^2 r + \sinh^2 r - 2\sinh r \cosh r \cos\theta], \\ \Delta\hat{a}_2^2 &= \frac{1}{4}[\cosh^2 r + \sinh^2 r + 2\sinh r \cosh r \cos\theta], \end{aligned}$$

- for $\theta = 0$, we have

$$\Delta\hat{a}_1^2 = \frac{1}{4}e^{-2r}, \quad \text{and} \quad \Delta\hat{a}_2^2 = \frac{1}{4}e^{+2r},$$

  and squeezing exists in the $\hat{a}_1$ quadrature.

- for $\theta = \pi$, the squeezing will appear in the $\hat{a}_2$ quadrature.

Quadrature Operators

- define a rotated complex amplitude at an angle $\theta/2$

$$\hat{Y}_1 + i\hat{Y}_2 = (\hat{a}_1 + i\hat{a}_2)e^{-i\theta/2} = \hat{a}e^{-i\theta/2},$$

where

$$\begin{pmatrix} \hat{Y}_1 \\ \hat{Y}_2 \end{pmatrix} = \begin{pmatrix} \cos\theta/2 & \sin\theta/2 \\ -\sin\theta/2 & \cos\theta/2 \end{pmatrix} \begin{pmatrix} \hat{a}_1 \\ \hat{a}_2 \end{pmatrix}$$

- then $\hat{S}^\dagger(\xi)(\hat{Y}_1 + i\hat{Y}_2)\hat{S}(\xi) = \hat{Y}_1 e^{-r} + i\hat{Y}_2 e^{r}$,
- the quadrature variance

$$\Delta\hat{Y}_1^2 = \frac{1}{4}e^{-2r}, \quad \Delta\hat{Y}_2^2 = \frac{1}{4}e^{+2r}, \quad \text{and} \quad \Delta\hat{Y}_1\Delta\hat{Y}_2 = \frac{1}{4},$$

- in the complex amplitude plane the coherent state error circle is squeezed into an *error ellipse* of the same area,
- the degree of squeezing is determined by $r = |\xi|$ which is called the squeezed parameter.

Generations of Squeezed States

- Generation of quadrature squeezed light are based on some sort of *parametric process* utilizing various types of nonlinear optical devices.
- for degenerate parametric down-conversion, the nonlinear medium is pumped by a field of frequency ω_p and that field are converted into pairs of identical photons, of frequency $\omega = \omega_p/2$ each,

$$\hat{H} = \hbar\omega\hat{a}^\dagger\hat{a} + \hbar\omega_p\hat{b}^\dagger\hat{b} + i\hbar\chi^{(2)}(\hat{a}^2\hat{b}^\dagger - \hat{a}^{\dagger 2}\hat{b}),$$

  where b is the pump mode and a is the signal mode.
- assume that the field is in a coherent state $|\beta e^{-i\omega_p t}\rangle$ and approximate the operators $\hat{b}$ and $\hat{b}^\dagger$ by classical amplitude $\beta e^{-i\omega_p t}$ and $\beta^* e^{i\omega_p t}$, respectively,
- we have the interaction Hamiltonian for *degenerate parametric down-conversion*,

$$\hat{H}_I = i\hbar(\eta^*\hat{a}^2 - \eta\hat{a}^{\dagger 2}),$$

  where $\eta = \chi^{(2)}\beta$.

Generations of Squeezed States

- we have the interaction Hamiltonian for *degenerate parametric down-conversion*,

$$\hat{H}_I = i\hbar(\eta^* \hat{a}^2 - \eta \hat{a}^{\dagger 2}),$$

where $\eta = \chi^{(2)}\beta$, and the associated evolution operator,

$$\hat{U}_I(t) = \exp[-i\hat{H}_I t/\hbar] = \exp[(\eta^* \hat{a}^2 - \eta \hat{a}^{\dagger 2})t] \equiv \hat{S}(\xi),$$

with $\xi = 2\eta t$.

- for degenerate four-wave mixing, in which two pump photons are converted into two signal photons of the same frequency,

$$\hat{H} = \hbar\omega \hat{a}^\dagger \hat{a} + \hbar\omega \hat{b}^\dagger \hat{b} + i\hbar\chi^{(3)}(\hat{a}^2 \hat{b}^{\dagger 2} - \hat{a}^{\dagger 2} \hat{b}^2),$$

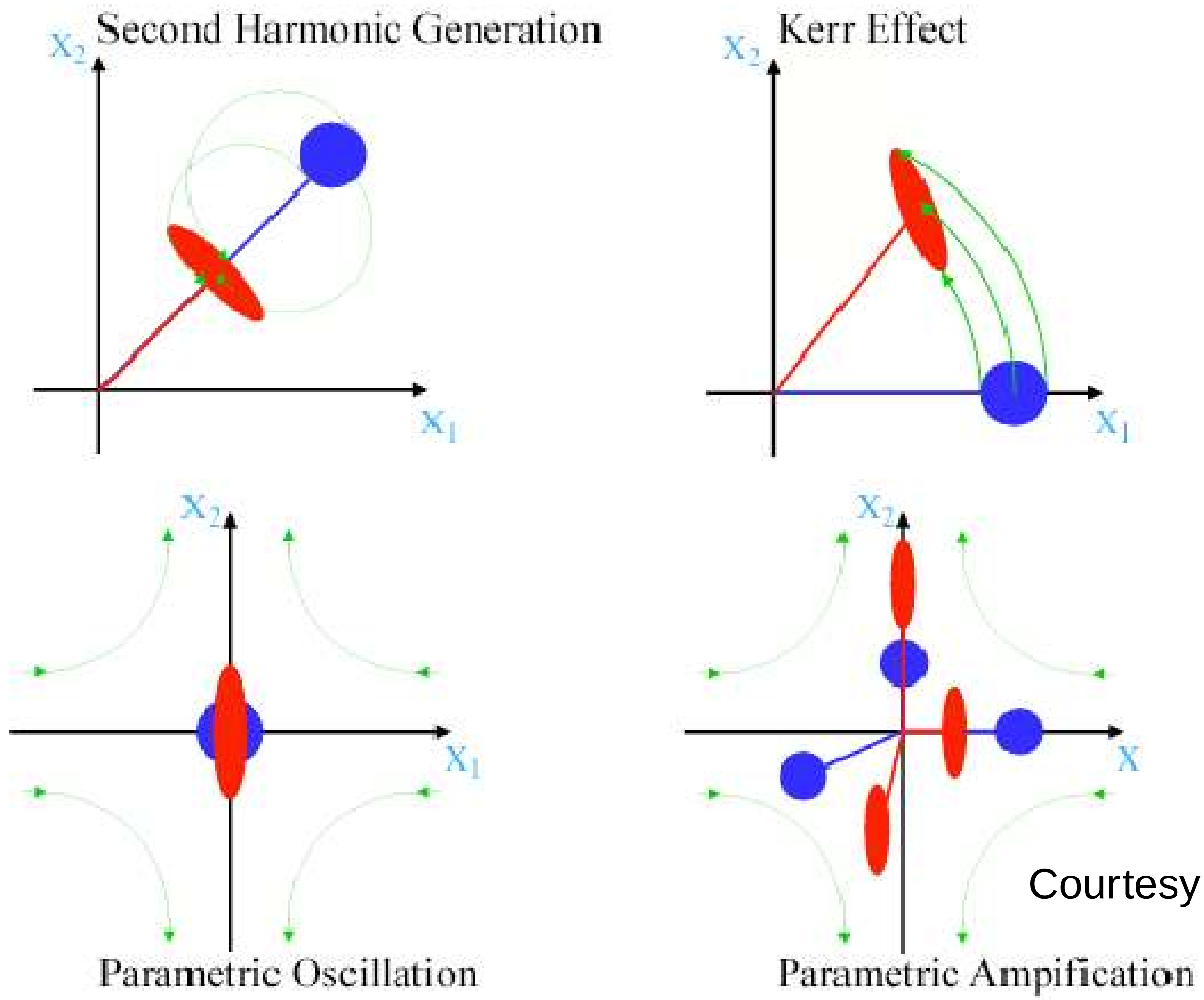
- the associated evolution operator,

$$\hat{U}_I(t) = \exp[(\eta^* \hat{a}^2 - \eta \hat{a}^{\dagger 2})t] \equiv \hat{S}(\xi),$$

with $\xi = 2\chi^{(3)}\beta^2 t$.

Generations of Squeezed States

Nonlinear optics:



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Squeezing in an Optical Parametric Oscillator

- when the nonlinear medium is placed within an optical cavity, oscillations build up inside and we have an *optical parametric oscillator* (OPO).
- this is a preferred method to generate squeezing, since the interaction is typically very weak and confining the light in a cavity helps to sizable effect by increasing the interaction time,
- loss from the cavity mirrors should be considered now,

$$\hat{H}_I = i\hbar(\eta^* \hat{a}^2 - \eta \hat{a}^{\dagger 2}),$$

- the dissipation and fluctuation should be included,

$$\begin{aligned} \frac{\mathrm{d}}{\mathrm{d}t}\hat{a} &= -\eta \hat{a}^\dagger - \frac{\Gamma}{2}\hat{a} + \hat{F}(t), \\ \frac{\mathrm{d}}{\mathrm{d}t}\hat{a}^\dagger &= -\eta \hat{a} - \frac{\Gamma}{2}\hat{a}^\dagger + \hat{F}^\dagger(t), \end{aligned}$$

where Γ represents the cavity decay and $\hat{F}(t)$ is the associated noise operator,

Squeezing in an Optical Parametric Oscillator

- for OPO,

$$\frac{\mathrm{d}}{\mathrm{d}t}\hat{a} = -\eta\hat{a}^\dagger - \frac{\Gamma}{2}\hat{a} + \hat{F}(t),$$
$$\frac{\mathrm{d}}{\mathrm{d}t}\hat{a}^\dagger = -\eta\hat{a} - \frac{\Gamma}{2}\hat{a}^\dagger + \hat{F}^\dagger(t),$$

  where Γ represents the cavity decay and $\hat{F}(t)$ is the associated noise operator,

- again the expectation value of the noise operator is zero, but with non-zero variances,

$$\langle\hat{F}_a(t)\rangle_R = \langle\hat{F}_a^\dagger(t)\rangle_R = 0,$$
$$\langle\hat{F}_a^\dagger(t)\hat{F}_a^\dagger(t')\rangle_R = \langle\hat{F}_a(t)\hat{F}_a(t')\rangle_R = 0,$$
$$\langle\hat{F}_a^\dagger(t)\hat{F}_a(t')\rangle_R = \Gamma\delta(t-t'),$$

Optical Parametric Oscillator in steady state

- the expectation values $\langle \hat{a} \rangle$ and $\langle \hat{a}^\dagger \rangle$ for OPO,

$$\frac{d}{dt}\langle \hat{a} \rangle = -\eta\langle \hat{a}^\dagger \rangle - \frac{\Gamma}{2}\langle \hat{a} \rangle,$$
$$\frac{d}{dt}\langle \hat{a}^\dagger \rangle = -\eta\langle \hat{a} \rangle - \frac{\Gamma}{2}\langle \hat{a}^\dagger \rangle,$$

where we have used $\langle \hat{F}(t) \rangle = \langle \hat{F}^\dagger \rangle = 0$, and the solution of this set of coupled equations is

$$\langle \hat{a}(t) \rangle = [\langle \hat{a}(0) \rangle \cosh \eta t - \langle \hat{a}^\dagger(0) \rangle \sinh \eta t] e^{-\Gamma t/2},$$
$$\langle \hat{a}^\dagger(t) \rangle = [\langle \hat{a}^\dagger(0) \rangle \cosh \eta t - \langle \hat{a}(0) \rangle \sinh \eta t] e^{-\Gamma t/2},$$

- it is clear that for an OPO operating below threshold, $\Gamma/2 > \eta$, in the steady state we have

$$\langle \hat{a} \rangle_{SS} = \langle \hat{a}^\dagger \rangle_{SS} = 0,$$

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Optical Parametric Oscillator in steady state

- next we look at the bilinear quantities $\langle \hat{a}^2 \rangle$, $\langle \hat{a}^{\dagger 2} \rangle$, and $\langle \hat{a}^\dagger \hat{a} \rangle$,
- define

$$A_1 = \langle \hat{a}^2 \rangle, \quad A_2 = \langle (\hat{a}\hat{a}^\dagger + \hat{a}^\dagger \hat{a}) \rangle, \quad A_3 = \langle \hat{a}^{\dagger 2} \rangle,$$

which satisfy the following set of equations,

$$\begin{aligned}
\frac{\mathrm{d}}{\mathrm{d}t} A_1 &= \frac{\mathrm{d}}{\mathrm{d}t} \langle \hat{a}^2 \rangle = -\eta A_2 - \Gamma A_1 + \langle (\hat{a}\hat{F} + \hat{F}\hat{a}) \rangle, \\
\frac{\mathrm{d}}{\mathrm{d}t} A_2 &= \frac{\mathrm{d}}{\mathrm{d}t} \langle (\hat{a}\hat{a}^\dagger + \hat{a}^\dagger \hat{a}) \rangle = -2\eta A_3 - 2\eta A_1 - \Gamma A_2 + \langle (\hat{a}\hat{F}^\dagger + \hat{F}^\dagger \hat{a} + \hat{a}^\dagger \hat{F} + \hat{F}\hat{a}^\dagger) \rangle \\
\frac{\mathrm{d}}{\mathrm{d}t} A_3 &= \frac{\mathrm{d}}{\mathrm{d}t} \langle \hat{a}^{\dagger 2} \rangle = -\eta A_2 - \Gamma A_3 + \langle (\hat{a}^\dagger \hat{F}^\dagger + \hat{F}^\dagger \hat{a}^\dagger) \rangle,
\end{aligned}$$

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Optical Parametric Oscillator in steady state

- for OPO,

$$\frac{\mathrm{d}}{\mathrm{d}t}\hat{a} = -\eta\hat{a}^\dagger - \frac{\Gamma}{2}\hat{a} + \hat{F}(t),$$
$$\frac{\mathrm{d}}{\mathrm{d}t}\hat{a}^\dagger = -\eta\hat{a} - \frac{\Gamma}{2}\hat{a}^\dagger + \hat{F}^\dagger(t),$$

- in order to determine the quantities involving the noise operators $\hat{F}$ and $\hat{F}^\dagger$, we rewrite above equations in the matrix form

$$\frac{\mathrm{d}}{\mathrm{d}t}\mathbf{A} = -\mathbf{M}\mathbf{A} + \mathbf{F},$$

where

$$\mathbf{A} = \begin{bmatrix} \hat{a} \\ \hat{a}^\dagger \end{bmatrix}, \quad \mathbf{M} = \begin{bmatrix} \frac{\Gamma}{2} & \eta \\ \eta & \frac{\Gamma}{2} \end{bmatrix}, \quad \mathbf{F} = \begin{bmatrix} \hat{F} \\ \hat{F}^\dagger \end{bmatrix},$$

Optical Parametric Oscillator in steady state

- for OPO,

$$\frac{\mathrm{d}}{\mathrm{d}t}\mathbf{A} = -\mathbf{M}\mathbf{A} + \mathbf{F},$$

which has a formal solution,

$$\mathbf{A}(t) = e^{-\mathbf{M}t}\mathbf{A}(0) + \int_0^t \mathrm{d}t' e^{-\mathbf{M}(t-t')}\mathbf{F}(t'),$$

- assume at the initial time $t = 0$, the field operators are statistically independent of the fluctuations, i.e. $\langle \hat{a}(0)\hat{F}(t)\rangle = 0$ etc., we obtain

$$\langle \mathbf{F}^\dagger(t)\mathbf{A}(t)\rangle = \begin{pmatrix} \langle \hat{F}^\dagger \hat{a}\rangle & \langle \hat{F}\hat{a}\rangle \\ \langle \hat{F}^\dagger \hat{a}^\dagger\rangle & \langle \hat{F}\hat{a}^\dagger\rangle \end{pmatrix} = \frac{\Gamma}{2}\begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix},$$

- in a similar manner,

$$\langle \mathbf{A}^\dagger(t)\mathbf{F}(t)\rangle = \begin{pmatrix} \langle \hat{a}^\dagger \hat{F}\rangle & \langle \hat{a}^\dagger\hat{F}^\dagger\rangle \\ \langle \hat{a}\hat{F}\rangle & \langle \hat{a}\hat{F}^\dagger\rangle \end{pmatrix} = \frac{\Gamma}{2}\begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix},$$

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Optical Parametric Oscillator in steady state

- in order to determine the quantities involving the noise operators $\hat{F}$ and $\hat{F}^\dagger$, we rewrite above equations in the matrix form

$$\begin{aligned}
\frac{\mathrm{d}}{\mathrm{d}t}A_1 &= \frac{\mathrm{d}}{\mathrm{d}t}\langle \hat{a}^2\rangle = -\eta A_2 - \Gamma A_1 + \langle(\hat{a}\hat{F} + \hat{F}\hat{a})\rangle, \\
\frac{\mathrm{d}}{\mathrm{d}t}A_2 &= \frac{\mathrm{d}}{\mathrm{d}t}\langle(\hat{a}\hat{a}^\dagger + \hat{a}^\dagger\hat{a})\rangle = -2\eta A_3 - 2\eta A_1 - \Gamma A_2 + \langle(\hat{a}\hat{F}^\dagger + \hat{F}^\dagger\hat{a} + \hat{a}^\dagger\hat{F} + \hat{F}\hat{a}^\dagger)\rangle \\
\frac{\mathrm{d}}{\mathrm{d}t}A_3 &= \frac{\mathrm{d}}{\mathrm{d}t}\langle \hat{a}^{\dagger 2}\rangle = -\eta A_2 - \Gamma A_3 + \langle(\hat{a}^\dagger\hat{F}^\dagger + \hat{F}^\dagger\hat{a}^\dagger)\rangle,
\end{aligned}$$

- all the correlation functions involving the noise operators above are zero except $\langle\hat{F}\hat{a}^\dagger\rangle = \langle\hat{a}\hat{F}^\dagger\rangle = \Gamma/2$, then

$$\begin{aligned}
\frac{\mathrm{d}}{\mathrm{d}t}A_1 &= -\eta A_2 - \Gamma A_1, \\
\frac{\mathrm{d}}{\mathrm{d}t}A_2 &= -2\eta A_3 - 2\eta A_1 - \Gamma A_2 + \Gamma, \\
\frac{\mathrm{d}}{\mathrm{d}t}A_3 &= -\eta A_2 - \Gamma A_3,
\end{aligned}$$

Optical Parametric Oscillator in steady state

- the steady state solutions for

$$\frac{\mathrm{d}}{\mathrm{d}t}A_1 = -\eta A_2 - \Gamma A_1,$$
$$\frac{\mathrm{d}}{\mathrm{d}t}A_2 = -2\eta A_3 - 2\eta A_1 - \Gamma A_2 + \Gamma,$$
$$\frac{\mathrm{d}}{\mathrm{d}t}A_3 = -\eta A_2 - \Gamma A_3,$$

are

$$A_1 = \langle \hat{a}^2 \rangle_{SS} = \frac{-\Gamma\eta}{4[(\Gamma/2)^2 - \eta^2]},$$
$$A_2 = \langle (\hat{a}\hat{a}^\dagger + \hat{a}^\dagger \hat{a}) \rangle_{SS} = \frac{\Gamma^2}{4[(\Gamma/2)^2 - \eta^2]},$$
$$A_3 = \langle \hat{a}^{\dagger 2} \rangle_{SS} = \frac{-\Gamma\eta}{4[(\Gamma/2)^2 - \eta^2]},$$

Optical Parametric Oscillator in steady state

- to see the squeezing, define the rotating quadrature operators,

$$\hat{X}_1 = \frac{1}{2}(\hat{a}e^{-i\theta/2} + \hat{a}^\dagger e^{i\theta/2}), \quad \hat{X}_2 = \frac{1}{2i}(\hat{a}e^{-i\theta/2} - \hat{a}^\dagger e^{i\theta/2}),$$

- the variances of these operators in the steady state are,

$$\begin{aligned}\langle \Delta \hat{X}_1 \rangle_{SS} &= \frac{1}{4}\langle(\hat{a}\hat{a}^\dagger + \hat{a}^\dagger\hat{a} + \hat{a}^2 e^{-i\theta} + \hat{a}^2 e^{i\theta})\rangle - \frac{1}{4}\langle(\hat{a}e^{-i\theta/2} + \hat{a}^\dagger e^{i\theta/2})\rangle^2, \\ &= \frac{1}{8}\frac{\Gamma}{\frac{\Gamma}{2} + \eta}, \\ \langle \Delta \hat{X}_2 \rangle_{SS} &= \frac{1}{8}\frac{\Gamma}{\frac{\Gamma}{2} - \eta},\end{aligned}$$

where we have taken $\theta = 0$,

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Optical Parametric Oscillator in steady state

- the best squeezing in an OPO is achieved on the oscillation threshold, $\eta = \Gamma/2$, giving

$$\langle \Delta \hat{X}_1 \rangle_{SS} = \frac{1}{8},$$

- this however represents only 50% squeezing below the vacuum level,
- in OPO, pairs of correlated (signal and idler) photons are producing,
- but the cavity mirror lets some single photon escape form each pair,
- so that some of the quantum correlation (and with it the squeezing) is lost,
- a theoretical limit of 50% squeezing is unattractive but the situation is different with the field outside the cavity,

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Spectrum of squeezing for the parametric oscillator

- below the threshold, the Hamiltonian for a parametric oscillator is

$$\hat{H}_S = \hbar\omega_0 \hat{a}^\dagger \hat{a} + \frac{i\hbar}{2}(\epsilon \hat{a}^{\dagger 2} - \epsilon^* \hat{a}^2),$$

then

$$[\mathbf{A} + (i\omega - \frac{\gamma}{2})\mathbf{1}]\mathbf{a}(\omega) = -\sqrt{\gamma}\mathbf{a}_I(\omega),$$

$$[\mathbf{A} + (i\omega + \frac{\gamma}{2})\mathbf{1}]\mathbf{a}(\omega) = +\sqrt{\gamma}\mathbf{a}_O(\omega),$$

where

$$\mathbf{A} = \begin{pmatrix} -i\omega_0 & \epsilon \\ \epsilon^* & i\omega_0 \end{pmatrix},$$

- the Fourier components for the output field is

$$\hat{a}_O(\omega) = \frac{1}{[\frac{\gamma}{2} - i(\omega - \omega_0)^2 - |\epsilon|^2]}\{[(\frac{\gamma}{2})^2 + (\omega - \omega_0)^2 + |\epsilon|^2]\hat{a}_I(\omega) + \epsilon\gamma\hat{a}_I^\dagger(-\omega)\},$$

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Spectrum of squeezing for the parametric oscillator

- the Fourier components for the output field is

$$\hat{a}_O(\omega) = \frac{1}{[\frac{\gamma}{2} - i(\omega-\omega_0)^2 - |\epsilon|^2]}\{[(\frac{\gamma}{2})^2 + (\omega-\omega_0)^2 + |\epsilon|^2]\hat{a}_I(\omega) + \epsilon\gamma\hat{a}_I^\dagger(-\omega)\},$$

- define the quadrature operators,

$$\hat{X}_1 = \frac{1}{2}(\hat{a}_O e^{-i\theta/2} + \hat{a}_O^\dagger e^{i\theta/2}), \quad \hat{X}_2 = \frac{1}{2i}(\hat{a}_O e^{-i\theta/2} - \hat{a}_O^\dagger e^{i\theta/2}),$$

  where θ is the phase of the pump,

- we find the following correlations,

$$\begin{aligned} \langle : \hat{X}_1(\omega), \hat{X}_1(\omega') : \rangle &= \frac{2\gamma|\epsilon|}{(\frac{\gamma}{2} - |\epsilon|)^2 + \omega^2}\delta(\omega+\omega'), \\ \langle : \hat{X}_2(\omega), \hat{X}_2(\omega') : \rangle &= \frac{-2\gamma|\epsilon|}{(\frac{\gamma}{2} + |\epsilon|)^2 + \omega^2}\delta(\omega+\omega'), \end{aligned}$$

  where $\langle : \hat{U}, \hat{V} : \rangle = \langle \hat{U}\hat{V}\rangle - \langle \hat{U}\rangle\langle\hat{V}\rangle$, and the input field $\hat{a}_I$ has been taken to be in the vacuum,

Spectrum of squeezing for the parametric oscillator

- we find the following correlations,

$$\langle : \hat{X}_1(\omega), \hat{X}_1(\omega') : \rangle = \frac{2\gamma|\epsilon|}{(\frac{\gamma}{2} - |\epsilon|)^2 + \omega^2}\delta(\omega + \omega'),$$
$$\langle : \hat{X}_2(\omega), \hat{X}_2(\omega') : \rangle = \frac{-2\gamma|\epsilon|}{(\frac{\gamma}{2} + |\epsilon|)^2 + \omega^2}\delta(\omega + \omega'),$$

- the δ function may be removed by integrating over ω' to give the normally ordered spectrum $S_1(\omega)$,

$$S_1(\omega) = \frac{2\gamma|\epsilon|}{(\frac{\gamma}{2} - |\epsilon|)^2 + \omega^2},$$

- the maximum squeezing occurs at the threshold for parametric oscillation, $|\epsilon| = \gamma/2$, where

$$S_1(\omega) = (\frac{\gamma}{\omega})^2,$$

- the light generated in parametric oscillation is said to be phase squeezed,

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Four-wave mixing

- Four-wave mixing is through the nonlinear susceptibility, $\chi^{(3)}$,
- two pump waves and the probe wave couple to produce the fourth wave, which is proportional to the spatial complex conjugate of the probe wave,
- for a probe wave,

$$E(r,t) = Re\{\mathbf{E}(r)\exp(i(\mathbf{k}\cdot\mathbf{r}-\omega t)\},$$

- the fourth wave is a phase conjugate wave,

$$E_{pc}(r,t) = Re\{\mathbf{E}^*(r)\exp(-i(\mathbf{k}\cdot\mathbf{r}+\omega t)\},$$

- equivalently, the spatial part of $E(r,t)$ remains unchanged and the sign of t is reversed,
- the phase conjugation is thus equivalent to *time reversal*,

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Classical four-wave mixing

- consider two intense pump waves E_2 and $E_{2'}$ traveling in opposite direction,
- and two weak fields E_1 and E_3 as the probe and conjugate waves,

$$E_j(r,t) = \frac{1}{2}\mathbf{E}_j(r)e^{i(k_j\cdot r-\omega t)} + \text{c.c.}, \quad \text{for} \quad (j = 1, 2, 2', 3),$$

- the wave directions imply,

$$k_1 + k_3 = 0, \quad k_2 + k_{2'} = 0,$$

- from the wave equation,

$$\nabla^2 E - \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P}{\partial t^2},$$

where $E = E_1 + E_2 + E_{2'} + E_3$, and $P = \chi^{(3)} E$,

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Classical four-wave mixing

- with the slowly varying envelope approximation, i.e.

$$|\frac{\mathrm{d}^2 E_i}{\mathrm{d}z^2}| \ll |k_i \frac{\mathrm{d}E_i}{\mathrm{d}z}|,$$

we have the following coupled-amplitude equations,

$$\begin{aligned} \frac{\mathrm{d}E_1}{\mathrm{d}z} &= (\frac{i\omega}{\epsilon c})P_1 = i\kappa_1 E_1 + i\kappa E_3^*, \\ \frac{\mathrm{d}E_3}{\mathrm{d}z} &= -(\frac{i\omega}{\epsilon c})P_3 = -i\kappa_1 E_3 - i\kappa E_1^*, \end{aligned}$$

where

$$\begin{aligned} P_1 &= \frac{3\chi^{(3)}}{8}(E_1^2 E_1^* + 2E_1 E_3 E_3^* + 2E_1 E_2 E_2^* + 2E_1 E_{2'} E2'^* + 2E_2 E_{2'} E_3^*), \\ P_3 &= \frac{3\chi^{(3)}}{8}(E_3^2 E_3^* + 2E_3 E_1 E_1^* + 2E_3 E_2 E_2^* + 2E_3 E_{2'} E2'^* + 2E_2 E_{2'} E_1^*), \\ \kappa &= \frac{3\omega\chi^{(3)}}{4\epsilon_0 c} E_2 E_{2'} \quad \kappa_1 = \frac{3\omega\chi^{(3)}}{4\epsilon_0 c}(|E_2|^2 + |E_{2'}|^2) \end{aligned}$$

Classical four-wave mixing

- with the slowly varying envelope approximation,

$$\frac{dE_1}{dz} = (\frac{i\omega}{\epsilon c})P_1 = i\kappa_1 E_1 + i\kappa E_3^*,$$
$$\frac{dE_3}{dz} = -(\frac{i\omega}{\epsilon c})P_3 = -i\kappa_1 E_3 - i\kappa E_1^*,$$

- define $\tilde{E}_1 = E_1 e^{-i\kappa_1 z}$ and $\tilde{E}_3 = E_3 e^{i\kappa_1 z}$, we have

$$\frac{d\tilde{E}_1}{dz} = i\kappa \tilde{E}_3^*,$$
$$\frac{d\tilde{E}_3}{dz} = -i\kappa \tilde{E}_1^*,$$

- for a nonlinear crystal with length L, the solutions are

$$\tilde{E}_1^*(z) = -\frac{i|\kappa|\sin(|\kappa|z)}{\kappa\cos(|\kappa|L)}\tilde{E}_3(L) + \frac{\cos[|\kappa|(z-L)]}{\cos(|\kappa|L}\tilde{E}_1^*(0),$$
$$\tilde{E}_3(z) = \frac{\cos[|\kappa|(z-L)]}{\cos(|\kappa|L}\tilde{E}_3(L) - \frac{i|\kappa|\sin(|\kappa|z)}{\kappa\cos(|\kappa|L)}\tilde{E}_1^*(0),$$

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Squeezing four-wave mixing

- if we replace the field variables $\tilde{E}_1$ and $\tilde{E}_3$ by the operators $\hat{a}_1$ and $\hat{a}_3$, then

$$\begin{aligned} \frac{\mathrm{d}\hat{a}_1}{\mathrm{d}z} &= i\kappa\hat{a}_3^\dagger, \\ \frac{\mathrm{d}\hat{a}_3}{\mathrm{d}z} &= -i\kappa\hat{a}_1^\dagger, \end{aligned}$$

- for a nonlinear crystal with length L, the solutions for $\kappa = |\kappa|$ are

$$\begin{aligned} \hat{a}_1(L) &= i\tan(\kappa L)\hat{a}_3^\dagger(L) + \sec(\kappa L)\hat{a}_1(0), \\ \hat{a}_3(0) &= \sec(\kappa L)\hat{a}_3(L) + i\tan(\kappa L)\hat{a}_1^\dagger(0), \end{aligned}$$

- define the quadrature components for the signal and the conjugate fields,

$$\hat{a}_{j1} = \frac{1}{2}(\hat{a}_j + \hat{a}_j^\dagger), \quad \hat{a}_{j2} = \frac{1}{2i}(\hat{a}_j - \hat{a}_j^\dagger), \quad \text{for} j = 1, 3,$$

- assume the input fields $\hat{a}_1(0)$ and $\hat{a}_3(L)$ to be in the coherent state, then

$$\Delta\hat{a}_{1i}^2(L) = \Delta\hat{a}_{3i}^2(0) = \frac{1}{4}[1 + 2\tan^2(\kappa L)], \quad \text{for} i = 1, 2,$$

Squeezing four-wave mixing

- assume the input fields $\hat{a}_1(0)$ and $\hat{a}_3(L)$ to be in the coherent state, then

$$\Delta\hat{a}_{1i}^2(L) = \Delta\hat{a}_{3i}^2(0) = \frac{1}{4}[1 + 2\tan^2(\kappa L)], \quad \text{for} i = 1, 2,$$

- the output fields are amplified as well as noisy,
- this is another manifold of the *dissipation-fluctuation* theory,

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Squeezing four-wave mixing

- define linear combination of the input modes,

$$\hat{d} = \frac{1}{\sqrt{2}}(\hat{a}_1 + \hat{a}_3)e^{i\theta},$$

and the canonically conjugate Hermitian amplitude operators,

$$\hat{d}_1 = \frac{1}{2}(\hat{d} + \hat{d}^\dagger), \quad \hat{d}_2 = \frac{1}{2i}(\hat{d} - \hat{d}^\dagger),$$

- the variance of the operator $\hat{d}_1$ and $\hat{d}_2$ are,

$$\Delta\hat{d}_1^2 = \frac{1}{4}[\sec(\kappa L) - \tan(\kappa L)]^2,$$
$$\Delta\hat{d}_2^2 = \frac{1}{4}[\sec(\kappa L) + \tan(\kappa L)]^2,$$

when $\theta = \pi/4$,

- as κL grows, the fluctuations in $\hat{d}_1$ are reduced below $1/4$, and eventually vanish as $\kappa L \to \pi/2$, the amplitude $\hat{d}_1$ is squeezed,

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