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Consecutive three-wave interactions in nonlinear optics of periodically inhomogeneous media

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Abstract. A brief review of recent advances in the studies of two coupled three-wave nonlinear optical processes with multiple frequencies, which possess a number of special properties compared to conventional three-wave processes, is presented. The consecutive interactions of co-propagating and counter-propagating light waves in a $LiNbO_3$ crystal with a regular domain structure are considered. The energy exchange upon consecutive interactions of the waves with frequencies ω, 2ω and 3ω is analysed. The prospects of using consecutive nonlinear optical processes in quantum electronics and nonlinear optics are discussed.

1. Introduction

It seems that the term 'consecutive interactions' was introduced for the first time by S A Akhmanov and R V Khokhlov in their book 'Problems of Nonlinear Optics' [1], where they noted on page 198 that 'the number of parametric effects in a quadratic medium can be substantially extended if its dispersion properties admit consecutive three-frequency interactions'. They also pointed out in this book that the consideration of two consecutive three-frequency interactions should involve the interaction of the waves with frequencies ω_1, ω_2, ω_3, and ω_4, which satisfy the relations

$$\omega_1 + \omega_2 = \omega_{\rm p},$$
$$\omega_1 + \omega_{\rm p} = \omega_3, \qquad (1)$$
$$\omega_2 + \omega_{\rm p} = \omega_4,$$

where $\omega_{\rm p}$ is the pump-wave frequency. The first of these relations corresponds to the parametric amplification upon high-frequency pump and the two others to the parametric frequency conversion. Note that these processes are conventional and well studied individually.

These interactions have been studied in the radio-frequency range in the late 1950s – early 1960s (see, for example, [2 – 6]). In particular, the conditions of parametric amplification upon low-frequency pump have been found (see also [1]). However, such a parametric process cannot be realised upon the three-wave mixing only, i.e., in the absence of consecutive interactions.

In Ref [7] excitation of the third optical harmonic in a homogeneous quadratically nonlinear crystal using consecutive processes of the second harmonic generation $\omega + \omega = 2\omega$ and optical frequency mixing $2\omega + \omega = 3\omega$ was considered. However, the conditions of collinear phase matching cannot be simultaneously satisfied in a homogeneous nonlinear crystal for the above processes and processes involving waves with multiple frequencies (see below), because this requires the identity of the phase velocities of the three waves with multiple frequencies (see p. 194 in [1]).

At the same time, the realisation of consecutive interactions in nonlinear optics appears attractive because in this

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case the number of generated frequencies is increased without the use of additional cascades of nonlinear optical conversion.

It appears that consecutive interactions have been realised for the first time in nonlinear homogeneous crystals in Ref. [8]. The authors [8] observed simultaneously the nondegenerate parametric generation, the signal-wave frequency doubling, and the difference frequency generation in a $LiNbO_3$ crystal with the phase-matching angle of 46.7° at 120°C pumped at 1.065 μm. In this case, the second-harmonic wavelength could be tuned from 0.925 to 0.95 μm by changing the crystal temperature and rotating the crystal.

In Ref. [9], similar consecutive collinear interactions were observed in a $LiNbO_3$ crystal at room temperature, the signal-wave wavelength being 1.889 μm and that of the difference-frequency emission being 2.436 μm. However, these experiments performed with homogeneous nonlinear crystals can be considered as exotic.

In the early 1990s, the authors [10] showed theoretically that the energy of the intense pump wave with frequency 3ω can be completely converted to that of the wave with frequency 2ω in consecutive three-wave processes with multiple frequencies ω, 2ω, and 3ω. In this case, processes of the type $3\omega = 2\omega + \omega$ and $\omega + \omega = 2\omega$ simultaneously occur.

As mentioned above, the conditions of collinear phase matching are not satisfied simultaneously for these processes in homogeneous crystals. On the other hand, as has been first shown in Ref. [11], the conditions of quasi-phase matching can be simultaneously satisfied for processes

$$2\omega = \omega + \omega, \quad \omega + 2\omega = 3\omega. \tag{2}$$

in a periodically polarised $LiNbO_3$ crystal [a crystal with the 180-degree regular domain structure (RDS)]. This possibility for $LiNbO_3$ and KTP crystals has been also noted in Ref. [12].

Therefore, by varying the modulation period of the nonlinear susceptibility and (or) choosing the phase matching order in RDS crystals, one can compensate simultaneously for the phase mismatch in two coupled three-wave processes of the type (2). In particular, the possibility of a highly efficient parametric amplification in RDS crystals upon low-frequency pump in processes of the type (2) was demonstrated in Ref. [13].

The dynamics of energy exchange upon consecutive quasi-phase-matched third harmonic and third subharmonic generation, as well as the conversion of the energy of the wave with frequency 3ω to that of the wave with frequency 2ω were later studied in Refs [14, 15]. Note also that similar studies in homogeneous nonlinear media were performed in Refs [16, 17].

By now, consecutive quasi-phase-matched interactions of light waves in RDS crystals were observed in Refs [12, 18–20], where the third harmonic generation was studied upon the interaction between co-propagating waves. The consecutive quasi-phase-matched third harmonic generation upon the interaction between counter-propagating waves was observed in Refs [19, 20]. The consecutive interaction between counter-propagating waves upon parametric amplification in the low-frequency pump field was theoretically studied in Ref. [21].

The aim of this review is to consider the physical properties of the energy exchange between the waves with multiple frequencies in two consecutive three-wave interactions.

2. Conventional quasi-phase-matched interactions of light waves

The idea of using periodic modulation of the quadratic susceptibility to compensate for the mismatch of the wave vectors of interacting waves belongs to Bloembergen [22, 23]. Later, such processes were called quasi-phase-matched processes. The principle of quasi-phase matching can be conveniently explained by analysing the second harmonic generation. In the presence of the phase mismatch $\Delta k_2 = k_2 - 2k_1$ ($k_j = k(j\omega)$ is the wave number of the jth harmonic), the second-harmonic amplitude changes, in the given field approximation, as (see, for example, [1])

$$\frac{dA_2}{dz} = -i\beta' A_{10}^2 \exp(i\Delta k_2 z), \tag{3}$$

where β' is the coefficient of nonlinear coupling of the waves and A_{10} is the complex amplitude of the fundamental wave of frequency ω. From (3), we obtain

$$A_2(z) = -i\beta' A_{10}^2 \frac{\sin(\Delta k_2 z/2)}{\Delta k_2/2} \exp(-i\Delta k_2 z/2). \tag{4}$$

The second-harmonic intensity $I_2 = |A_2|^2$ achieves the maximum value at the length $z = l_c$ (where $l_c = \pi/|\Delta k_2|$ is the so-called coherence length). In this case, the phase incursion caused by the mismatch is equal to π. If we locate behind the first crystal the second nonlinear crystal of length l_c, which has the same mismatch Δk_2 in the direction of propagation of the interacting waves, then the second-harmonic amplitude at the output of the second crystal will be

$$A_2(z) = -(\beta' - \beta'') A_{10}^2 \frac{\sin(\Delta k_2 z/2)}{\Delta k_2/2}. \tag{5}$$

Here, β'' is the coefficient of nonlinear coupling of the waves for the second crystal. It is obvious that the maximum amplitude of the second harmonic is achieved for $\beta' = -\beta''$. Therefore, a change in the sign of nonlinearity of the second crystal compensates for the destructive phase incursion.

In this case, the amplitude and intensity of the second harmonic can be represented in the form

$$A_2 = -\frac{2}{\pi} \beta'(2l_c) A_{10}^2, \tag{6}$$

$$I_2 = \left[\frac{2}{\pi} \beta'(2l_c)\right]^2 I_{10}^2, \quad I_{10} = |A_{10}|^2. \tag{7}$$

According to (7), the second-harmonic intensity changes upon quasi-phase-matched interaction of the waves as in a homogeneous medium with the effective nonlinear coefficient $\beta_{eff} = 2\beta'/\pi$. Comparison of (4) and (6) shows that the second-harmonic phase changes from layer to layer in contrast to a homogeneous medium where, in the absence of the second harmonic at the input, the phase takes the constant value at once.

Expression (5) corresponds to the first order of quasi-phase matching. In a more general case, when the thickness of an individual layer of the crystal is $l = m l_c$ (Fig. 1), the quasi-phase matching condition has the form

$$\Delta k_2 = m\pi/l_c, \quad m = \pm 1, \pm 3, \ldots. \tag{8}$$

In this case, $\beta_{eff} = 2\beta'/\pi|m|$, i.e., the effective nonlinear coef-

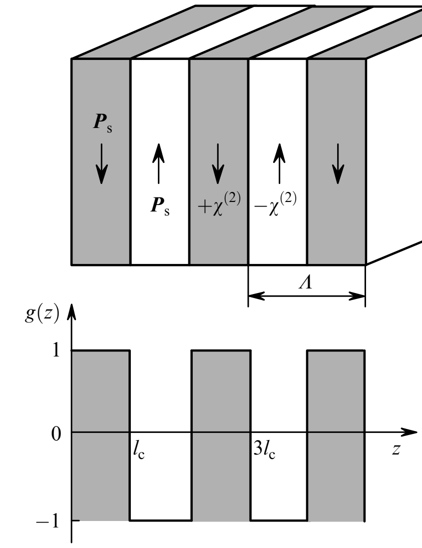

Figure 1. Nonlinear material with the RDS (a) and the modulation function of the nonlinear susceptibility (b); $\boldsymbol{P}_s$ is the polarisation vector and $\chi^{(2)}$ is the nonlinear susceptibility.

ficient decreases with increasing order of the quasi-phase matching.

The condition (8) admits another clear interpretation. Let us represent the coefficient of nonlinear coupling in the form $\beta = \beta_2 g(z)$, where β_2 is the coefficient modulus and $g(z)$ is an alternating periodic function (Fig. 1) equal to $+1$ or -1 at the layer thickness l. The period of a grating appearing in this case is $\Lambda = 2l$.

Let us expand the function $g(z)$ in a series

$$g(z) = \sum_{m=-\infty}^{\infty} g_m \exp(-\mathrm{i}mKz),$$

where $g_m = 2/(\pi|m|)$, and $K = 2\pi/\Lambda$ is the modulus of the vector of a reciprocal lattice. The substitution of (9) into (3) yields

$$\frac{\mathrm{d}A_2}{\mathrm{d}z} = -\mathrm{i}\beta_2 A_{10}^2 \sum_{m=-\infty}^{\infty} g_m \exp[\mathrm{i}(\Delta k_2 - mK)z]. \qquad (9)$$

The term, for which $\Delta k_2 = mK$, makes the maximum contribution to the right-hand side of expression (9). It is in this case that the phase mismatch is compensated by the vector of a reciprocal lattice in the mth order.

The above approach is applicable when the so-called nonlinear length is $L_{\rm nl} = (\beta_2|A_{10}|)^{-1} \gg l_c$. If $L_{\rm nl} \simeq l_c$, we can use the undepleted intensity approximation, which is valid for the second-harmonic conversion efficiency up to 0.5 [24]. Note also that approximate analytic methods cannot correctly describe the behaviour of the phase relations between interacting waves in RDS crystals [25, 26]. The phase relations undergo oscillations in such crystals, whereas the phases of the interacting waves in homogeneous nonlinear media change monotonically.

The use of quasi-phase matching nonlinear interactions in nonlinear optics offers some advantages over the use of phase-matched interactions realised due to birefringence in crystals. In the case of quasi-phase-matched interactions, the influence of birefringence on the efficiency of the nonlinear optical process can be excluded, while in the case of the degenerate three-frequency interaction, the condition of group phase matching can be realised [26].

In aperiodically polarised nonlinear crystals, the compression of ultrashort light pulses can be also fulfilled. Thus, in Ref. [27], the duration of a frequency-doubled chirped pulse in $LiNbO_3$ crystals with the aperiodic structure was shortened by a factor of 150. The periodic modulation of nonlinear optical coefficients is accompanied by the modulation of electrooptical coefficients, which can eliminate the influence of the photorefractive effect on nonlinear processes [28]. Note that in the case of RDS crystals, the conditions of optimal focusing upon the second harmonic generation drastically change compared to those for homogeneous crystals [29].

However, the most important from the practical point of view is the fact that upon quasi-phase-matched interactions one can use the maximum nonlinearity coefficient by choosing polarisations of the interacting wave in an appropriate way. For example, in a periodically polarised lithium niobate crystal, the ee – e interaction is used (all the waves are extraordinary). This interaction is determined by the component d_{33} of the nonlinear susceptibility, which exceeds other components in this crystal by an order of magnitude (Fig. 2).

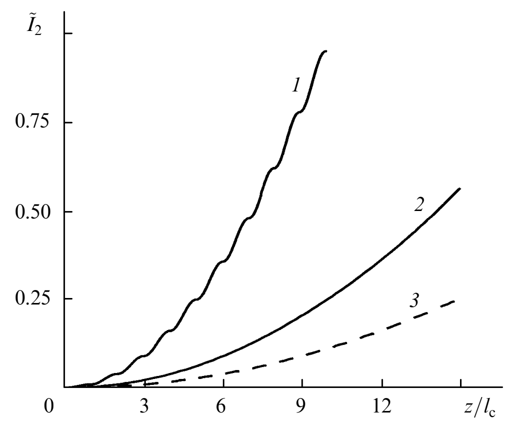

Figure 2. Dependences of the relative intensity $\tilde{I}_2$ of the second harmonic in a $LiNbO_3$ crystal on the reduced interaction length z/l_c for the first-order quasi-phase-matched ee-e interaction (*1*), and phase-matched oo – e (*2*) and oe – e (*3*) interactions.

At present, conventional phase-matched interactions of light waves are widely used for the second harmonic generation, generation of sum and difference frequencies, and parametric generation of light (see, for example, reviews [30, 31]). In this way, coherent radiation in the spectral range from the IR to UV region is produced. In these experiments, $LiNbO_3$, KTP, $LiTaO_3$, and RTA crystals with RDS structures are used.

Quite recently, in Refs. [32, 33], the quasi-phase-matched frequency doubling was obtained in actively nonlinear RDS crystals, i.e., along with lasing, the frequency doubling of this emission was observed.

3. Nonlinear optical media with a regular domain structure

The main feature of RDS crystals is the periodic change in the direction of the crystal polar axis resulting in the modulation of nonlinear properties characterised by a periodic change in the sign of nonlinear susceptibility (Fig. 1a). The modulation of the sign of nonlinear susceptibility and, correspondingly, nonlinear coupling coefficients of the interacting waves from layer to layer produces a 'nonlinear' grating. In the quasi-phase-matched process, the phase mismatch of the interacting waves is compensated due to the vector of a reciprocal lattice.

At present, several methods are used for the formation of RDSs in nonlinear media: the diffusion method, the after-growth method (the action of an alternate electric field on a crystal that is pulled through a furnace with the temperature gradient), the 'high-voltage' method, the electron beam method, and the growth method.

The method of chemical diffusion consists in the following. A crystal with a periodic mask applied by the lithography technique is placed in a medium whose reagents diffuse into the crystal producing the concentration gradient in a near-surface layer and the inversion of domains. As a result, a high-quality periodic structure with a period of 3 – 8 μm and 1 – 2 μm in depth appears [34, 35].

Another method for producing RDSs consists in the aftergrowth electrothermal processing when a crystal is placed in an alternate electric field during its pulling through a furnace with the temperature gradient. This method can be applied for producing bulk structures with a minimum period of a few tens of micrometers.

The crystal polarisation switching at room temperature caused by an electric field produced by periodic electrodes deposited on the crystal surface is a comparatively new method [36]. This method allows one to manufacture structures with a minimum period of 1.7 μm. However, it can be applied only for polarisation switching in thin samples of thickness from 200 to 500 μm. A substantial drawback of the methods discussed above is a small thickness of the structures obtained, which precludes the possibility of noncoplanar nonlinear optical interactions.

There also exists the method of electron beam polarisation switching in which the local inversion of domains is produced under the action of an electron beam on the crystal surface [37, 38]. This method allows one to produce domains of thickness 500 μm.

The method for producing bulk RDSs directly during the crystal growth is promising. The growth layered domain structure is inherent in a number of ferroelectrics grown by the Czochralski method [39 – 41]. This structure repeats the so-called growth banded structure – a growth defect, which appears because of oscillations in the growth rate and represents local variations in the crystal chemical composition.

The growth rate can be modulated, for example, by periodically changing temperature at the crystal growth front. The obvious advantage of the bulk RDS produced is its large size, whereas its disadvantage is the period instability, which restricts the effective length of nonlinear optical interactions by several millimetres. The growth method permits the formation of RDSs with flat and thin boundaries with periods varying in a broad range. The volume of a crystal containing RDSs can reach a few cubic centimetres.

The features of formation of a periodical structure by modulating crystal chemical composition during the crystal growth by the Czochralski method have been considered in Refs [42, 43]. When the symmetry axis of the thermal field does not coincide with the rotation axis of the crystal, the temperature at the crystal growth front changes periodically, resulting in the modulation of the crystal chemical composition. Such an inhomogeneous crystal composition results in the formation of the so-called rotational growth bands. During cooling, which is accompanied by the transition through the Curie point, ferroelectric domains attach to the growth bands. Their shape corresponds to that of the crystallisation isotherm.

The domain boundaries in crystals grown in the directions of axes X, Y, Z are curved because the growth front is not planar in this case, which is a drawback of the method. To grow a structure with plane boundaries, crystals are sometimes grown along the normal to a closely packed crystal face (the so-called face crystal) (Fig. 3b). Along with smooth and plane domain walls, the 'face' structure possesses a high degree of periodicity and has no such defects as microdomains and monodomains.

However, in this case, the direction of spontaneous polarisation makes an angle of 33° with domain walls, which leads to the presence of a coupled charge on them, resulting in a jump in the linear refraction coefficient. Such jumps can cause phase interruption of the waves involved in nonlinear optical interaction and decrease its efficiency. This feature is absent in crystals grown along the X-axis (Fig. 3a) because the direction of spontaneous polarisation in them is parallel to domain walls, while the role of the linear refractive index grating is insignificant.

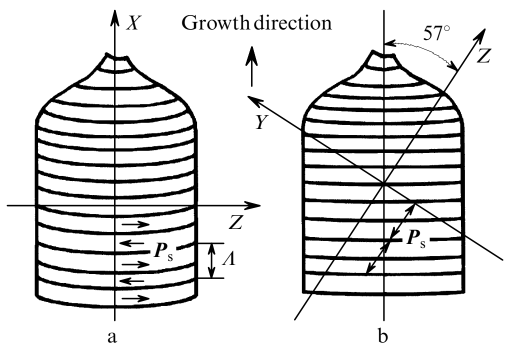

Figure 3. Schematic views of a crystal grown along the X axis (a) and a 'face' crystal (b) [43].

Organic polymers are promising materials for parametric conversion of light and generation of harmonics. The main advantage of these materials is the large second-order nonlinearity, which is a few times larger than that in conventional crystals. The polarisation switching and quasi-phase-matched second harmonic generation have been observed in Ref. [44].

The producing of ideal semiconductor structures with the crystal-axis orientation that periodically changes along the propagation of radiation attracts great interest in the last years [45, 46]. Such semiconductors are transparent in the range between 1 and 12 μm, they have the large nonlinear coefficient $d_{14} = 90$ pm V^{-1} (whereas the maximum nonlinear coefficient of $LiNbO_3$ is $d_{33} = 34$ pm V^{-1}), and have a high threshold of the optical breakdown. This allows one to use such semiconductor structures for quasi-phase-matched conversion of radiation in the near- and middle-IR ranges [47, 48].

4. Simultaneous realisation of two quasi-phase-matched processes. Interactions of co-propagating and counter-propagating waves

Let us analyse first the interaction of co-propagating waves. Consider two collinear processes (2), which proceed with the phase mismatches

$$\Delta k_3 = k_3 - k_2 - k_1, \quad \Delta k_2 = k_2 - 2k_1. \tag{10}$$

Under the conditions of simultaneous quasi-phase matching, it is necessary that

$$\Delta k_3 = 2\pi m_2/\Lambda, \quad \Delta k_2 = 2\pi m_1/\Lambda, \tag{11}$$

where m_1 and m_2 are quasi-phase matching orders. Expressions (11) can be reduced to the form

$$\Lambda = \frac{m_1\lambda}{3n(3\omega) - 2n(2\omega) - n(\omega)} = \frac{m_2\lambda}{2[n(2\omega) - n(\omega)]}, \tag{12}$$

where $\lambda = 2\pi c/\omega$ and $n(j\omega)$ is the refractive index for the wave with the frequency $j\omega$. For $m_1 = m_2$, it follows from (12) that

$$4n(2\omega) = 3n(3\omega) + n(\omega). \tag{13}$$

The corresponding expressions can be readily obtained for $m_1 \neq m_2$.

The fulfilment of expressions (11) was demonstrated for the first time for the waves propagating at an angle of 90° to the optic axis of a $LiNbO_3$ crystal [11]. The condition (13) is valid, for example, for $\lambda = 0.355$ μm, the modulation period $\Lambda = 8.2$ μm, and the crystal temperature equal to 24.5°C.

Another way to obtain relations (11) is a variation of the angle between the crystal optic axis and the normal to the modulation grating of the nonlinear susceptibility. This angle can be called the quasi-phase matching angle [15]. The tuning quasi-phase matching curves for the interaction of co-propagating waves are presented in Fig. 4. The curves are constructed for the case when the wave with frequency ω has ordinary polarisation, whereas the waves with frequencies 2ω and 3ω have extraordinary polarisation. Note that the waves in the short-wavelength region require higher orders of quasi-phase matching. Note also that the tuning range proves to be substantially narrower for the waves with the same polarisation.

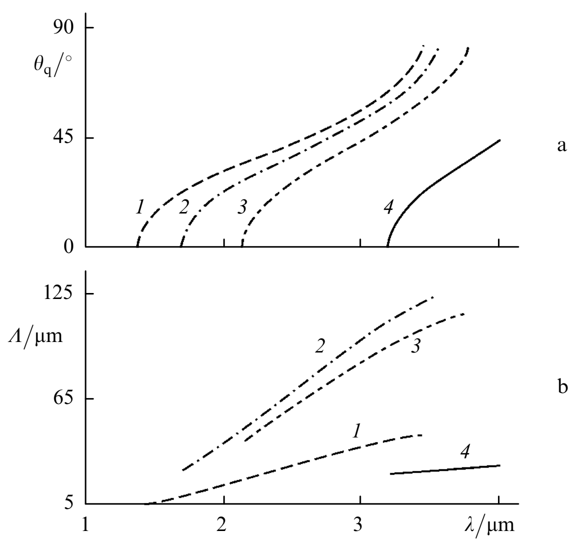

Figure 4. Dependences of the quasi-phase matching angle θ_q (a) and the modulation period Λ (b) on the wavelength $\lambda = 2\pi c/\omega$ for the interaction of co-propagating waves with frequencies $\omega, 2\omega$, and 3ω in a Mg : Y : *LiNbO*$_3$ crystal for quasi-phase matching orders $m_1 = 1$, $m_2 = 3$ (*1*), $m_1 = 3$, $m_2 = 7$ (*2*), $m_1 = 3$, $m_2 = 5$ (*3*), and $m_1 = m_2 = 1$ (*4*).

The conditions of quasi-phase matching for processes (2) in a $LiNbO_3$ crystal can be realised not only for the consecutive interaction of co-propagating waves but also for the interaction of counter-propagating waves. This requires the fulfilment of the following conditions [21]

$$\pm k_3 \mp k_2 \mp k_1 = 2\pi m_2/\Lambda, \quad \pm k_2 \mp 2k_1 = 2\pi m_1/\Lambda. \tag{14}$$

The upper and lower signs in (14) refer to the waves propagating in the positive and negative directions along the z axis, respectively. At least one of the interacting counter-propagating waves should be backward. For the backward wave with frequency ω, the relations (14) can be simultaneously satisfied when $m_1 = 2m_2$ and $n(2\omega) = n(3\omega)$. In the case of the backward wave with frequency 2ω, the conditions (14) are fulfilled for $m_1 = -m_2$ and $n(\omega) = n(3\omega)$.

When the frequency of the backward wave is equal to 3ω, the conditions (14) are satisfied if the degenerate three-frequency process proceeds synchronously $[n(\omega) = n(2\omega)]$, while the mixing process occurs quasi-synchronously. In this case, the quasi-phase matching order is $m_2 = -3[n(\omega) + n(3\omega)] \times\Lambda/\lambda$ ($|m_2| \gg 1$, because usually $\Lambda \gg \lambda$).

5. Consecutive three-frequency interactions of the waves with multiple frequencies

The three-frequency processes under study (2) are described in the general case by the following truncated equations [21]

$$\pm\frac{dA_1}{dz} = -i\beta_3 g(z) A_3 A_2^* \exp(-i\Delta k_3 z) - i\beta_2 g(z) A_2 A_1^* \exp(-i\Delta k_2 z),$$
$$\pm\frac{dA_2}{dz} = -2i\beta_3 g(z) A_3 A_1^* \exp(-i\Delta k_3 z) - i\beta_2 g(z) A_1^2 \exp(i\Delta k_2 z), \tag{15}$$
$$\pm\frac{dA_3}{dz} = -3i\beta_3 g(z) A_1 A_2 \exp(i\Delta k_3 z),$$

where A_j is the complex amplitude of the wave with frequency $j\omega$ ($j = 1, 2, 3$) and β_2 and β_3 are the moduli of nonlinear coupling coefficients of the waves. The coefficients β_3 and β_2 are related to the nondegenerate and degenerate three-frequency interaction, respectively.

The system of equations (15) can be solved in the general form only numerically. The relation between the wave intensities $I_j = |A_j|^2$

$$\pm I_3^{\pm}(z) \mp I_2^{\pm}(z) \mp I_1^{\pm}(z) = \text{const} \tag{16}$$

is useful for the understanding of the features of processes considered below. The upper index $+(-)$ is related to the wave propagating in the positive (negative) direction along the z-axis.

The relation (16) represents the law of conservation of the difference of the intensities of co- and counter-propagating waves. In the case of the conventional nondegenerate three-frequency process ($\beta_2 = 0$), along with relation (16), the another relation

$$\pm 2I_1^{\pm}(z) \mp I_2^{\pm}(z) = \text{const} \qquad (17)$$

can be readily obtained. Expressions (16) and (17) are in fact the Manly – Row relations for the process under study. It follows from these expressions that pump, for example, at the frequency 3ω is transformed to a co-propagating wave with the frequency ω and the intensity conversion coefficient equal to $1/3$. Such restrictions in the case of consecutive interactions are absent because the relation (17) is not satisfied in this case. In this respect, consecutive interactions fundamentally differ from conventional three-frequency interactions.

Comparison of the energy properties of processes described by equations (15) with conventional three-frequency processes suggests that the term 'consecutive interactions' [1] adequately reflects the essence of these interactions. Note that at present the term 'consecutive interactions' is used, for example, in the analysis of the spectral enrichment of a shock wave in weakly dispersing media [49].

The nature of energy exchange between interacting waves is determined, of course, by initial conditions at the input and output of a nonlinear crystal:

$$A_j(z=0) = A_{j0}, \quad A_j(z=L) = A_{jL}.$$

One can distinguish three types of consecutive interactions between co-propagating waves:

(1) The generation of higher harmonics:

$$A_{10} \neq 0,\ A_{20} = A_{30} = 0;$$

(2) the frequency down-conversion:

$A_{30} \neq 0,\ |A_{30}| \gg |A_{10}|, |A_{20}|$; and

(3) the parametric amplification at low-frequency pump:

$$A_{20} \neq 0,\ \ |A_{20}| \gg |A_{10}|, |A_{30}|.$$

Below, we consider the dynamics of energy exchange between interacting waves for these processes. Note that the phase relations between the interacting waves have a complicated and irregular nature [15]: the phase jumps are observed in the region of a strong energy exchange, and when the phase relations at the input of a periodically inhomogeneous medium are not optimal, they do not tend to a stable value inside the medium.

5.1. Third harmonic generation

Consider first the consecutive quasi-phase-matched third harmonic generation. When an intense wave of frequency ω is incident on a quadratically nonlinear crystal, first a wave with frequency 2ω is generated in the crystal. At the next stage, provided the corresponding quasi-phase matching conditions are satisfied, the waves with frequencies ω and 2ω excite a wave with frequency 3ω. As a result, the energy of the wave with frequency ω is converted to that of the third harmonic [14, 15]. The latter is absent at all upon conventional direct frequency multiplication in homogeneous quadratically nonlinear crystals.

Fig. 5 shows the intensity of the third harmonic upon its consecutive quasi-phase-matched excitation for different ratios $r = m_1\beta_3/m_2\beta_2$ of the effective nonlinear coupling coefficients of the waves. Note first of all that the pump wave phase, as in homogeneous media, does not affect the coefficient of conversion into the third harmonic. At the same time, the maximum efficiency of conversion to the third harmonic strongly depends on the ratio of the nonlinear coupling coefficients of the waves and quasi-phase matching orders used.

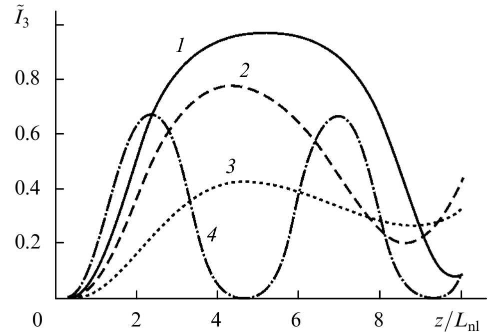

Figure 5. Dependences of the relative intensity $\tilde{I}_3$ of the third harmonic on the reduced length $z/L_{\rm nl}$ for the ratio of effective nonlinearity coefficients $r = 0.66$ (*1*), 0.5 (*2*), 0.3 (*3*), and 1.0 (*4*).

The dynamics of energy exchange between the waves for the optimal ratio of nonlinear coefficients ($r \approx 0.67$) is shown in Fig. 6. Here, the pump-wave energy is almost completely transformed to that of the third harmonic. Note that in this case, unlike direct excitation of the third harmonic in a medium with cubic nonlinearity, the self-action and cross interaction are absent. It is for this reason, that the 100-% conversion efficiency of the third harmonic can be achieved upon its consecutive generation.

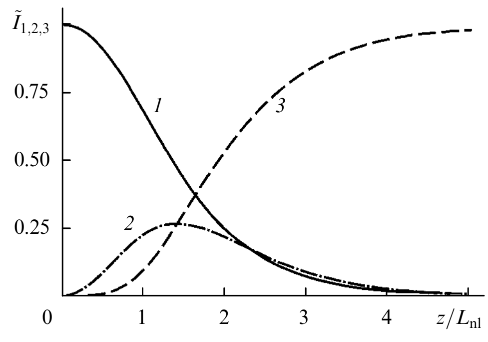

Figure 6. Dependences of the relative intensities $\tilde{I}_1$ (*1*), $\tilde{I}_2$ (*2*), and $\tilde{I}_3$ (*3*) of the waves with frequencies ω, 2ω, and 3ω, respectively, on $z/L_{\rm nl}$ upon the third harmonic generation for the optimal ratio of effective nonlinearities $r = 0.67$.

The nonlinear length $L_{\rm nl}$ in experiments with lithium niobate crystals can be a few centimetres.

5.2. Third subharmonic generation

This process is one of the processes of frequency down-conversion upon pumping at frequency 3ω. The dynamics of conversion of an intense wave of frequency 3ω to the wave with frequency ω is unstable [14, 15]. If the phase relations at the input to a nonlinear medium are not optimal for this interaction, the intensity of the waves oscillates. Upon consecutive generation of the third subharmonic, there exist the optimal initial phase relation between the pump and subharmonic and the optimal relation between the effective nonlinear coefficients r for the complete pump energy transfer to the subharmonic wave energy. The optimal value of r is the same as that for the third harmonic generation ($r = 0.67$).

The energy transfer from the wave with frequency 3ω to the wave with frequency ω occurs most efficiently when the initial phases of the interacting waves are $\varphi_{20} = \varphi_{30} = 0$ and the initial phase of the pump wave is $\varphi_{10} = \pi/2$. This optimal phase relation differs from that for homogeneous media: in a homogeneous medium with the cubic nonlinearity there are several optimal phases φ_{10} that differ by $2\pi/3$ in the case of the third subharmonic generation [51]. Fig. 7 shows the behaviour of the intensities of interaction waves for optimal conditions of the third subharmonic generation in a RDS crystal (the number of medium layers on the nonlinear length is $N = 10$). Weak oscillations in the curves are caused by high orders of quasi-phase matching: $m_1 = 3$, $m_2 = 5$. Let us emphasise once more that, unlike a medium with cubic nonlinearity, where energy transfer to the third subharmonic is also possible, the process under study involves the quadratic nonlinearity.

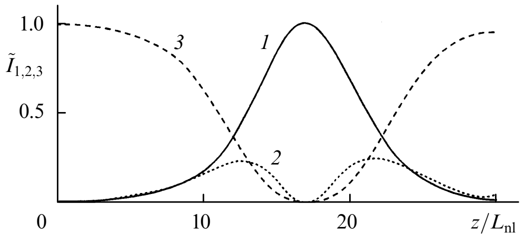

Figure 7. Dependences of the relative intensities $\tilde{I}_1$ (*1*), $\tilde{I}_2$ (*2*), and $\tilde{I}_3$ (*3*) of the waves with frequencies ω, 2ω, and 3ω, respectively, on $z/L_{\rm nl}$ upon the third subharmonic generation for $A_{30} = 1$, $A_{10} = {\rm i}0.1$, and $A_{20} = 0.1$.

5.3. Parametric frequency conversion

Consider now energy conversion from the wave of frequency 3ω to the wave of frequency 2ω. This process has been studied in detail in homogeneous media in Refs [10, 16, 17]. In Ref. [10], it was shown for the first time that the energy of an intense wave with frequency 3ω can be completely converted to that of the wave with frequency 2ω. It was found in Ref. [14] that in spite of the quasi-phase-matched type of interactions in RDS crystals, the behaviour of plane waves (for a given interaction length) with increasing number of layers tends to that for a homogeneous medium.

Fig. 8 shows the dependences of the wave intensities on a spatial coordinate in the consecutive quasi-phase-matched process under study for the optimal initial phase relation of the waves, $\beta_2/\beta_3 = 1$, $m_1 = m_2 = 1$ and the number of layers $N = 30$ on the nonlinear length $L_{\rm nl}$, where $L_{\rm nl} = 1/\beta_3|A_{30}|$.

The energy transfer to the wave with frequency 2ω in a RDS crystal, unlike the case of a homogeneous medium, is not aperiodic and can be highly efficient when the number of layers is limited. In principle, the 100-% conversion efficiency can be achieved when the number of layers is very great.

5.4. Parametric amplification at low-frequency pump

Consider now the quasi-phase-matched parametric amplification of a wave with frequency 3ω in the pump-wave field

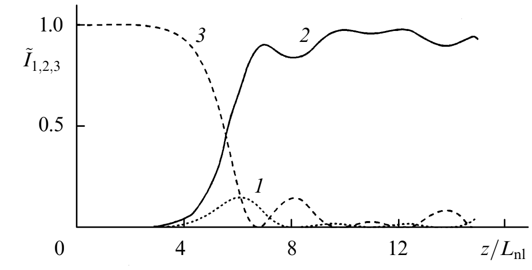

Figure 8. Dependences of the relative intensities $\tilde{I}_1$ (*1*), $\tilde{I}_2$ (*2*), and $\tilde{I}_3$ (*3*) of the waves with frequencies ω, 2ω, and 3ω, respectively, on $z/L_{\rm nl}$ upon conversion of the energy of the wave with frequency 3ω to that of the wave with frequency 2ω for $A_{30} = -1$, $A_{10} = 0.01$, and $A_{20} = 0$.

with frequency 2ω. Its dynamics depends on many parameters such as the ratio of intensities of the pump wave at frequency 2ω and the signal wave, the phase relations between the interacting waves, the ratio of the effective nonlinear coefficients, etc. [13 – 15]. The parametric amplification at low-frequency pump in a homogeneous medium has been studied in Refs [2, 5, 6]. The efficient conversion of the pump-wave energy with frequency 2ω to the energy of the wave with frequency 3ω in a periodically inhomogeneous nonlinear medium has been first demonstrated in Ref. [13].

Figs 9 – 11 show the dynamics of energy transfer upon parametric amplification. The energy exchange between the waves has the oscillatory nature: once the pump energy at frequency 2ω has been almost completely converted to the energy of the wave with frequency 3ω, the reverse energy transfer to the pump wave begins. Fig. 9 shows the behaviour of the waves upon frequency up-conversion ($A_{10} \neq 0$, $A_{30} = 0$) in the case of the efficient interaction for the optimal initial phase relation $\beta_2/\beta_3 = 1$, $m_1 = m_2 = 1$ and the number of layers on the nonlinear length $N = 10$ (here, $L_{\rm nl} = 1/\beta_2|A_{20}|$).

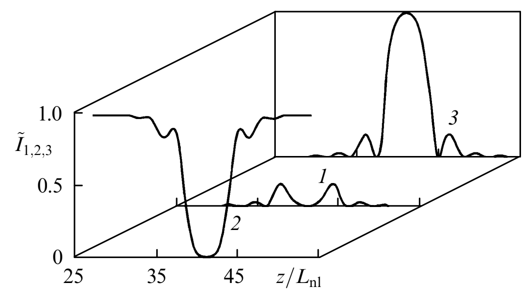

Figure 9. Dependences of the relative intensities $\tilde{I}_1$ (*1*), $\tilde{I}_2$ (*2*), and $\tilde{I}_3$ (*3*) of the waves with frequencies ω, 2ω, and 3ω, respectively, on $z/L_{\rm nl}$ at the parametric amplification in the low-frequency pump field with frequency 2ω for $A_{20} = 1$, $A_{10} = 4.27 \times 10^{-5}$, and $A_{30} = 0$.

In the case of parametric amplification at low-frequency pump, i.e., for $A_{30} \neq 0$ and $A_{10} = 0$, the maximum conversion efficiency at frequency 3ω is achieved at larger interaction lengths than at frequency up-conversion [14]. This is explained by the fact that in a nonlinear medium the wave at the difference frequency ω is first excited and then the degenerate parametric amplification ($2\omega = \omega + \omega$)) and up-conversion ($\omega + 2\omega = 3\omega$) occur.

The dynamics of the waves at the initial stage of the interaction depends on the ratio of moduli β_2 and β_3, which are responsible for the conventional high-frequency parametric amplification and frequency mixing, and on the initial phase relation. In the undepleted pump-wave field approximation and assuming that a nonlinear medium is homogeneous, a change in the amplitude of the wave with frequency 3ω is described by the expression (cf. section 6)

$$A_3(z) = \sum_{j=1}^{2} B_j \sinh \Gamma_j z + C_j \cosh \Gamma_j z, \tag{18}$$

where B_j and C_j are specified by the initial conditions for $z = 0$ [13] and

$$\Gamma_{1,2} = \frac{1}{2}\left[\beta_2 \pm \left(\beta_2^2 - 12\beta_3^2\right)^{1/2}\right]|A_{20}| \tag{19}$$

is the increment of the amplitude increase. One can see that for $\beta_2 \sim \beta_3$, the real part of $\Gamma_{1,2}$ is determined by the coefficient β_2, which is responsible for the degenerate parametric amplification at high-frequency pump. Note that the structure of expression (18) is typical for parametric ampification at high-frequency pump.

By varying the parameters of signal waves with frequencies ω and 3ω at the input to a periodically modulated nonlinear medium, one can substantially change the dynamics of energy exchange between the waves. Figs 10 and 11 show the dynamics of a parametrically amplified signal as a function of the ratio of the intensities and phases of the input signals. The curves were constructed for $N = 10$, $m_1 = m_2 = 1$ and $\beta_2/\beta_3 = 1$. One can see that the dynamics of this process can be controlled by varying the signal parameters at the input to a nonlinear medium. A weak change in the initial amplitude or phase of a signal can result either in a complete energy conversion to the energy of the wave with frequency 3ω at the given length in the medium or in the absence of a signal at this frequency, i.e., the optical switching from one frequency to another can be completely performed. The dependence of the signal intensity on these parameters exhibits also a certain periodicity (see [16]).

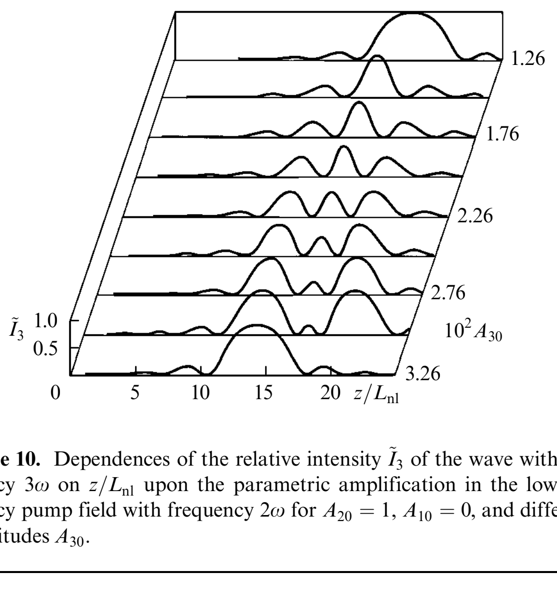

Figure 10. Dependences of the relative intensity $\tilde{I}_3$ of the wave with frequency 3ω on z/L_{nl} upon the parametric amplification in the low-frequency pump field with frequency 2ω for $A_{20} = 1$, $A_{10} = 0$, and different amplitudes A_{30}.

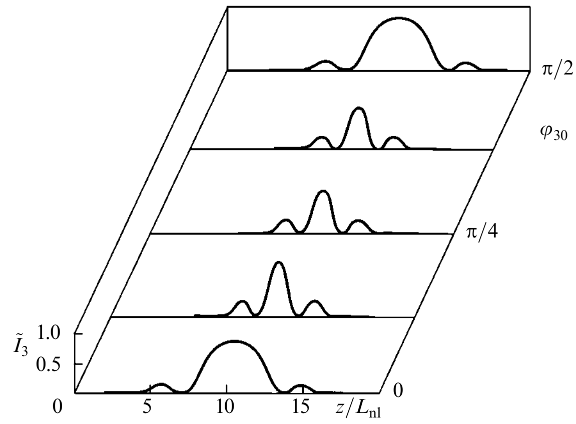

Figure 11. Dependences of the relative intensity $\tilde{I}_3$ of the wave with frequency 3ω on z/L_{nl} upon the parametric amplification in the low-frequency pump field with frequency 2ω for $A_{20} = 1$, $A_{10} = 0$, $|A_{30}| = 3.26 \times 10^{-2}$, and different phases φ_{30}.

5.5. Interactions of counter-propagating waves

Consider now the quasi-phase-matched parametric amplification for the case of the low-frequency backward pump wave [21], i.e., when the pump wave with frequency 2ω comes on the crystal output ($z = L$). In this case, we obtain from (16) the relation

$$I_1^+(0) + I_3^+(0) - I_2^-(0) = I_1^+(L) + I_3^+(L) - I_2^-(L)\,. \tag{20}$$

for the intensities of the interacting waves. In the absence of a signal at the medium input ($I_3^+(0) = 0$), we obtain from (20)

$$I_3^+(L) = I_2^-(L) - I_1^+(L) + I_1^+(0) - I_2^-(0). \tag{21}$$

If the conditions $I_1^+(L) = 0$ and $I_1^+(0) \simeq I_2^-(0)$ are satisfied, the energy of the wave with frequency 2ω converts almost completely to that of the wave with frequency 3ω. This is illustrated in Fig. 12 where the results of calculations are given for $N = 500$ at the nonlinear length $L_{nl} = 1/\beta_3|A_{20}|$ and $m_2 = 1$. One can see that during propagation of the wave with frequency 2ω in a nonlinear medium, the energy of the wave converts to that of the wave with frequency 3ω at rather weak input intensities of the waves at frequencies ω and 3ω. In accordance with Eqn (21), the efficient energy exchange

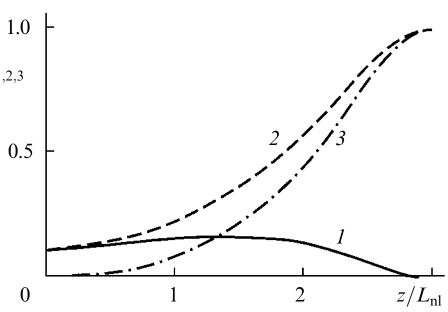

Figure 12. Dependences of the relative intensities $\tilde{I}_1$ (*1*), $\tilde{I}_2$ (*2*), and $\tilde{I}_3$ (*3*) of the waves with frequencies ω, 2ω, and 3ω, respectively, on z/L_{nl} upon the parametric amplification of counter-propagating waves in the low-frequency pump field with frequency 2ω. The wave with frequency 2ω propagates toward the waves with frequencies ω and 3ω.

between counter-propagating waves with frequencies 2ω and 3ω can also takes place when $I_2^-(0) = 0$ and $I_1^+(L) \simeq I_1^+(0)$.

It follows from analysis [21] of the parametric amplification at low-frequency pump that a complete energy conversion from the wave with frequency 2ω to the counter-propagating wave with frequency 3ω occurs at smaller lengths than in the case of co-propagating waves, and the role of phase relations is greater in the former case. In such processes, unlike the interaction of co-propagating waves, it is impossible, for example, to convert completely the energy of an intense wave with frequency 3ω to that of the wave with frequency 2ω.

6. Generation of nonclassical light at parametric amplification in the low-frequency pump field

The conventional three-frequency processes of parametric amplification in the high-frequency pump field represent sources of nonclassical or squeezed light (see, for example, [50 – 54]). The specific features of using quasi-phase-matched processes for generation of squeezed light were considered in review [55]. At present, the applications of squeezed light in various precision optical and physical measurements and in systems for optical data communication and processing are extensively studied.

The parametric amplification of light upon low-frequency pump can be used to produce nonclassical light whose properties differ from those of the light obtained upon parametric amplification in the low-frequency pump field. Let us analyse the quantum properties of the light generated upon parametric amplification during the interaction of co-propagating waves in the low-frequency pump field. We will follow paper [56], assuming that the conditions of quasi-phase matching are satisfied, whereas a nonlinear medium is nevertheless homogeneous.

In the approximation of a undepleted classical low-frequency pump field, this parametric process is described by the expressions

$$\frac{\mathrm{d}a_1}{\mathrm{d}z} = -\mathrm{i}\varkappa_3^* a_3 - \mathrm{i}2\varkappa_2 a_1^+,$$
$$\frac{\mathrm{d}a_3}{\mathrm{d}z} = -\mathrm{i}\varkappa_3 a_1, \qquad (22)$$

where $a_j(z)$ and $a_j^+(z)$ are the operators of creation and annihilation of a photon with frequency $j\omega$, which obey the commutation relations $[a_j, a_k^+] = \delta_{jk}$ and $[a_j, a_k] = 0$; δ_{jk} is the Kronecker symbol; $\varkappa_j = \gamma_j D_2$; γ_2 and γ_3 are effective nonlinear coupling coefficients; and D_2 is the classical amplitude of the pump wave.

A solution of equations (22) has the form that is similar to expression (18), in which complex amplitudes should be replaced by operators (see [56]). Note that this solution is also similar to the solution obtained in the case of the quantum description of a nonlinear asymmetric coupler of the waves with frequencies ω and 2ω upon intense pump at the second-harmonic frequency [57].

It was shown in section 5.4 that upon parametric amplification in the low-frequency pump field with frequency 2ω, photons with frequency ω are first generated, which are added with pump photons to produce photons with frequency 3ω. The first process represents the degenerate three-frequency parametric amplification at low-frequency pump in which nonclassical (quadrature-squeezed light) is generated at frequency ω [51 – 54]. For this reason, the field at frequency 3ω in the second process proves to be in a nonclassical state.

Consider the behaviour of fluctuations of the quadrature components at frequencies ω and 3ω:

$$X_j(\theta_j) = a_j \exp(\mathrm{i}\theta_j) + a_j^+ \exp(-\mathrm{i}\theta_j),$$
$$Y_j(\theta_j) = \mathrm{i}[a_j \exp(\mathrm{i}\theta_j) - a_j^+ \exp(-\mathrm{i}\theta_j)] \quad (j = 1, 3), \qquad (23)$$

where θ_j is the phase of the heterodyne wave, which is used for measuring the jth quadrature component. The operators $X_j(\theta_j)$ and $Y_j(\theta_j)$ satisfy the commutation relations $[X_j(\theta_j), Y_j(\theta_j)] = -2\mathrm{i}$.

Analysis showed [56] that the dynamics of quadratures depends on many parameters of the problem. In particular, it was found that the fluctuations of quadratures X_1 and X_3 decrease during the interaction of the waves under the conditions

$$3\theta_1 = \theta_3, \quad \varphi_2 + 2\theta_1 = -\pi/2, \qquad (24)$$

where $\varphi_2 = \arg D_2$. In this case, the quadrature components are determined by the expressions

$$X_1(z, \theta_1) = K_1(z) X_1 + K(z) X_3,$$
$$X_3(z, \theta_3) = -K(z) X_1 + K_3(z) X_3, \qquad (25)$$

where

$$X_j = X_j(z = 0);$$
$$K_1(z) = \left(\cosh\gamma z - \frac{|\varkappa_2|}{\gamma}\sinh\gamma z\right)\exp(-|\varkappa_2| z);$$
$$K_3(z) = \left(\cosh\gamma z + \frac{|\varkappa_2|}{\gamma}\sinh\gamma z\right)\exp(-|\varkappa_2| z);$$
$$K(z) = \frac{|\varkappa_3|}{\gamma}\exp(-|\varkappa_2| z)\sinh\gamma z;$$
$$\gamma = \left(|\varkappa_2|^2 - |\varkappa_3|^2\right)^{1/2}.$$

The function $K(z)$ is related to the mutual influence of fluctuations at the generated frequencies. For the initial fields at frequencies ω and 3ω in the vacuum or coherent state, the dispersion of quadratures is

$$V_j = \langle X_j^2 \rangle - \langle X_j \rangle^2 = Q_j(z), \qquad (26)$$

where

$$Q_{1,3}(z) = \frac{1}{\gamma^2}\left(|\varkappa_2|^2\cosh 2\gamma z \mp |\varkappa_2|\gamma\sinh 2\gamma z - |\varkappa_3|^2\right)\exp(-2|\varkappa_2| z). \qquad (27)$$

The upper sign in (27) is related to $Q_1(z)$ and the lower one, to $Q_3(z)$. The quadrature dispersion at the input to a nonlinear medium is $V_0 = 1$ (the vacuum or coherent field).

For $|\varkappa_3| = 0$, i.e., upon the conventional parametric amplification in the low-frequency pump field, the function $Q_1(z) = \exp(-4|\varkappa_2| z)$ and $Q_3(z) = 1$. In this case, the dispersion of the quadrature $X_1(z)$ in a nonlinear medium is smaller than the amplitude of vacuum oscillations, whereas the dispersion of the quadrature $X_3(z)$ is equal to the amplitude of vacuum fluctuations. In accordance with the quantum-mechanical uncertainty relation, the dispersion of the quadrature Y_1, which is not discussed, exceeds the amplitude of vacuum fluctuations.

When $|\varkappa_3| \neq 0$, the parametric amplification takes place both upon low-frequency and high-frequency pump. In this case, because of the phase relations chosen, the dispersions of quadratures $X_1(z)$ and $X_3(z)$ in a nonlinear medium decrease, according to (26) and (27), whereas the dispersions of quadratures $X_1(z)$ and $X_3(z)$ obviously increase.

The behaviour of quadratures $X_1(z)$ and $X_3(z)$ in a nonlinear medium is demonstrated in Fig. 13 for different ratios $|\varkappa_3|/|\varkappa_2|$ of nonlinearities. One can see that the dispersions of quadratures $X_1(z)$ and $X_3(z)$ decrease with the distance propagated by the waves, i.e., the suppression of fluctuations of the quadratures is correlated. However, the fluctuations of the quadrature $X_3(z)$ are always greater than the fluctuations of the quadrature $X_1(z)$. The dispersion of the quadrature $X_3(z)$ decreases with increasing the nonlinearity coefficient $\varkappa_3$, whereas the dispersion of the quadrature $X_1(z)$ somewhat increases.

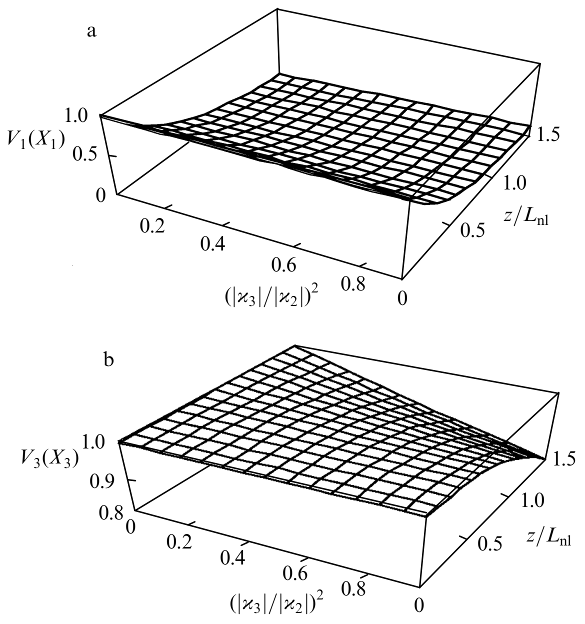

Figure 13. Dispersions $V_1(X_1)$ (a) and $V_3(X_3)$ (b) of quadrature components at frequencies ω and 3ω, respectively, for the parametric amplification of co-propagating waves in the low-frequency pump field with frequency 2ω as functions of z/L_{nl} and the ratio of nonlinearity coefficients $(|\varkappa_3|/|\varkappa_2|)^2$.

Thus, it is possible to suppress the quantum fluctuations of quadrature components at multiple frequencies in consecutive three-frequency processes coupled by common pump. In this case, a conventional degenerate parametric amplification is realised at one of the frequencies and a parametric amplification upon low-frequency pump is realised at another frequency. In the case of conventional method for generation of higher harmonics based on frequency mixing of coherent emission, no nonclassical light is generated at the sum frequency.

7. Experiments on generation of higher optical harmonics

Studies of consecutive interactions between light waves in nonlinear optics have been started comparatively recently and they are predominantly theoretical because of the difficulties encountered upon phase matching of two three-wave processes. However, due to the progress in the fabrication of structures with a periodic modulation of nonlinear susceptibility, experimental observations of consecutive interactions have already been reported in a few papers. These studies are concerned so far with the generation of higher optical harmonics using consecutive interactions [12, 18–20].

The authors [18] studied the third harmonic generation using the second harmonic generation ($\omega + \omega = 2\omega$) and the subsequent wave mixing ($\omega + 2\omega = 3\omega$) in the 9th and 33th quasi-phase matching orders, respectively. In experiments, a Y : $LiNbO_3$ crystal was used, which was grown by the Czochralski technique and had the RDS formed during its growth. The modulation period of the nonlinear susceptibility was 60 μm and the crystal length was 5 mm. The crystal was pumped by a 1.064-μm quasi-cw Nd : YAG laser. The laser had an average power of about 1 W, a pulse length of 100 ns, and a repetition rate of 1 kHz. The eee/eee interactions were realised, which allowed the use of the maximum nonlinear coefficient d_{33}.

The simultaneous generation of the second and third harmonics has been also obtained in Ref. [12]. The authors [12] used a 3.6-μm, 195-mW CO laser, whose radiation was focused on a lithium niobate crystal which was periodically polarised with a period of 31.5 μm. The efficiency of conversion to the second and third harmonics in the eee/eee interactions was 5.4×10^{-4} W^{-1} and no more than 10^{-6} W^{-2}, respectively.

The second and third harmonic generation upon the consecutive interaction of counter-propagating waves as a result of the second harmonic generation and the consecutive wave mixing was observed in papers [19, 20]. A $KTiOPO_4$ crystal waveguide of length 2.6 mm was repolarised with a period of 4 μm. The 9-ns pump pulse at 1.230 μm produced the intensity of about 16.2 GW cm^{-2} inside the waveguide. The second and third harmonic generation was achieved for $m = 24$ and 13, respectively. The efficiency of conversion to the third harmonic was about 0.4 %.

8. Conclusions

We considered a new class of nonlinear optical interactions - consecutive interactions of the waves with multiple frequencies. Such interactions can be realised in periodically inhomogeneous crystals, in particular, RDS crystals, owing to the fulfilment of quasi-phase matching conditions for two three-wave processes proceeding on the same 'nonlinear' grating.

Note that RDS crystals have uniform linear optical properties. They substantially differ in this respect from the so-called photonic crystals whose linear and nonlinear properties vary periodically in space.

Our detailed treatment of quasi-phase-matched interactions of the type (2) shows that the energy of the intense pump wave with frequency ω_p can be efficiently converted to the energy of the wave with one of the frequencies $\omega_p/3$, $2\omega_p/3$, $3\omega_p/2$ and $3\omega_p$.

In RDS crystals, consecutive interactions of the waves with multiple frequencies of the type [12, 15]

$$\omega + \omega = 2\omega, \quad 2\omega + 2\omega = 4\omega$$

can also occur. In this case, along with the well-known processes proceeding in a medium with the quadratic nonlinearity, the energy of the pump wave with frequency ω_p can be efficiently converted to the energy of the waves with frequencies $\omega_p/4$ or $4\omega_p$. To generate such waves in the case of direct conversion, the fourth-order nonlinearity is required. We emphasise that a highly efficient conversion of the pump radiation to radiation at other frequencies in the consecutive processes does not depend on these frequencies. In this respect, these processes substantially differ from conventional ones.

The experiments on the third harmonic generation performed upon consecutive quasi-phase-matched interactions is only the beginning of experimental studies of a new class of interactions in nonlinear optics. In this respect, the possibility of realisation of the parametric amplification upon low-frequency pump appears the most interesting in our opinion. At present it was shown theoretically that this process can occur upon the interaction of both co-propagating and counter-propagating waves. The parametric amplification upon low-frequency pump can be also of interest for generating nonclassical light. As shown in section 6, in this process, the quadrature-squeezed light is generated at frequencies above and below the pump frequency. In this case, the quantum fluctuations of quadratures with different frequencies prove to be correlated, which is important for various applications.

The question of the effect of random deviations from the periodicity is important for the efficient conversion in consecutive quasi-phase-matched interactions of the waves. On the other hand, it seems that stochastic consecutive interactions can be realised in statistically inhomogeneous media with a random spatial variation of the nonlinear coupling coefficient of the waves.

From the point of view of practical applications of consecutive interactions, the most important is undeniably the development of highly efficient laser radiation frequency converters. For example, using the consecutive parametric amplification at low-frequency pump by a 1.064-μm Nd:YAG laser, one can obtain a source of coherent radiation emitting simultaneously at three wavelengths of 1.064, 2.128, and 0.709 μm.

Another important application of the consecutive processes is their use in optical switches. This application is based on the phase sensitivity of these processes, when a change in the phase of one of the interacting waves results in substantial changes in the amplitudes of the waves involved in the process. A detailed study of these effects in consecutive quasi-phase-matched interactions is a subject of our further investigations.

The consecutive interactions should be distinguished from cascade quasi-phase-matched processes, which are realised using either two crystals located one after another and differing in the grating nonlinearity period [58] or two nonlinear gratings in one crystal [59].

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Note added in proof

Recently, a paper of Chao Zhang et al. was published (*Opt. Lett.* **25** 436 (2000)) in which the consecutive third harmonic generation in a homogeneous medium was theoretically studied. The authors have paid the main attention to the conditions of the efficient conversion of the fundamental radiation to the third harmonic. The ratio of the nonlinear coupling coefficients for the interacting waves at which the complete conversion occurs obtained by them coincides, with an accuracy to the notation, with the results obtained earlier in our papers [14, 15] (see section 5.1).

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