# Physics 581 - Quantum Optics II
## Lecture 3: Nonlinear Optics and Nonclassical Light

How do we produce nonclassical light? To better understand this, we need to revisit light-matter interactions. The light produced from natural sources - stars, lamps, etc. is thermal in nature. We have seen that such a state is represented by Bose-Einstein statistics (in the discrete photon variables) and Gaussian statistical fluctuations of the wave amplitude (continuous variable)

Thermal state.

$$\hat{\rho} = \bigotimes_k \hat{\rho}_k, \quad \hat{\rho}_k = \sum_{n_k} \frac{(\bar{n}_k)^{n_k}}{(\bar{n}_k+1)^{n_k+1}} |n_k\rangle\langle n_k| = \int d^2\alpha_k \, \frac{e^{-\frac{|\alpha_k|^2}{\bar{n}_k}}}{\pi \bar{n}_k} |\alpha_k\rangle\langle\alpha_k|$$

The state of the electromagnetic field arising from "engineered sources" that produce a stable wave with well defined amplitude and phase (such as radio wave produced by an antenna, or a laser field) is the coherent state $|\{\alpha_k\}\rangle$. Such states are created in quantum theory when the quantized field interacts with a classical current. The fundamental interaction Hamiltonian (in the interaction picture)

$$\hat{H}_{int} = -\int d^3r \, \frac{1}{c} \hat{\vec{J}}(\vec{r},t) \cdot \hat{\vec{A}}(\vec{r},t) \approx -\int d^3r \, \hat{\vec{P}}(\vec{r},t) \cdot \hat{\vec{E}}(\vec{r},t)$$

$$\hat{\vec{E}}(\vec{r},t) = \sum_{\vec{k},\lambda} \sqrt{\frac{2\pi\hbar\omega_k}{V}} \, \vec{\epsilon}_{\vec{k},\lambda} \, \hat{a}_{\vec{k},\lambda} \, e^{i(\vec{k}\cdot\vec{r} - \omega_k t)} + h.c.$$

$\hat{\vec{J}}(\vec{r},t)$ = Current operator $= \dfrac{\partial \hat{\vec{P}}(\vec{r},t)}{\partial t}$ (where polarization current dominates)

$$\hat{\vec{P}}(\vec{r},t) = \sum_i \hat{\vec{d}}_i(t) \, \delta(\vec{r} - \vec{r}_i(t))$$

When we can replace $\hat{\vec{d}}_i(t) \Rightarrow \langle \vec{d}_i \rangle$ (quantum fluctuations negligible)

$$\hat{\vec{P}}(\vec{r},t) \Rightarrow \vec{P}(\vec{r},t) \quad \text{(classical polarization current)}$$

$$\Rightarrow \hat{H}_{int} = \sum_{\vec{k},\lambda} \sqrt{\frac{2\pi\hbar\omega_k}{V}} \left( P^*_{\vec{k}\lambda}(t)\, e^{-i\omega_k t}\, \hat{a}_{\vec{k}\lambda} + P_{\vec{k}\lambda}(t)\, e^{+i\omega_k t}\, \hat{a}^\dagger_{\vec{k}\lambda} \right)$$

$$\int d\vec{r}\, \vec{P}(\vec{r},t)\cdot \vec{\epsilon}^{\,*}_{\vec{k}\lambda}\, e^{-i\vec{k}\cdot\vec{r}}$$

State evolved from vacuum $|\psi\rangle = \hat{U}(t)\,|0\rangle$

Since $[\hat{H}(t), \hat{H}(t')] =$ c-number we can exponentiate (up to overall phase)

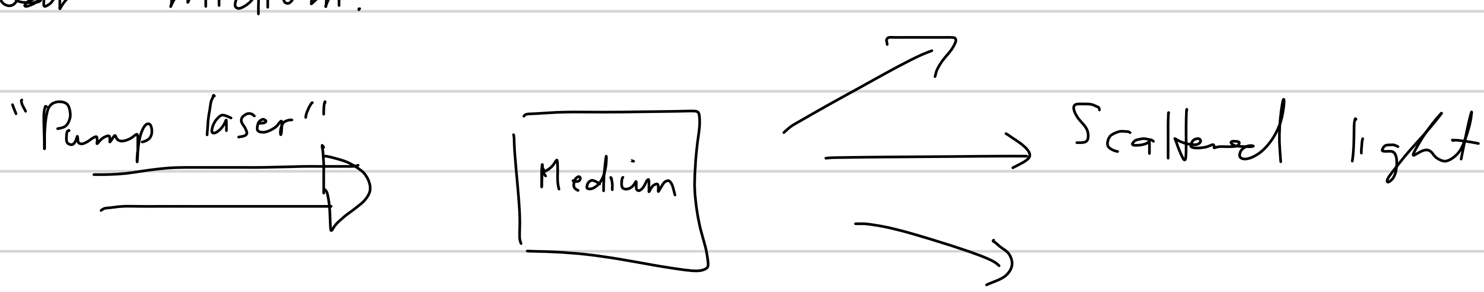
$$\hat{U}(t) = e^{-\frac{i}{\hbar}\int_0^t \hat{H}_{int}(t')dt'}\,|0\rangle = \prod_{\vec{k},\lambda} e^{\alpha_{\vec{k}\lambda}\hat{a}^\dagger_{k\lambda} - \alpha^*_{\vec{k}\lambda}\hat{a}_{k\lambda}}$$

$$\Rightarrow |\psi(t)\rangle = \prod_{\vec{k}\lambda} D(\alpha_{\vec{k}\lambda})\,|0\rangle = |\{\alpha_{k\lambda}\}\rangle$$

$$\alpha_{\vec{k}\lambda} = -i\sqrt{\frac{2\pi\omega_k}{\hbar V}} \int_0^t P_{\vec{k}\lambda}(t')\, e^{+i\omega_k t'} =$$ Radiated field of a dipole distribution

$\Rightarrow$ Classical current is the source of coherent state. A classically noisy current yield a statistical mixture of $|\{\alpha_k\}\rangle$.

To produce nonclassical light, we must drive atom dynamics that are not describable by a classical current. A route to implement this is nonlinear optics, where the intense field of a laser beam induces a nonlinear response of the medium. This is a kind of scattering problem, in which we scatter a laser field from a nonlinear medium:



In this picture, the medium mediates interactions between modes of the electromagnetic field. In steady-state, the medium's polarization density is determined by the field. In a perturbative regime, we can expand in power series in the field amplitude

$$P = \chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \cdots$$

At a given position $E(t) = E^{(+)}(t) + E^{(-)}(t)$, $\quad E^{(+)}(t) = \int_0^\infty d\omega\, \tilde{E}(\omega)\, e^{-i\omega t}$

⇒ Linear optics, $P^{(+)}(t) = \int_0^\infty d\omega\, \underbrace{\chi^{(1)}(\omega)\, \tilde{E}(\omega)}\, e^{-i\omega t}$

$\tilde{P}(\omega) =$ Frequency response at same $\omega$

⇒ Nonlinear, e.g., Second order

$$P^{(+)}(\vec{r}, t) = \int_0^\infty d\omega_1 d\omega_2\, \chi^{(2)}(\omega_3 = \omega_1 + \omega_2)\, \tilde{E}(\vec{r}; \omega_1)\, \tilde{E}(\vec{r}; \omega_2)\, e^{-i(\omega_1 + \omega_2)t}$$

$$+ \int_0^\infty d\omega_1 d\omega_2\, \chi^{(2)}(\omega_3 = \omega_1 - \omega_2)\, \tilde{E}(\vec{r}; \omega_1)\, \tilde{E}^*(\vec{r}; \omega_2)\, e^{-i(\omega_1 - \omega_2)t} \quad + \text{c.c.}$$

The "beat note" between the two field oscillates at the sum and difference frequencies. If the material responds to the force we can generate fields at $\omega_3 = \omega_1 \pm \omega_2$

The nonlinear wave equation that describes these processes is as follows. The wave equation with source

index of refraction

$$\left(\nabla^2 - \frac{n^2}{c^2} \frac{\partial^2}{\partial t^2}\right) E = \frac{4\pi}{c^2} \frac{\partial^2 P^{NL}}{\partial t^2}$$

Polarization Due to nonlinearity

We look for evolution of the field at a given frequency $\omega$. The evolution is taken only over one-dimension, $z$, for simplicity

Helmholtz eqn: $(\nabla^2 + k^2)\tilde{E}(z,\omega) = -4\pi \frac{\omega^2}{c^2} \tilde{P}^{NL}(z,\omega)$ $\qquad k(\omega) = n(\omega)\frac{\omega}{c}$

$\tilde{E}(z,\omega) = \tilde{\mathcal{E}}(z,\omega) e^{i(kz-\omega t)}$ $\qquad$ slowly varying envelope: $\frac{d\tilde{\mathcal{E}}}{dz} \ll k\mathcal{E}$

Frequency addition: $\omega_1, \omega_2 \Rightarrow \omega_3 = \omega_1 + \omega_2$

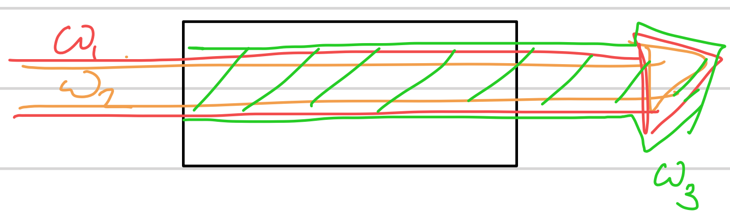
$$\tilde{P}(z,\omega_3) = \tilde{\chi}^{(2)}_{(\omega_3=\omega_1+\omega_2)} \tilde{\mathcal{E}}(z,\omega_1)\tilde{\mathcal{E}}(z,\omega_2) e^{i(k_1+k_2)z}$$

$$i2k_3 \frac{\partial \tilde{\mathcal{E}}(z,\omega_3)}{\partial z} e^{ik_3 z} = -\frac{4\pi\omega_3^2}{c^2}\chi^{(2)}(\omega_3=\omega_1+\omega_2)\tilde{\mathcal{E}}(z,\omega_1)\tilde{\mathcal{E}}(z,\omega_2) e^{i(k_1+k_2)z}$$

$$\Rightarrow \frac{d\tilde{\mathcal{E}}(z,\omega_3)}{dz} = +i\frac{2\pi\omega_3}{cn_3}\tilde{\chi}^{(2)}(\omega_3)\tilde{\mathcal{E}}(z,\omega_1)\tilde{\mathcal{E}}(z,\omega_2) e^{i\Delta k z}$$

$$\Delta k = k_1 + k_2 - k_3$$

Example of three-wave mixing




Often, the nonlinear effect is weak, and we can treat the "pump" input lasers as "undepleted", i.e., the amount of energy that is transferred from $\omega_1, \omega_2 \Rightarrow \omega_3$ is negligible, so $\tilde{\mathcal{E}}(z,\omega_1)$ and $\tilde{\mathcal{E}}(z,\omega_2)$ are approximately unchanged.

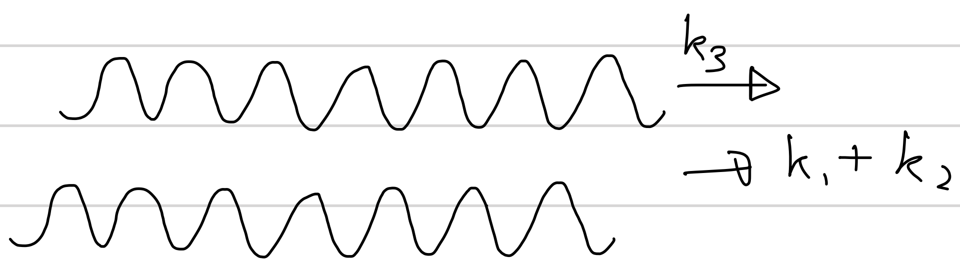
$$\Rightarrow \tilde{\mathcal{E}}(z,\omega_3) \approx \frac{2\pi\omega_3}{cn_3}\tilde{\chi}^{(2)}\tilde{\mathcal{E}}(\omega_1)\tilde{\mathcal{E}}(\omega_2)\frac{e^{i\Delta k z}-1}{\Delta k}$$

$\Rightarrow$ Intensity at output $z=L$

$$I_3 = \left(\frac{2\pi\omega_3}{cn_3}\tilde{\chi}^{(2)}\right)^2 I_1 I_2 \operatorname{sinc}^2\left(\frac{\Delta k L}{2}\right)\frac{L}{2}$$

The amount of sum-frequency generation depends on $\Delta k = k_3 - (k_1 + k_2)$

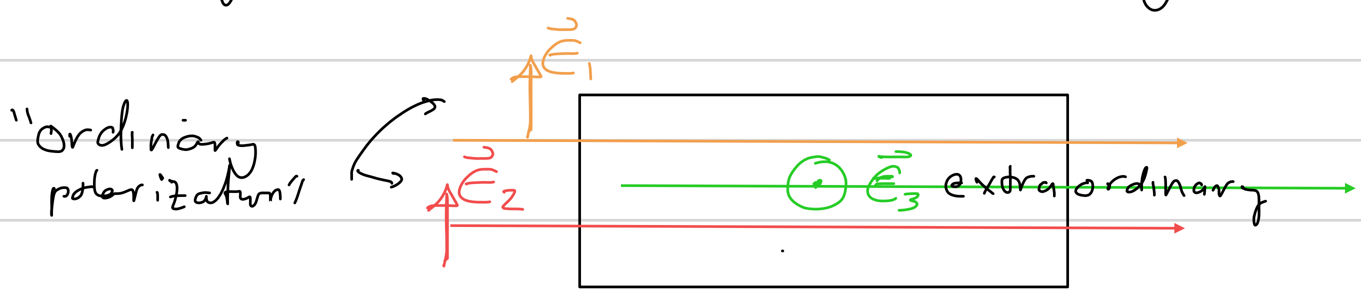
If $\Delta k = 0$, the nonlinear process is said to be "phase matched."
A phase-mismatch implies the "polarization wave" does not stay in phase with the wave $\mathcal{E}(\omega_3)\, e^{i(k_3 z - \omega_3 t)}$, so energy conversion is not efficient:




Phase matching: $k_1 + k_2 = k_3 \Rightarrow n(\omega_1)\,\omega_1 + n(\omega_2)\,\omega_2 = n(\omega_3)\,\omega_3$

Also $\omega_3 = \omega_1 + \omega_2$

Since the nonlinear crystal is generally dispersive, we need to play some tricks to achieve phase matching. We can use a birefringent crystal which has different indices of refraction for two orthogonal polarizations

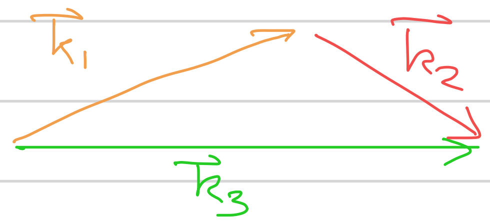



This configuration is known as "type-I" phase matching:
$\omega_1$ and $\omega_2$ have the same polarization, $\omega_3$ is orthogonal

$$\Rightarrow n_o(\omega_1)\,\omega_1 + n_o(\omega_2)\,\omega_2 = n_e(\omega)\,\omega_3$$

The same argument generalizes to non-collinear propagation


Phase matching




## General Three-Wave Mixing

When the phase matching conditions are satisfied:

$$\vec{k}_3 = \vec{k}_1 + \vec{k}_2 \,, \qquad \omega_3 = \omega_1 + \omega_2$$

The three waves $\mathcal{E}_i \, e^{i(\vec{k}_i\cdot\vec{r}-\omega_i t)}$, $i = 1, 2, 3$ are coupled according to

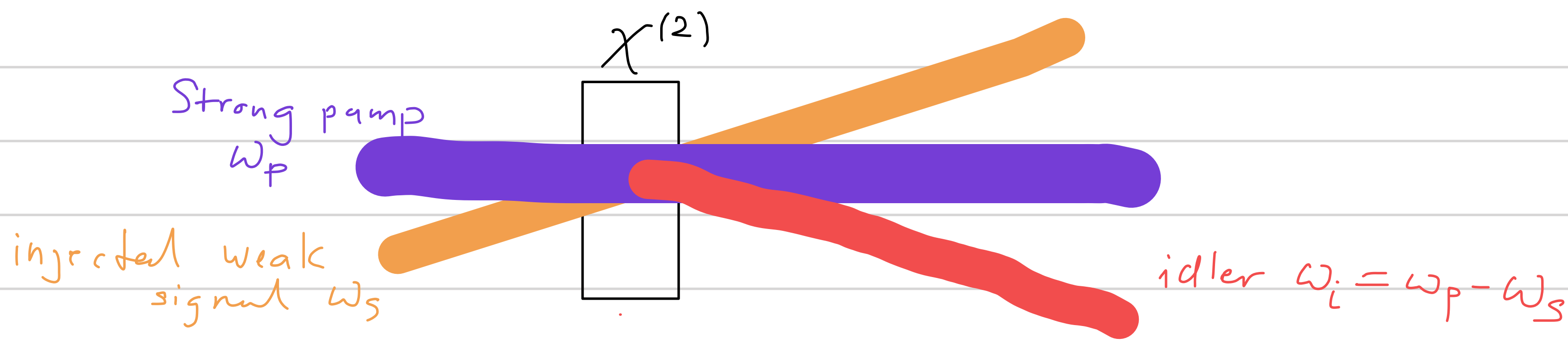
$$\frac{d\tilde{\mathcal{E}}_3}{dz} = i\frac{2\pi\omega_3}{cn_3}\tilde{\chi}^{(2)}\tilde{\mathcal{E}}_1\tilde{\mathcal{E}}_2$$

$$\frac{d\tilde{\mathcal{E}}_1}{dz} = i\frac{2\pi\omega_3}{cn_3}\tilde{\chi}^{(2)}\tilde{\mathcal{E}}_3\tilde{\mathcal{E}}_2^*$$

$$\frac{d\tilde{\mathcal{E}}_2}{dz} = i\frac{2\pi\omega_3}{cn_3}\tilde{\chi}^{(2)}\tilde{\mathcal{E}}_3\tilde{\mathcal{E}}_1^*$$

The first equation represents the sum frequency generation described above. The second two represent difference-frequency generation, where we input, e.g. frequency $\omega_3$ and $\omega_1$ and generate $\omega_2 = \omega_3 - \omega_1$.

Of particular interest is the case where $\omega_3$ is a strong "pump field" and $\omega_1$ is a weak field; $\omega_2$ is not injected. This configuration leads to "optical parametric amplification," (OPA), where we can amplify $\omega_1$ (typically referred to as the "signal" at $\omega_s$) transferring energy from the pump at $\omega_p$. Because this this a three-wave interaction, in the process we also generate a new wave at frequency $\omega_i = \omega_p - \omega_s$, known as the "idler."




Because the process is weak, we can work in the "nondepleted pump" limit, and treat the pump amplitude as constant. We are then left with two-coupled equations for signal and idler:

$$\frac{d\tilde{\mathcal{E}}_s}{dz} = \kappa\, \tilde{\mathcal{E}}_i^*, \qquad \frac{d\tilde{\mathcal{E}}_i}{dz} = \kappa\, \tilde{\mathcal{E}}_s^*$$

$$\kappa = i\frac{2\pi\omega_3}{cn_3}\tilde{\chi}^{(2)}\tilde{\mathcal{E}}_p = |\kappa| e^{2i\theta} \; ; \quad 2\theta = \phi_p + \frac{\pi}{2}$$

$$\Rightarrow \left.\begin{aligned} \tilde{\mathcal{E}}_s(z) &= \cosh(|\kappa| z)\,\tilde{\mathcal{E}}_s(0) + e^{i2\theta}\sinh(|\kappa| z)\,\tilde{\mathcal{E}}_i^*(0) \\ \tilde{\mathcal{E}}_i(z) &= \cosh(|\kappa| z)\,\tilde{\mathcal{E}}_i(0) + e^{-2i\theta}\sinh(|\kappa| z)\,\tilde{\mathcal{E}}_s^*(0) \end{aligned}\right\} \text{Bogoliubov transf.}$$

The solution with input conditions $\tilde{\mathcal{E}}_s(0) \neq 0 \quad \tilde{\mathcal{E}}_i(0) = 0$

$$\tilde{\mathcal{E}}_s(z) = \underbrace{\cosh(|\kappa| z)}_{\text{"Parametric Gain"}}\tilde{\mathcal{E}}_s(0), \qquad \tilde{\mathcal{E}}_i(z) = e^{-2i\theta}\sinh(|\kappa| z)\,\tilde{\mathcal{E}}_s^*(0)$$

"Phase Conjugate Amplification"

## Quantum theory

The fundmental Hamiltonian at hand

$$\hat{H} = -\int d^3r \, \hat{\vec{P}}(\vec{r}) \cdot \hat{\vec{E}}(\vec{r})$$

The dominant (steady-state) processes are seen in the RWA

$$\hat{H} = -\int d^3r \, \vec{P}^{(-)}(\vec{r}) \cdot \vec{E}^{(+)}(\vec{r}) \quad + h.c.$$

Example: Linear Optics $\quad \vec{P}^{(+)} = \chi^{(1)} \vec{E}^{(+)}$

$$\hat{H} = \sum_{ij} h_{ij} \, \hat{a}_i^\dagger \hat{a}_j \qquad (i = \vec{k}\lambda, \; j = \vec{k}'\lambda' \text{ short hand}) \quad |\vec{k}| = |\vec{k}'|$$

$$h_{ij} = -\int \frac{d^3r}{V} \, \chi^{(1)}(\vec{r}) \, 2\pi\hbar\omega_{k_i} \, e^{i(\vec{k}_i - \vec{k}_j)} = -2\pi \, \tilde{\chi}^{(1)}(\vec{q} = \vec{k}_i - \vec{k}_j) \, \hbar\omega_k$$

For linear (non absorbing) optics the photon number and frequency are conserved. The photons are annihilated in a mode and created in a mode with the same frequency.

The scattering matrix generated by this Hamiltonien

$$\hat{U} = e^{-\frac{i}{\hbar}\Delta t \hat{H}} \quad , \text{ where } \Delta t = \text{interaction time (e.g. } \tfrac{L}{c} \text{ for 1D)}$$

$$\Rightarrow \hat{U} = e^{-i\sum_{ij} \Theta_{ij} \, \hat{a}_i^\dagger \hat{a}_j}$$

$$\Theta_{ij} = -2\pi \, \tilde{\chi}^{(1)}(\vec{q} = \vec{k}_i - \vec{k}_j) \, \omega_k \, \Delta t$$

There are two kinds of processes. Consider only two modes

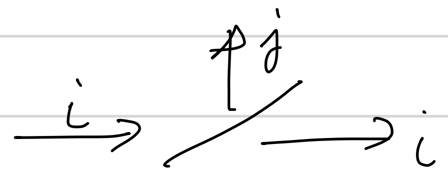
- $i = j \Rightarrow \hat{U}^{(i)} = e^{-i\theta_i \hat{a}_i^\dagger \hat{a}_i}$ : Phase space rotation (phase shift)

$$\hat{U}^{(i)\dagger} \hat{a}_i \hat{U}^{(i)} = e^{-i\theta_i} \hat{a}_i$$

- $i \neq j \Rightarrow \hat{U}^{(ij)} = e^{-i(\theta_{ij} \hat{a}_i^\dagger \hat{a}_j + \theta_{ij}^* \hat{a}_j^\dagger \hat{a}_i)}$

$$\Rightarrow \hat{U}^{(ij)\dagger} \hat{a}_i \hat{U}^{(ij)} = \cos\frac{\theta_{ij}}{2} \hat{a}_i + i \sin\frac{\theta_{ij}}{2} \hat{a}_j \quad (\text{here } \theta_{ij} \text{ real})$$

Scattering, e.g. beam splitter




Generally, for linear optics $U^\dagger \hat{a}_i U = \sum_j \underset{\substack{\uparrow \\ \text{classical scattering matrix}}}{\Lambda_{ij}} \hat{a}_j$

## Example: Three-wave mixing and parametric amplification

We consider the interaction of 3 modes: A pump, signal, and idler

$$\hat{\vec{E}}^{(+)}_{p,s,i}(\vec{r},t) = \sqrt{\frac{2\pi\hbar\omega_{p,s,i}}{V}}\, \hat{a}_{p,s,i}\, \vec{\epsilon}_{p,s,i}\, e^{i(\vec{k}_{p,s,i}\cdot\vec{r} - \omega_{p,s,i} t)},$$

$$\boxed{\text{Phase-matched} \quad \omega_p = \omega_s + \omega_i, \quad \vec{k}_p = \vec{k}_s + \vec{k}_i}$$

The induced polarization wave: $\hat{\vec{P}}(r,t) = \chi^{(2)} \hat{\vec{E}}(\vec{r},t) \hat{\vec{E}}(\vec{r},t)$

$$\Rightarrow \hat{H} = -\int d^3r\, \chi^{(2)} \vdots\, \vec{E}_p^{(+)}(\vec{r},t)\, \vec{E}_s^{(-)}(\vec{r},t)\, \vec{E}_i^{(-)}(\vec{r},t) + h.c.$$

$$= (\quad) \underbrace{\hat{a}_p \hat{a}_s^\dagger \hat{a}_i^\dagger}_{\text{Parametric down-conversion and amplification}} + (\quad) \underbrace{\hat{a}_p^\dagger \hat{a}_s \hat{a}_i}_{\text{sum frequency generation}}$$

The first term corresponds to frequency down conversion (known as parametric downconversion) whereby a pump photon is annihated and a signal and a signal and idler photon are created. The second term represents sum frequency generation whereby a signal and idler photon are annihilated and a pump photon is created.

In the case that we input a strong pump, and in the non-depletion limit, we can replace $\vec{E}_p^{(+)}$ with a classical mean value

$$\Rightarrow \hat{H} = K^* \hat{a}_s \hat{a}_i + K \hat{a}_i^\dagger \hat{a}_s^\dagger \quad , \text{ where } K^* \propto \chi^{(2)} \mathcal{E}_p^*$$

For example, in the degenerate case, $\hat{a}_s = \hat{a}_i = \hat{a}$

$\vec{k}_s$ $\vec{k}_i$

$\vec{k}_p$ (Collinear phase matching)

$\hat{H} = K\hat{a}^2 + K^* \hat{a}^{\dagger 2}$ : This Hamiltonian is the generator of the squeezing operator: $\hat{S} = e^{-i\frac{L}{c}\hat{H}} = \exp\left\{\frac{1}{2}(\zeta^* \hat{a}^2 - \zeta \hat{a}^{\dagger 2})\right\}$

$$\zeta = \frac{|K|L}{2}$$

This evolution generates squeezed vacuum. To increase the interaction strength, the three-wave mixing process enhanced in an optical cavity. Such a system is known as an optical parametric oscillator OPO. An OPO is the standard source of squeezed vacuum.

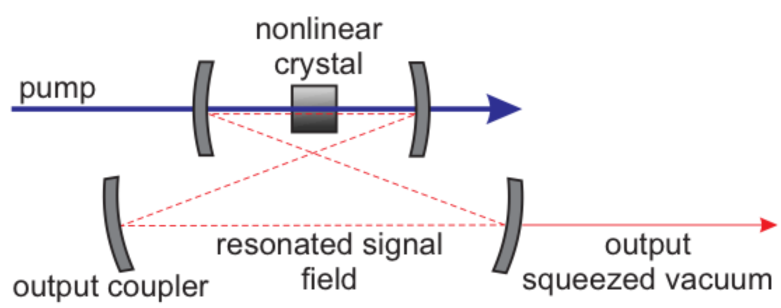



FIG. 12. Squeezing in an OPA cavity. The cavity mirrors are reflective to the signal field, but transparent to the pump.

From "Squeezed Light" arXiv: 1401.4118 A. Lvovsky