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NONLINEAR OPTICAL PHENOMENA

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Applied nonlinear optics in the journal 'Quantum Electronics'

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Abstract. **A brief historical review of the experimental and theoretical works on nonlinear optical frequency conversion (generation of harmonics, up- and down-conversion, parametric oscillation), which have been published in the journal 'Quantum Electronics' for the last 40 years, is presented.**

Keywords: *nonlinear frequency conversion of laser radiation, generation of harmonics, optical parametric interaction, thermal self-interaction, nonlinear crystals, nonclassical light generation.*

1. Introduction

The rapid development of laser physics, which began in the 1960s, was accompanied by intensive and extensive development of nonlinear optics, in particular, nonlinear optical laser frequency converters. This progress was caused by the need for coherent light sources with wavelengths from UV to mid-IR range and different parameters in various practical applications. THz radiation sources, which have been intensively developed in the last years, are also highly demanded. Nonlinear optical laser frequency converters solve actually the following problem: to cover the optical range using various radiation sources and obtain light fields with desired parameters (from single photons to superstrong light fields and from continuous radiation to attosecond pulses).

Fifty years have passed since the observation of the first nonlinear optical phenomenon, related to the conversion of carrier optical frequency at second-harmonic generation. Many theoretical and experimental studies devoted to nonlinear optical processes and their applications have been carried out during this period. The achievements in nonlinear optics have been described in a number of reviews and monographs. Of special note is the book [1], the Appendix of which presents the remarkable history of nonlinear optics.

About 700 papers devoted to the development of the theory of nonlinear optical frequency converters, their experimental realisation, and different applications have been published in the journal 'Quantum Electronics' for the last four decades. In this review on applied nonlinear optics, which is devoted to the 40th anniversary of the first issue of 'Quantum Electronics', we focused mainly on these studies. Thus, our review is in essence bibliographic. When considering a particular problem, we tried to focus on the works where it was first stated or, in our opinion, made a significant contribution to its solution. Obviously, we are far from considering all the works that are absent in the reference list as making no or little contribution to the nonlinear optics development. Also, we could not completely exclude references to publications in other journals. The works on stimulated Raman scattering, stimulated Brillouin scattering, phase conjugation, and self-focusing are beyond the scope of this review.

The review is organised as follows. First, the achievements in the theory of nonlinear optical frequency conversion and in the technique of laser frequency multipliers are presented. Then the works on nonlinear optical frequency conversion in the presence of thermal self-interaction are considered. Separate sections are devoted to the studies of nonlinear optical crystals and the analysis of the works on optical frequency conversion in periodically polsed nonlinear crystals (PPNCs), in which quasi-phase-matched wave conversions are implemented. A separate section considers lasers with intracavity generation of harmonics. It is followed by the analysis of works on the laser frequency conversion in gaseous media and on the generation of high-order harmonics. The parametric optical frequency conversion is discussed only briefly, although the number of studies devoted to this process is almost 20% of the total number of works on applied nonlinear optics. The review is ended with brief consideration of the studies of statistical characteristics of nonlinear optical processes.

2. Development of the theory and technique of optical frequency conversion

Some theoretical results were obtained at the beginning of the 1960s, which showed the fundamental possibility of reaching 100% efficiency of nonlinear optical frequency conversion in quadratically nonlinear media and the necessity of providing equal phase velocities of interacting waves (phase-matching condition [2, 3]) for obtaining this conversion coefficient. At the same time, it was clear that the dispersion of an isotropic medium does not make it possible to obtain equal phase velocities in it in the transparency window. In 1962 Giordmain [4] proposed to use anisotropic crystals. A possibility of providing the phase-matching condition was revealed in an experiment with a KDP crystal, and second-harmonic generation was implemented. The studies carried out by the mid-

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1960s showed that complete nonlinear optical conversion of laser radiation to another frequency cannot be obtained in real experiments. It was necessary to reveal the effects that manifest themselves at nonlinear conversion and the limitations to which they lead and determine the requirements to the parameters of laser radiation and nonlinear optical crystals. The efficiency of nonlinear frequency conversion was found to be determined by a set of spatial (angular), temporal (spectral), and energy laser characteristics and the nonlinear optical, dispersion, and dissipative properties of crystals.

The general theory of nonlinear conversion of laser beam frequency had been developed well by the beginning of the 1970s. This theory is based on the parabolic nonlinear wave equation, derived by the method of slowly varying amplitudes (Khokhlov method [5]). The diffraction effect in anisotropic media was described in [6], and the influence of diffraction at second-harmonic generation (SHG) by focused laser beams was taken into account in [7]. The role of the effect of light beam drift (spatial dispersion of refractive indices), caused by the birefringence of crystals under the phase-matching condition was studied in [8]. By that time the existence of phase matching had been established for different types of wave interactions and for different nonlinear optical processes (generation of harmonics, sum- and difference-frequency generation, parametric interactions). In addition, the limitations following from the crystal properties (angular, spectral, and temperature phase-matching widths) had been revealed. Due to this progress, conversion of pulsed laser radiation to the second harmonic with efficiency of more than 50% was experimentally obtained.

In the beginning of the 1960s, the sum-frequency generation [9] and the third-harmonic generation (THG) [10] were important experimental achievements. At that time, fourth-harmonic generation (FHG) of laser radiation was also realised [11]. To date, sixth-harmonic generation (at a wavelength of 177 nm) of Nd^{3+} laser radiation has been obtained at nonlinear optical frequency conversion in crystals, and radiation with a wavelength as short as 155 nm was produced upon sum-frequency generation. Radiation in the mid-IR range with a wavelength of several tens of micrometers was obtained in the regime of difference-frequency generation.

The theory of nonlinear optical frequency conversion was first developed in the plane-wave approximation for light beams and in the quasi-stationary approximation for pulsed radiation. Furthermore, with allowance for the mechanisms limiting the conversion efficiency, the description was based on the most adequate system of nonlinear wave equations, which take into account diffraction, effect of wave shift, group delay, and dispersion spreading (frequency dispersion) of laser pulses.

In the 1970s, a number of studies devoted to the influence of different related nonlinear optical phenomena on the laser frequency conversion efficiency were published in 'Quantum Electronics'. Primarily, the influence of concurrent Brillouin scattering [12], degenerate parametric process [13], optical detection [14], cubic nonlinearity [15], two-photon absorption [16], etc. was considered by the example of SHG.

The method of undepleted-field approximation (where the initial-radiation amplitude and phase are assumed to remain the same throughout the entire length of the medium) played a significant role in establishing the physics of nonlinear wave interaction. It allowed one to obtain simple and descriptive analytical solutions for a large number of important particular cases. All phase-matching widths were determined specifically in this approximation. The next step in developing the analytical methods was the method of undepleted-intensity approximation, where only the real amplitude of the laser radiation field is assumed to be invariable [17]. Some other methods for solving systems of coupled nonlinear wave equations were also developed, which made it possible to take into account more thoroughly the properties of nonlinear crystals. Along with the widely used finite-difference and finite-element methods, which take into account the spatial dispersion in the first approximation and the frequency dispersion in the second approximation, the use of spectral methods made it possible to take into account more completely the spatial [18] and frequency [19] (all dispersion orders of refractive indices and absorption coefficient) properties of nonlinear crystals.

The requirements to the spatial, spectral, and energy parameters of laser radiation for effective nonlinear frequency conversion have been determined. The complex work on the development of laser devices generating radiation with required characteristics was a success. By the mid-1980s, the following limiting conversion efficiencies had been achieved: more than 90% for SHG, more than 80% for THG, and 92% for FHG [20, 21]. Engineering design techniques for frequency converters that are widely used in practice were proposed even in the beginning of the 1970s [22, 23]; these techniques give satisfactory agreement with the experimental results. Laser physics made it possible to form extremely short pulses. One can speak about three types of nonstationarity at nonlinear optical frequency conversion: (1) nonstationary establishment of nonlinear polarisation, (2) dispersion spread of pulses, and (3) group velocity mismatch for the interacting pulses.

The first-type nonstationarity manifests itself only at pulses shorter than 0.1 fs, whereas the second-type nonstationarity contributes at femtosecond pulses. The third type of nonstationarity is observed even at subnanosecond pulses. The action of these mechanisms determines the corresponding model for describing nonlinear processes. Note that even in the beginning of the 1960s S.A. Akhmanov and R.V. Khokhlov paid attention to the spatiotemporal analogy between the oscillations in nonlinear lumped-parameter systems and the wave processes in nonlinear dispersive media [2]. The generalisation of this analogy to nonlinear wave problems showed (see, for example, [24]) the existence of the following pairs of similar processes: diffraction–dispersion spread and shift–group velocity mismatch. The study of time-dependent nonlinear optical processes confirmed the validity of this analogy.

Currently, light pulses with a duration of several femtoseconds have been obtained. At these times all types of nonstationarity manifest themselves in full measure; therefore, to perform effective nonlinear optical conversion, one has to find a direction in the crystal in which both phase and group matchings are provided, as well as the minimum dispersion spread for all interacting pulses. Unfortunately, this can be done with only some crystals. Even at relatively low energies this pulsed radiation has a record intensity: at a level of several tera- or gigawatts per square centimeter. The optimal nonlinear-crystal length is several hundred micrometers. At these radiation intensities the action of many related competing and limiting mechanisms manifests itself most completely. The nonlinear optical frequency conversion at high intensities has some specific features, for example, self-compression pulses [25] and distortion of the generated radiation spectrum

[26]. Optimisation of the frequency converter made it possible to reach high conversion efficiencies of 110-fs Cr-forsterite laser radiation to the second and third harmonics: 69% and 26%, respectively [26].

3. Nonlinear optical frequency conversion under thermal self-interaction

Although the average powers of the first lasers were rather low, the problems of thermal self-interaction of radiation in nonlinear optical crystals have attracted the attention of the researchers since the beginning of the 1970s. This interest was caused by rather large absorption coefficients of these crystals and the problems of frequency conversion for the lasers operating in cw and quasi-cw regimes. A theory of frequency conversion under thermal self-interaction conditions was developed; it showed that in the case of inhomogeneous temperature distribution over the crystal cross section as a result of volume heat release and boundary cooling, the phase-matching directions change inhomogeneously over the laser beam cross section. Depending on the crystal orientation, the phase-matching conditions are implemented not for the entire beam but for its separate parts: from the centre to the annular peripheral region [27]. Along with the total decrease in the nonlinear conversion efficiency, this process is accompanied by an increase in the phase-matching widths. The main mechanism of violation of this process is the temperature change in the refractive index, Δn; photorefraction, nonlinear absorption, and other effects also contribute.

When cylindrical focusing is implemented and the ratio of the transverse crystal and beam sizes is rather large, the temperature difference and, accordingly, the effect of thermal self-interaction decrease [28]. At high average radiation powers the temperature-field inhomogeneity leads to the formation of thermoelastic stresses. For the crystals belonging to the symmetry point groups 32, 3m, and $\bar{3}$m, the corresponding results of the analysis of thermoelastic stresses were reported in [29]. For $LiNbO_3$ crystals (3m), which are widely used in various applications, the Δn_σ value is about $10^{-5}\Delta T$, where ΔT is the temperature difference. Thus, the contribution of thermoelastic stresses must be taken into account at large ΔT values (i.e., high average radiation powers).

One of the parameters of a nonlinear optical crystal, which determines the maximum average power of converted radiation and, therefore, the allowable temperature difference over the crystal cross section, is the temperature phase-matching width. It was traditionally believed for uniaxial crystals that phase matching is always critical with respect to this parameter. At the same time, according to the data of [30, 31], biaxial crystals allow for temperature-noncritical phase matching. Hence, one can obtain phase matching with an unprecedently large temperature width. For example, a temperature phase-matching width more than 200 °C was obtained for SHG at 1.064 μm in a KTP crystal [30, 31]. In the case of THG, for 1.064-μm radiation, there is a direction in LBO crystals, along which the temperature width exceeds 70 °C [32]. For the processes with interaction efficiency determined by the difference in the wave vectors, it was also shown that a temperature-noncritical regime can be implemented in biaxial crystals, for example, temperature-noncritical birefringence [33–35]. This regime can also be implemented upon acousto-optic interaction, induced Brillouin scattering, Raman scattering, temperature scattering, etc.

Under thermal self-interaction, the temperature change in the refractive index is not the only significant factor. The thermal strains in a crystal, which are due to the anisotropy of its thermal expansion, can also play an important role. These strains change the shape of the crystal and cause rotation of its optical axes, as a result of which the exact phase-matching conditions are violated. A proper choice of the way of fixing a nonlinear crystal makes it possible to compensate for the changes that are due to thermal strains by the temperature change in the refractive indices, which allows one to increase the temperature phase-matching width. For example, different ways of fixing an LBO crystal at SHG made it possible to change the temperature phase-matching width by a factor of more than 1.5.

In the first experiments the optical loss in crystals was as large as 0.1–0.05 cm^{-1}. Currently, this value has been reduced to 0.001–0.0003 cm^{-1} for many modern crystals. However, since the average laser powers have increased significantly in the last years, the problem of nonlinear laser frequency conversion in the presence of thermal self-interaction remains urgent.

4. Nonlinear optical crystals

The problems of the synthesis of nonlinear media and study of their properties, as well as the analysis of the competing processes at nonlinear optical frequency conversion, remain one of the most important directions of research. Initially nonlinear optics was developed based on uniaxial crystals, which were applied in linear optics and acoustooptics in the pre-laser period. The first nonlinear optical crystals were characterised by nonuniform distribution of refractive index. A theory was developed for optically inhomogeneous crystals, and the influence of optical inhomogeneities on nonlinear frequency conversion was analysed [36–38].

As a result of intensive research in the synthesis of nonlinear media, more than 100 types of nonlinear optical crystals have been obtained; among them, biaxial crystals are most demanded. A number of nonlinear optical crystals make it possible to implement conversion in the near-IR and visible ranges (KTP, KTA, CTA, RTA, RTP, etc.), UV and VUV ranges (LBO, LB4, KBBF, BBO, BiBO, SBBO, CBO, CLBO, KABO, KBBF), and in the mid-IR range (LIS, LISe, ZGP, LGS, LGSe, AGS, AGGS).

In the first crystals the effective nonlinearity coefficient was about 0.5 pm V^{-1}. In modern crystals it reaches several pm V^{-1}, due to which high-efficiency conversion at low-intensity light can be implemented. The beam damage threshold of modern crystals for single-shot radiation ranges from several units to several tens of GW cm^{-2}. The development of high-energy lasers calls for large crystals of high optical quality. The first review on nonlinear optical properties of crystals was published in 'Quantum Electronics' in 1977 [39]. The further advances in this field led to the publication of the world-wide known monograph [40]. We should also note the high-quality handbook of crystals [41].

The synthesis of biaxial crystals of high optical quality and with small loss opened new possibilities for solving the problems of nonlinear optical frequency conversion. In uniaxial crystals the phase-matching directions form an axisymmetric cone, the axis of which coincides with the $c(z)$ axis, whereas in biaxial crystals these directions form a fourth-order conical surface. All possible phase-matching directions for SHG in uniaxial crystals were classified for the first time in [42]. For the more general case of generation of sum and difference frequencies, the diagram of phase-matching directions was reported in [43]. However, the analysis in [42, 43] was performed on the assumption of low birefringence dispersion, which was obviously caused by insufficient amount of data on the optical properties of biaxial crystals. In [44] a diagram of phase-matching directions for SHG in biaxial crystals was presented for the most general case, with allowance for the dispersion properties.

Active nonlinear media, which act simultaneously as laser active media (i.e., have activator properties) and as a nonlinear optical converter (matrix properties) [45] are promising for designing compact multifrequency radiation sources. However, these sources are still rather exotic and, correspondingly, insufficiently studied.

The recent years showed a rapid development of a new field of nonlinear optics: nonlinear frequency conversion in photonic crystals [46–49] (one-, two-, and three-dimensional periodic structures having a 'band gap' for all propagation directions of electromagnetic field). One of the advantages of photonic crystals is the significant increase in the radiation intensity near the edge of the photonic band gap, which makes it possible to reduce the crystal length that is necessary to attain effective nonlinear interaction. In this case, phase-matched wave interactions may occur for nonlinear processes of different types. For example, it was shown in [46] that crystals with alternating quarter-wave layers implement the conditions under which the optical frequency conversion efficiency for the interaction of counterpropagating waves exceeds that for copropagating waves. Devices with complex properties can be designed based on photonic crystals. The frequency conversion in an optical fiber with a photonic crystal cladding was investigated in [47]. The significant expansion of the spectrum in the case of phase self-modulation and synchronous interaction makes it possible to generate pulses with a duration of several femtoseconds. With the possibility of controlling phase-matching properties in such structures, one can try to generate efficiently THz radiation [48, 49].

5. Optical frequency conversion in PPNCs and quasi-phase-matched wave interactions

One of the methods for implementing high-efficiency nonlinear frequency conversion is based on the use of sandwiched crystalline plates, with oppositely oriented polar axes in neighbouring plates (proposed by H. Bloembergen, [3]) or on the application of PPNCs (they are also referred to as nonlinear photonic crystals). The change in the sign of the effective nonlinearity coefficient is equivalent to the change in the phase relations between the interacting waves by π. This change allows one to compensate for the phase mismatch of the same value, which is acquired on the plate (domain) length or on the crystal (structure) containing many domains, and to implement efficient nonlinear frequency conversion, because the phase mismatch in the crystal is compensated for by the reciprocal nonlinear 'lattice' vector. The surge of interest in this method in the 1980s was stimulated by the development of the technology of electric polarisation reversal of crystals with periods from several tens to several hundreds of micrometers. This technique has been developed most thoroughly for $LiNbO_3$ and KTP crystals. One of its drawbacks is that the thickness of obtained PPNCs does not exceed 1.0–1.5 mm; thus, they can only be used to focus laser radiation (note that PPNC structures can be designed in the waveguide version to increase the interaction length). Crystals for frequency conversion in the case of wide-aperture laser beams can be grown by the Czochralski method [50]. It was established experimentally [50] that there is a refractive index inhomogeneity at the domain boundary, which is caused by the bound charge and a jump of spontaneous polarisation.

The specific features of doubling optical frequency by focused beams in a PPNC were studied for the first time in [51]. During preparation of domain structure its periodicity can be violated; the character of these violations is related to the PPNC technology. The condition of the so-called stochastic (random) quasi-phase-matching was found for a disordered random domain structure [52].

Applying PPNCs, one can implement quasi-phase-matched wave interactions in the entire transparency range of nonlinear crystals (this is impossible for most of uniform crystals, where the phase-matching range is much narrower than the transparency range) and implement a nonlinear process using the components of the nonlinear susceptibility tensor that have maximum values. In $LiNbO_3$ and KTP crystals the maximum coefficient of nonlinear wave coupling is implemented for the interactions of the eee- and fff-types, respectively. For example, in a homogeneous $LiNbO_3$ crystal the 'working' effective nonlinearity coefficient is about 6 pm V^{-1}, whereas in the PPNC structure it is larger by a factor of 6. In KTP crystals the ratio of these coefficients is 4. A large effective nonlinearity coefficient makes it possible to implement frequency self-doubling in a periodically polsed active nonlinear neodymium-doped $LiNbO_3$ crystal [53].

Due to the large ratio of nonlinear coefficients, PPNCs can be used not only as optical-frequency converters but also as electro-optical *Q* switches [54]. In addition, PPNCs are promising for frequency conversion of femtosecond laser pulses, because they provide quasi-phase-matching conditions in the directions that are noncritical with respect to interaction wavelengths.

Periodically poled crystals have another remarkable property: they allow for effective nonlinear processes in different quasi-phase-matching orders [55]. On the one hand, this circumstance allows one to implement several processes successively or simultaneously (for example, SHG and THG). On the other hand, the additional process can be competing and undesirable for the main one. This question was analysed in [56–59], where multiharmonic generation in crystals with different orders of quasi-phase-matching and simultaneous frequency conversion at phase-matched and quasi-phase-matched interactions were considered. Generation of the second and third harmonics at quasi-phase-matching of the 9th and 33rd orders, respectively, was observed in [56]. Periodically poled crystals allow for simultaneous generation of harmonics in both copropagating and counterpropagating directions, successive parametric frequency down-conversion, and sum-frequency generation in the field of one pump wave (in this case degenerate parametric amplification under low-frequency pumping is implemented) [55, 59]. Note that nondegenerate parametric amplification under low-frequency pumping may occur in nonlinear optical crystals with an intentionally formed aperiodic domain structure.

6. Lasers with intracavity harmonic generation

Since the laser radiation intensity in the cw and quasi-cw regimes (which are widely used in laser technology) is low, it is most reasonable to perform intracavity frequency conversion, i.e., to use a frequency converter as an output mirror [60, 61]. The reason is that the frequency converter is a part of cavity, and its transfer (angular and spectral) characteristics, as well as the nonlinear character of conversion, affect the formation of the laser mode spectrum. The output power of converted radiation in lamp-pumped lasers is only few watt, whereas for semiconductor-pumped lasers, in view of much smaller thermo-optical distortions in the active element, the output power of the second and third harmonics exceeds 100 W. Recently, a variety of conversion schemes have been developed: ‘linear’, ‘angular’, ‘Z-scheme’, ‘annular’, etc. [1]; each of them has specific advantages. The high sensitivity of nonlinear process to the phase difference of interacting waves makes it possible to control the radiation parameters [62].

Intracavity frequency conversion is used not only in cw and quasi-cw lasers, but also in mode-locked lasers with pulse repetition rates from several tens to several hundreds of MHz [63, 64]. Schemes of intracavity generation of harmonics were used for sum-frequency generation and parametric generation [65]. Dual-wavelength lasing with frequency conversion was implemented in [66]. Note also paper [67], where the theory of intracavity frequency conversion in a laser based on periodically poled active nonlinear medium was developed. Many theoretical studies were aimed at analysing the formation of the energy and time parameters of radiation, the generation stability, and the influence of phase effects on the intracavity generation [68]. Nevertheless, paradoxical as it may seem, the complex analysis of the formation of the energy, temporal, and spatial parameters of radiation in such schemes has not been performed.

7. Optical frequency conversion in waveguides

The development of fibre lasers as radiation sources with an unprecedently high average power has put a problem of designing waveguide frequency converters that could be simply matched with a laser. The frequency conversion of fibre laser radiation in homogeneous crystals cannot yield a desired result, because the effective interaction length is limited by the large radiation divergence (because of the finiteness of angular phase-matching width), and the low radiation intensity does not allow one to perform nonlinear conversion with high efficiency. The propagation of radiation in a waveguide forms prerequisites for generating a spatial spectrum of radiation without a significant quadratic component of phase distribution, for which the phase-matching (or, more specifically, quasi-phase-matching, which is determined by the contributions of the material and waveguide dispersions) conditions are retained on a large interaction length. SHG in doped fibres, which was found experimentally in the end of the 1980s, stimulated further studies. The analysis of the mechanisms leading to the formation of quadratic nonlinearity in an initially centrosymmetric medium was performed, in particular, in [69–72]. Quadratic nonlinearity is induced in core bulk using heat treatment in the presence of a dc electric field. In this case, a space charge is induced to form the necessary nonsymmetricity of the medium, and periodically poled regions arise in the fiber. Optical frequency conversion occurs upon quasi-phase-matched interaction.

Currently, the maximum conversion efficiency to the second harmonic in germanosilicate fibers is 20%. Along with optical fibres, these processes are implemented in channel, planar, and hollow waveguides, as well as in the surface layers of hollow or gas-filled waveguides. Generation of sum frequency in coupled waveguides (coupled second-harmonic generators with a periodic domain structure), where radiation from two laser diodes is introduced into different fibres, was described in [73]. Nonlinear fibre optics was considered in review [74]; the main attention was paid to four-wave interactions and to SRS and SBS.

8. Laser frequency conversion in gaseous media; generation of high-order harmonics

The upper limit of the UV transparency range of nonlinear crystals is 150–170 nm. Gaseous media make it possible to shift this limit to shorter wavelengths. In the 1980s and at the beginning of 1990, a number of studies on resonant nonlinear optical processes in vapour of different metals (Na, Cs, Ta, Hg, etc.) were published in ‘Quantum Electronics’. The processes of parametric generation, frequency summation, and phase conjugation were considered [75, 76]. Generation of harmonics, scattering, and other processes were analysed in other studies. A number of papers published at that time were devoted to THG in gaseous media (in particular, laser plasma). The purpose of these studies was, on the one hand, to increase the generation efficiency, and, on the other hand, to analyse THG in order to probe fast processes in different media (see, for example, [77]).

Since the mid-1990s many theoretical works devoted to the generation of high-order harmonics (HOHs) and attosecond UV and soft X-ray pulses in gas jets have been published in ‘Quantum Electronics’. The spatial and temporal structure of the total HOH field was investigated, methods for selecting single attosecond pulses from this field and techniques for controlling the angular structure of individual harmonics were developed, and some other important questions were considered (some of these works were reviewed in [78]). HOH generation was investigated in bichromatic fields [79] and in relativistically strong fields [80]. The studies on the HOH generation in jets are closely related to the works on generation of harmonics (in particular, HOHs) in gas-filled hollow waveguides. For example, the possibility of satisfying phase-matching conditions during HOH generation in waveguides was considered in [81].

9. Parametric optical frequency conversion

More than 20% of studies on nonlinear optical frequency conversion published in ‘Quantum Electronics’ were devoted to the problems of optical parametric oscillation (OPO). Unfortunately, the volume of this review does not make it possible to analyse in detail the results of studying this nonlinear process. Therefore, we will primarily note the research teams that were leaders in the number of publications on the optical parametric process in ‘Quantum Eectronics’. These are teams headed by A.S. Piskarskas (Vilnius University), G.I. Freidman (Institute of Applied Physics, Russian Academy of Sciences), and A.I. Kholodnykh (Moscow State University). We also recommend review [82] and monographs [1, 83, 84] to a reader.

Using the parametric frequency down-conversion, one can obtain tunable generation in an unprecedently wide frequency range: from UV to mid-IR [82, 83]. A strong pump wave is decomposed into signal and idler waves, the wavelengths of which are determined by the energy and momentum conservation conditions in a nonlinear crystal. Changing the phase-matching condition (by changing the crystal orientation or temperature) or the pump wavelength, one can tune the generated radiation frequency in a wide range.

The first optical parametric loscillator, which confirmed the fundamental possibility of an OPO, did not have a cavity. It operated in the travelling wave regime; however, its energy and spectral characteristics were unsatisfactory. As a result of the development of cavity OPO schemes, devices with high conversion efficiency were designed and transform-limited radiation pulses were obtained. The conversion in the case of a noncavity parametric light generator is equivalent to some extent to difference-frequency generation, whereas in cavity schemes it occurs under the conditions providing formation and interaction of cavity modes and can be implemented for any combination of interacting waves or for all waves simultaneously.

In the case of homogeneous and periodically poled crystals, parametric light generation can be implemented practically in all lasing regimes, both beyond and inside the laser cavity and upon both scalar and vector wave interactions.

Various aspects of the OPO theory have been considered in many studies, where the gain and phase – frequency characteristics were determined; generation thresholds were established for different schemes; the requirements to the time, energy, and spectral parameters of pump radiation were formulated; and a comparative analysis of different generation schemes and their functional possibilities was performed. Pulse narrowing in both linear and nonlinear regimes of parametric wave interaction in the presence of group velocity mismatch in both matched and mismatched cavities was considered in [85].

Unfortunately, complex analysis of the formation of energy, temporal, spectral, and spatial parameters of radiation at the OPO has not been performed. We should only note the study [83] with qualitative results on light beam evolution, which were obtained based on a spatial and temporal analogy. A transformation of the spatial spectrum of the radiation formed was observed in [86].

10. Statistical phenomena in nonlinear optical processes; generation of nonclassical light

The systematic study of the statistical effects in nonlinear optics began with the analysis of fluctuations upon SHG [87, 88]. The power of the second harmonic excited by a laser operating in the spike regime (the first pulse-pumped lasers operated specifically in this regime) exhibited the so-called excess fluctuations. They manifested themselves as follows: spikes of the same laser power corresponded to spikes of different second-harmonic power [2]. This effect was explained by simultaneous generation of several longitudinal modes with a fluctuation spread of mode phases. In this case, the SHG for multimode radiation was accompanied by a twofold increase in the conversion efficiency (statistical gain) in comparison with the generation of single-mode radiation of the same power. It was shown for the first time in [89] that, for laser radiation with random mode phases, the statistical gain η increases with an increase in the harmonic number n or the order n of the multiphoton process; at a large number of modes N, the gain $\eta = n![1 - n(n-1)/N + ...]$. Furthermore the results of [89] were used to interpret the experimental data on multiphoton ionisation of atoms in a high-power laser field.

Later studies were focused on the specific features of generation of the sum and difference frequencies, parametric amplification, and stimulated Raman scattering in a random multimode pump field. In particular, the parametric frequency conversion in an incoherent pump field was investigated in [90 – 94]. We restrict ourselves to these publications and recommend a reader the monograph [24] to gain a deeper insight into this problem. In [90, 94], parametric image conversion from IR to visible range was considered and the statistical characteristics of images were investigated (see review [95]). We should also note the study [96], where parametric light conversion was used to measure the absolute brightness of thermal source radiation.

In the 1980s – 1990s a number of studies on the propagation of incoherent intense laser beams, accompanied by self-interaction, were published in 'Quantum Electronics'. A specific feature of nonlinear propagation of this radiation is that a strong light field changes the refractive index; fluctuations of this change correlate with the initial-field fluctuations. This causes, in turn, additional modulation of the propagating initial radiation, which can be accompanied by a significant change in the angular and frequency spectra. Numerical and approximate analytical methods were used to analyse the self-interaction of incoherent light beams. The results obtained in this field were reported in the reviews [97, 98]. In addition, we should note that the propagation of incoherent light beams was also considered for random-inhomogeneous nonlinear media in [98]. Thus, the main properties of nonlinear propagation of random light beams can be considered as established by this time.

Processes of parametric interaction and self-interaction are known to produce nonclassical light; they can be adequately described using only quantum-mechanical methods. Among the studies on nonclassical-light generation that were published in 'Quantum Electronics', the works [99 – 101] are of particular importance: they stimulated research in the corresponding fields. From the quantum point of view, optical parametric amplification in a high-frequency pump field is actually a process producing correlated photons. This process generates light in entangled quantum state and squeezed light, i.e., light with suppressed quantum fluctuations in one of the quadrature components. S.A. Akhmanov et al. [99] applied for the first time a parabolic equation to quantum description of squeezed-light generation upon parametric interaction. Based on this approach, it was shown in [102] that quantum fluctuations in space can be suppressed. This finding initiated a new research direction in optics, which was referred to as quantum imaging [103].

It was established for the first time in [101] that a polarisation state of intense light for which the level of quantum fluctuations in one of the Stokes parameters is lower than in the coherent state can be formed in an optical fiber upon self-interaction. This light was referred to as polarisation-squeezed. To date, some other ways for obtaining polarisation-squeezed light have been proposed and implemented. The general quantum formalism for describing light polarisation states was developed by V.P. Karasev (see, for example, [104]).

11. Conclusions

This review shows clearly the diversity of studies in the field of nonlinear optics. The development of new pump sources and media with new nonlinear optical properties put new problems before the researchers. Solution of these problems expands the application range of nonlinear optical laser frequency converters. It is impossible to dwell on all problems considered in different papers devoted to nonlinear optical frequency conversion and present a complete list of studies in this field. The thematic set of papers on the problems of nonlinear optical frequency conversion, which were published in ‘Quantum Electronics’, can be found at the site www.qe.ru/NLO-Ref.pdf of the editorial board of the journal.

One of the authors, Valentin Georgievich Dmitriev, died during the work on this paper. He was one of the pioneers of nonlinear optics and the author of fundamental studies on the theory of nonlinear optical frequency conversion, including monograph [1], which became a handbook for students, postgraduates, and experts in this field. Dmitriev was among the founders of nonlinear optics; his candidate’s dissertation was the first in this field. The life of Valentin Georgievich Dmitriev was entirely devoted to science.

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