Lecture 2. Classical description of nonlinear quadratic interactions.

Harmonic generation, sum frequency generation, difference frequency generation. Phase matching. Three-wave interactions: frequency up-conversion, parametric amplification (frequency down-conversion).

1. *Classical and quantum description. Maxwell's equations.*

Not all nonlinear optical effects can be described in the framework of the classical theory. This is similar to the atomic transitions: stimulated transitions both 'up' and 'down' can be described semiclassically (quantum matter, classical light), but spontaneous transitions 'down' need quantum mechanical description. A 'trick' can be made by assuming 'quantum vacuum field', having the brightness equivalent to 1 photon per mode. But in this lecture, we will only consider effects that can be described in the framework of classical electrodynamics. All we will need is the nonlinear polarization (discussed in the last lecture) and Maxwell's equations.

We start with the latter. In the case of materials without induced changes and without currents, Maxwell's equations have the form

$$\begin{aligned} &\vec{\nabla}\times\vec{E} = -\dot{\vec{B}};\\ &\vec{\nabla}\times\vec{H} = \dot{\vec{D}};\\ &(\vec{\nabla}\cdot\vec{D}) = \rho = 0;\\ &(\vec{\nabla}\cdot\vec{B}) = 0. \end{aligned} \tag{1}$$

Here, $\vec{H}$ is magnetization field, $\vec{B}$ magnetic field, $\vec{D}$ displacement, $\vec{E}$ electric field, and dot denotes time differentiation. In a non-magnetic material, $\vec{B} = \mu_0\vec{H}$, but we should keep the relation between the displacement and the electric field: $\vec{D} = \varepsilon_0\vec{E} + \vec{P}$, where for $\vec{P}$ the nonlinear expansion in powers of the field is valid. We multiply the first equation in (1) vectorially by $\vec{\nabla}$ and differentiate the second one; then we get

$$\vec{\nabla}\times\vec{\nabla}\times\vec{E} = -\mu_0\ddot{\vec{D}}.$$

Then we use an identity from the vector algebra, $\vec{\nabla}\times\vec{\nabla}\times\vec{A} = \vec{\nabla}(\vec{\nabla}\cdot\vec{A}) - \nabla^2\vec{A}$, where $\vec{A}$ is any vector and $\nabla^2 \equiv \Delta \equiv \frac{d^2}{dx^2} + \frac{d^2}{dy^2} + \frac{d^2}{dz^2}$ is the Laplace operator. If this identity is applied to $\vec{E}$, the term $\nabla(\vec{\nabla}\cdot\vec{E})$ vanishes for a plane wave (the field is transverse). Then, replacing μ_0 by $1/\varepsilon_0 c^2$, we get the *wave equation*, or the *Helmholtz equation*,

$$\nabla^2\vec{E} = \frac{1}{\varepsilon_0 c^2}\ddot{\vec{D}}.$$

It is convenient to split here $\vec{D}$ in the linear and nonlinear parts: $\vec{D} = \varepsilon_0\vec{E} + \vec{P}^L + \vec{P}^{NL}$, and write $\varepsilon_0\vec{E} + \vec{P}^L = \varepsilon_0\varepsilon\vec{E}$, $\varepsilon = n^2$ (this is how refractive index is introduced). Then, the wave equation takes the form

$$\nabla^2\vec{E} - \frac{\varepsilon}{c^2}\ddot{\vec{E}} = \frac{1}{\varepsilon_0 c^2}\ddot{\vec{P}}^{NL}. \tag{2}$$

This *Helmholtz equation* describes the field induced by nonlinear polarization. If the right-hand side is zero (no nonlinearity), any plane wave of the form $\vec{E}(\vec{r},t)=\vec{E}_0 e^{-i\omega t+i\vec{k}\vec{r}}$, $k^2=\frac{\varepsilon\omega^2}{c^2}$ will be a solution. You can imagine a pendulum having its own frequencies. The nonlinear polarization plays the role of the force driving this pendulum.

Note that here and further, we describe everything in terms of complex 'analytic signal', or positive-frequency field, because it is used in statistical and quantum optics. It is introduced by expanding the real field into a Fourier integral, then taking only the positive-frequency part.

Already from the general form of the Helmholtz equation (2) we can derive some important conclusions. Indeed, let us represent both the field and the polarization in terms of their frequency components:

$$\vec{E}^{(+)}(\vec{r},t)=\sum_n \vec{E}_n(\vec{r})e^{-i\omega_n t},\ \vec{P}^{NL(+)}(\vec{r},t)=\sum_n \vec{P}_n^{NL}(\vec{r})e^{-i\omega_n t}\ . \qquad (3)$$

Note that the field amplitudes $\vec{E}_n(\vec{r})$ are complex. Because the Helmholtz equation (2) is uniform in field and polarization, it will split into equations for each frequency component,

$$\nabla^2\vec{E}_n(\vec{r})+\frac{\varepsilon\omega_n^2}{c^2}\vec{E}_n(\vec{r})=-\frac{\omega_n^2}{\varepsilon_0 c^2}\vec{P}_n^{NL}(\vec{r}), \qquad (4)$$

i.e., the n-th frequency component of the polarization is related only to the n-th frequency component of the induced field.
At the same time, because, for an m-th order nonlinear effect, involving m fields, $\vec{P}_n^{NL}(\vec{r})e^{-i\omega_n t}=\varepsilon_0\chi^{(m)}\vec{E}_{\pm1}(\vec{r})\vec{E}_{\pm2}(\vec{r})...\vec{E}_{\pm m}(\vec{r})e^{-i(\pm\omega_1\pm\omega_2\pm...\omega_m)}$ (plus or minus, depending on whether the positive-frequency or negative-frequency field is involved), we can write that

$$\omega_n=\pm\omega_1\pm\omega_2\pm....\pm\omega_m, \qquad (5)$$

i.e., the frequency of the output field is a combination of the input field frequencies. In terms of photons, this equation can be considered as the energy conservation law.

As an example, consider quadratic nonlinearity, and two plane-wave input fields along the z axis, and we will also omit the vectors:

$$\begin{aligned} E_1(\vec{r},t)&=E_{10}e^{-i\omega_1 t+ik_1 z}+c.c.\equiv E_1^{(+)}+E_1^{(-)},\\ E_2(\vec{r},t)&=E_{20}e^{-i\omega_2 t+ik_2 z}+c.c.\equiv E_2^{(+)}+E_2^{(-)}. \end{aligned} \qquad (6)$$

The polarization is then

$$\begin{aligned} P^{(2)}&=\varepsilon_0\chi^{(2)}(E_1^{(+)}+E_1^{(-)}+E_2^{(+)}+E_2^{(-)})^2=\\ &=\varepsilon_0\chi^{(2)}[E_{10}^2e^{-i2\omega_1 t+i2k_1 z}+E_{20}^2e^{-i2\omega_2 t+i2k_2 z}+E_{10}E_{20}e^{-i(\omega_1+\omega_2)t+i(k_1+k_2)z}+\\ &+E_{10}E_{20}^*e^{-i(\omega_1-\omega_2)t+i(k_1-k_2)z}+c.c.]. \end{aligned} \qquad (7)$$

The first two terms describe second-harmonic generation from both fields, the third one sum-frequency generation, and the fourth one difference-frequency generation. The 'c.c.' describes then the negative-frequency part of the polarization, and hence the induced field; we will further omit it.

We already see from here that for the second-harmonic generation, the frequency of the output field will be $2\omega_1$ (or $2\omega_2$), for the sum-frequency generation, $\omega_1+\omega_2$, and for the difference-frequency generation, $\omega_1-\omega_2$ (if $\omega_1\geq\omega_2$).

In what follows, we will consider how different nonlinear effects arise from equation (2). The right-hand part will be different for second-order, third-order, etc. effects.

2. *Second-order effects (no depletion).*

These effects are related to the second-order polarization, $\vec{P}^{(2)} = \chi^{(2)}\vec{E}$. In the tensor form, it is written as

$$P_i^{(2)} = \varepsilon_0 \chi_{ijk}^{(2)} E_j E_k. \qquad (8)$$

The second-order susceptibility $\chi_{ijk}^{(2)}$ is therefore a tensor of rank 3.

Materials with $\chi^{(2)}$. The susceptibility or any other macroscopic property (refractive index, Raman tensor etc.) obeys the von Neumann principle: its symmetry cannot be lower than the symmetry of the material structure. It means that in a material with centrosymmetric structure, $\chi_{ijk}^{(2)}$ does not change under inversion. At the same time, any rank 3 tensor (and, in general, any odd-rank tensor) should change its sign under inversion: $\chi_{ijk}^{(2)} \to -\chi_{ijk}^{(2)}$. Therefore, in any material with inversion symmetry, $\chi_{ijk}^{(2)} = 0$.

Another way to prove it: under inversion, $\vec{P} \to -\vec{P}, \vec{E} \to -\vec{E}, \chi^{(2)} \to \chi^{(2)}$. Then (8) holds only with $\chi_{ijk}^{(2)} = 0$.

This means that among bulk materials, only some crystals have nonzero $\chi^{(2)}$. These are: quartz, lithium niobate, lithium tantalate, KDP, KTP, BBO, ZnS, CdS, CdSe and many others. Certainly, there are ways to obtain second-order nonlinear effects in centrosymmetric materials. It can be done by somehow breaking the symmetry. One way is to use an interface between two materials. The other is to apply electric field.

Consider first *sum-frequency generation* (SFG) and, as a special case, *second-harmonic generation* (SHG). For a start, we assume that there are two *strong fields* of the form (6). It means that *the fields will be considered undepleted*. The nonlinear polarization has the form (7), and we will only consider its third term,

$$P^{(2)} = \varepsilon_0 \chi^{(2)} [E_{10} E_{20} e^{-i(\omega_1+\omega_2)t + i(k_1+k_2)z} + c.c.].$$

The positive-frequency part of the nonlinear polarization $P^{(2)}$, according to Eq. (2), induces a new field (labelled 3) with the positive-frequency part

$$E_3^{(+)}(\vec{r},t) = E_{30}(z) e^{-i\omega_3 t + ik_3 z}.$$

Note that the frequency dependence we already know from (4): $\omega_3 = \omega_1 + \omega_2$. But about the spatial dependence, we will only assume that it is a wave propagating along z axis (Fig.1). Moreover, in the absence of the nonlinear polarization it will be a monochromatic wave, $E_{30}(z) = const$.

If the nonlinear interaction is not too strong, $E_{30}(z)$ will be a slowly varying amplitude.

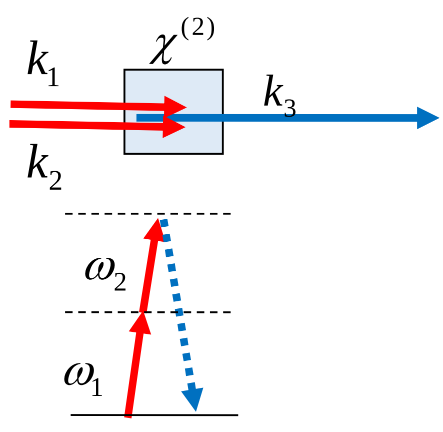

Fig.1

The Helmholtz equation (2) then yields

$$(\nabla^2 + k^2) E_{30}(z) e^{ik_3 z} = \frac{-\omega_3^2}{c^2} \chi^{(2)} E_{10} E_{20} e^{i(k_1+k_2)z}, \quad k_3 = \frac{n_3 \omega_3}{c}.$$

By differentiation we obtain

$$(\frac{d^2}{dz^2}+2ik_3\frac{d}{dz})E_{30}(z)=\frac{-\omega_3^{\ 2}}{c^2}\chi^{(2)}E_{10}E_{20}e^{i(k_1+k_2-k_3)z}.$$

Because $E_{30}(z)$ is a slowly varying amplitude, the first term can be ignored. (This will be the case if $\left|\frac{d^2E_{30}(z)}{dz^2}\right| << k_3\left|\frac{dE_{30}(z)}{dz}\right|$, i.e., the relative variation of $E_0(z)$ on a distance of a wavelength is small. This is a reasonable assumption.)

Then we introduce the wavevector mismatch, $\Delta k \equiv k_1+k_2-k_3$, and obtain the differential equation

$$\frac{dE_{30}(z)}{dz}=\frac{i\omega_3^{\ 2}}{2k_3c^2}\chi^{(2)}E_{10}E_{20}e^{i\Delta kz}. \qquad (9)$$

The solution is obtained by differentiating over the crystal length L,

$$E_{30}(L)=\frac{i\omega_3^{\ 2}}{2k_3c^2}\chi^{(2)}E_{10}E_{20}\int_0^L dze^{i\Delta kz}=\frac{\omega_3}{2n_3\Delta kc}\chi^{(2)}E_{10}E_{20}(e^{i\Delta kL}-1)=e^{i\Delta kL/2}\frac{\omega_3L}{2n_3c}\chi^{(2)}E_{10}E_{20}\mathrm{sinc}(\frac{\Delta kL}{2}).$$

The resulting intensity I_3 will scale as $\left|E_{30}(L)\right|^2$, and we can write

$$I_{SFG}=\kappa L^2[\chi^{(2)}]^2I_1I_2\mathrm{sinc}^2(\frac{\Delta kL}{2}), \qquad (10)$$

where κ is the proportionality coefficient.

We will point out three things here.

1. The role of phase matching. The sinc-squared function is unity at exact phase matching, when $\Delta k=0$. Then the intensity of the sum-frequency generation is maximal and the whole length of the crystal contributes. The phase matching equation, in terms of photons, can be considered as the momentum conservation law.

Without any special ‘tricks’, the phase matching cannot be satisfied. Consider, for instance, SHG. Then the phase matching implies that $2k(\omega)=k(2\omega)$. From the k-vector definition, it follows that the refractive index should be equal for the pump and the SH: $n(\omega)=n(2\omega)$. Using the Selmeier equations (fitting the refractive-index dispersion) you can see that this is impossible.

The phase matching can be satisfied by using the crystal anisotropy (some tasks will be solved in the problem class). However, in the absence of phase matching the nonlinear effect will be still present, but instead of the crystal length L, the intensity will be determined by the so-called coherence length $L_{coh}=2/\Delta k$. In some cases, it will be enough.

2. The role of susceptibility. The output intensity scales as $[\chi^{(2)}]^2$. Therefore it matters which components of susceptibility are used. The value entering (10) and other equations is, according to the definition of nonlinear polarization, the tensor $\chi^{(2)}_{ijk}$ contracted with the unity vectors defining the direction of the fields: $\chi^{(2)}=\chi^{(2)}_{ijk}e_i^3e_j^1e_k^2$, where the upper indices 1,2,3 denote the interacting waves (1,2 are input waves and 3 is the output wave). Note that in several nonlinear crystals such as lithium niobate, lithium tantalate, and KTP, the $\chi^{(2)}_{zzz}$ value exceeds the other ones by about an order of magnitude. Therefore in many cases the

polarizations along the z axis are chosen for all waves. The question is of course how to satisfy the phase matching, but there are methods.

3. The output intensity scales as $I_1 I_2$, and as I_1^2 for second-harmonic generation. If the intensities vary in time simultaneously (pulsed laser), the time-averaged $\langle I_1 I_2 \rangle$ will be higher than the product of the mean intensities. This is why pulsed lasers are good for nonlinear optical effects. The same relates to tight focusing.

Higher harmonic generation.

$$I(n\omega_1) \sim I_1{}^n [\chi^{(n)}]^2 \mathrm{sinc}^2(\Delta k \frac{L}{2}), \quad \Delta k = nk_1 - k.$$

Difference-frequency generation. Let us now take the fourth term in (7), $P^{(2)} = \varepsilon_0 \chi^{(2)} [E_{10} E_{20}^* e^{-i(\omega_1-\omega_2)t + i(k_1-k_2)z} + c.c.]$. Everything will be very similar, and we will have, similarly to (10),

$$I_{DFG} = \kappa L^2 [\chi^{(2)}]^2 I_1 I_2 \mathrm{sinc}^2(\frac{\Delta k L}{2}), \tag{11}$$

only now with $\Delta k \equiv k_1 - k_2 - k$.

All these cases (SHG, SFG, DFG) are about the interaction of two or three waves where only one is varying in space, the other(s) undepleted. Much more interesting is the case where two waves can exchange energy.

3. Three-wave interactions.

We will assume now that not only for the wave that emerges, but also for the other two waves, the amplitudes can vary (slowly). Instead of (9), we get

$$\frac{dE_{30}(z)}{dz} = \frac{i\omega_3}{2n_3 c} \chi^{(2)} E_{10}(z) E_{20}(z) e^{i\Delta k z}. \tag{12}$$

It is convenient to introduce for each wave, instead of intensity, photon flux density $F = \frac{I}{\hbar\omega}$ [photons/(s cm^2)] and its amplitude a, $|a|^2 = F$. Then (12) takes the form

$$\frac{da_3(z)}{dz} = i\beta a_1(z) a_2(z) e^{i\Delta k z}, \tag{13}$$

with the coupling constant $\beta \sim \chi^{(2)}(\omega_3 = \omega_1 + \omega_2)\sqrt{\omega_1\omega_2\omega_3}$.

Because equations similar to (12) can be written for $E_{10}(z)$ and $E_{20}(z)$ as well, we obtain a system of equations

$$\begin{aligned} \frac{da_1(z)}{dz} &= i\beta a_2^*(z) a_3(z) e^{-i\Delta k z}, \\ \frac{da_2(z)}{dz} &= i\beta a_1^*(z) a_3(z) e^{-i\Delta k z}, \\ \frac{da_3(z)}{dz} &= i\beta a_1(z) a_2(z) e^{i\Delta k z}. \end{aligned} \tag{14}$$

Here, for simplicity we will consider one wave (pump) as undepleted. The other two will then exchange energy. We assume that phase matching is satisfied: $\Delta k = 0$.

Frequency up-conversion. Assume that wave 1 is the strong pump: $a_1 = a_{10} = a_{10}{}^* = const$. Then we get from (14):

$$\frac{da_2}{dz} = i\gamma a_3,$$
$$\frac{da_3}{dz} = i\gamma a_2, \qquad (15)$$
$$\gamma \equiv \beta a_{10}.$$

By differentiating the first one and substituting the second one, we obtain

$$\frac{d^2 a_2}{dz^2} = -\gamma^2 a_2,$$

the solution being sines and cosines:

$$a_2 = a_{20}\cos(\gamma z) + C_2 \sin(\gamma z),$$
$$a_3 = a_{30}\cos(\gamma z) + C_3 \sin(\gamma z). \qquad (16)$$

The constants are found from (15):

$$-\gamma a_{20}\sin(\gamma z) + \gamma C_2 \cos(\gamma z) = i\gamma(a_{30}\cos(\gamma z) + C_3 \sin(\gamma z)),$$
$$-\gamma a_{30}\sin(\gamma z) + \gamma C_3 \cos(\gamma z) = i\gamma(a_{20}\cos(\gamma z) + C_2 \sin(\gamma z)).$$

At z=0, we get $C_2 = ia_{30}, C_3 = ia_{20}$.

Then, the solution is

$$a_2 = a_{20}\cos(\gamma z) + ia_{30}\sin(\gamma z),$$
$$a_3 = a_{30}\cos(\gamma z) + ia_{20}\sin(\gamma z). \qquad (17)$$

We see that the waves (modes) 2 and 3 are exchanging energies, like coupled pendulums, periodically. It means that if we want to transfer energy from wave 3 to wave 2 or vice versa, we can do it with 100% probability.

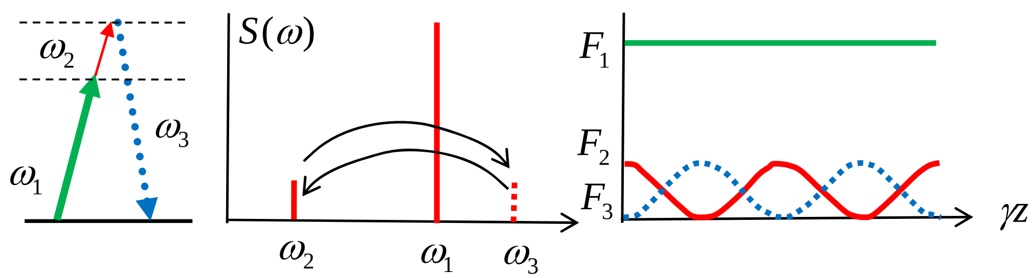

Fig.2

If initially only mode 2 is present at the output (Fig.2 left, middle), then $a_2 = a_{20}\cos(\gamma z), a_3 = ia_{20}\sin(\gamma z),$ and the photon fluxes exchange as

$$F_2 = F_{20}\cos^2(\gamma z), F_3 = F_{20}\sin^2(\gamma z). \qquad (18)$$

Parametric amplification. Now, assume that the strong pump is mode 3 having the highest frequency (Fig.3, left panel).

Then we get from (14):

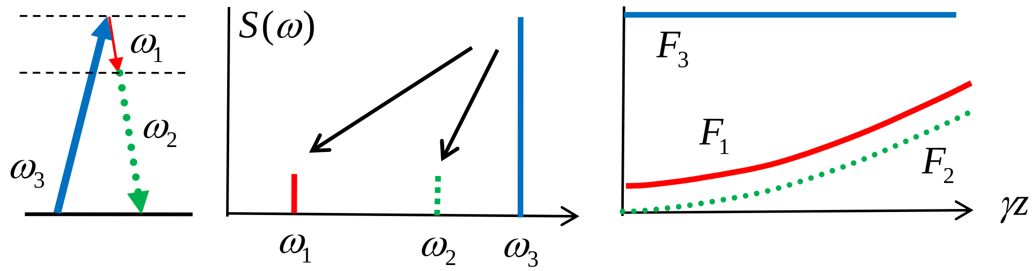

Fig.3

$$\frac{da_1}{dz} = i\gamma a_2^*,$$
$$\frac{da_2}{dz} = i\gamma a_1^*, \qquad (19)$$
$$\gamma \equiv \beta a_{30}.$$

By differentiating the first one and substituting the complex conjugated second one, we obtain

$$\frac{d^2 a_1}{dz^2} = \gamma^2 a_1,$$

the solution being hyperbolic sines and cosines (because $\gamma \to i\gamma$). If we also assume that initially, only wave 1 is present, then

$$a_1 = a_{10}\cosh(\gamma z),$$
$$a_2 = i a_{10}^* \sinh(\gamma z). \qquad (20)$$

For the photon fluxes, we obtain

$$F_1 = F_{10}\cosh^2(\gamma z),\ F_2 = F_{10}\sinh^2(\gamma z). \qquad (21)$$

It means that due to the pump energy, wave 1 is amplified (signal, parametric amplification) but also, wave 2 emerges (idler). The energy goes then from wave 3 to both waves 1 and 2 (Fig.3, central panel). The evolution of the photon fluxes is shown in the right panel. The dimensionless value $G = \gamma z$ is called the parametric gain (sometimes, $\sinh^2(\gamma z)$ is called parametric gain instead). Note that in the absence of wave 1, classical description does not predict any radiation at the output. The emission at the output will however appear as a result of quantum description.

Home task: A 3 mm BBO crystal pumped by strong pulses at 355 nm provides phase-matched parametric amplification of radiation at 1064 nm with the gain $\gamma z = 5$. Estimate the thickness of BBO needed for the 100% up-conversion of radiation at 1064 nm with the same pump (532 nm), assuming that the phase matching is satisfied with the same effective value of $\chi^{(2)}(\omega_3 = \omega_1 + \omega_2)$.

Books:

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