Lecture 9.

Nonlinear optical effects for producing nonclassical light.

1. Nonlinear polarization and nonlinear effects originating from it

If an electric field $\vec{E}$ is incident on the matter, the response (described by polarization $\vec{P}$) is not linear but has the general form

$$\vec{P} = \varepsilon_0 \chi^{(1)} \vec{E} + \varepsilon_0 \chi^{(2)} \vec{E}\vec{E} + \varepsilon_0 \chi^{(3)} \vec{E}\vec{E}\vec{E} + ... \qquad (1)$$

Here, the expansion coefficients $\chi^{(n)}$ are called nonlinear susceptibilities. In the matrix form, the same equation reads

$$P_i = \varepsilon_0 \chi^{(1)}_{ij} E_j + \varepsilon_0 \chi^{(2)}_{ijk} E_j E_k + \varepsilon_0 \chi^{(3)}_{ijkl} E_j E_k E_l + ...$$

(repeating indices imply summation). This means that $\chi^{(n)}$ is a tensor of rank n+1. The elements of these tensors are determined by the symmetry of the material. For instance, in a material with the inversion symmetry all even susceptibilities are zero, $\chi^{(2n)} = 0$ - otherwise, inversion would change the polarization vector on the left to its opposite but leave the right-hand side unchanged.

Hierarchy of the nonlinearities. Usually, the effect of every next-order susceptibility is smaller than the one of the previous one (an obvious exception is a centrosymmetric material with $\chi^{(2)} = 0$ but $\chi^{(3)} \neq 0$). The ratio between susceptibilities of n-th and n+1-st – orders is given by the atomic field, which is very large:

$$\chi^{(n+1)} \sim \chi^{(n)} / E_a .$$

Therefore, each next term in (1) is smaller than the previous one by a factor E / E_a.

Let us estimate this atomic field:

$$E_a \sim \frac{\hbar\omega}{e a_0},$$

where $\hbar\omega \sim 10^{-19} J$ is the photon energy, $e = 1.6 \cdot 10^{-19} C$, and $a_0 = 5.3 \cdot 10^{-11} m$. We get $E_a \sim 10^{11} V/m$, which corresponds to about $10^{16} W/cm^2$. This is why high-order nonlinear effects are hard to observe.

But it is still possible. The strongest laser we have in the lab is pulsed Ti-sapphire, after a regenerative amplifier, and it has mean power 3W, and the duty cycle 5x10^{-9}. This means a peak power of GW, and if this beam is focused into 10 um=10^{-3} cm, then the peak intensity will be only about an order of magnitude less than the atomic one.

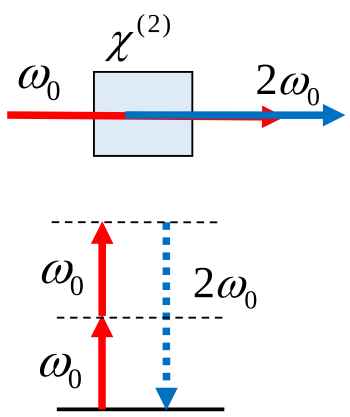

Fig.1

We will start therefore with the effects corresponding to quadratic nonlinearity, $\chi^{(2)}$.

Second harmonic generation. Like atomic transitions 'up', such effects can be described in the framework of the classical theory. In an experiment on second-harmonic generation, there is a real field E incident on a nonlinear material (usually a crystal without centre of symmetry). The field oscillates at frequency ω_0, then polarization

P oscillates at frequency $2\omega_0$, and induces a new field, also at frequency $2\omega_0$ (Fig.1). One can write (ignoring vectors)

$$E^{(+)}(\vec{r},t) = E_0 e^{-i\omega_0 t + i\vec{k}\vec{r}},$$

$$P_2 = \varepsilon_0 \chi^{(2)} (E^{(+)} + c.c)^2 = \varepsilon_0 \chi^{(2)} [E_0^2 e^{-i2\omega_0 t + i2\vec{k}\vec{r}} + E_0^{*2} e^{i2\omega_0 t - i2\vec{k}\vec{r}} + 2|E_0|^2].$$

The first terms describe second-harmonic generation, the last one optical rectification. The nonlinear polarization P_2 induces a new field E_2 according to the Helmholz equation:

$$\nabla^2 E_2 - \frac{n^2}{c^2}\ddot{E}_2 = \frac{4\pi}{\varepsilon_0 c^2}\ddot{P}_2.$$

The Helmholz equation is linear in E_2 and P_2; it means that the SHG intensity will be quadratic in the pump intensity:

$$I(2\omega_0) \sim [I(\omega_0)]^2.$$

Sum- and difference-frequency generation. Let us now have fields at ω_1 and ω_2, and the nonlinear polarization will contain terms oscillating with frequencies $\omega_1 + \omega_2$ and $\omega_1 - \omega_2$. There will be fields at these sum and difference frequencies, whose intensities will scale as the product of the initial intensities:

$$I(\omega_1 \pm \omega_1) \sim I(\omega_1) I(\omega_2).$$

Phase matching condition. The nonlinear polarization, for instance, in the sum-frequency generation, will necessarily have the wavevector equal to the sum of the two input wavectors, $\vec{k} = \vec{k}_1 + \vec{k}_2$. However, it is possible that this wavevector does not satisfy the dispersion relation, $k = k(\omega_1 + \omega_2)$. Then the electric field will not be efficiently excited. For the efficient excitation of the electric field, it is necessary that the phase matching condition is satisfied, $k(\omega_1) + k(\omega_2) = k(\omega_1 + \omega_2)$. For the SHG, similarly, $2k(\omega_0) = k(2\omega_0)$.

Parametric down-conversion. Here, at the input of the crystal there is field at frequency ω_p with the wavevector $\vec{k}_p$ (further called ‘the pump’). At the output, we will get light at less than this frequency, but how can one derive it? It turns out that there is no classical explanation. This is similar to the situation with the atomic transition ‘down’. And similarly, we can use a trick. At Lecture 5, we mentioned ‘zero-point vacuum fluctuations’: even in the absence of photons, there was some energy in every radiation mode. Let us imagine that these fluctuations, which are present at every frequency and at any wavevector direction, also exist at frequency $\omega < \omega_p$. Then (Fig.2), there will be difference-frequency generation with this wave. One can say ‘scattering by zero-point vacuum fluctuations’.

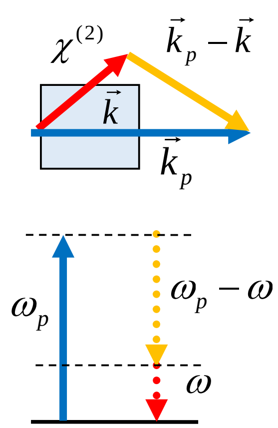

Fig.2

But at the next lecture, there will be full quantum theory of PDC.

Let us now pass to third-order effects.

Kerr effect. Taking the first and third terms in (1), we see that the linear susceptibility gets an additional term quadratic in the field: $\chi^{(1)} \to \chi^{(1)} + \chi^{(3)}\vec{E}\vec{E}$. This means that there is additional refractive index, proportional to the intensity, $n = n_0 + 2n_2|E|^2$.

Four-wave mixing. Let two fields be present at the input of a nonlinear medium (crystal, glass, fibre, liquid, atoms, …): the pump E_p and the signal E_s. Then, there will be nonlinear polarization

$$P_3 = \varepsilon_0 \chi^{(3)} (E_p^{(+)} + E_s^{(+)} + c.c.)^3.$$

There will be a lot of terms here, for instance, third-harmonic generation, but only some of them will be such that phase matching will be satisfied. Now we are interested in the term $\varepsilon_0 \chi^{(3)} [E_p^{(+)}]^2 E_s^{(-)}$. It will oscillate at frequency $2\omega_p - \omega_s$ and have the wavevector $2k(\omega_p) - k(\omega_s)$. It will correspond to the four-photon diagram shown in Fig.3. Similar to the difference-frequency generation, this effect is described classically.

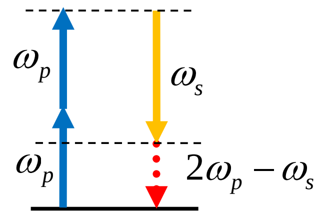

Fig.3

However, if the signal beam is absent, there will still be radiation at the output (Fig.4). This effect, *spontaneous four-wave mixing*, will be considered quantum mechanically at the next lecture.

Third harmonic generation. This is similar to second harmonic generation, perfectly classical effect. More interesting is its inverse, photon triplet generation, a purely quantum effect that has never been observed before.

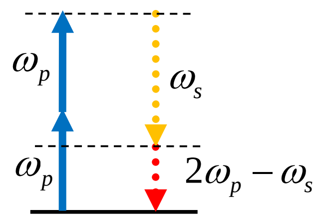

Fig.4

2. Obtaining nonclassical light

Now we will see how various nonclassical types of light can be obtained through nonlinear effects. Strictly speaking, there is one definition of nonclassicality: the negativity or singularity of the Glauber-Sudarshan quasiprobability. But it is impossible to measure directly; therefore, one uses various witnesses (sufficient conditions) for nonclassicality. We will list them here, the weakest ones first:

- Squeezing: the uncertainty in some variable less than the one for a coherent state. For instance, quadrature squeezing or Stokes variable squeezing (polarization squeezing).
- Sub-Poissonian statistics of photocounts, $\Delta m^2 < \langle m \rangle$, or photocurrent in electron units, $\Delta i^2 < \langle i \rangle$. This means that the Fano factor is less than unity, $F \equiv \Delta m^2 / \langle m \rangle < 1$.
- Anti-bunching: $g^{(2)}(0) < 1$. These two conditions are related because $(g^{(2)}(0) - 1)\langle m \rangle = F - 1$. But at large mean photon numbers, it is easier to observe F<1 while at small photon numbers, g<1.
- Higher-order analogues of anti-bunching:
  $g^{(k-1)}(0) g^{(k+1)}(0) < [g^{(k)}(0)]^2$. (*)

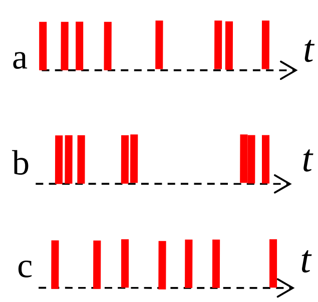

Fig.5

Weaker conditions are simply $g^{(k)}(0) < 1$ - they follow from (*).

- Wigner function negativity.

Sub-Poissonian light from SHG. Figure 5 illustrates the three cases of photon statistics: Poissonian, $g^{(2)} = 1$ (a), bunched, $g^{(2)} > 1$ (b) and anti-bunched, $g^{(2)} < 1$ (c). In these cases, the Fano factor will be also =1, >1, and <1, respectively. In SHG, mean photon number is always large. Due to SHG, pairs or groups of photons will be removed from the pump beam, and the pump will become anti-bunched. The same effect can be illustrated by a simple diagram on the phase plane (Fig.6). The initial pump state (blue) becomes squeezed in the amplitude, because intensity peaks are suppressed.

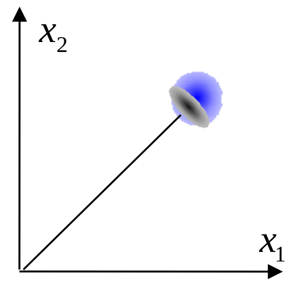

Fig.6

Kerr squeezing. The intensity-dependent refractive index can lead to squeezing (Fig.7). To see this, let us consider the evolution of a coherent state in the course of its propagation through a fibre. Points corresponding to higher amplitudes will have larger refractive index and larger phase delays. As a result, the uncertainty region will evolve and become compressed (grey ellipse) in some direction, corresponding to a certain quadrature (red arrows). To obtain amplitude squeezing, one can overlap this beam with some other one and shift the ellipse (shown in the figure). In practice, one observes polarization squeezing.

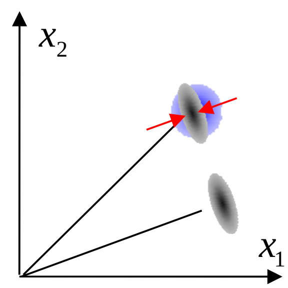

Fig.7

Parametric down-conversion creates pairs of photons. At the next lecture we will see how it leads to super-bunching and quadrature squeezing. *Spontaneous four-wave mixing* (next lecture) creates the same effects. *Photon triplet generation* can be discussed similarly.

Home task: check the higher-order anti-bunching condition (*) for coherent light and thermal light. Hint: the correlation functions for both types of light can be calculated classically and were indeed calculated (or at least given) in Lecture 3.

Books:

1. Boyd, Nonlinear optics, 1.1-1.2
2. Klyshko, Physical foundations of quantum electronics, 6.5.3, 6.5.5, 6.5.9