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.
define the squeezed state as

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

where the unitary squeeze operator

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

where $\xi = r \exp(i\theta)$ 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

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

- for $\theta = 0$, we have

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

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 $\omega_p$ and that field are converted into pairs of identical photons, of frequency $\omega = \omega_p/2$ each,

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

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

Second Harmonic Generation

Kerr Effect

Parametric Oscillation

Parametric Amplification

Courtesy of P. K. Lam
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,

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

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

where $\Gamma$ represents the cavity decay and $\hat{F}(t)$ is the associated noise operator,
Squeezing in an Optical Parametric Oscillator

for OPO,

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

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

where \(\Gamma\) 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,

\[
\langle \hat{F}_a(t) \rangle_R = \langle \hat{F}_a^\dagger(t) \rangle_R = 0,
\]

\[
\langle \hat{F}_a^\dagger(t) \hat{F}_a(t') \rangle_R = \langle \hat{F}_a(t) \hat{F}_a(t') \rangle_R = 0,
\]

\[
\langle \hat{F}_a^\dagger(t) \hat{F}_a(t') \rangle_R = \Gamma \delta(t - t'),
\]
Optical Parametric Oscillator in steady state

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

\[
\begin{align*}
\frac{d}{dt} \langle \hat{a} \rangle &= -\eta \langle \hat{a}^\dagger \rangle - \frac{\Gamma}{2} \langle \hat{a} \rangle, \\
\frac{d}{dt} \langle \hat{a}^\dagger \rangle &= -\eta \langle \hat{a} \rangle - \frac{\Gamma}{2} \langle \hat{a}^\dagger \rangle,
\end{align*}
\]

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

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

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

\[
\langle \hat{a} \rangle_{SS} = \langle \hat{a}^\dagger \rangle_{SS} = 0,
\]
Optical Parametric Oscillator in steady state

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

define

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

which satisfy the following set of equations,

$$\frac{d}{dt} A_1 = \frac{d}{dt} \langle \hat{a}^2 \rangle = -\eta A_2 - \Gamma A_1 + \langle (\hat{a}\hat{F} + \hat{F}\hat{a}) \rangle,$$

$$\frac{d}{dt} A_2 = \frac{d}{dt} \langle (\hat{a}\hat{a}^{\dagger} + \hat{a}^{\dagger}\hat{a}) \rangle = -2\eta A_3 - 2\eta A_1 - \Gamma A_2 + \langle (\hat{a}\hat{F}^{\dagger} + \hat{F}^{\dagger}\hat{a} + \hat{a}^{\dagger}\hat{F} + \hat{F}\hat{a}^{\dagger}) \rangle,$$

$$\frac{d}{dt} A_3 = \frac{d}{dt} \langle \hat{a}^{\dagger} \hat{a}^2 \rangle = -\eta A_2 - \Gamma A_3 + \langle (\hat{a}^{\dagger}\hat{F}^{\dagger} + \hat{F}^{\dagger}\hat{a}^{\dagger}) \rangle,$$
Optical Parametric Oscillator in steady state

for OPO,

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

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

\[
\frac{d}{dt} A = -MA + F,
\]

where

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

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Optical Parametric Oscillator in steady state

for OPO,

\[ \frac{d}{dt} A = -MA + F, \]

which has a formal solution,

\[ A(t) = e^{-Mt}A(0) + \int_0^t dt' e^{-(t-t')} F(t'), \]

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

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

in a similar manner,

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

\[
\frac{d}{dt} A_1 = \frac{d}{dt} \langle \hat{a}^2 \rangle = -\eta A_2 - \Gamma A_1 + \langle (\hat{a} \hat{F} + \hat{F} \hat{a}) \rangle,
\]

\[
\frac{d}{dt} A_2 = \frac{d}{dt} \langle (\hat{a} \hat{a}^{\dagger} + \hat{a}^{\dagger} \hat{a}) \rangle = -2\eta A_3 - 2\eta A_1 - \Gamma A_2 + \langle (\hat{a} \hat{F}^{\dagger} + \hat{F}^{\dagger} \hat{a} + \hat{a}^{\dagger} \hat{F} + \hat{F} \hat{a}^{\dagger}) \rangle,
\]

\[
\frac{d}{dt} A_3 = \frac{d}{dt} \langle \hat{a}^{\dagger^2} \rangle = -\eta A_2 - \Gamma A_3 + \langle (\hat{a}^{\dagger} \hat{F}^{\dagger} + \hat{F}^{\dagger} \hat{a}^{\dagger}) \rangle,
\]

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

\[
\frac{d}{dt} A_1 = -\eta A_2 - \Gamma A_1,
\]

\[
\frac{d}{dt} A_2 = -2\eta A_3 - 2\eta A_1 - \Gamma A_2 + \Gamma,
\]

\[
\frac{d}{dt} A_3 = -\eta A_2 - \Gamma A_3,
\]
Optical Parametric Oscillator in steady state

The steady state solutions for

\[
\frac{d}{dt} A_1 = -\eta A_2 - \Gamma A_1,
\]
\[
\frac{d}{dt} A_2 = -2\eta A_3 - 2\eta A_1 - \Gamma A_2 + \Gamma,
\]
\[
\frac{d}{dt} A_3 = -\eta A_2 - \Gamma A_3,
\]

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,

\[ \langle \Delta \hat{X}_1 \rangle_{SS} = \frac{1}{4} \langle (\hat{a}\hat{a}^\dagger + \hat{a}^\dagger \hat{a} + \hat{a}^2 e^{-i\theta} + \hat{a}^2 e^{i\theta}) \rangle - \frac{1}{4} \langle (\hat{a}e^{-i\theta/2} + \hat{a}^\dagger e^{i\theta/2}) \rangle^2, \]

\[ = \frac{1}{8} \frac{\Gamma}{\Gamma_2 + \eta}, \]

\[ \langle \Delta \hat{X}_2 \rangle_{SS} = \frac{1}{8} \frac{\Gamma}{\Gamma_2 - \eta}, \]

where we have taken \( \theta = 0 \),
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 from 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,
below the threshold, the Hamiltonian for a parametric oscillator is

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

then

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

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

where

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

the Fourier components for the output field is

\[ \hat{a}_O(\omega) = \frac{1}{[\frac{\gamma}{2} - i(\omega - \omega_0)^2 - |\epsilon|^2] \left\{ \left[ (\frac{\gamma}{2})^2 + (\omega - \omega_0)^2 + |\epsilon|^2 \right] \hat{a}_I(\omega) + \epsilon \gamma \hat{a}_I^\dagger(-\omega) \right\}}, \]
Spectrum of squeezing for the parametric oscillator

the Fourier components for the output field is

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

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 $\theta$ is the phase of the pump,

we find the following correlations,

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

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

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,
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 \(\delta\) function may be removed by integrating over \(\omega'\) 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

\[
S_1(\omega) = \left(\frac{\gamma}{\omega}\right)^2,
\]

the light generated in parametric oscillation is said to be phase squeezed,
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, 
  
  $$ E(r, t) = \text{Re}\{E(r)\exp(i(k \cdot r - \omega t)\}, $$
  
- the fourth wave is a phase conjugate wave, 
  
  $$ E_{pc}(r, t) = \text{Re}\{E^*(r)\exp(-i(k \cdot r + \omega t)\}, $$
  
- 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*,
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,

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

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 $P = \chi^{(3)} E$, 
with the slowly varying envelope approximation, i.e.
\[ \left| \frac{d^2 E_i}{dz^2} \right| \ll \left| k_i \frac{dE_i}{dz} \right|, \]
we have the following coupled-amplitude equations,
\[
\frac{dE_1}{dz} = \left( \frac{i\omega}{\epsilon c} \right) P_1 = i\kappa_1 E_1 + i\kappa E_3^*, \\
\frac{dE_3}{dz} = -\left( \frac{i\omega}{\epsilon c} \right) P_3 = -i\kappa_1 E_3 - i\kappa E_1^*,
\]
where
\[
P_1 = \frac{3\chi^{(3)}}{8} (E_1^2 E_1^* + 2E_1 E_3 E_3^* + 2E_1 E_2 E_2^* + 2E_1 E_2' E_2'^* + 2E_2 E_2' E_3^*), \\
P_3 = \frac{3\chi^{(3)}}{8} (E_3^2 E_3^* + 2E_3 E_1 E_1^* + 2E_3 E_2 E_2^* + 2E_3 E_2' E_2'^* + 2E_2 E_2' E_1^*), \\
\kappa = \frac{3\omega\chi^{(3)}}{4\epsilon_0 c} E_2 E_2', \quad \kappa_1 = \frac{3\omega\chi^{(3)}}{4\epsilon_0 c} (|E_2|^2 + |E_2'|^2)
\]
Classical four-wave mixing

with the slowly varying envelope approximation,

\[
\frac{dE_1}{dz} = \left( \frac{i\omega}{\epsilon c} \right) P_1 = i\kappa_1 E_1 + i\kappa E_3^*,
\]

\[
\frac{dE_3}{dz} = -\left( \frac{i\omega}{\epsilon c} \right) P_3 = -i\kappa_1 E_3 - i\kappa E_1^*,
\]

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

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

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

\[
\frac{d\hat{a}_1}{dz} = i\kappa \hat{a}_3^\dagger, \\
\frac{d\hat{a}_3}{dz} = -i\kappa \hat{a}_1^\dagger,
\]

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

\[
\hat{a}_1(L) = i \tan(\kappa L) \hat{a}_3^\dagger(L) + \sec(\kappa L) \hat{a}_1(0), \\
\hat{a}_3(0) = \sec(\kappa L) \hat{a}_3(L) + i \tan(\kappa L) \hat{a}_1^\dagger(0),
\]

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

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

the output fields are amplified as well as noisy,

this is another manifold of the *dissipation-fluctuation* theory,
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 \( \kappa L \) 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,