Lecture 7. From nonlinear optical effects to photon correlations and anti-bunching.

Correlations from PDC and FWM, heralded preparation of single photons. Calculation and experiments. Two-photon amplitude, methods of measurement.

1. Photon correlations.

In our previous lectures, we have derived pair-producing Hamiltonians, which can be realized through four-wave mixing and parametric down-conversion. In both cases, the Hamiltonian has the form

$$\hat{H} = i\hbar\Gamma a_1^+ a_2^+ + h.c. \qquad (1)$$

The difference between a quadratic nonlinear effect (PDC) and a cubic one (FWM) is that in the first case, $\Gamma \sim \chi^{(2)} E_0 L$ and in the second one, $\Gamma \sim \chi^{(3)} E_0{}^2 L$, where E_0 is the pump field amplitude. The modes 1 and 2 can coincide [then $\hat{H} = i\hbar\Gamma (a^+)^2 h.c.$] or be different.

If $\Gamma t \equiv G$ (parametric gain) is low, we can write the state at the output using the perturbation theory (see Lecture 4) as

$$|\Psi\rangle = |vac\rangle + G a_1^+ a_2^+ |vac\rangle \equiv |0\rangle_1 |0\rangle_2 + G|1\rangle_1 |1\rangle_2, \quad G << 1, \qquad (2)$$

in the two-mode case and

$$|\Psi\rangle = |0\rangle + \sqrt{2} G |2\rangle, \quad G << 1, \qquad (2')$$

in the single-mode one. Terms with more than two creation operators are absent here.

We see that light contains only vacuum or photon pairs; this is by itself unusual, but what rigorous conclusion can one make if we do not calculate the higher-order terms? Obviously all we can calculate is the mean number of photons and the second-order correlation functions; cross-correlation for (2) and auto-correlation for (2'). For instance, the latter, which can be measured through the HBT experiment, is

$$g^{(2)} = \frac{\langle 2|2|G|^2 (a^+)^2 a^2 |2\rangle}{\langle 2|2|G|^2 a^+ a |2\rangle^2} = \frac{2|G|^2 \cdot 2}{4|G|^4 \cdot 4} = \frac{1}{4|G|^2} >> 1. \qquad (3)$$

We see there is bunching ($g^{(2)} > 1$), one can call it even super-bunching because it is stronger than for thermal light; but is it nonclassical?

To answer, we need the exact calculation of the correlation function, without the assumption of low gain. This can be done using the Heisenberg approach and the Bogolyubov transformations.

Single-mode case. We have

$$a = a_0 \cosh G + a_0^+ \sinh G,$$

then $\langle N\rangle = \langle [a_0^+ \cosh G + a_0 \sinh G][a_0 \cosh G + a_0^+ \sinh G]\rangle = \sinh^2 G$ and

$$\langle : N^2 : \rangle = \langle [a_0^+ \cosh G + a_0 \sinh G][a_0^+ \cosh G + a_0 \sinh G][a_0 \cosh G + a_0^+ \sinh G][a_0 \cosh G + a_0^+ \sinh G]\rangle =$$

$$= \sinh^2 G \langle [a_0 a_0^+ \cosh G + (a_0)^2 \sinh G][a_0 a_0^+ \cosh G + (a_0^+)^2 \sinh G]\rangle = \sinh^2 G[\cosh^2 G + 2\sinh^2 G].$$

From this,

$$g^{(2)} = \frac{\langle : N^2 : \rangle}{\langle N\rangle^2} = \frac{\sinh^2 G[\cosh^2 G + 2\sinh^2 G]}{\sinh^4 G} = 2 + \frac{\cosh^2 G}{\sinh^2 G} = 3 + \frac{1}{\sinh^2 G} = 3 + \frac{1}{\langle N\rangle}. \qquad (4)$$

So the lower $\langle N \rangle$, the higher the correlation function – the more bunching one observes. For bright PDC, $g^{(2)} = 3$.

But so far, there is nothing nonclassical. A high $g^{(2)}$ does not yet mean nonclassicality. We have to go further and calculate $g^{(3)}$.

$$\langle : N^3 : \rangle = \left\langle \begin{matrix} [a_0^+ \cosh G + a_0 \sinh G][a_0^+ \cosh G + a_0 \sinh G][a_0^+ \cosh G + a_0 \sinh G] \times \\ \times [a_0 \cosh G + a_0^+ \sinh G][a_0 \cosh G + a_0^+ \sinh G][a_0 \cosh G + a_0^+ \sinh G] \end{matrix} \right\rangle =$$

$$= \sinh^2 G \langle [a_0 a_0^+ \cosh G + (a_0)^2 \sinh G][a_0^+ \cosh G + a_0 \sinh G][a_0 \cosh G + a_0^+ \sinh G][a_0 a_0^+ \cosh G + (a_0^+)^2 \sinh G] \rangle =$$

$$= \sinh^2 G \langle [\cosh G + (a_0)^2 \sinh G][a_0^+ \cosh G + a_0 \sinh G][a_0 \cosh G + a_0^+ \sinh G][\cosh G + (a_0^+)^2 \sinh G] \rangle =$$

$$= \sinh^2 G \langle [(a_0)^2 a_0^+ \sinh G \cosh G + a_0 \sinh G \cosh G + (a_0)^3 \sinh^2 G][a_0 (a_0^+)^2 \sinh G \cosh G + a_0^+ \sinh G \cosh G + (a_0^+)^3 \sinh^2$$

$$= \sinh^2 G [\langle (a_0)^2 a_0^+ a_0 (a_0^+)^2 \rangle \sinh^2 G \cosh^2 G + \langle (a_0)^2 (a_0^+)^2 \rangle \sinh^2 G \cosh^2 G + \langle (a_0)^2 (a_0^+)^2 \rangle \sinh^2 G \cosh^2 G + \sinh^2 G \cosh^2$$

$$+ \langle (a_0)^3 (a_0^+)^3 \rangle \sinh^4 G] = \sinh^4 G[(\langle (a_0)^2 a_0^+ a_0 (a_0^+)^2 \rangle + 2\langle (a_0)^2 (a_0^+)^2 \rangle + 1) \cosh^2 G + \langle (a_0)^3 (a_0^+)^3 \rangle \sinh^2 G] =$$

$$= \sinh^4 G [9 \cosh^2 G + 6 \sinh^2 G].$$

Then,

$$g^{(3)} = \frac{\langle : N^3 : \rangle}{\langle N \rangle^3} = \frac{\sinh^4 G [9 \cosh^2 G + 6 \sinh^2 G]}{\sinh^6 G} = 6 + 9 \frac{\cosh^2 G}{\sinh^2 G} = 15 + \frac{1}{\sinh^2 G} = 15 + \frac{1}{\langle N \rangle}. \quad (5)$$

Therefore, at low $\langle N \rangle$ both $g^{(2)}$ and $g^{(3)}$ scale as $1/\langle N \rangle$, and unless $\langle N \rangle$ is too large, $[g^{(2)}]^2 > g^{(3)}$ (a higher-order analog of anti-bunching). At $\langle N \rangle >> 1$, it is not valid any more.

Two-mode case. We have then

$$a_1 = a_{10} \cosh G + a_{20}^+ \sinh G,$$

$$a_2 = a_{20} \cosh G + a_{10}^+ \sinh G.$$

Then, $\langle N_1 \rangle = \langle N_2 \rangle = \langle [a_{10}^+ \cosh G + a_{20} \sinh G][a_{10} \cosh G + a_{20}^+ \sinh G] \rangle = \sinh^2 G$, and

$$\langle : N^2 : \rangle = \langle [a_{10}^+ \cosh G + a_{20} \sinh G][a_{10}^+ \cosh G + a_{20} \sinh G][a_{10} \cosh G + a_{20}^+ \sinh G][a_{10} \cosh G + a_{20}^+ \sinh G] \rangle =$$

$$= \sinh^2 G \langle [a_{20} a_{10}^+ \cosh G + (a_{20})^2 \sinh G][a_{10} a_{20}^+ \cosh G + (a_{20}^+)^2 \sinh G] \rangle = 2 \sinh^4 G,$$

$$\langle N_1 N_2 \rangle = \langle [a_{10}^+ \cosh G + a_{20} \sinh G][a_{10} \cosh G + a_{20}^+ \sinh G][a_{20}^+ \cosh G + a_{10} \sinh G][a_{20} \cosh G + a_{10}^+ \sinh G] \rangle =$$

$$= \sinh^2 G \langle [a_{10} a_{20} \cosh G + a_{20} a_{20}^+ \sinh G][a_{10}^+ a_{20}^+ \cosh G + a_{10} a_{10}^+ \sinh G] \rangle = \sinh^2 G [\cosh^2 G + \sinh^2 G],$$

and for the correlation functions we have

$$g_{11}^{(2)} = g_{11}^{(2)} = \frac{\langle : N^2 : \rangle}{\langle N \rangle^2} = 2,$$

$$g_{12}^{(2)} = \frac{\langle N_1 N_2 \rangle}{\langle N \rangle^2} = \frac{\sinh^2 G [\cosh^2 G + \sinh^2 G]}{\sinh^2 G} = 1 + \frac{\cosh^2 G}{\sinh^2 G} = 2 + \frac{1}{\sinh^2 G} = 2 + \frac{1}{\langle N \rangle}.$$

Then,

$$g_{11}^{(2)} g_{22}^{(2)} = 4,$$

$$[g_{12}^{(2)}]^2 = 4 + \frac{2}{\langle N \rangle} + \frac{1}{\langle N \rangle^2}, \quad (6)$$

and the Cauchy-Schwarz inequality $g_{11}^{(2)} g_{22}^{(2)} \geq [g_{12}^{(2)}]^2$ is violated at any $\langle N \rangle$. But of course at larger $\langle N \rangle$ it is more difficult to notice.

This strong correlation of single photons is used in the absolute calibration of single-photon detectors.

2. Two-photon amplitude and entanglement.

So far we considered only a single mode, or two modes 1,2. In reality, both PDC and FWM lead to the generation of photon pairs within a certain frequency range (which can be broad); PDC in bulk crystals also has a broad angular width, therefore a large range of transverse wavevectors. As a result, the Hamiltonian contains integration over the frequencies and transverse wavevectors of the two photons. For instance, the transverse wavevector integration follows from assuming a finite transverse size of the pump beam (there was such a task after Lecture 3). Then, the Hamiltonian will be written as

$$\hat{H} \sim \iint dk_1 dk_2 F(k_1, k_2) a^+(k_1) a^+(k_2) + h.c. \qquad (7)$$

Here, k_1 and k_2 are transverse k-vectors (momenta) of the photons, and the two-photon amplitude $F(k_1, k_2)$ is introduced, which is the probability amplitude that a pair with such wavevectors (angles) has been created.

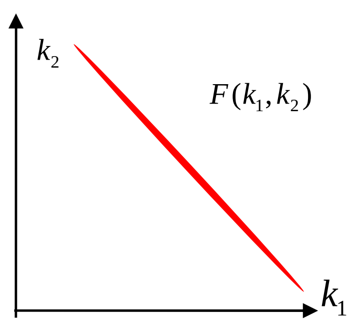

Fig. 1

The state is written, as usual, in the first order of the perturbation-theory expansion:

$|\Psi\rangle = |0\rangle_1 |0\rangle_2 + G \iint dk_1 dk_2 F(k_1, k_2) a^+(k_1) a^+(k_2) |0\rangle_1 |0\rangle_2 .$

Usually, $F(k_1, k_2)$ is stretched and tilted as shown in Fig.1, i.e., the transverse wavevectors of the two photons are anti-correlated. This immediately means that the state cannot be written as the product of the states of photons 1,2 separately:

$$|\Psi\rangle \neq |\Psi_1\rangle |\Psi_2\rangle . \qquad (8)$$

This is the definition of *entanglement (verschränkung)* and it is interesting both from the fundamental viewpoint and from the viewpoint of applications. As shown in the figure, entanglement of the photon pair (here, in the transverse momentum) means that each photon taken separately, has the transverse momentum very uncertain; at the same time, if a transverse momentum k_1 is measured for photon 1, the other photon has a very definite transverse momentum k_2.

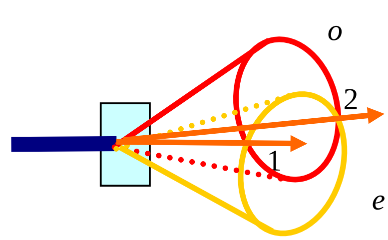

Fig.2

Better known is *polarization entanglement* of single photons. To obtain it, one should realize a special type of phase matching for PDC, in which two polarizations of the down-converted photons are possible. It is called type-II phasematching and it implies that the daughter photons are polarized orthogonally: one like an ordinary beam, the other one extraordinary. (The pump usually has extraordinary polarization). The situation is shown in Fig.2: o and e radiation is emitted into two cones displaced along the optic axis. The cones intersect along two lines (shown by arrows). In these directions, polarization can be either o (horizontal) or e (vertical), and the two-photon part of the state emitted via SPDC can be written as

$$|\Psi\rangle = \frac{1}{\sqrt{2}}\{|H\rangle_1|V\rangle_2 + |V\rangle_1|H\rangle_2\}.$$

In other words, photon 1 can be polarized horizontally or vertically; but if photon 1 is polarized horizontally, photon 2 is polarized vertically. This type of a state is called a Bell state, because it violates the Bell inequality, and there are four of them:

$$|\Psi^{(+)}\rangle = \frac{1}{\sqrt{2}}\{|H\rangle_1|V\rangle_2 + |V\rangle_1|H\rangle_2\}, \quad |\Psi^{(-)}\rangle = \frac{1}{\sqrt{2}}\{|H\rangle_1|V\rangle_2 - |V\rangle_1|H\rangle_2\},$$

$$|\Phi^{(+)}\rangle = \frac{1}{\sqrt{2}}\{|H\rangle_1|H\rangle_2 + |V\rangle_1|V\rangle_2\}, \quad |\Phi^{(-)}\rangle = \frac{1}{\sqrt{2}}\{|H\rangle_1|H\rangle_2 - |V\rangle_1|V\rangle_2\}.$$

For each of these states, each photon is not polarized, but as long as photon 1 is registered with some definite polarization (H), photon 2 is registered also with a definite polarization (H for $|\Phi^{(\pm)}\rangle$ states and V for $|\Psi^{(\pm)}\rangle$) states.

3. Heralded preparation of single photons.

From PDC and FWM, one can also prepare anti-bunched (ideally, single-photon) light. The experimental scheme is shown in Fig.3. Photon 2 of the state (2) is registered by a detector, and its 'click' (a count) opens the gate and lets photon 1 through; then the gate is closed again. This strategy eliminates the vacuum term in Eq. (2), because one only chooses the cases where there is a photon in arm 2. It also enables the suppression of higher-order terms (higher-order Fock states) omitted in Eq. (2) if the gate is closed again fast enough.

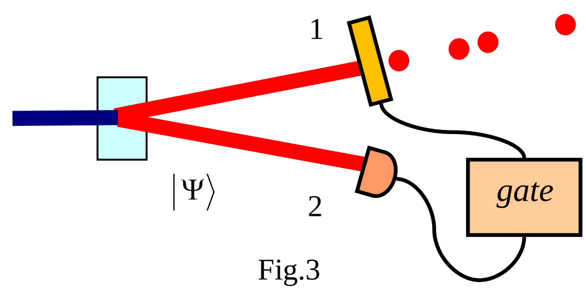

Fig.3

Let us try to estimate the bunching parameter of the obtained state. The initial state should be written at least in the second order of the expansion, to take into account the 'multiphoton' events:

$$|\Psi\rangle = |0\rangle_1|0\rangle_2 + G|1\rangle_1|1\rangle_2 + G^2|2\rangle_1|2\rangle_2, \quad G \ll 1.$$

The density matrix is then

$$\rho = |0\rangle_1|0\rangle_2\langle 0|_2\langle 0|_1 + |G|^2|1\rangle_1|1\rangle_2\langle 1|_2\langle 1|_1 + |G|^4|2\rangle_1|2\rangle_2\langle 2|_2\langle 2|_1.$$

The first term is excluded by discarding events where detector 2 registers no photon, and then the state in channel 1 is given by

$$\rho_1 \sim |G|^2|1\rangle_1\langle 1|_1 + |G|^4|2\rangle_1\langle 2|_1 \sim |1\rangle_1\langle 1|_1 + |G|^2|2\rangle_1\langle 2|_1$$

(the last equality follows from normalization, Tr=1, $|G|^2 \ll 1$.)

Then, the mean photon number in channel 1 is $\langle N_1\rangle = \langle 1|a_1^+a_1|1\rangle + |G|^2\langle 2|a_1^+a_1|2\rangle = 1 + 2|G|^2$,

and the second moment is $\langle :N_1^2:\rangle = \langle 1|(a_1^+)^2a_1^2|1\rangle + |G|^2\langle 2|(a_1^+)^2a_1^2|2\rangle = 4|G|^2$. As a result, we get

$$g^{(2)} = \frac{4|G|^4}{[1+2|G|^2]^2} \approx 4|G|^4 << 1. \quad (9)$$

One can also measure the Wigner function for this state and show that it has a negative area (see Lecture 6).

Single photons obtained this way can be used for quantum key distribution (QKD), based on the impossibility to measure the state of a single photon.

Home task: calculate the shape of the two-photon amplitude for PDC in a 3 mm long BBO crystal pumped at 800 nm with the pump beam waist 100 um.

Books:

1. Mandel, Wolf, Optical Coherence and Quantum Optics.