Lecture 3. Classical description of nonlinear cubic interactions. Hamiltonians leading to the generation of nonclassical light

Cubic interactions: Kerr effect, four-wave mixing, third harmonic generation. Hamiltonians producing photon pairs.

1. *Third-order nonlinear effects.*

These effects are related to the third-order polarization, $\vec{P}^{(3)} = \chi^{(3)}\vec{E}\vec{E}\vec{E}$. In the tensor form, it is written as

$$P_i^{(3)} = \varepsilon_0 \chi_{ijkl}^{(3)} E_j E_k E_l. \tag{1}$$

The third-order susceptibility $\chi_{ijkl}^{(3)}$ is a tensor of rank 4. It is present in all materials, although of course it can be higher or weaker. We will now consider several effects originating from it.

Kerr effect. Taking the first and third terms in the general equation

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

we see that the linear susceptibility gets an additional term quadratic in the field: $\chi^{(1)} \to \tilde{\chi}^{(1)} = \chi^{(1)} + \chi^{(3)}\vec{E}\vec{E}$. This means that the displacement can be represented as

$$\vec{D} = \varepsilon_0 \vec{E} + \vec{P} = \varepsilon_0 (1 + \tilde{\chi}^{(1)})\vec{E} = \varepsilon_0 \tilde{n}^2 \vec{E},$$

and one can introduce the ‘corrected’ refractive index $\tilde{n}$ quadratic in the field.

$$\tilde{n} = n + n_2 I. \tag{3}$$

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

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

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 $\chi^{(3)}[E_p^{(+)}]^2 E_s^{(-)}$. 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.1. Similar to the difference-frequency generation, this effect is described classically. The pump can be a single one or two different beams at two different frequencies as well.

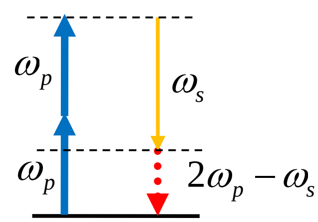

Fig.1

However, if the signal beam is absent, there will still be radiation at the output (Fig.2). This effect, *spontaneous four-wave mixing*, can be only considered quantum mechanically.

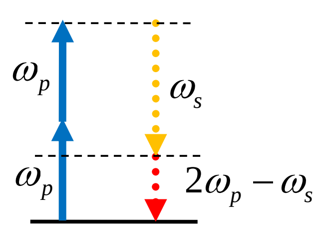

Fig.2

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.

2. *Pair-creating Hamiltonians.*

A Hamiltonian in quantum mechanics is just the energy, but as any observable in quantum mechanics, it is an operator. So the strategy will be to write the energy of the nonlinear interaction but to keep quantum what should be kept quantum.

In the dipole approximation, the energy of interaction between the matter and light is

$$H = -\vec{d}\vec{E}, \tag{4}$$

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

$$\vec{d} = \int d^3 r \vec{P}(r)\,. \tag{5}$$

Parametric down-conversion. Consider first quadratic nonlinear interaction and the second-order nonlinear polarization,

$$\vec{P}^{(2)} = \varepsilon_0 \chi^{(2)} \vec{E}\vec{E}. \tag{6}$$

After substituting (5) and (6) into (4), 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 (labeled with 0), signal (1), idler (2). Then,

$$E^3 = (E_0 e^{i\omega_0 t - i\vec{k}_0\vec{r}} + E_1 e^{i\omega_1 t - i\vec{k}_1\vec{r}} + E_2 e^{i\omega_2 t - i\vec{k}_2\vec{r}} + c.c.)^3 \tag{7}$$

We are interested only in the term

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

Now we have to take into account that the fields are actually quantum fields. We have to recall quantization of the field, which means that each field mode (for simplicity, a plane-wave mode given by the wavevector $\vec{k}$) is similar to a harmonic oscillator. It can be populated by a certain (or, rather, uncertain) number of quanta. There are photon creation and annihilation operators $a^+_{\vec{k}}, a_{\vec{k}}$, which increase and decrease the number of quanta by one. And then the positive- and negative-frequency fields are also operators; they can be written as a superposition of mode contributions, each of them containing a photon annihilation or creation operator:

$$E^{(+)}(\vec{r},t) = \sum_{\vec{k}} c_{\vec{k}} a_{\vec{k}} e^{i\omega(\vec{k})t - i\vec{k}\vec{r}}, \quad E^{(-)}(\vec{r},t) = \sum_{\vec{k}} c_{\vec{k}} a^+_{\vec{k}} e^{-i\omega(\vec{k})t + i\vec{k}\vec{r}}. \tag{8}$$

Therefore, we have an operator a instead of the analytic signal. Recall that the operators $a^+_{\vec{k}}, a_{\vec{k}}$ satisfy the commutation relations

$$[a_{\vec{k}}, a^+_{\vec{k}}] = 1\,.$$

It is also useful to recall the quadratures, or position and momentum operators, which are defined as the Hermitian and anti-Hermitian parts of $a^+_{\vec{k}}, a_{\vec{k}}$:

$$\hat{q}_{\vec{k}} = (a^+_{\vec{k}} + a_{\vec{k}})/2,\ \hat{p}_{\vec{k}} = (a^+_{\vec{k}} - a_{\vec{k}})/2i, \quad [\hat{q}_{\vec{k}}, \hat{p}_{\vec{k}}] = \frac{1}{2i}.$$

But if we try to show the operator, as the field, as a point on the phase plane (Fig.3), the quadratures being its real and imaginary parts, we face a problem. Quantum mechanics teaches us that such a single point makes no sense: position and momentum cannot be defined simultaneously, there is the uncertainty relation $\Delta\hat{q}_{\vec{k}}\Delta\hat{p}_{\vec{k}} \geq \frac{1}{2}$. Therefore, any state will look like a set of points, or an area. For instance, the vacuum will look not like a point at

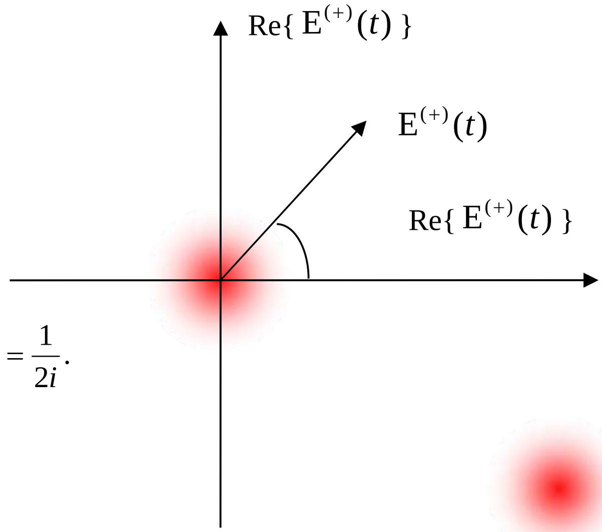

Fig.3

the center but as a blurred 'circle'. For this reason, in Eq. (7) we substitute fields 1,2 with expressions (8). For simplicity, we will only take into account one term in the sum over the modes. At the same time, the pump is strong and can be considered as classical light. In Fig.3, it will be a blurred circle displaced VERY far from the origin. (We will discuss later what all this means, in connection with the Wigner function.) Then the Hamiltonian is proportional to

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

Here we introduced frequency and wavevector mismatches, $\Delta\omega = \omega_0 - \omega_1 - \omega_2$, $\Delta\vec{k} = \vec{k}_0 - \vec{k}_1 - \vec{k}_2$. 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.4), there is the length of the crystal, and the integral is

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

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

$$\hat{H} \sim \chi^{(2)} E_0 L a_1^+ a_2^+ + h.c. \equiv i\hbar\Gamma a_1^+ a_2^+ + h.c., \qquad (9)$$

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).

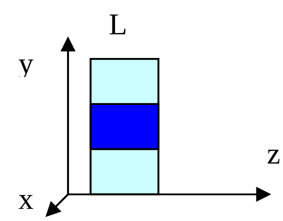

Fig.4

We have obtained an interesting operator: it creates photons only in pairs. And the probability of pair creation depends on the quadratic susceptibility, pump field, and the length of the crystal. In the next lecture, we will calculate the mean number of these pairs and their various properties.

To satisfy the phase matching, one can simply use a birefringent crystal and choose different polarizations for the pump and for the signal/idler photons. For instance, in type-I PDC the pump is polarized as an extraordinary beam while the down-converted photons are ordinary. For degenerate phase matching, one should then satisfy $k_{eff}(2\omega) = 2k_o(\omega)$, or $n_{eff}(2\omega) = n_o(\omega)$, where n_{eff} is the effective refractive index (was considered in the problem class).
One can also distinguish type-II phase matching, for which signal and idler photons are polarized orthogonally.
Quasi phase matching. It is also possible to phase match PDC with all photons (pump, signal, idler) polarized the same way. It can be useful, for instance, because one can then take advantage of the strongest quadratic susceptibility component $\chi^{(2)}_{zzz}$, which is the largest in most crystals. In a periodically poled crystal, $\chi^{(2)}$ is periodically modulated. In reality it just changes sign, but for simplicity we will assume that it varies sinusoidally: $\chi^{(2)}(z) = \chi_0 e^{\pm iKz} + c.c.$ Then, in the Hamiltonian, instead of the integral $\int_{-L}^{0} dz e^{-i\Delta k_z z}$ we will

obtain $\int_{-L}^{0} dz e^{-i\Delta k_z z \pm iKz}$. This immediately means that the mismatch will be $\Delta\tilde{k}_z = k_0 - k_1 - k_2 \pm K$. By choosing an appropriate poling period $\Lambda = \frac{2\pi}{K}$, one can satisfy the phase matching. Of course the $\chi^{(2)}$ dependence on z is not sinusoidal but rather meander-type, but this simply means that only a single harmonic will play a role.

Four-wave mixing/modulation instability. Consider now cubic nonlinear interaction and the third-order nonlinear polarization,

$$\vec{P}^{(3)} = \varepsilon_0 \chi^{(3)} \vec{E}\vec{E}\vec{E}. \qquad (10)$$

Similarly to the previous case, we see that $H = -\varepsilon_0 \int d^3 r \chi^{(3)}(r) \vec{E}\vec{E}\vec{E}\vec{E}$.

We omit the vectors again, and again assume only three fields: the pump (labeled with 0), signal (1), idler (2). Then, the Hamiltonian will contain

$$E^4 = (E_0 e^{i\omega_0 t - i\vec{k}_0 \vec{r}} + E_1 e^{i\omega_1 t - i\vec{k}_1 \vec{r}} + E_2 e^{i\omega_2 t - i\vec{k}_2 \vec{r}} + c.c.)^4, \qquad (11)$$

And we will be interested in the term

$$\sim E^2 e^{2i\omega_0 t - 2i\vec{k}_0 \vec{r}} E_1^* e^{-i\omega_1 t + i\vec{k}_1 \vec{r}} E_2^* e^{-i\omega_2 t + i\vec{k}_2 \vec{r}} + c.c.$$

We proceed the same way; the only difference from the case of PDC is that here, two pump fields enter the Hamiltonian. The latter then takes the form (9) with $\Gamma \sim \chi^{(3)} E_0^2 L$ and the frequency and wavevector mismatches being $\Delta\omega = 2\omega_0 - \omega_1 - \omega_2, \Delta\vec{k} = 2\vec{k}_0 - \vec{k}_1 - \vec{k}_2$.

Phase matching in four-wave mixing and modulation instability. Because these $\chi^{(3)}$ effects are usually observed in optical fibers, one cannot use birefringence to provide the phase matching. Periodic poling also does not work, since poling does not change $\chi^{(3)}$.

In this case, it is important whether the pump wavelength is below or above the 'zero-dispersion wavelength', ZDW $\lambda_0 = 2\pi c / \omega_0$ (Fig.5). At λ_0, the group velocity dispersion is zero and the dispersion dependence is flat.

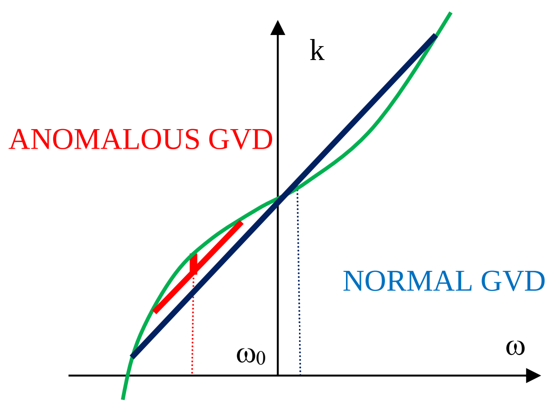

Fig.5

Above ω_0 (normal group-velocity dispersion range), it is possible to satisfy the relation $k(\lambda_1) + k(\lambda_1) = 2k(\lambda_0)$, where the indices 0,1,2 still mean the pump, signal, and idler. This is shown by the blue line. The pump wavelength should not be far below the ZDW, though. This regime is called four-wave mixing.

But below ω_0 (anomalous dispersion range) the pump wavevector cannot be the mean arithmetic of the signal and idler wavevectors. And here the Kerr effect helps. Indeed, if the pump is strong, its refractive index changes due to the Kerr effect (red vertical bar in the figure). And the stronger the pump, the larger the nonlinear change in the refractive index, and hence the further apart the signal and idler wavelengths. This regime is called modulation instability.

Fig.6 shows the signal and idler wavelengths as functions of the pump wavelength for different pump powers. We see that as the pump power increases, in the anomalous group-velocity dispersion range, the signal and idler wavelengths get separated due to the Kerr effect. In the normal dispersion range, their wavelengths are very different and do not depend on the pump power.

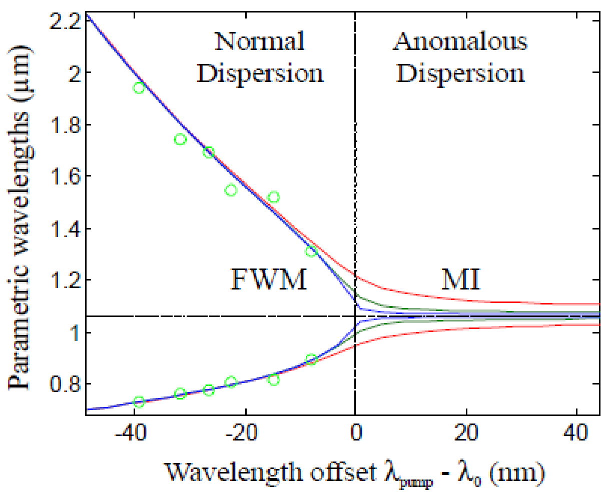

Fig.6 (from Wadsworth et al., Op. Ex. 12, 299 (2004)).

Home task: What will happen if the pump beam is not infinitely broad, but has a Gaussian shape with a waist w?

Books:

1. Boyd, Nonlinear optics
2. Klyshko, Physical foundations of quantum electronics