Lecture 10. Generation of nonclassical light through parametric down-conversion and four-wave mixing

Parametric down-conversion: Hamiltonian, perturbation theory, Heisenberg approach. Spontaneous four-wave mixing. Nonclassical features.

1. Hamiltonian of parametric down-conversion

At the last lecture we saw how parametric down-conversion can be derived 'almost classically'. Today we will derive it quantum-mechanically.

In the dipole approximation, the interaction between the matter and light has the energy

$$H = -\vec{d}\vec{E},$$

where $\vec{E}$ is the electric field, and the dipole moment can be written as the volume integral of the polarization,

$$\vec{d} = \int d^3 r \vec{P}(r).$$

Here, we take the second-order nonlinear polarization:

$$\vec{P} = \varepsilon_0 \chi^{(2)} \vec{E} \cdot \vec{E}. \qquad (1)$$

After substituting all this, we see that $H = -\varepsilon_0 \int d^3 r \chi^{(2)}(r) \vdots \vec{E}\vec{E}\vec{E}.$

We omit the vectors, and assume first that there are three real fields: the pump, signal, idler

$$E^3 = (E_0 e^{i\omega_0 t - i\vec{k}_0 \vec{r}} + E_1 e^{i\omega_1 t - i\vec{k}_1 \vec{r}} + E_2 e^{i\omega_2 t - i\vec{k}_2 \vec{r}} + c.c.)^3$$

We are interested only in the term

$$\sim E_0 e^{i\omega_0 t - i\vec{k}_0 \vec{r}} E_1^* e^{-i\omega_1 t + i\vec{k}_1 \vec{r}} E_2^* e^{-i\omega_2 t + i\vec{k}_2 \vec{r}} + c.c.$$

In the quantum picture, the pump field is classical but for waves 1,2 we have photon creation and annihilation operators. And then the Hamiltonian is (we omit the unimportant constants)

$$\hat{H} \sim \int d^3 r \chi^{(2)}(\vec{r}) E_0 e^{i(\omega_0 - \omega_1 - \omega_2)t - i(\vec{k}_0 - \vec{k}_1 - \vec{k}_2)\vec{r}} a_1^+ a_2^+ + h.c. \equiv \int d^3 r \chi^{(2)}(\vec{r}) E_0 e^{i\Delta\omega t - i\Delta\vec{k}\vec{r}} a_1^+ a_2^+ + h.c.$$

Here we introduced frequency and wavevector mismatches. The volume integral is over the space in the crystal occupied by the pump. The transverse size of the pump, for simplicity, can be assumed to be infinite. This leads to a factor $\delta(\Delta k_x)\delta(\Delta k_y)$ in the Hamiltonian. Along the z axis (Fig.1), there is the length of the crystal, and the integral is

$$\int_{-L}^{0} dz e^{-i\Delta k_z z} = \left. \frac{e^{-i\Delta k_z z}}{-i\Delta k_z} \right|_{-L}^{0} = \frac{2\sin(\Delta k_z L/2)}{\Delta k_z} e^{-i\Delta k_z L/2} =$$

$$= L \operatorname{sinc}(\Delta k_z L/2) e^{-i\Delta k_z L/2} \equiv L F(\Delta k_z)$$

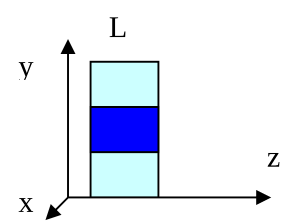

Fig.1

Up to the factor $F(\Delta k_z)$, the Hamiltonian becomes simple:

$$\hat{H} \sim \chi^{(2)} E_0 L a_1^+ a_2^+ e^{i\Delta\omega t} + h.c.$$

The Hamiltonian becomes time-independent (why do we need this?) if $\Delta\omega = 0$.
Then, the Hamiltonian can be written as

$$\hat{H} \sim \chi^{(2)} E_0 L a_1^+ a_2^+ + h.c. \equiv i\hbar\Gamma a_1^+ a_2^+ + h.c., \qquad (2)$$

where we deliberately dragged out the Planck constant and included all relevant parameters (crystal length, quadratic nonlinearity, field of the pump, etc.) into Γ (coupling parameter).

2. Perturbation theory and photon pairs

In the Schroedinger approach, we can describe the evolution of the state vector:

$$i\hbar \frac{d}{dt}\left|\Psi\right\rangle = \hat{H}\left|\Psi\right\rangle;$$

$$\left|\Psi\right\rangle = e^{\frac{1}{i\hbar}\int \hat{H}dt}\left|\Psi\right\rangle_0$$

The initial state is the vacuum and the Hamiltonian is taken in the form (2). Then,

$$\left|\Psi\right\rangle = e^{\Gamma t a_1^+ a_2^+}\left|vac\right\rangle \approx \left|vac\right\rangle + \Gamma t a_1^+ a_2^+ \left|vac\right\rangle + \frac{1}{2}\Gamma^2 t^2 a_1^{+2} a_2^{+2}\left|vac\right\rangle + \ldots \qquad (3)$$

In the first order of the perturbation theory, we only keep the first two terms. This describes the generation of the two-photon Fock state or an entangled state of two photons but only in superposition with the vacuum.

3. Spontaneous four-wave mixing

As mentioned in Lecture 9, SFWM is very similar to PDC – only two pump photons take part instead of one. This can be illustrated by the diagrams in Fig.2.

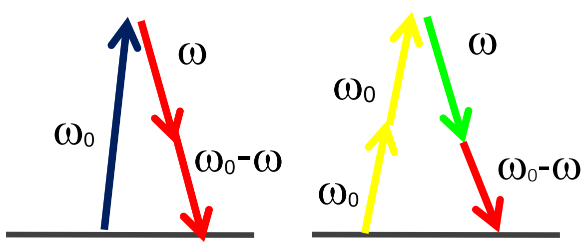

Fig.2

To derive the Hamiltonian for SFWM, we have to take, instead of (1), the cubic polarization,

$$\vec{P} = \varepsilon_0 \chi^{(3)} \vec{E}\cdot\vec{E}\cdot\vec{E}.$$

After choosing the proper terms (and which ones are proper, depends on the phase matching), the Hamiltonian takes the form

$$\hat{H} \sim \chi^{(3)} E_0^{\ 2} L a_1^+ a_2^+ + h.c. \equiv i\hbar\Gamma a_1^+ a_2^+ + h.c. \qquad (4)$$

It creates photon pairs, similarly to PDC, but has quadratic dependence on the pump field; also, there is cubic nonlinearity instead of the quadratic one. For this reason, SFWM can be realized not only in chi-2 materials but in all materials (chi-3 is present everywhere).
For PDC, $\Gamma \sim \chi^{(2)} E_0 L$ and for FWM $\Gamma \sim \chi^{(3)} E_0^2 L$.

4. Single-mode squeezed vacuum

What happens when the interaction becomes strong?
The Hamiltonian is the same,

$$H = i\hbar\Gamma a_1^+ a_2^+ + h.c. \qquad (5)$$

It describes both PDC and SFWM.
But the perturbation theory does not work any more. We cannot do expansion (3). Or, rather, we can but we cannot keep only the first term.

We have to write the whole state:

$$|\Psi\rangle = e^{\Gamma t a_1^+ a_2^+}|vac\rangle = |vac\rangle + \Gamma t a_1^+ a_2^+ |vac\rangle + \frac{1}{2}\Gamma^2 t^2 a_1^{+2} a_2^{+2}|vac\rangle + ...$$

This is what is called **squeezed vacuum**. The reason why it is called so will be clear from what follows.

We are going to describe what happens due to the Hamiltonian (5) in the case of strong interaction. In fact, we are not so much interested in the state, but rather in the values of observables. It means that we can use the Heisenberg approach, in which we will write the Heisenberg equations and get the evolution of the operators. Then, by averaging the operator expressions over the vacuum state, we will calculate everything we need.

Consider first the case where the modes 1 and 2 coincide (this will be so under certain phase matching conditions):

$$H = i\hbar\Gamma a^{+2} + h.c.$$

We immediately see that this will result in **light with even photon numbers**,

$$|\Psi\rangle = c_0|0\rangle + c_2|2\rangle + c_4|4\rangle + ...$$

For calculating the evolution of the operators, we write the Heisenberg equations:

$$i\hbar\frac{da}{dt} = [a, H].$$

We get

$$i\hbar\frac{da}{dt} = 2i\hbar\Gamma a^+,$$

And let us pass to quadratures, $\hat{x}_1 = (a + a^+)/2, \quad \hat{x}_2 = (a - a^+)/2i$. For the quadratures, we get very simple equations,

$$\frac{d\hat{x}_1}{dt} = 2\Gamma\hat{x}_1, \frac{d\hat{x}_2}{dt} = -2\Gamma\hat{x}_2.$$

The solutions are $\hat{x}_1 = e^{2\Gamma t}\hat{x}_{10}, \hat{x}_2 = e^{-2\Gamma t}\hat{x}_{20}$. This evolution is described by hyperbolas in the phase space: $\hat{x}_1\hat{x}_2 = \hat{x}_{10}\hat{x}_{20} = const$. We can draw these hyperbolas if we assume that we are allowed to draw points in the phase space. In reality, we cannot draw points but rather, 'areas' (Fig.3).

One can see that an input coherent state (C) becomes a squeezed coherent state (SC) but a vacuum (V) becomes a squeezed vacuum (SV, this is where the name comes from). Further, we will only consider SV.

We see that one quadrature becomes squeezed. Why is this good? Because the noise in this quadrature is reduced at the cost of increasing the noise in the other quadrature.

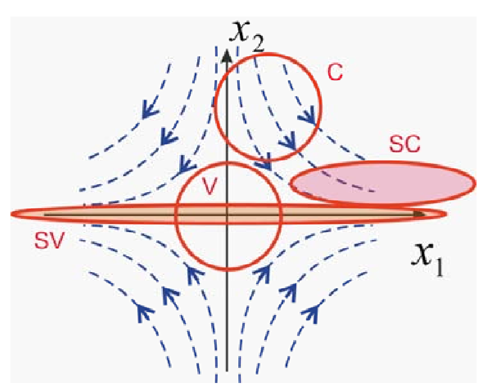

Fig.3

We can further look into the equations; to calculate everything for this light it is reasonable to return to the photon creation and annihilation operators. Then we get the transformations

$$\hat{x}_1 = e^{2\Gamma t}\hat{x}_{10},\ \hat{x}_2 = e^{-2\Gamma t}\hat{x}_{20}$$
$$a = \hat{x}_1 + i\hat{x}_2 = e^{2\Gamma t}\hat{x}_{10} + ie^{-2\Gamma t}\hat{x}_{20} = e^{2\Gamma t}(a_0 + a_0^+)/2 + ie^{-2\Gamma t}(a_0 - a_0^+)/2i = a_0\cosh(\Gamma t) + a_0^+\sinh(\Gamma t).$$

This is called the single-mode Bogolyubov transformations. The combination $\Gamma t \equiv G$ is often called the parametric gain. This parameter determines all properties of SV.

For instance, we can find the mean photon number per mode:
$$\langle N\rangle = \langle a^+a\rangle = \langle (a_0{}^+\cosh G + a_0\sinh G)(a_0\cosh G + a_0^+\sinh G)\rangle = \langle a_0\sinh G a_0^+\sinh G\rangle = \sinh^2 G.$$
At small gain, this is a linear function of the pump power, but at high gain, it becomes nonlinear.

A bit more complicated is calculation of correlation functions. For instance,
$$G^{(2)} = \langle (a^+)^2a^2\rangle = \langle (a_0{}^+\cosh G + a_0\sinh G)^2(a_0\cosh G + a_0^+\sinh G)^2\rangle =$$
$$= \langle a_0\sinh G(a_0{}^+\cosh G + a_0\sinh G)(a_0\cosh G + a_0^+\sinh G)a_0^+\sinh G\rangle =$$
$$= \sinh^2 G\langle (\cosh G + a_0^2\sinh G)(\cosh G + a_0^{+2}\sinh G)\rangle = \sinh^2 G(\cosh^2 G + 2\sinh^2 G) =$$
$$= \sinh^2 G(1 + 3\sinh^2 G).$$
Then, the normalized correlation function is
$$g^{(2)} = 3 + \frac{1}{\langle N\rangle}. \qquad (6)$$
A similar, just more bulky calculation, gives
$$g^{(3)} = 15 + \frac{9}{\langle N\rangle}. \qquad (7)$$

5. Two-mode squeezed vacuum.

If there are two modes, and the Hamiltonian has the form (5), the Heisenberg equations
$$i\hbar\frac{da_1}{dt} = [a_1, H],$$
$$i\hbar\frac{da_2}{dt} = [a_2, H].$$
We obtain
$$\frac{da_1}{dt} = \Gamma a_2^+,$$
$$\frac{da_2}{dt} = \Gamma a_1^+.$$
The solutions (check) lead to the two-mode Bogolyubov transformations:
$$a_1 = a_{10}\cosh G + a_{20}^+\sinh G,$$
$$a_2 = a_{20}\cosh G + a_{10}^+\sinh G.$$

From them, we can find that not only the mean numbers in modes 1,2 are the same, $\langle N_1\rangle = \langle N_2\rangle = \sinh^2 G$, but also that the variance of the photon-number difference does not fluctuate:

$$Var(\hat{N}_1 - \hat{N}_2) = \left\langle (\hat{N}_1 - \hat{N}_2)^2 \right\rangle - \left\langle \hat{N}_1 - \hat{N}_2 \right\rangle^2 = \left\langle (\hat{N}_1 - \hat{N}_2)^2 \right\rangle = \left\langle (\hat{N}_1)^2 \right\rangle + \left\langle (\hat{N}_2)^2 \right\rangle - 2\left\langle \hat{N}_1 \hat{N}_2 \right\rangle.$$

Due to symmetry, it will be

$$Var(\hat{N}_1 - \hat{N}_2) = 2\left\langle (\hat{N}_1)^2 \right\rangle - 2\left\langle \hat{N}_1 \hat{N}_2 \right\rangle.$$

$$\left\langle (\hat{N}_1)^2 \right\rangle = \left\langle \begin{matrix} (a_{10}{}^{+} \cosh G + a_{20} \sinh G)(a_{10} \cosh G + a_{20}^{+} \sinh G) \times \\ \times (a_{10}{}^{+} \cosh G + a_{20} \sinh G)(a_{10} \cosh G + a_{20}^{+} \sinh G) \end{matrix} \right\rangle =$$

$$= \left\langle a_{20} \sinh G (a_{10} \cosh G + a_{20}^{+} \sinh G)(a_{10}{}^{+} \cosh G + a_{20} \sinh G) a_{20}^{+} \sinh G \right\rangle =$$

$$= \sinh^2 G \left\langle (a_{10} a_{20} \cosh G + \sinh G)(a_{10}{}^{+} a_{20}^{+} \cosh G + \sinh G) \right\rangle = \sinh^2 G (\sinh^2 G + \cosh^2 G).$$

$$\left\langle \hat{N}_1 \hat{N}_2 \right\rangle = \left\langle \begin{matrix} (a_{10}{}^{+} \cosh G + a_{20} \sinh G)(a_{10} \cosh G + a_{20}^{+} \sinh G) \times \\ \times (a_{20}{}^{+} \cosh G + a_{10} \sinh G)(a_{20} \cosh G + a_{10}^{+} \sinh G) \end{matrix} \right\rangle =$$

$$= \left\langle a_{20} \sinh G (a_{10} \cosh G + a_{20}^{+} \sinh G)(a_{20}{}^{+} \cosh G + a_{10} \sinh G) a_{10}^{+} \sinh G \right\rangle =$$

$$= \sinh^2 G \left\langle (a_{10} a_{20} \cosh G + \sinh G)(a_{10}{}^{+} a_{20}^{+} \cosh G + \sinh G) \right\rangle = \sinh^2 G (\sinh^2 G + \cosh^2 G).$$

Therefore,

$$Var(\hat{N}_1 - \hat{N}_2) = 0. \quad (8)$$

6. Nonclassical features.

Let us check what nonclassical features we can see for SV.

1. *Squeezing* in one or another quadrature (Fig.3; take into account that the picture also rotates). Squeezing will be noticeable only at large parametric gain.

2. Super-bunching. Let us consider the conditions for normalized correlation functions. At small gain, we will have

$$\frac{g^{(3)}}{\left[g^{(2)}\right]^2} = \frac{15 + \dfrac{9}{\langle N\rangle}}{9 + \dfrac{6}{\langle N\rangle} + \dfrac{1}{\langle N\rangle^2}} \approx 9\langle N\rangle << 1 .$$

Therefore, only at small gain, when all CFs boost, the CF nonclassicality condition can be satisfied. So super-bunching itself is not yet nonclassical, but when *the growth of g2 is anomalously fast compared to g3*, this is nonclassical.

3. *Sub-shot-noise correlations in photon numbers – two-mode squeezing*. For a coherent beam split in 2, or even for two thermal beams split in 2 equally, or for two independent beams, the variance of the difference photon number is equal to the mean photon number:

$$Var(\hat{N}_1 - \hat{N}_2) = \left\langle \hat{N}_1 + \hat{N}_2 \right\rangle.$$

This is called noise at the shot-noise level.

Therefore, (8) indicates sub-shot-noise correlations.

Home task: calculate the normalized 2nd-order CF for two-mode squeezed vacuum.