Experiment. Introduction. Part 2.

Nonlinear polarization.

As it has been discussed, the electric field of a light wave propagating in a medium induces charge displacement (deformation of electron cloud) of atoms and/or molecules of the medium. This results in polarization of molecules, i.e. generation of the induced dipole moment of the molecules. If the electric field strength of light wave is not high, the dipole moment is proportional to the field strength. In this case, the polarizability of a molecule or an atom is independent of the light wave intensity and, therefore, all optical parameters of the material are independent of the intensity of light.

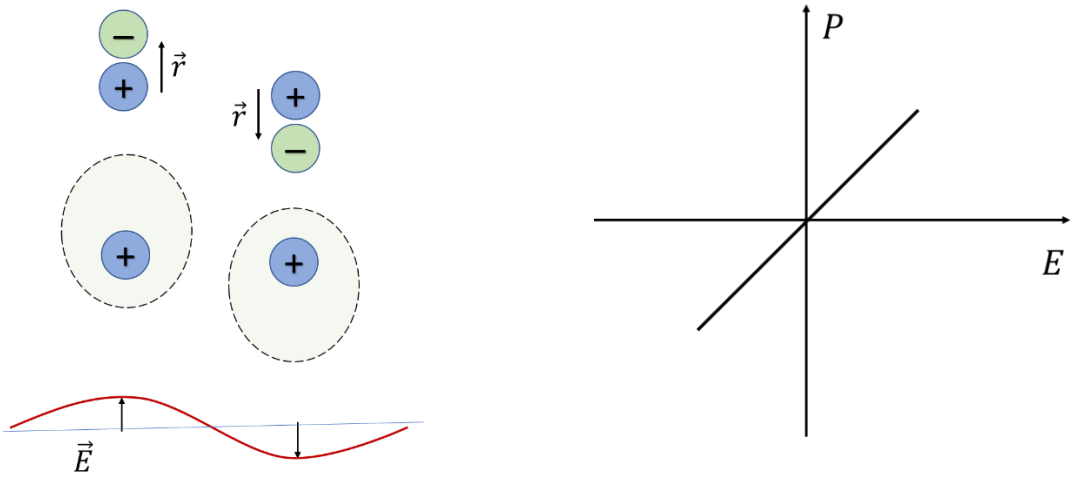


Fig. 1. Generation of linear polarization in matter.

In this case, the induced dipole moment of a single atom is directly proportional to the electric field of the incident light wave. The combined dipole momenta of a given volume of material is called the electric polarization of the medium or polarization density (not to be confused with polarization of light wave!). Therefore, one can write that the polarization density $\vec{P}$ of the medium is directly proportional to the electric field $\vec{E}$ (Fig. 1):

$$\vec{P} = \chi^{(1)}\vec{E}, \tag{1}$$

where $\chi^{(1)}$ – is the dielectric susceptibility of the medium. For simplicity, we will further disregard the vector nature of polarization equations – this is not going to obstruct the description of the phenomena under consideration. The approach considered see Eq.(1) allows one to describe completely the phenomena arising in propagation of weak light waves in non-absorbing media such as reflection, refraction, etc.

As a measure of strength of the electric field of light one usually adopts intraatomic electric field. Its strength is 10^8.. 10^9 V/cm by the order of magnitude, while the field of a regular light

source (not a laser) is only 0,1..10 V/cm, i.e. the electric field of a light wave E is much less than an intraatomic electric field E_a:

$$E \ll E_a \tag{2}$$

Therefore, it is safe to use Equation (1) for describing optical effects caused by weak light waves. In this case, the effects are called linear.

Consider a light wave which electric field strength does not meet the inequality (2). When such a field is applied to even an isotropic medium, the resulting displacement of electron clouds of molecules (or atoms) ceases to depend linearly on the electric field.

However, since the medium under study is isotropic and has no asymmetry (such a medium is called centrosymmetric), a deviation from the linear relation (1) for the induced dipole moment and, correspondingly, for the polarization density will be symmetric under inversion of the sign of *E* (Fig. 2).

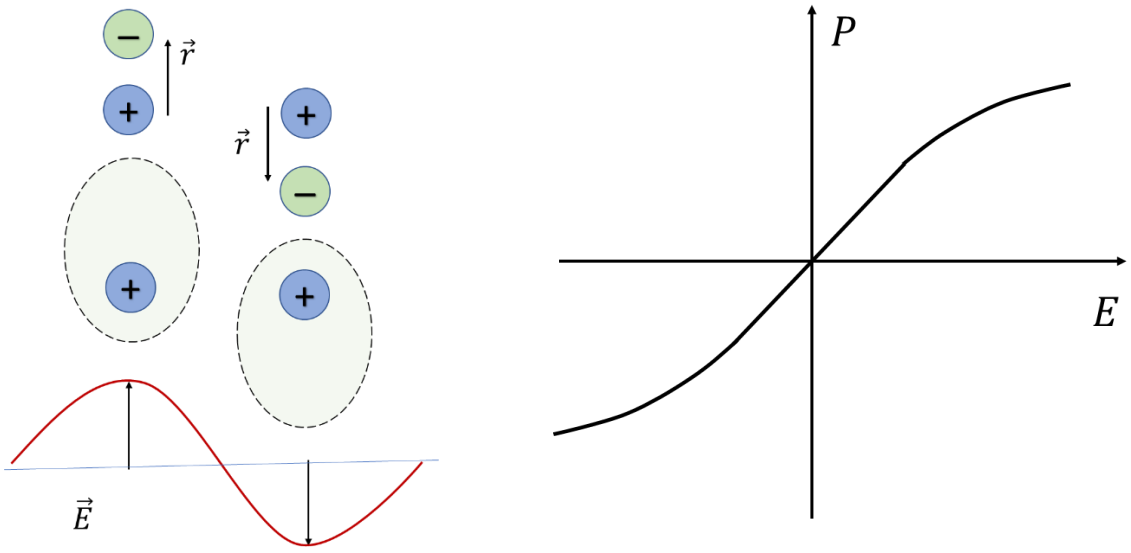


Fig. 2.Emergence of nonlinear polarization in isotropic medium.

In this case, the dependence of the polarization density *P* on the electric field *E* of a light wave can be written as a power series including only the terms of odd orders:

$$P = \chi^{(1)}E + \chi^{(3)}E^3 + \chi^{(5)}E^5 + \cdots \tag{3}$$

If the light wave is incident on an anisotropic medium, e.g. on a crystal with a certain symmetry (non-centrosymmetric medium), the electron cloud will be displaced differently in different directions (see Fig.3).

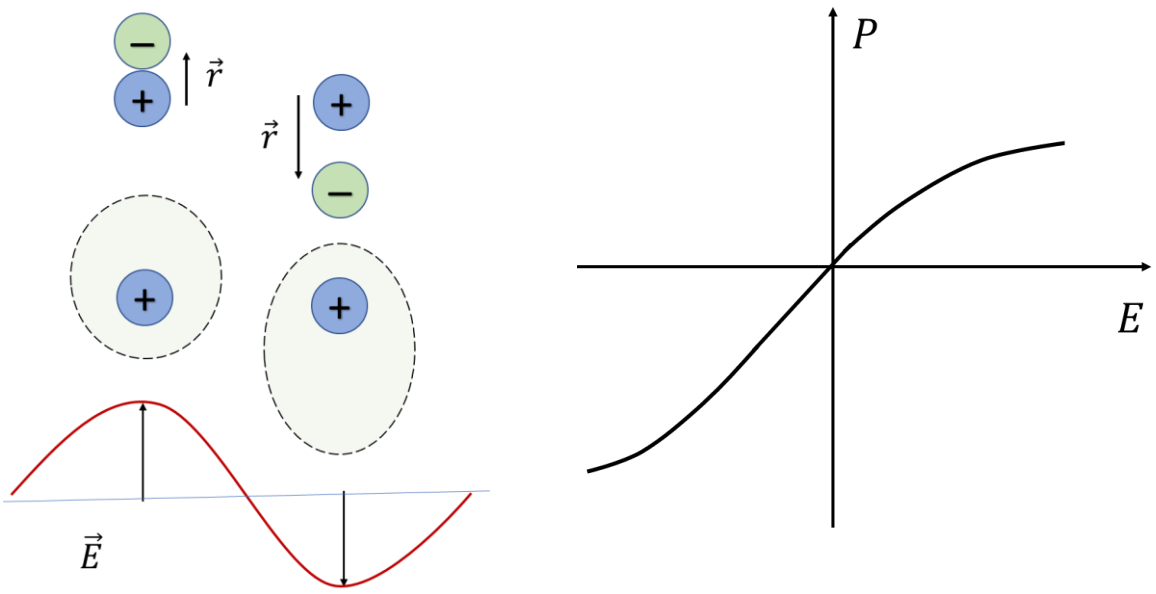


Fig. 3.Emergence of nonlinear polarization in asymmetric medium.

In this case, the dependence of the polarization density P on the electric field E of a light wave can be written as a power series including the terms of both odd and even orders:

$$P = \chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \chi^4E^4 + \cdots. \quad (4)$$

In the above equations, $\chi^{(1)}$ is the linear dielectric susceptibility of a material, $\chi^{(2)}$ is the nonlinear susceptibility of the second order, $\chi^{(3)}$ is the nonlinear susceptibility of the third order, etc.

The total field-induced polarization is also written as

$$P = P_{Л} + P_{НЛ}^{(2)} + P_{НЛ}^{(3)} + \cdots, \quad (5)$$

where P_{L} is a linear polarization, $P_{\text{NL}}^{(2)}$ is a quadratic nonlinear polarization, $P_{\text{NL}}^{(3)}$ is a cubic nonlinear polarization, etc.

Notice that in Equation (4):

$$\frac{\chi^{(n)}}{\chi^{(1)}} \sim \left(\frac{1}{E_a}\right)^{n-1}, \quad (6)$$

therefore, for weak fields the nonlinear terms in (4) can be neglected (when $E \ll E_a$).

Suppose that a plane harmonic wave with a strong electric field E is incident on such a medium,

$$E = E_0 \cos(\omega t - kx) \tag{7}$$

Substituting this equation in (4), using the notations introduced in (5), and applying well-known trigonometric formulae, one obtains:

$$\begin{aligned} P_{\text{Л}} &= \chi^{(1)} E_0 \cos(\omega t - kx) \sim \cos(\omega t - kx), \\ P_{\text{НЛ}}^{(2)} &= \chi^{(2)} (E_0 \cos(\omega t - kx))^2 \sim \cos(2\omega t - 2kx), \\ P_{\text{НЛ}}^{(3)} &= \chi^{(3)} (E_0 \cos(\omega t - kx))^3 \sim \cos(3\omega t - 3kx) \end{aligned} \tag{8}$$

and so on…

One can see that the polarization density now has not only the components oscillating at the fundamental frequency ω, but at the frequencies 2ω, 3ω, 4ω , etc. as well. And since the polarization density is a combination of dipole momenta of atoms and molecules of a medium, the medium itself will reemit electromagnetic field of all these frequencies. This is called generation of optical harmonics of the fundamental wave (of a frequency ω) of doubled (2ω), tripled (3ω), etc. frequencies (see Fig.4).

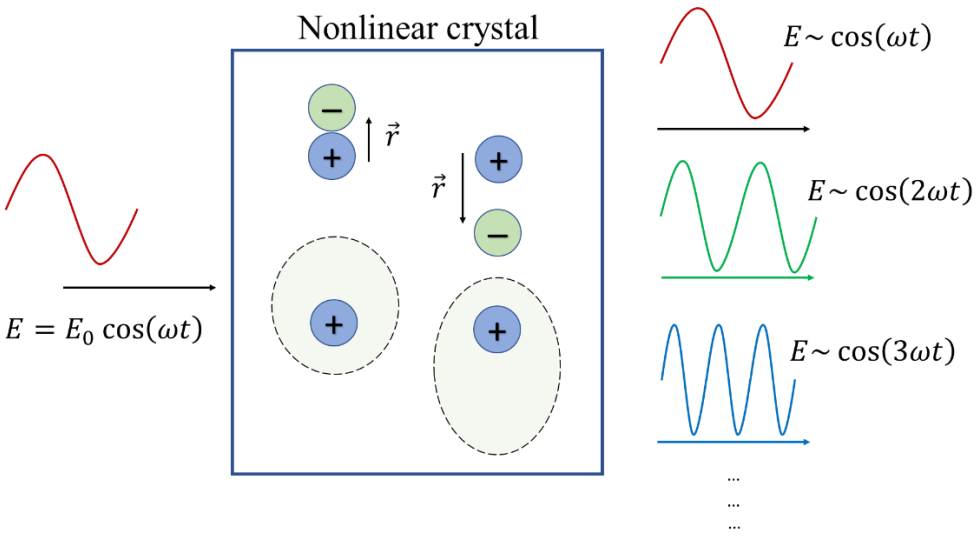


Fig. 4. Generation of optical harmonics in nonlinear medium.

Now consider separately the nonlinear polarization of the second order and the related nonlinear processes. Suppose two collinear light waves of different frequencies, $E_1(x,t) = E_{10} \cos(\omega_1 t - k_1 x)$ and $E_2(x,t) = E_{20} \cos(\omega_2 t - k_2 x)$, so that $E = E_1 + E_2$, propagate unidirectionally in a medium with a quadratic nonlinearity. In these equations, ω_i is a frequency

and $k_i = \frac{n_i(\omega_i)\omega_i}{c}$ (i = 1, 2). Using Equations (4) and (5) for quadratic polarization, one can write in a simplified form:

$$
\begin{aligned}
&P_{\text{НЛ}}^{(2)} = \chi^{(2)}E^2 = \chi^{(2)}[E_{10}\cos(\omega_1 t - k_1 x) + E_{20}\cos(\omega_2 t - k_2 x)]^2 = \\
&= \tfrac{1}{2}\chi^{(2)}E_{10}^2\cos[2(\omega_1 t - k_1 x)] + \tfrac{1}{2}\chi^{(2)}E_{20}^2\cos[2(\omega_2 t - k_2 x)] + \\
&+ \chi^{(2)}E_{10}E_{20}\cos[(\omega_1 + \omega_2)t - (k_1 + k_2)x] + \\
&+ \chi^{(2)}E_{10}E_{20}\cos[(\omega_1 - \omega_2)t - (k_1 - k_2)x] + \\
&+ \tfrac{1}{2}\chi^{(2)}(E_{10}^2 + E_{20}^2)\,.
\end{aligned}
\tag{9}
$$

Therefore, in the case at hand the nonlinear polarization contains components of doubled frequencies, of the sum and the difference of the frequencies, and the zero frequency component as well. The waves of polarization induced in the crystal generate radiation of the corresponding electromagnetic waves and, under certain conditions discussed below, it is possible that a significant energy of the incident wave will be transferred to the waves generated by the quadratic polarization.

Generation of the second harmonic.

Now consider the factors influencing the efficiency of nonlinear optical processes related to the quadratic polarization using generation of the second optical harmonic as an example. To this end, let us imagine an idealized situation when a plane wave of a frequency ω passes through an unbounded crystal layer of a thickness L. Let us determine the strength of the field at the frequency of the second harmonic at the exit surface of the crystal. The electric field of the fundamental frequency at a point with coordinate x (see Fig.5) is $E_0\cos(\omega t - k_1 x)$; according to (4) the polarization at this point is:

$$P_{\text{NL}}^{(2)} = \tfrac{1}{2}\chi^{(2)}E_0^2(1 + \cos(2\omega t - 2k_1 x)), \tag{10}$$

and at the doubled frequency (discarding the constant term) is:

$$P_{\text{NL}}^{(2)}(2\omega) = \tfrac{1}{2}\chi^{(2)}E_0^2\cos(2\omega t - 2k_1 x). \tag{11}$$

It should be noted here that the polarization wave of a doubled frequency propagates at the phase velocity $v_{P(2\omega)} = \frac{2\omega}{2k_1} = \frac{\omega}{k_1} = v_{E(\omega)}$ which is equal to the phase velocity at the fundamental

frequency, while the electromagnetic wave of the second harmonic frequency propagates in the crystal at the phase velocity $v_{E(2\omega)} = \frac{2\omega}{k_2}$, where $k_2 = n(2\omega)\frac{2\omega}{c}$ is the wavenumber of electromagnetic field of the second harmonic in the crystal.

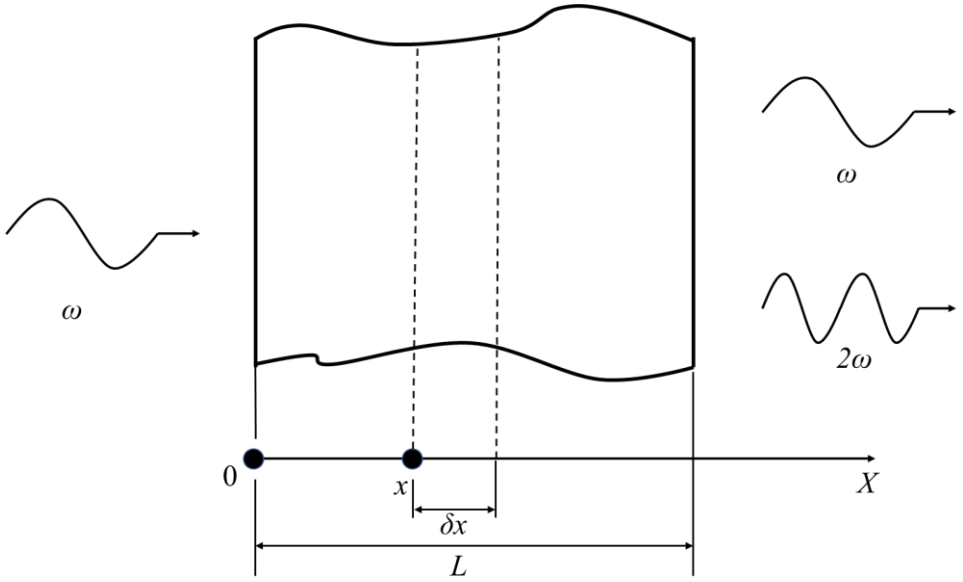


Fig. 5. Generation of the second harmonic in nonlinear crystal.

The electric field of the harmonic $\delta E(2\omega)$ emitted by the harmonic polarization $P_{\text{НЛ}}^{(2)}(2\omega)\delta x$ by a layer of thickness δx at the exit surface of the crystal is:

$$\delta E(2\omega) \sim E_0^2 \cos\big(2\omega t - 2k_1 x - k_2(L-x)\big)\delta x, \tag{12}$$

where $k_2(L-x)$ is the additional phase acquired by the wave of the second harmonic upon propagation inside the crystal from the layer δx (coordinate x) to the exit surface of the crystal (coordinate L).

Integrating Equation (12) over the crystal length, one obtains the magnitude of electric field of the second harmonic frequency at the exit surface of the crystal:

$$E_L(2\omega) = \int_0^L dE(2\omega) \sim \frac{E_0^2}{2k_1 - k_2} \sin\left[\frac{L}{2}(2k_1 - k_2)\right] \cos\left[k_1 L + \frac{k_2 L}{2} - 2\omega t\right]. \tag{13}$$

The corresponding intensity of the second harmonic is:

$$I(2\omega) = \langle E_L^2(2\omega)\rangle \sim E_0^4 \left[\frac{\sin\left(\frac{L}{2}(2k_1 - k_2)\right)}{2k_1 - k_2}\right]^2 = I_0^2 \frac{L^2}{4}\left(\frac{\sin u}{u}\right)^2, \quad (14)$$

where $u = \frac{(2k_1 - k_2)L}{2} = \frac{\Delta kL}{2}$.

Equation (14) and the derivations to follow are made under assumption that the magnitude of the fundamental wave propagating in a nonlinear medium does not increase by generation of the second harmonic and remains constant. This assumption is called the fixed field approximation. Of course, this is not strictly true because the energy transferred to the second harmonic is supplied by the fundamental radiation wave which intensity thereby decreases. However, for a small conversion efficiency (less than 10 %) the fixed field approximation describes the experimental results fairly well. The conversion efficiency to the second harmonic η is defined as

$$\eta = \frac{I(2\omega)}{I(\omega)} \times 100\%. \quad (15)$$

An exact expression for the intensity of the second harmonic wave exiting the crystal of a thickness L can be obtained in the fixed field approximation (i.e. when the conversion efficiency of the second harmonic is small, no more than 10%) by solving the so-called reduced equations for the amplitudes of interacting waves $E(\omega)$ and $E(2\omega)$. Up to a common factor, the solution is:

$$I(2\omega) = C\frac{d^2L^2I^2(\omega)}{n_1^2 n_2 \lambda_2^2}\left[\frac{\sin u}{u}\right]^2, \quad (16)$$

where C is a numerical factor,

d is a quantity specifying the quadratic nonlinear polarizability of the crystal (directly proportional to $\chi^{(2)}$),

$I(\omega)$ is the intensity of the field of a frequency ω entering the crystal (the fundamental radiation wave),

n_1 and n_2 are the refractive indices of the fundamental wave (of the frequency ω) and of the second harmonic (of the frequency 2ω),

λ_2 is the wavelength of the second harmonic,

L is the length of nonlinear crystal.

Equations (14) and (16) exhibit some typical features of the process of generation of the second harmonic.

1. For $u \neq 0$, i.e. $\Delta k = 2k_1 - k_2 \neq 0$, the intensity of the second harmonic depends on the length L of nonlinear crystal as $(\sin(\Delta kL/2))^2$ (see Fig.6). Here, the maximum possible intensity is such that it could be generated by a crystal of a length $L_{coh} = \frac{\lambda_1}{4(n_1 - n_2)}$ (follows from $u = \pi/2$), called the coherent length. This length in transparent crystals equals several tens of wavelengths, i.e. of the order of 10^{-3} cm. Therefore, the coherent length is much less than the length of a crystal which can be used for generation of intensive radiation of the second harmonic.

2. For $u = 0$, i.e. when

$$\Delta k = 2k_1 - k_2 = 0, \tag{17}$$

the intensity of the second harmonic (see Fig.6) grows like the squared length of nonlinear crystal (because $\lim_{u \to 0} \frac{\sin u}{u} = 1$), of course, as long as the fixed field approximation remains valid. Equation (17) is called the phase matching condition for generation of the second harmonic.

3. Intensity of the radiation of the second harmonic is proportional to the squared intensity of the fundamental radiation, i.e. $I(2\omega) \sim I^2(\omega)$.

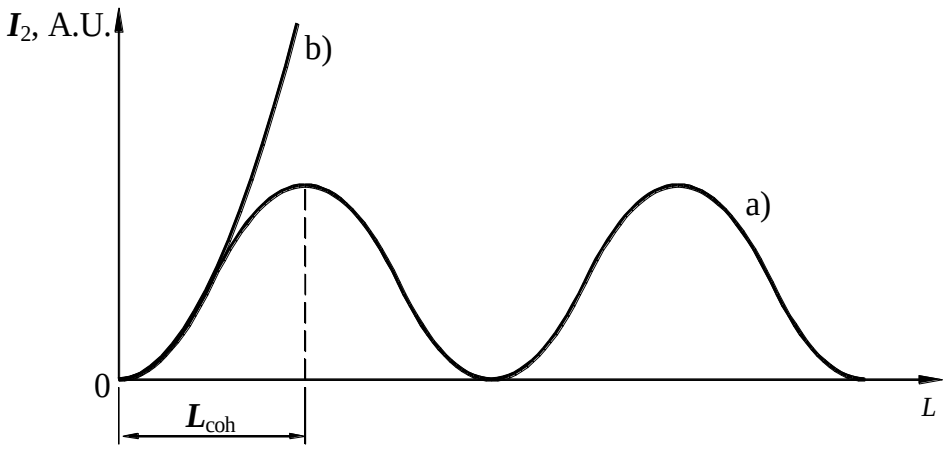


Fig. 6. Intensity of the second harmonic I_2 versus length L of nonlinear crystal:

a) $\Delta k \neq 0, \quad I_2 \sim (sin(\Delta kL/2))^2;$ *b)* $\Delta k = 0, \quad I_2 \sim L^2$

The phase matching condition

Therefore, effective generation of the second optical harmonic is possible only if the condition (17) is met, which is true if refractive indices for the waves of the fundamental frequency and the

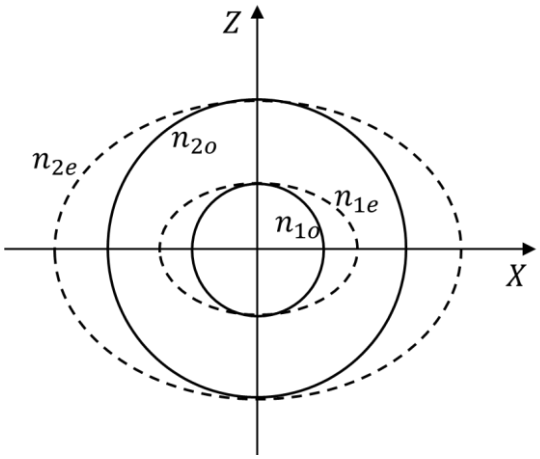
second harmonic are equal: $n_1 = n_2$ (since $k_i = \frac{n_i(\omega)\omega_i}{c}$). This condition cannot be met for an isotropic medium because its refractive index in the transparency range monotonically increases with frequency (normal dispersion) and $n_2 > n_1$ always.

This obstacle can be circumvented by using interaction of waves of different polarization (ordinary and extraordinary) propagating in birefringent crystals. In some crystals, it turns out, there are directions of propagation in which, e.g. the refractive index of ordinary wave of the fundamental frequency equals the refractive index of extraordinary wave at the second harmonic frequency.

The diagram in Fig.7 depicts cross sections of refractive index surfaces at frequencies ω and 2ω for a crystal of quartz (a) and a crystal of potassium dihydrogen phosphate KH_2PO_4 (KDP) (b). Both crystals are uniaxial (the optical axis is denoted as *Z*). Quartz crystal is positive: the principal value of the refractive index for extraordinary wave is greater than for ordinary wave ($n_e > n_o$). KDP crystal is negative: $n_e < n_o$.

When direction of propagation of extraordinary wave relative to the optical axis varies (angle Θ), the refractive index for this wave in the crystal changes between n_o and n_e (see Fig.7). By this reason, in some crystals there are such directions Θ_{pm} of propagation in which (e.g. for a negative crystal) $n_{1o} = n_{2e}(\Theta_{\text{pm}})$, i.e. the condition (17) is met.

a) Positive crystal of quartz ($n_e > n_o$)



b) Negative crystal of KDP ($n_e < n_o$)

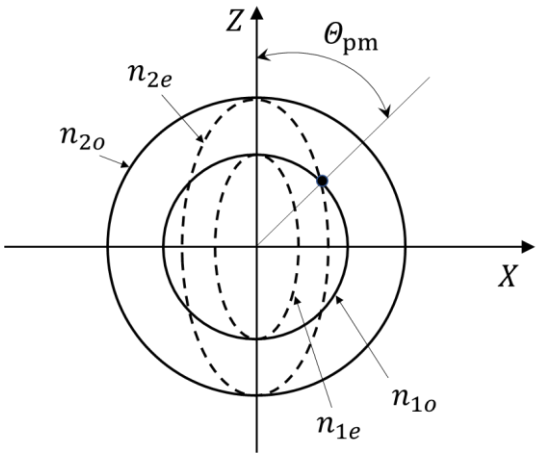


Fig. 7. Cross sections of refractive index surfaces for positive a) and negative b) crystals.

In spite of different directions of polarization of the waves, a nonlinear interaction in such crystals is possible. Suppose, e.g. an ordinary wave is incident on the crystal. The electric field vector of the wave is perpendicular to the principal optical plane and, correspondingly, to *Z*-axis. Due to complicated interaction of the incident wave and the crystal, the wave will generate oscillation of the displaced charges at a doubled frequency in other directions thereby generating the second harmonic wave polarized parallel to the principal optical plane, i.e. the extraordinary wave. Wise choice of the incident wave direction (at the angle Θ_c relative to *Z*) makes it possible to transfer a significant fraction of energy to the second harmonic.

A KDP crystal has a valuable feature: its surfaces $n_{1o}(\Theta)$ and $n_{2e}(\Theta)$ intersect in a wide interval of frequencies ω of fundamental radiation (in particular, the interval includes the frequencies of ruby and neodymium lasers). One can see in Fig.7 (b) that $n_{1o} = n_{2e}(\Theta_{\text{pm}})$ for the waves of frequencies ω and 2ω propagating at some angle Θ_c to the optical axis of the crystal. This angle is called the angle of phase matching. The phase velocities of waves propagating at this angle are equal at frequencies ω and 2ω.

The angle of phase matching Θ_c can be calculated if the principal refractive indices n_{1o}, n_{1e}, n_{2o}, and n_{2e} are known.

Types of phase matching.

The condition (17) of phase matching for collinear waves can be written as

$$k_{1o} + k_{1o} = k_{2e}, \tag{18}$$

where k_{1o} is the wavenumber of ordinary wave of the frequency ω and k_{2e} is the wavenumber of extraordinary wave of the second harmonic frequency 2ω.

The condition (18) is called the type I phase matching condition (or oo-e). The notation “oo-e” means that two ordinary waves of the fundamental wave interact with an extraordinary wave of

the second harmonic. By the two waves of fundamental radiation wave we imply that generation of the second harmonic can be considered as a generation of the sum of frequencies $\omega + \omega = 2\omega$ of two waves of the same frequency ω. Wherein other specifications of the waves (orientation of polarization plane, wavevectors, and amplitudes) can be different.

Under "oo-e" interaction the polarization planes of the two waves of fundamental radiation wave coincide (both waves are ordinary) and if their amplitudes, phases, and wavevectors are equal, the waves become indistinguishable; however, one should keep in mind that this is only a particular case.

Another type of phase matching interaction for generation of the second harmonic is possible for a KDP crystal, namely, the interaction of ordinary and extraordinary waves of the fundamental radiation with an extraordinary wave of the second harmonic (the so-called type II interaction or oe-e). The phase matching condition for this interaction in one-dimensional, or scalar, case (i.e. when all waves propagate in the same direction) is:

$$k_{1o} + k_{1e} = k_{2e}, \tag{19}$$

whence the refractive indices for two waves are related as:

$$\frac{n_{1o} + n_{1e}}{2} = n_{2e}. \tag{20}$$

Generation of the second harmonic in real optical beams

The results described above were derived for plane waves and under the assumption that the radiation incident on the crystal was monochromatic. Often, the laser radiation can be regarded as monochromatic (i.e. its light field vector oscillates harmonically). And in what follows, we will not take into account phenomena related to non-monochromaticity.

When using a real beam of a certain diameter and a finite divergence, it is important to match the parameters of laser radiation and the parameters of crystals used as frequency doublers. To describe specifications of non-linear optical converters when using real light beams, the concept of angular width of phase matching is introduced.

As it was discussed above, the phase matching condition (17) for the type I interaction and for generation of the second harmonic is reduced to $n_{1o} = n_{2e}(\Theta_{\text{pm}})$. It is evident that the quantity$\Delta k = 2k_1 - k_2$ is a function of the angle Θ because the refractive index n_{2e} of extraordinary wave depends on the angle. At the exact phase matching ($\Delta k = 0$) and a subsequent variation of the angle the conversion efficiency decreases (according to (14) and (16)) and the quantity I_2 changes as $\left(\frac{\sin u}{u}\right)^2$ (see Fig.8).

It is not difficult to calculate a quantity $\Delta\theta$, such that $I_2\left(\Theta_{\text{pm}} + \Delta\theta\right) = \frac{1}{2} I_2(\Theta_{\text{pm}})$. The angular width of phase matching for a given interaction type is then a quantity $\Delta\Theta_{\text{pm}} = 2\Delta\theta$. Since the

argument of $\sin(u)/u$ is $\Delta kL/2$, the angular width of phase matching is inversely proportional to the crystal length. In reference literature the angular width of phase matching is given for the length of one centimeter. The quantity $\Delta\Theta_c$ for various crystals spans a wide range between several minutes and several degrees.

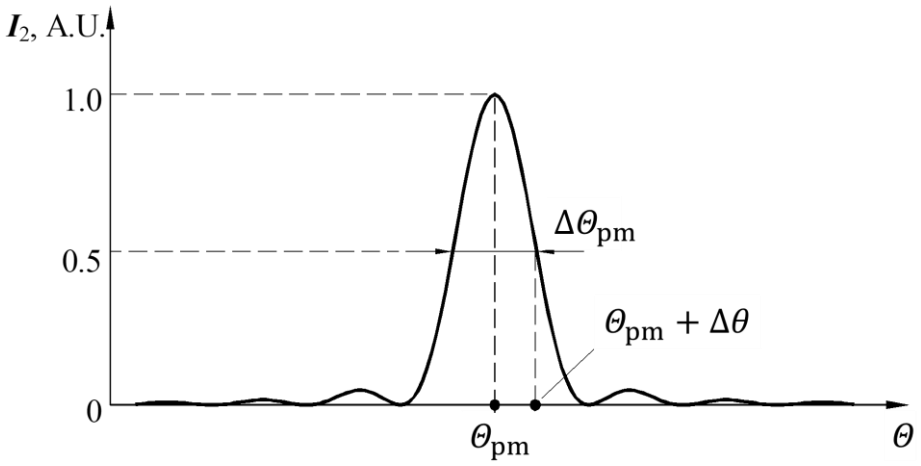


Fig. 8. Intensity of the second harmonic versus angle.

It is important to understand that such a dependence exists for the incident plane wave with a single vector $\vec{k}$. The angular divergence of this wave is zero (see Fig.9(a)). A real beam can be represented by a sum of many wavevectors of various directions and magnitudes; the angular distribution of laser beam intensity usually has a bell-like shape of a certain nonzero divergence $\Delta\varphi$ (see Fig.9 (b)). One should keep in mind that when a real beam interacts with a nonlinear crystal, the conversion into the second harmonic illustrated in Fig.8 occurs to all beam vectors $\vec{k}_i$.

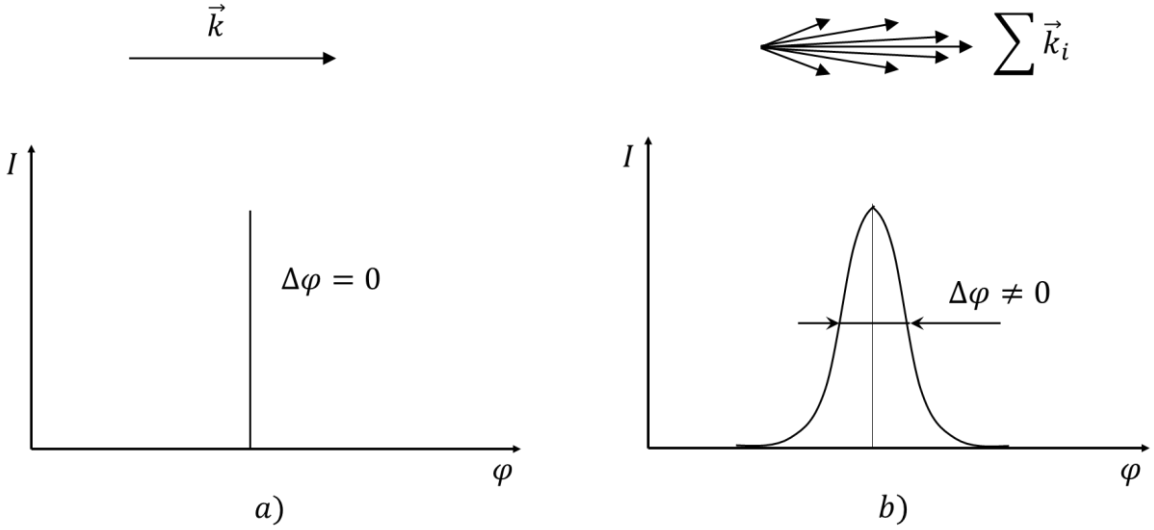


Fig. 9. Angular divergence of plane wave (a) and real beam (b).

When choosing the frequency converter for a specific laser system it is necessary to ensure that the divergence of the laser beam does not exceed (or, preferably, is much less) the corresponding $\Delta\Theta_{pm}$ of the chosen converter.

Generation of the third harmonic

As it follows from Equations (4) and (8), the third harmonic can be generated by using nonlinearity of the third order. The third harmonic occurs even in a centrosymmetric medium. In this case however, the conversion efficiency is low since, according to (6), the nonlinear susceptibility is significantly less, about a factor of 10^9 than the nonlinear susceptibility of the second order. Besides, it is challenging to meet the phase matching condition for effective generation of the third harmonic by using the third order nonlinearity.

To enhance the effectiveness of conversion of the fundamental radiation into the third harmonic, the method of the so-called cascade generation can be employed. The process consists of the following steps: a laser radiation of a frequency ω enters the first nonlinear crystal with a quadratic nonlinearity which generates the second harmonic of a frequency 2ω. Thus, two waves exit the first crystal: a wave of the fundamental frequency ω, and a wave of the doubled frequency 2ω. Then these two waves enter the second crystal with a quadratic nonlinearity where the frequencies are being add up: $\omega + 2\omega = 3\omega$. According to (9) this process is possible. Moreover, an exact calculation shows that in this case the intensity of the third harmonic I_3 depends on the intensities of the first and the second harmonics as:

$$I_3 \equiv I(3\omega) = C\frac{d^2L^2I(\omega)I(2\omega)}{n_1n_2n_3\lambda_3^2}\left[\frac{\sin u}{u}\right]^2, \tag{21}$$

where C is a numerical factor,

d is a quadratic nonlinear polarizability of the crystal (directly proportional to $\chi^{(2)}$),

$I(\omega)$ is an intensity of the wave of frequency ω entering the crystal (the fundamental wave),

$I(2\omega)$ is an intensity of the wave of frequency 2ω entering the crystal (the wave of doubled frequency),

n_1, n_2, and n_3 are the refractive indices for the fundamental wave ω, the second harmonic 2ω, and the third harmonic 3ω, respectively,

λ_3 is a wavelength of radiation of the third harmonic, and

L is the crystal length.

In Equation (21) $u = \Delta kL/2$, but now Δk is a phase mismatch of the three waves:

$$\Delta k = k_1 + k_2 - k_3. \tag{22}$$

When using uniaxial negative nonlinear crystals such a cascade process is possible providing the first nonlinear crystal employs type I interaction (oo-o) for conversion to the second harmonic and the second crystal employs type II interaction (oe-e or eo-o) for summing up the frequencies

ω and 2ω. In so doing, the type of the nonlinear crystal determines which wave (ω or 2ω) entering the crystal will be ordinary and/or extraordinary. The process of cascade generation of the third harmonic is sketched in Fig.10.

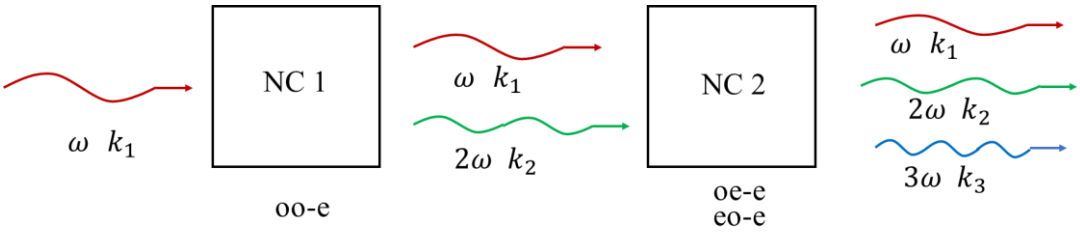


Fig. 10.Cascade generation of the third harmonic.

It should be also noted that if the type I phase matching (oo-e) is used, the nonlinear crystals are oriented and cut so that the incident linearly polarized wave propagates only as «o»-wave in the medium. If type II phase matching is employed (oe-e or eo-e) the crystal is oriented so that the incident wave corresponds either to «o»-wave or «e»-wave of the crystal.