Lecture 1. Introduction and brief history of the subject.

The purpose of this course. Brief parallel history of nonlinear and quantum optics. How they merged. Quantum optics without nonlinear optics: atoms and solid-state emitters. Basics of nonlinear optics: parametric and non-parametric processes; nonlinear polarization; mean photon number per mode (brightness).

1. The purpose of this course.

This course is planned to be at the boundary of nonlinear and quantum optics. Quantum states of light that are studied by quantum optics and used in light-related quantum information technology are mostly produced through nonlinear optical effects. Examples are: entangled photon pairs, single photons and multiphoton states prepared by 'heralding', squeezed vacuum, squeezed coherent states. It is important to know how these states are generated. In addition to nonlinear optical methods for producing quantum light, there are methods for its detection such as up-conversion. The course is for those who already studied nonlinear optics, and maybe even quantum optics, but if necessary, we will fill some gaps in the understanding. (Or maybe make new ones, which is always useful.)

There will be 10 lectures, every two lectures followed by a problem class (led by Cameron Okoth) where the problems will be strongly related to the content of the lectures. The focus will be on experiments, and on making estimates. As part of the course, we will organize a lab tour where we will show harmonic generation, sum-frequency generation, high-gain parametric down-conversion. All this will be a good illustration to the course.

2. Brief parallel history of nonlinear and quantum optics.

Early frequency conversion experiments. Any frequency conversion is a nonlinear effect. Using only linear optical elements, you cannot obtain 'red from blue' or vice versa, you cannot change the spectrum of light. In this sense, fluorescence is definitely a nonlinear effect, and it is known from the 19th century, the first experiments made by Herschel in 1845 and by Stokes in 1852 (Fig.1). In fact, the important step made by Stokes was that he used filters to select the short-wavelength part of the excitation radiation (church blue glass transmitting only UV) and the long-wavelength fluorescence (wine NOT transmitting the UV). The conclusion was that fluorescence is red-shifted. Furthermore, Raman scattering, discovered experimentally in 1928, is also a type of inelastic scattering and can be formally treated in terms of nonlinear optics. The Raman tensor, characterizing the strength of Raman scattering, is in one-to-one correspondence with the cubic nonlinear susceptibility. But traditionally, only the 1961 experiment by Franken on the second-harmonic generation is considered as the start of nonlinear optics.

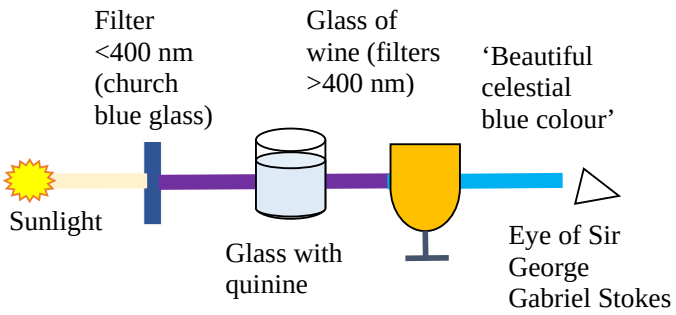

Fig.1

Franken's experiment

The experiment was possible only after the appearance of lasers. The first laser (at that time, called *optical maser*) was made by Maiman in 1960, but in 1961 Franken already used a *commercially available* pulsed ruby laser! Certainly, the nonlinear optics of microwaves was

already well studied, so there was a search for analogous effects in optics. The harmonic was observed after focusing red light into a quartz crystal (actually, one of the worst materials to observe second harmonic generation), as a spot on a photographic plate at the output of a prism spectrograph. Fig.2 shows the experiment, and one can notice the resemblance to the Stokes experiment: filters were used before and after the object under study, but this time the emission was blue shifted. Of course, in this experiment a different source was used and a different detector. Today, the equipment is even better.

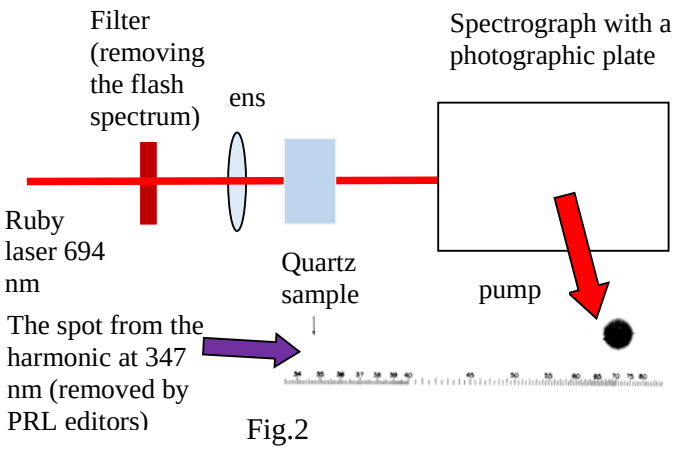

Fig.2

After the second harmonic generation, an avalanche of nonlinear optics experiments started. In fact, all these phenomena have been already observed in the microwave range, and now they were transferred into the optical range. Here just a few of them are listed:
1961: Kaiser and Garrett, two-photon fluorescence
1962: Woodbury, stimulated Raman effect and Raman laser
1965: Maker and Terhune, third-harmonic generation in calcite.
1965: Akhmanov et al.; Giordmaine and Miller, optical parametric amplification.
1967: New et al., third-harmonic generation in gas.
1967: Magde&Mahr; Akhmanov et al.; Harris et al., parametric down-conversion.

Hanbury Brown and Twiss experiment. Quantum optics is believed to have started from the Hanbury Brown-Twiss (HBT) experiment (1956). In this experiment, or rather a series of experiments, Robert Hanbury Brown and Richard Twiss observed intensity correlations for the radiation of a mercury lamp and some bright stars. After a beamsplitter (in the case of a mercury lamp) or at two spatially separated (but not too much) points the intensities measured by two detectors were fluctuating, and these fluctuations were correlated. The correlation can be quantified by the correlation function of the intensities measured by the two detectors,

$$g^{(2)} = \frac{\langle I_1 I_2 \rangle}{\langle I_1 \rangle \langle I_2 \rangle},$$

which was larger than 1 in these experiments.
The experiments were immediately explained in terms of photons ('light consists of photons'): it looked like in the radiation of thermal sources, such as stars and gas discharge lamps, photons 'bunch' into groups (pairs, as one could naively infer from second-order correlation results). For this reason, $g^{(2)}$

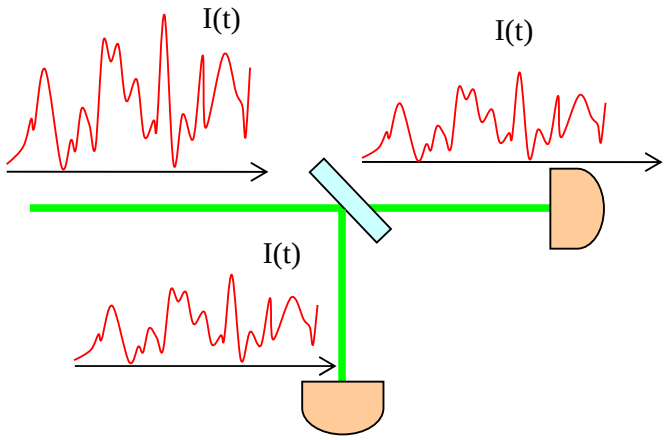

Fig.3

is called the bunching parameter.

In fact, HBT experiments with thermal light have perfectly classical explanation, without any photons: one has just to know that the intensity has fluctuations in time, with a certain probability distribution. In particular, for thermal sources (and most sources are thermal), the distribution is negative-exponential,

$P(I) = \frac{1}{\langle I \rangle} e^{-I/\langle I \rangle}$. This fully explains the results of HBT experiments.

Photons. In the 1960-s both 'nonlinear optics community' and 'quantum optics community' spoke of *photons*. Both were interested in *multiphoton effects*, where several photons are absorbed or emitted. In this connection, a benchmark was Roy Glauber's paper of 1963, where it was shown that the absorption of n photons is mathematically described by the n-th order normally ordered correlation function. Together with the coherent-state representation (another 1963 paper by Glauber), it formed the basis of quantum optics theory. But experimental quantum optics really started when so-called nonclassical light sources were discovered. Nonclassical light (which will be considered in detail further) requires quantum theory for its description. Historically, the first example of such light was photon pairs (Fig.4a). The first experiment was performed in 1967 (Kocher and Commins, calcium atoms). Later (1982), even Bell's inequalities were tested using this source.

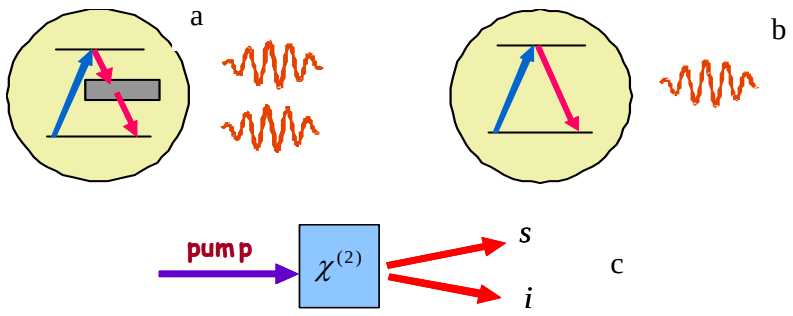

Fig.4

Single photons were obtained for the first time only in 1977, in experiments on the fluorescence of single atoms (Fig.4c).

Nonlinear optics and quantum optics merged in 1967, when parametric down-conversion was discovered (Fig.4c). This effect was used in 1970 for producing photon pairs, in 1986 for producing single photons and in 1987 for producing squeezed light.

3. Quantum optics without nonlinear optics.

Links to other fields. As we see from the history, in the first experiments nonclassical light was obtained from atoms. This direction has been developing since those first experiments, and starting from the 2000-s, various solid-state emitters started to replace atoms. These solid-state emitters include self-assembled quantum dots, later colloidal quantum dots, organic molecules in a solid-state matrix, and color centers in diamond, including diamond nanocrystals. At first, these solid-state emitters were used for producing single photons. Sometimes they are called 'single-photon emitters'. But later cascaded transitions were

discovered, similar to the ones in atoms. This whole field is sometimes referred to as ‘solid-state quantum optics’.

We see that together with atom optics and nonlinear optics, solid-state physics forms the basis of quantum optics as it describes one possible source of quantum states of light. And quantum optics, in its turn, has such applications as quantum metrology and quantum information. This structure is schematically shown in Fig.5. Without the nonlinear optics (and atom optics), quantum optics lacks a lot.

4. Basics of nonlinear optics.

Parametric and non-parametric effects.
The most important nonlinear optical effects are schematically shown in Fig.6. These are: second-harmonic generation, sum-frequency generation, difference-frequency generation, parametric down-conversion, spontaneous four-wave mixing, Stokes and anti-Stokes Raman scattering, fluorescence, two-photon fluorescence. Solid horizontal lines are real levels, dashed lines are virtual levels. Thick lines up or down denote external light fields, thin lines are generated fields. The first four effects are called parametric and the other four, non-parametric: in parametric effects, the matter returns into its initial state. In this course we will only study parametric effects.

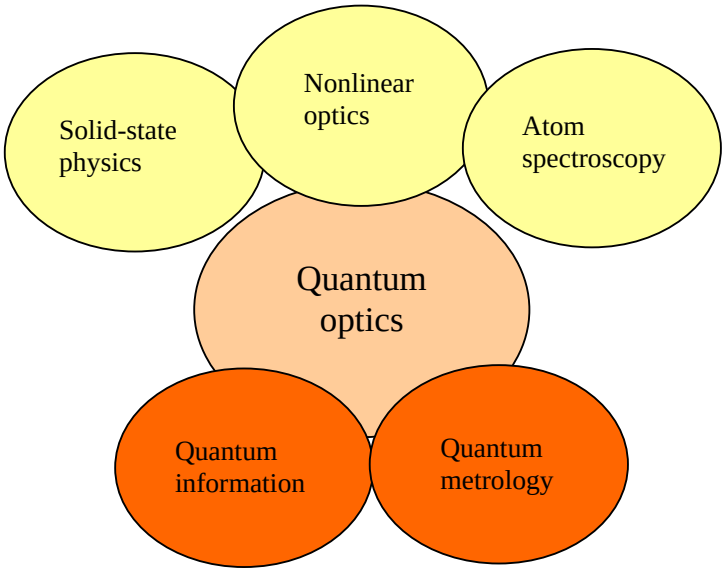

Fig. 5

Nonlinear polarization and nonlinear susceptibilities.

For the simple reason that ‘nothing is linear’, polarization, or the dipole moment of a unit volume, is nonlinear in the field. However, there exist microscopic models of the nonlinear susceptibilities.

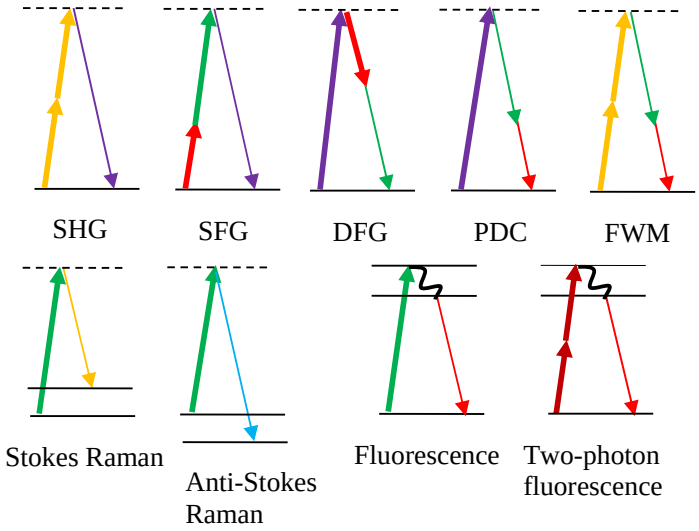

Fig. 6

In the vector form, the polarization is written as the power expansion of the electric field:

$$\vec{P} = \varepsilon_0 \hat{\chi}^{(1)} \cdot \vec{E} + \varepsilon_0 \hat{\chi}^{(2)} : \vec{E}\vec{E} + \varepsilon_0 \hat{\chi}^{(3)} \vdots \vec{E}\vec{E}\vec{E} + ... \quad (1)$$

This equation is written in SI units; this is why the vacuum permittivity enters. For simplicity, in what follows we will omit the vectors and tensors.

This expansion, as any powers expansion, is only valid when each next term is much smaller than the previous one. An example where it is not valid: a resonance leading to an extremely high value of some susceptibility.

We can also introduce nonlinear polarization of different orders,

$$P^{(2)} = \varepsilon_0 \chi^{(2)} E^2, \quad P^{(3)} = \varepsilon_0 \chi^{(3)} E^3, ... \quad (2)$$

Further, we will consider separately second-order nonlinear effects and third-order nonlinear effects (we won't discuss higher-order effects). It is important that even-order susceptibilities, for symmetry reasons, cannot differ from zero in materials with a center of symmetry (liquids, gases, glasses). Therefore, second-order effects are absent there. But one can remove this symmetry – for instance, apply electric field or consider an interface between two materials. Then second-order effects will appear.

One can see from (1) that nonlinear susceptibilities are all of different dimensionalities: $\chi^{(1)}$ is dimensionless, the dimensionality of $\chi^{(2)}$ is inverse electric field, and so on. One can assume that the ratio $\chi^{(n)} / \chi^{(n+1)}$ is given by some quantity of the dimensionality of the field. It turns out that if the susceptibilities are of the electronic origin, then this field is the so-called atomic field, which can be estimated as the field created by the electron charge at a distance given by the 'atom radius', or 'Bohr's radius' a_0,

$$E_{at} = \frac{e}{4\pi\varepsilon_0 a_0^2},$$

where the Bohr radius is

$$a_0 = \frac{4\pi\varepsilon_0 \hbar^2}{me^2},$$

and e, m are the charge of the electron and the mass of the atom. The resulting atomic field turns out to be

$$E_{at} = 5 \cdot 10^{11} \frac{\mathrm{V}}{\mathrm{m}}.$$

It follows that for the n-th -order nonlinear effect to be as strong as the n-1-st one, the laser we use should have the field on the order of the atomic field.

I will try to stick to SI units. Then, the intensity corresponding to the atomic field is

$$I_{at} = \frac{1}{2} \varepsilon_0 c E_{at}^2 = 0.5 \cdot 8.85 \cdot 10^{-12} \cdot 3 \cdot 10^8 \cdot 25 \cdot 10^{22} \,\mathrm{m} \frac{\mathrm{F} \cdot \mathrm{V}^2}{\mathrm{m}^2 \mathrm{m} \cdot \mathrm{s}} = 3.3 \cdot 10^{20} \frac{\mathrm{W}}{\mathrm{m}^2} = 3.3 \cdot 10^{16} \frac{\mathrm{W}}{\mathrm{cm}^2}.$$

Let us compare this to the powers of some typical lasers we have.

1. Continuous-wave Verdi laser, emitting about 10 W. Focusing the beam into 10 micrometers, we get $I = 10^7 \frac{\mathrm{W}}{\mathrm{cm}^2}$. (Many materials will burn at this intensity.)
2. Pulsed femtosecond Ti-sapphire laser, emitting about 1W mean power, but as 100 fs pulses with the repetition rate 80 MHz. The peak intensity I_p will be related to the mean intensity I as $I_p = I / D,$ where the duty cycle $D = T_p / \Delta t,$ T_p being the pulse duration and Δt the interval between the pulses. We get for this laser $D = 7 \cdot 10^{-6}$ and $I_p = 1.4 \cdot 10^{11} \frac{\mathrm{W}}{\mathrm{cm}^2},$ if the beam is focused, again, into 10 micrometers. This is already higher. The mean intensity will be then only about $I = 10^4 \frac{\mathrm{W}}{\mathrm{cm}^2}.$ This is not high at all.
3. Pulsed Ti-sapphire laser after a regenerative amplifier, mean power 3W. Now, the pulse is 1 ps long and the repetition rate is 5 kHz. The duty cycle is $D = 5 \cdot 10^{-9}$ and $I_p = 6 \cdot 10^{14} \frac{\mathrm{W}}{\mathrm{cm}^2},$ if the beam is focused, again, into 10 micrometers. We see that for this laser, so tightly focused, the peak field is only about 7 times less than the atomic one. With this laser one can even expect the expansion (1) to be not quite valid.

4. Brightness in photon numbers per mode.

In connection with what will be discussed in the next lectures, it is useful to consider not only the field or intensity of various sources, but the mean number of photons per mode.

A mode, or a coherence volume, is the volume in space within which the radiation is coherent. (Obviously it is coherent within some, sufficiently small, volume.) Assuming that we know the coherence length and the coherence radius, we can imagine a part of space where radiation is coherent. It can be viewed (Fig.7) as a rectangular cylinder with the length l_{coh} and the transverse size ρ_{coh}. The volume of this cylinder is called the *coherence volume*.

$V_{coh} = \rho_{coh}{}^2 l_{coh}$

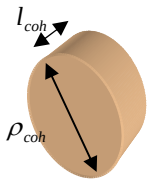

Fig.7

What is the coherence radius for a beam of sunlight? Coherence length? Coherence volume?

So we can divide all space into ‘elementary cells’ as shown in Fig.7 and assume that the radiation inside each cell is coherent while the radiation in different cells is incoherent. Such a cell can be called ‘a mode’ (not necessarily modes should be chosen like this, but this is an option).

The number of photons per mode is the energy contained in the coherence volume divided by the energy of a single photon. For instance, if a source has the mean intensity I, then the energy flowing through the coherence area per unit time (the power) is $P = I\rho_{coh}^2$. Then, the energy in the coherence volume is obtained just by multiplying this by the coherence time,

$W_{coh} = I\rho_{coh}^2 \tau_{coh} = IV_{coh} / c$,
and the mean number of photons per mode is
$N_{mode} \equiv \frac{W_{coh}}{\hbar\omega} = \frac{IV_{coh}}{\hbar\omega c}$.

- It is called the degeneracy parameter because it shows the occupation number of a single ‚cell'; while for fermions this occupation number cannot exceed 1, for photons it can take any value.
- It is this number of photons per mode that is given by the Planck formula for the blackbody radiation.
- In nonlinear optics, the interaction efficiency is given by N_{mode}. Hence the degeneracy parameter of a light field also determines its ability to interact with other light fields.
- The degeneracy factor is also important because it tells whether the photon structure of light is pronounced ($N_{mode} \leq 1$) or not ($N_{mode} >> 1$).

Home task: Estimate the numbers of photons per mode for all three lasers discussed above. All lasers are assumed to be spatially coherent and all pulses are assumed to be Fourier-transform limited.

Books:

1. Boyd, Nonlinear optics
2. Shen, Principles of nonlinear optics
3. Klyshko, Physical foundations of quantum electronics